Environmental Toxicology and Chemistry, Vol. 29, No. 3, pp. 633–643, 2010

Printed in the USA
DOI: 10.1002/etc.96

MERCURY CONCENTRATIONS IN LANDLOCKED ARCTIC CHAR (*SALVELINUS ALPINUS*) FROM THE CANADIAN ARCTIC. PART II: INFLUENCE OF LAKE BIOTIC AND ABIOTIC CHARACTERISTICS ON GEOGRAPHIC TRENDS IN 27 POPULATIONS

NIKOLAUS GANTNER,*†‡ DEREK C. MUIR,†‡ MICHAEL POWER,§ DEBORAH IQALUK,|| JAMES D. REIST,# JOHN A. BABALUK,# MARKUS MEILI,†† HANS BORG,†† JOHAN HAMMAR,‡‡ WENDY MICHAUD,§ BRIAN DEMPSON,§§ and KEITH R. SOLOMON†

†Department of Environmental Biology, University of Guelph, Guelph, Ontario N1G 2W1, Canada
‡Water Science and Technology Directorate, Environment Canada, Burlington, Ontario L7R 4A6
§Department of Biology, University of Waterloo, Waterloo, Ontario N2L 3G1 Canada
||Hamlet of Resolute Bay, Resolute Bay, Nunavut X0A 0V0 Canada
#Freshwater Institute, Fisheries and Oceans Canada, Winnipeg, Manitoba, R3T 2N6
††Applied Environmental Science (ITM), SE-106 91, Stockholm University, Sweden
‡‡Institute of Freshwater Research, SE-178 93, Drottningholm, Sweden
§§Science Branch, Fisheries and Oceans Canada, St. John's, Newfoundland, A1C 5X1

(*Submitted* 29 *April* 2009; *Returned for Revision* 23 *June* 2009; *Accepted* 9 *October* 2009)

Abstract—Among-lake variation in mercury (Hg) concentrations in landlocked Arctic char was examined in 27 char populations from remote lakes across the Canadian Arctic. A total of 520 landlocked Arctic char were collected from 27 lakes, as well as sediments and surface water from a subset of lakes in 1999, 2002, and 2005 to 2007. Size, length, age, and trophic position ($\delta^{15}N$) of individual char were determined and relationships with total Hg (THg) concentrations investigated, to identify a common covariate for adjustment using analysis of covariance (ANCOVA). A subset of 216 char from 24 populations was used for spatial comparison, after length-adjustment. The influence of trophic position and food web length and abiotic characteristics such as location, geomorphology, lake area, catchment area, catchment-to-lake area ratio of the lakes on adjusted THg concentrations in char muscle tissue were then evaluated. Arctic char from Amituk Lake (Cornwallis Island) had the highest Hg concentrations (1.31 μg/g wet wt), while Tessisoak Lake (Labrador, 0.07 μg/g wet wt) had the lowest. Concentrations of THg were positively correlated with size, $\delta^{15}N$, and age, respectively, in 88, 71, and 58% of 24 char populations. Length and $\delta^{15}N$ were correlated in 67% of 24 char populations. Food chain length did not explain the differences in length-adjusted THg concentrations in char. No relationships between adjusted THg concentrations in char and latitude or longitude were found, however, THg concentrations in char showed a positive correlation with catchment-to-lake area ratio. Furthermore, we conclude that inputs from the surrounding environment may influence THg concentrations, and will ultimately affect THg concentrations in char as a result of predicted climate-driven changes that may occur in Arctic lake watersheds. Environ. Toxicol. Chem. 2010;29:633–643. © 2009 SETAC

Keywords—Spatial trend Arctic Lake catchment Food chain length Arctic char

INTRODUCTION

Despite recent reduced emissions and use of mercury (Hg) and Hg-containing products in some parts of the industrial world, mercury remains of critical concern to human populations relying on subsistence fisheries as a key source of dietary protein [1–3]. A recent global budget [4] indicates that the largest emitters of Hg are the rapidly growing economies in Asia (namely China and India), while North American and European emissions have declined since the 1990s. Overall, total global anthropogenic atmospheric emissions have remained constant since 1980, totaling 2,190 tons in the year 2000 [4]. Sources of Hg in the atmosphere range from emission by coal fired power plants, garbage incineration (anthropogenic), to volcanic eruption (natural) [4].

* To whom correspondence may be addressed (nikolaus.gantner@ec.gc.ca).
Published online 10 December 2009 in Wiley InterScience (www.interscience.wiley.com).

Long range atmospheric transport of Hg from these sources is believed to deliver atmospheric Hg to the Arctic, where it is deposited into the environment [5]. Transfer of Hg species to the Arctic and into Arctic lakes, as well as mechanisms of bioaccumulation and food web transfer of monomethyl Hg (MeHg) are outlined in the accompanying study [6]. The recent review by Munthe et al. [2] summarizes contributing pathways and recent trends in Hg concentrations in fish. Mercury in fish remains a dominant issue across the Arctic, where concentrations in predatory fish from Arctic and sub-Arctic regions of Canada are frequently found to exceed Health Canada consumption guidelines [7–9]. In some small lakes, concentrations in fish have been increasing over time [7]; however, in one of the largest Arctic lakes (Lake Hazen, Ellesmere Island, Nunavut, Canada), concentrations remain unchanged over a 16-year period (1990–2006) [10]. It has been suggested that documented increases of Hg concentrations in lake sediments maybe linked to recent climate changes in the Arctic [11] and other authors have speculated that warming might lead to greater accumulation of Hg ([12,13]; www.amap.no). Increases of Hg in Arctic lake sediments have been attributed to anthropogenic sources [14]. A combination of both climatic effects and the global distribution of Hg, due to its long residence time in the atmosphere (~1 year) [5], could be responsible for the lack of decline of concentrations in the Arctic, despite recent North American reductions. Studies of Alert on Ellesmere Island, Nunavut, have shown that there is no decline of gaseous elemental Hg (GEM) in the atmosphere [15,16], while evidence from sediment cores shows reductions in Hg deposition in lower latitude lakes in Canada [17].

A comparison of Hg species (organic + inorganic Hg fraction) in a single fish species over a wide geographical range (latitudinal and longitudinal) should improve our knowledge of biotic and abiotic factors contributing to Hg variability. However, spatial comparisons of Hg concentrations in fish are often constrained by the high variability among species, among populations within the same species, or among individuals within the same population [1,7,9]. Fish are exposed to Hg mainly through dietary uptake [18]. Thus, food web characteristics are important for any explanation of lake-to-lake variability, because biomagnification up the food chain appears to be lake specific and longer food chains may result in higher Hg concentrations [6]. Other factors such as trophic position, fish size, and age [1,10,19–23], contribute to the variability for Hg and other (organic) bioaccumulative contaminants [24,25]. Once we evaluate the role of biological factors on fish tissue Hg concentrations, we can investigate differences among sites and regions due to climate or other abiotic factors. Abiotic characteristics such as watershed-lake area ratio have been shown to affect Hg cycling in Alaskan Arctic lakes [26], and mid-latitude lakes [2,27] and were also used to contrast spatial differences in Hg.

Landlocked Arctic char (*Salvelinus alpinus* L.) is widely abundant in the Canadian Arctic [28], and is a keystone species throughout the Arctic in the lakes where it is found [29]. The distribution of Arctic char extends farther north than that of any other freshwater fish, and it is often the only species present in high latitude Arctic lakes [29–32]. Its circumpolar distribution within the Arctic allows widespread monitoring and large scale spatial comparisons of contaminant concentrations. Previous studies have shown that THg concentrations in Arctic char muscle tissue frequently exceed Health Canada guidelines (subsistence consumption threshold of 0.2 μg/g wet wt), with landlocked populations typically having greater THg concentrations than migratory populations [1,7,33,34]. There have been no previous large-scale spatial comparisons of Hg tissue concentrations in landlocked Arctic char, apart from reports on Hg data from anadromous char populations [1]. The earlier study of Muir et al. [7] investigated spatial variability only at local scales, using six lakes located near Resolute Bay, Nunavut, and did not include consideration of possible food web-related effects on Hg concentrations. Here, we evaluate large-scale spatial differences of Hg concentrations along longitudinal and latitudinal gradients in the Canadian Arctic after adjusting data for the effects of trophic position, fish length, and/or age.

MATERIALS AND METHODS

Site selection

We selected landlocked Arctic char populations from lakes in geographically distinct Arctic or subarctic regions, attempting to cover the largest possible latitudinal range (Fig. 1, Table 1). With the exception of Ruggles River (Lake Hazen, Ellesmere Island), all outflows of the study lakes do not allow for migration, and Lake Hazen char are confirmed to be lake resident [35]. Selection of sites was constrained by accessibility and guided by past sampling campaigns, and/or current monitoring programs. The four northernmost lakes were located in Quttinirpaaq National Park (~82°N) on Ellesmere Island (Nunavut, Canada). To our knowledge, three char populations

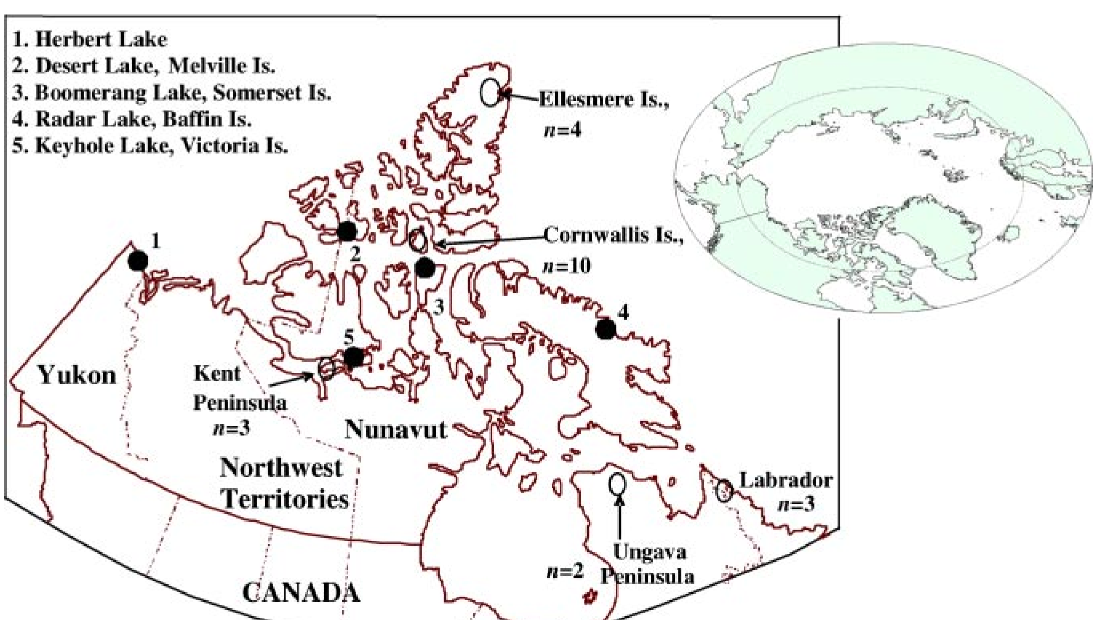

Fig. 1. Approximate sampling locations across the Canadian Arctic. Circles indicate main sampling areas (multiple lakes per area were sampled); dots represent single lakes of the dataset (refer to Table 1 for names and location). [Color figure can be seen in the online version of this article, available at www.interscience.wiley.com.]

Table 1. Lake, regions, locations, sampling years, and Arctic char (*n*, biodata, trophic signatures, and unadjusted total mercury [THg])[a]

	Location				Length (mm)		Weight (g)		Age (y)		$\delta^{15}N$ (‰)		$\delta^{13}C$ (‰)		[THg] (μg/g)	
Region/Lake	N	W	Years	*n*	M	SD	M	SD	M	SD	M	SD	M	SD	GM	SD
Ellesmere Is., Nunavut																
Lake D	82.11	67.48	2007	7	333	113	498	390	19	7	12.9	1.4	−27.5	2.7	0.08	0.09
Lake G	81.82	69.18	2007	34	287	73	260	460	19	3	9.0	1.0	−22.7	0.4	0.20	0.29
Lake F	81.81	69.35	2007	10	228	132	276	640	13	5	8.3	0.4	−23.2	1.4	0.11	0.19
Lake Hazen	81.83	70.42	2006–2007	54	331	117	413	367	17	7	10.3	1.8	−20.2	1.9	0.08	0.16
Cornwallis Is., Nunavut																
Amituk	75.05	93.75	2005–2007	24	419	87	657	440	20	3	11.3	1.2	−21.1	0.4	1.31	0.89
Aquiatusuk	74.70	94.23	2005–2007	10	441	43	741	259	31	18	9.8	0.9	−26.7	0.5	0.30	0.21
12 Mile	74.82	95.34	2005–2007	49	334	28	484	323	16	1	11.5	0.3	−21.4	0.4	0.11	0.04
9 Mile	74.81	95.20	2005–2007	34	321	26	225	54	16	3	10.3	0.4	−21.7	0.5	0.16	0.06
North	74.78	95.09	2005–2007	23	352	58	415	120	17	3	10.2	0.8	−23.8	0.3	0.26	0.13
Meretta	74.68	94.92	2005–2007	16	443	126	1261	713	7	2	11.8	0.4	−24.9	1.8	0.32	0.08
Char	74.70	94.88	2005 + 2007	13	401	118	867	770	19	6	11.6	1.2	−23.5	0.9	0.46	0.26
Resolute	74.68	94.88	2005–2007	40	374	66	475	218	19	5	12.2	1.2	−22.4	0.7	0.22	0.08
Small	74.76	95.06	2005–2007	28	390	55	492	283	16	3	10.1	1.1	−23.6	0.9	0.12	0.11
Teardrop	74.68	94.99	2007	4	109	6	11	2	—	—	10.8	0.1	−24.7	0.6	0.19	0.01
Somerset Is.																
Boomerang	73.94	92.89	2007	8	461	33	648	173	16	2	12.2	1.4	−20.7	0.7	0.28	0.11
Victoria Is.																
Keyhole	69.38	106.24	2006	7	427	100	1167	840	9	6	11.3	0.9	−27.7	0.7	0.09	0.10
Kent Peninsula, Nunavut																
Gavia Faeces	68.34	107.73	2006	8	275	98	211	201	12	5	8.1	1.3	−25.3	1.1	0.11	0.18
Little Nauyuk	68.35	107.75	2006	15	349	50	349	157	13	4	7.6	0.6	−22.2	0.3	0.11	0.06
Notgordie	68.35	107.66	2006	5	442	34	1134	337	9	1	10.4	2.7	−25.6	0.7	0.23	0.09
Labrador																
Tasialuk	56.74	62.69	2007	37	341	130	593	378	—	—	7.9	0.4	−25.0	1.4	0.11	0.07
Tessisoak	56.63	62.52	2007	8	113	28	17	15	—	—	10.9	0.7	−25.3	1.2	0.07	0.02
Coady's Pond	56.64	63.63	2007	33	308	111	430	356	—	—	6.9	0.4	−20.8	1.5	0.10	0.03
Ungava Peninsula, Nunavik																
Laflamme	61.32	73.71	2007	9	497	124	1563	875	14	5	9.5	0.8	−22.7	1.0	0.14	0.07
Pingualuk	61.28	73.66	2007	23[b]	400	129	532	495	19	5	12.1	1.1	−26.6	1.9	0.18	0.11
Yukon																
Herbert	69.42	139.63	1999	9	295	79	186	109	—	—	9.8	1.2	−29.9	1.1	0.46	0.70
Melville Is.																
Desert	75.03	107.87	1999	2	540	62	1538	651	—	—	14.9	0.3	−27.9	0.9	1.24	0.89
Baffin Is.																
Radar	68.42	66.83	1999	10	380	154	695	811	—	—	10.6	1.8	−28.4	1.4	0.73	0.62

[a] M = arithmetic mean; GM = Geomean; SD = standard deviation.
[b] Included are two Arctic char recovered from stomachs of two larger specimens.

in this area (lakes F, G, and D) have never been sampled before the present study. Information on limnological characteristics [36] and diatom assemblages [37] from nearby lakes and ponds has highlighted the characteristics of area lakes due to local climate effects that make the area an Arctic oasis. Twelve lakes were located near Resolute Bay (Nunavut, Canada) (~74°N), and were accessed either by all terrain vehicle or helicopter. Three populations in the Resolute Bay area (North, Small, and Resolute lakes) are known to be used by the local community (the only lakes in the study for which there was any recreational fishing). The food web of Char Lake also in the Resolute area has also been previously studied in detail [38], and information on several contaminants (Hg and dichlorodiphenyltrichloroethane, PCBs) in char is available for some lakes of the Resolute area [1,39]. Four lakes on the Kent Peninsula are located southwest of Cambridge Bay (~69°N) (NU, Canada), and their resident char populations have been extensively studied by Fisheries and Oceans Canada in the 1970s and 1980s [40,41]. Two lakes included in the present study form part of the Parc National de Pingualuit (~61°N) on the Ungava Peninsula (QC, Canada); Lake Pingualuk is a meteor-impact crater lake [42]. Three lakes from north Labrador (~56°N) represent the most eastern and southern lakes and were sampled in conjunction with a Canadian International Polar Year survey carried out by Department of Fisheries and Oceans (St. John's, Newfoundland, and Labrador) that are primarily focused on anadromous Arctic char. A report on Hg in Labrador fish species from 1977 to 1978 includes Arctic char data for Tasialuk Lake [43]. The most western lake (Herbert Lake, ~69°N, 139°W) is located in the Yukon Territory (Canada). A single lake (Radar Lake) is located on the north shore of eastern Baffin Island (~68°N, 66°W). With the exception of the most southern study sites, three north Labrador lakes and the Kent Peninsula lakes, all other lakes selected for the present study hosted exclusively landlocked Arctic char as the only fish species.

Field collections

Between July and August of 1999, landlocked Arctic char and zooplankton samples were collected from Radar, Herbert, and Desert lakes during the Tundra North West (TNW99) expedition. Samples collected during TNW99 were shipped to ITM (Stockholm University, Sweden) on ice, and stored there at −20°C until analyzed. Sampling was supported by local Inuit guides, who also provided general information on the fish population of the sites. For all other study lakes Arctic char and food web samples were obtained in July and August of 2005, 2006, and 2007, following procedures outlined in Gantner et al. [6], with the exception of Lake Pingualuk and Lac Laflamme (collection in May). Briefly, adult char were collected using gillnets (32–46 mm mesh size) or traditional ice-fishing methods and/or angling through ice cover. A maximum annual catch of 20 adult Arctic char per lake was used for conservation purposes, set by Fisheries and Oceans Canada, with exception of Labrador sites. Typically, collections at each site occurred on a single day. If fishing results were not sufficient ($n < 7$ adult char), a second sampling was added. All sampling conducted in the Resolute area was aided by the local Inuit. Sub-sampling and fish dissections were performed immediately after capture at the facilities of the Polar Continental Shelf Project in Resolute Bay, Nunavut. At all other sites, samples were dissected in the field, put on ice, or frozen whole. Samples collected between 2005 and 2007 were shipped frozen to the National Water Research Institute (NWRI, Burlington, ON, Canada), and stored there at −20°C until analyzed. Between 2 and 49 Arctic char were available from 27 lakes, sampled between 1999 and 2007, making a total of 520 samples for the present study (Table 1).

Surface water samples were obtained near the center of each lake using a precleaned 2 L Teflon® container and clean techniques. Duplicate samples were transferred into acid-washed and rinsed amber (MeHg) and clear (THg) glass bottles (250 ml), and 3 ml of HCl (0.5%) added to preserve the MeHg samples. Field blanks (2 per site) were brought to the field and were handled in accordance with the sampling procedure, except for filling of the bottles. All water samples and blanks were stored at 4°C in dark conditions until analyzed.

Laboratory analysis

All THg analyses were conducted using fish muscle, by use of the direct combustion method (Direct Mercury Analyser, Milestone Instruments) according to U.S. Environmental Protection Agency (U.S. EPA) method 7473 ([44]; http://www.epa.gov/osw/hazard/testmethods/sw846/pdfs/7473.pdf). A method detection limit (= 3 × standard deviation of the blank) of 2 to 5 ng/g dry weight and precision (=1 standard deviation) in homogenous samples (at 100 × detection limit) of ± 1–2% was determined for the processing of all TNW99 samples at (Department of Applied Environmental Science, ITM, Stockholm, Sweden). Equal precision, but lower detection limits (1–2 ng/g) were determined for samples analyzed at NWRI, Burlington laboratories. Arctic char muscle from the 2005 to 2007 collections was sub-sampled (~0.1–0.2 g wet wt) after a brief thawing period, and measured without prior drying, following U.S. EPA method 7473 [44]. Surficial sediment samples from 2005 to 2007 were analyzed for THg with identical methods, but freeze-dried before analysis. The TNW99 char samples were freeze-dried and moisture content recorded, with a 0.05 g dry weight were used for analysis. Resulting Hg concentrations (dry wt) in TNW99 char samples were converted to wet weight concentrations using the mean water content of individual fish (77 ± 6% wt). Consequently, all reported concentrations herein are based on wet weight. Standard reference materials (DORM1, DORM3, DOLT, all National Research Council of Canada) were run daily to check for method accuracy, instrument performance, and matrix effects. Data were only used if the reference materials were within ± 10% of their certified value, or samples were reanalyzed.

Stable isotope analysis of char muscle tissue and surficial sediments were completed following the methods described in the accompanying study [6]. Briefly, subsamples of muscle were freeze dried and homogenized to a fine powder using a ball-mill grinder before analysis. Sediment subsamples were acidified in Petri dishes using HCl (10%), then neutralized by adding MilliQ water (to pH ~6.5–7; Bedford, MA, USA), and dried at 60°. Trophic magnification factors (TMFs) and food chain lengths (FCL) of a subset of 18 lakes (derived from food webs presented in the companion study [6] were used here to infer their influence on THg concentrations in Arctic char muscle.

Total Hg in water samples was analyzed at NWRI, Burlington, using atomic fluorescence spectroscopy. Methylmercury in surficial sediments were determined using gas chromatography for separation and atomic fluorescence spectroscopy detection applying extraction methods outlined in Gantner et al. [6].

Statistical analysis

The confidence level (type I error rate) for statistical significance was set to $\alpha = 0.05$ for all analyses. All statistical analyses were conducted using software by Systat [45]. The relationships between the dependent variable THg and $\delta^{15}N$, fish length, or age were determined using linear regression analysis. Before analysis, THg concentrations were $\log_{10}$-transformed and normality of the data distribution was confirmed using Shapiro-Wilk test. The ANCOVA was performed to test for trophic, length, and age (fish) effects using a general linear model (GLM), following procedures described in Gantner et al. [10] and the resulting GLM model was used to adjust Hg concentrations for the effect of significant covariates. Adjusted Hg concentrations, back-calculated from least squares means, were then used for graphical presentation. Tukey's honestly significant difference test was used to detect significant differences within regions with more than two sites. Student's t test was applied, when only two lakes were compared.

RESULTS AND DISCUSSION

Unadjusted THg—27 populations

Means of unadjusted THg concentrations in Arctic char ranged from 0.07 μg/g (Tessisoak Lake) to 1.31 μg/g (Amituk Lake) (Table 1). Char from Amituk Lake had the highest individual THg concentrations as well, with one char reaching 3.4 μg/g of THg. Mean Hg concentrations in Arctic char from 14 lakes were below the Health Canada guideline for subsistence consumption (of 0.2 μg/g), while mean THg concentrations in char from 10 lakes exceeded that guideline (Table 1). The THg concentrations presented here are comparable to previously reported literature values for six lakes from the Resolute Bay area [7], and in Tasialuk Lake to concentrations reported in the late 1970s ($n = 24$; THg = 0.06–0.14 μg/g) [43]. Thus, the present study is the most comprehensive summary of current concentrations of THg available for landlocked Arctic

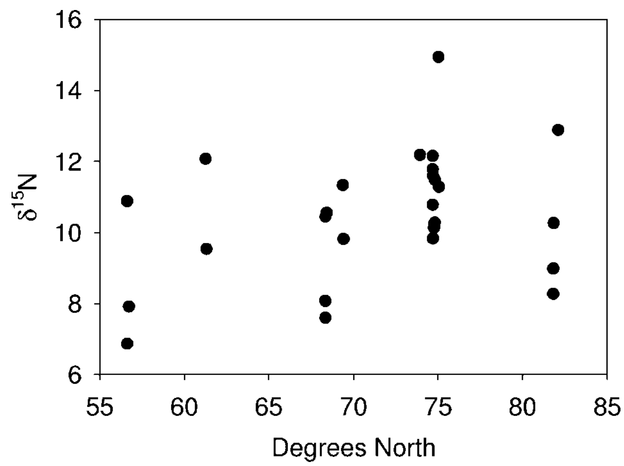

Fig. 2. Mean trophic signature ($\delta^{15}N_{unadjusted}$) in Arctic char versus latitude.

char across a large proportion of the geographic range of the Canadian Arctic.

Trophic signatures ($\delta^{15}N$)

Detailed analysis of $\delta^{15}N$ signatures in Arctic char, are presented in Gantner et al. [6] and Table 1, and will only be discussed briefly here. The average $\delta^{15}N$ signature of Arctic char ranged from 6.7‰ in Coady's Pond (Labrador; $n = 33$) to 14.9‰ in Desert Lake (Melville Islands, Nunavut; $n = 2$). For the 18 lakes of known baseline (periphyton signature), char $\delta^{15}N$ signatures were baseline corrected ($\delta^{15}N_{A.\ char\ adjusted} = \delta^{15}N_{A.\ char\ unadjusted} - \delta^{15}N_{periphyton}$). The $\delta^{15}N$ signatures in char (adjusted and unadjusted for baseline) were not related to latitude (linear regression $r^2 < 0.1$, Fig. 2). Food chain lengths were also not related to latitude or longitude ($p < 0.05$).

Relationships of THg in char with length, age, and $\delta^{15}N$

Linear regression revealed positive relationships between the unadjusted THg and covariates known to influence Hg concentration in fish: fork length, age, and $\delta^{15}N$ (Table 2). In most lakes (17 of 27), THg in Arctic char was positively correlated with $\delta^{15}N$ in char muscle (linear regression r^2 range from 0.33 in Resolute Lake to 0.96 in Gavia Faeces Lake). In 21 of 27 lakes (linear regression range of $r^2 = 0.11$ in 12 Mile Lake to 0.88 in Lake F), THg was predicted by fork length. Fish ages were not available for seven lakes; however, in the remaining 14 lakes, THg was related to fish age. Noteworthy is the oldest Arctic char collected during the sampling campaign at Aquiatusuk Lake (Cornwallis Island) in 2005; at 44+ years, it represents the oldest individual of this species aged by our age determination laboratory and as such may be the oldest recorded specimen for the species (J. Babaluk and R. Wastle, personal communication).

Size, age, and $\delta^{15}N$ were thus predictors of concentrations of THg for each lake, although no general rule could be established across all lakes of the present study region, likely due to the known variability of underlying food webs [6] and the ecological plasticity of the individual char populations. This poses an issue for spatial comparisons. While small-scale comparisons (of lakes within one region) may work well with either of

Table 2. Linear regression parameters of unadjusted total mercury (THg) and covariates for all Arctic char collected[a]

Lake		log[THg] vs $\delta^{15}N$		log[THg] vs length		log[THg] vs age		length vs $\delta^{15}N$	
Name	Abbreviation	$p < 0.05$	r^2	$p < 0.05$	r^2	$p < 0.05$	r^2	$p < 0.05$	r^2
Lake D	D	-	-	√	0.52	-	-	-	-
Lake G	G	√	0.67	√	0.69	√	0.17	√	0.82
Lake F	F	√	0.61	√	0.88	-	-	√	0.75
Hazen	Hz	√	0.51	√	0.71	√	0.31	√	0.39
Amituk	Am	√	0.82	√	0.49	√	0.26	√	0.7
Aquiatusuk	Aq	√	0.71	√	0.59	√	0.40	√	0.6
12 Mile	12 m	-	-	√	0.11	-	-	-	-
9 Mile	9 m	√	0.45	-	-	√	0.14	√	0.11
North	No	√	0.40	√	0.26	√	0.14	√	0.22
Meretta	M	-	-	√	0.33	√	0.25	√	0.79
Char	C	√	0.74	√	0.74	√	0.31	√	0.61
Resolute	Rl	√	0.33	√	0.13	√	0.24	-	-
Small	S	√	0.59	√	0.28	√	0.57	√	0.24
Teardrop	Td	-	-	-	-	-	-	-	-
Boomerang	B	√	0.83	√	0.62	-	-	√	0.45
Keyhole	K	√	0.54	√	0.46	-	-	-	-
Gavia Faeces	GF	√	0.96	√	0.78	√	0.89	√	0.81
Little Nauyuk	LN	√	0.26	-	-	√	0.53	-	-
Notgordie	Ng	-	-	-	-	-	-	-	-
Tasialuk	Ta	√	0.40	√	0.63	-	N/A	√	0.24
Tessisoak	Te	-	-	√	0.73	-	N/A	-	-
Coady's Pond	CP	-	-	√	0.38	-	N/A	-	-
Laflamme	L	-	-	√	0.47	√	0.72	-	-
Pingualuk	P	√	0.38	√	0.57	√	0.71	√	0.19
Herbert	He	-	0.52	-	-	-	N/A	√	0.37
Desert	Dt	-	-	-	-	-	N/A	-	-
Radar	Rd	√	0.75	√	0.69	-	N/A	√	0.87

[a] Abbreviation of lake names used as labels in Figure 4; hyphens (-) indicate not significant results; *n* of Table 1 apply.

the presented covariates (Table 2), large scale comparisons, such as conducted here require statistical adjustment for a covariate that is significant for a maximum number of populations. Hence, a statistically adjusted subset of char was compared across the study region.

Subset for spatial comparison

To reduce biologically related variation of THg concentrations for spatial comparisons, we identified relationships (Table 2) of THg concentrations for the overall dataset ($n = 520$) and selected covariates for adjustment using ANCOVA. As the slopes of the regression of THg concentration with $\delta^{15}N$, length, age, and weight varied considerably from lake to lake, the adjustment using ANCOVA failed when attempted for all Arctic char (interaction was observed $p < 0.05$). Varying sample size may have caused the interaction, as only nine of 27 lakes had >20 char, while 10 lakes had < 10 char. To standardize comparisons by lakes from all regions, we selected five to 10 similarly sized char from each lake, which were then adjusted using the ANCOVA model (see below). Data from three populations (Teardrop, Desert, and Tessisoak) were removed due to low sample size ($n = 2$ and 4) or non-common fish sizes (Tessisoak). The remaining 216 char (ranging from 173 to 639 mm fork length) from 24 lakes were used for the spatial comparison in the present study.

Fork length and $\delta^{15}N$ were evaluated as covariates in the adjustment using ANCOVA, as ages were not available for all lakes. The GLM model $\log[THg] = lake + \delta^{15}N + lake * \delta^{15}N$ revealed interaction ($p < 0.05$), indicating that there was no common slope of log[THg] and $\delta^{15}N$ to be found for the 24 lakes and thus $\delta^{15}N$ could not be used for ANCOVA adjustment. The same model was applied using baseline adjusted $\delta^{15}N$ values, again showing significant interaction ($p < 0.05$). However, the model log[THg] = lake + fork length + lake * fork length revealed no interaction ($p = 0.124$), and thus a common slope of log[THg] and length across lakes. The results also indicated a good fit for this model ($r^2 = 0.88$; $p < 0.05$). Subsequently, the GLM model was re-run without an interaction term (log[THg] = lake + fork length), and the resulting the least squares means from the model were used to back calculate fork length-adjusted THg concentrations. The resulting Hg concentrations are thus adjusted to the length estimated by the grand mean [46], rather than to a given length. Hereafter, the results from this adjustment with fork length as the covariate will be referred to as length-adjusted THg concentrations.

Spatial comparison

Ellesmere Island. In the most northern region (Ellesmere Island), length-adjusted THg concentrations in char varied significantly among the four lakes (ANCOVA $p < 0.05$) (Fig. 3). The highest ($p_{\text{Tukey's}} < 0.05$) concentrations of THg were found in Lake G (0.30 ± 0.04 μg/g), followed by the neighboring Lake F (0.23 ± 0.05 μg/g). Both sites had higher concentrations ($p_{\text{Tukey's}} < 0.05$) than Lake Hazen ($0.10 \pm 0.02$ μg/g) and Lake D (0.10 ± 0.02 μg/g). The latter two lakes did not differ significantly ($p_{\text{Tukey's}} > 0.05$). Lake D is located upstream of Lake Hazen and is in the same watershed, but further from Lake Hazen, than either lakes G and F. Lakes G and F drain into the outflow of Lake Hazen, and are thus not

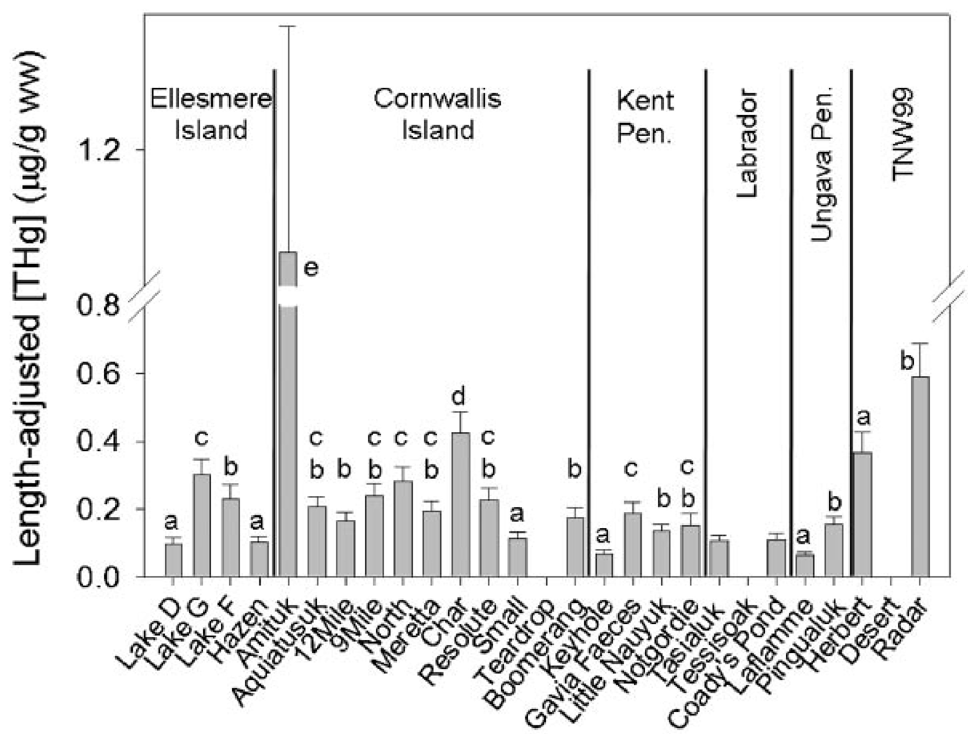

Fig. 3. Length-adjusted total Hg (THg) concentrations in 24 Arctic char populations. Bars represent THg concentrations (μg/g), back calculated from analysis of covariates least squares means. Significance (Tukey's test or Student's *t* test; $p < 0.05$) of differences between populations within each region is indicated by different lowercase associated letter(s). Labrador populations did not differ ($p = 0.49$). Teardrop, Tessisoak, and Desert lakes were removed due to small sample size before adjustment. Error bars represent 1 standard deviation. TNW99 indicates sites collected during the Tundra Northwest Expedition 1999. Two lakes (Pingualuk and Laflamme) are located on the Ungava Peninsula.

connected to the Hazen watershed. The similarity of lakes G and F is also apparent in their food chain lengths [6]. Of interest, we found the greatest concentrations of THg in char from the two smallest lakes (G and F) on Ellesmere Island, which is not the case for the other regions in the present study. These results suggest that lakes of the same watershed may have similar processes for delivery of Hg to the lake; this then leads to similar THg in char.

Cornwallis Island. Resolute Bay area char (includes Cornwallis Island, Somerset Island, Melville Island) showed the greatest length-adjusted THg concentrations in the char from the present study area. Concentrations differed among all populations (ANCOVA $p < 0.05$), and differences among lakes were found (Fig. 3). Amituk Lake char, the char with the greatest individual un-adjusted THg concentration (3.4 μg/g), also have the greatest mean length-adjusted THg concentrations (1.12 ± 0.17 μg/g) of lakes on Cornwallis Island, and of all populations in the present study. Next highest concentrations were found in Char and North lakes (0.43 ± 0.6, 0.28 ± 0.4 μg/g), respectively. Char from Small Lake had the lowest length-adjusted THg concentrations (0.11 ± 0.02 μg/g). Results of the present study concur with the previous studies [7], which also indicate that Amituk and Char lakes had the highest Hg concentrations in char. Although Hg release from upstream wetlands, input to, and export from Amituk Lake has been well studied [47], it remains a unique example of extraordinarily high char THg concentrations in a remote lake.

Kent Peninsula and Victoria Island. Concentrations of Hg in char on the Kent Peninsula were generally lower compared with the more northerly sites (Fig. 3). One explanation could be the more diverse benthic invertebrate communities (shorter FCL), or diverse feeding behavior (feeding on prey with lower Hg concentrations) among char within these lakes [6]. Among lakes in this region, char from Keyhole Lake on Victoria Island had the lowest concentration. Concentrations in Little Nauyuk and Notgordie Lake did not differ ($p_{\mathrm{Tukey's}} > 0.05$), neither did those of Notgordie and Gavia Faeces ($p_{\mathrm{Tukey's}} > 0.05$). Gavia Faeces char had higher concentrations than Little Nauyuk ($p_{\mathrm{Tukey's}} < 0.05$). Trophic magnification factors for Hg are high [6] and food web lengths are longer than in other regions of the present study (Fig. 4). The absence of cannibalism may be a further reason for lower mean THg concentrations. The fact that Keyhole Lake char had lower THg concentrations compared with Kent Peninsula sites may be an indication that regional factors, such as underlying geology, may have played a role.

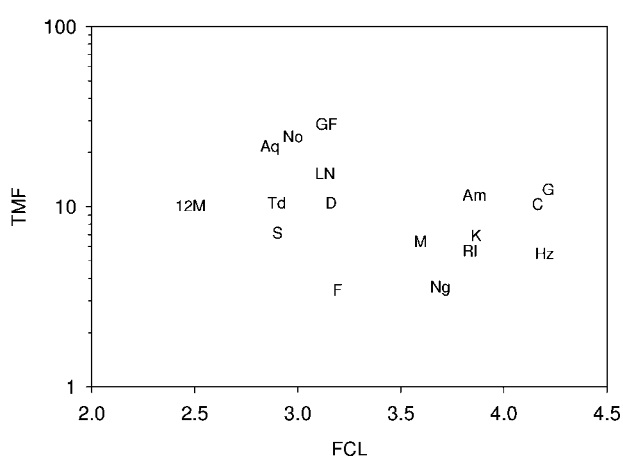

Fig. 4. Relationship of trophic biomagnification factors (TMF) and food chain length (FCL) in 17 lakes (9 Mile Lake TMF was identified as outlier). The first or first two letters of the lake name are used and codes are given in Table 2.

Easterly sites. Both sites on the northern Labrador coast, Coady's Pond and Tesialuk Lake had low length-adjusted Hg concentrations (Fig. 3). The adjusted Hg concentrations did not differ between lakes (Student's $t = -0.695$; $p = 0.496$). No food web information is available for these lakes. Because limited information on lake characteristics is available, these sites were excluded from most of the follow-on analysis. Lac Laflamme and Lake Pingualuk char from the Ungava Peninsula had low concentrations of THg compared with the other sites in the present study. Pingualuk char had higher concentrations than Laflamme (Student's $t = -9.567$; $p < 0.05$), which is explained by its FCL of approximately 4.2 (Fig. 4). The char population from Radar Lake on Baffin Island showed the second highest length-adjusted THg concentrations at 0.59 ± 0.1 μg/g wet weight in the present study region. Although this lake is located near an abandoned Distant Early Warning Line radar station, previous studies have shown no signs of contamination with Hg or other metals in this lake [33]. Food chain length in this lake was determined to be approximately 4.2 (using zooplankton as the baseline) [6], which is indicative of piscivory/cannibalism and could be the reason for higher Hg concentrations (Fig. 4).

Yukon - westerly site. Finally, Herbert Lake, the most western lake of the present study range, showed higher concentrations of THg compared with most lakes in the present study (0.37 ± 0.06 μg/g). This is in contrast to the low MeHg concentrations in zooplankton detected in this lake [6]. Other metals, especially cadmium (Cd), were previously measured in char muscle from this lake [33], and also exceeded concentrations found in other lakes of that study.

Food chain length and biomagnification

For an additional four sites, FCLs were determined for TNW99 lakes and Lake Pingualuk using zooplankton $\delta^{15}N$ as the baseline; however, no TMFs were available from these four lakes. Linear regression analysis showed no relationship between TMF and FCL (Fig. 4), after removing the outlier value (Studentized residual 5.554) of the TMF for Hg in 9 Mile Lake (64.3).

Surprisingly, FCL alone was not predictive of length-adjusted Hg concentrations (linear regression, $r^2 = 0.20$; $p = 0.251$) across our data set (22 lakes), see Figure 5. Trophic magnification factors also did not predict length-adjusted Hg in Arctic char ($p > 0.05$). Multiple regression of log[length-adjTHg] and TMFs and FCL was not significant ($p > 0.05$). This may be less surprising than the previously described lack of relationship with FCL, as TMFs are measures that integrate the uptake of Hg through whole lake food webs. High TMFs can only lead to high THg in fish, if the food chain is long. The linkage is supported by our findings in Amituk and Char lakes where Hg in char is high (length-adj THg = 1.12 and 0.42 μg/g wet wt, with FCL = 3.8 and 4.2, respectively) due to TMFs of approximately 10, whereas concentrations of THg in 9 Mile Lake char are low (0.24 μg/g wet wt, FCL = 2.1), despite high TMF (~64), due to a short food chain.

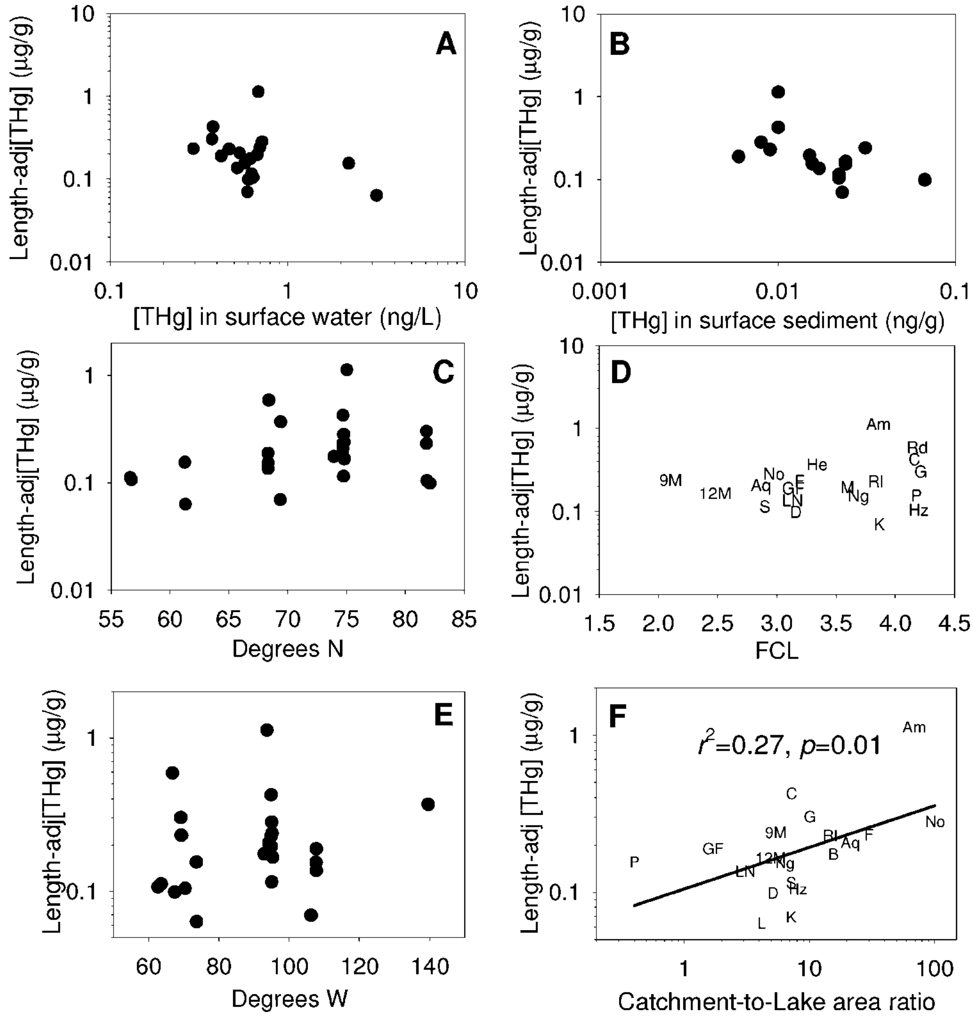

Fig. 5. Relationships among concentrations of total mercury (THg) in water, sediment (dry wt), and length-adjusted [THg] in char muscle (wet wt) (A,B), char [THg] with latitude (C), food chain length (FCL) (D), longitude (E), and catchment-to lake area ratio (F). Regression lines are shown for significant relationships only. The first or first two letters of the lake name are used and codes are given in Table 2.

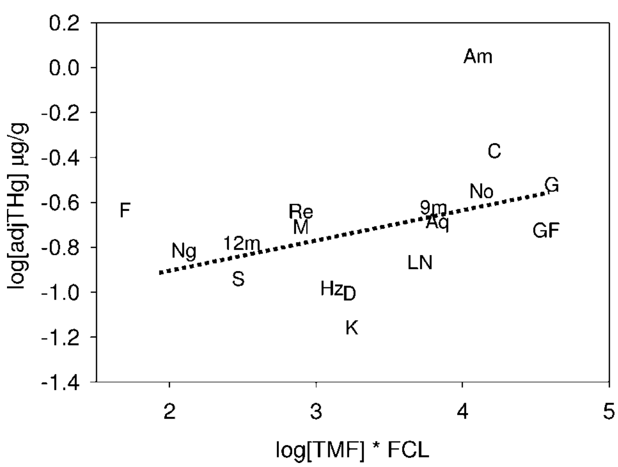

Fig. 6. Modeled relationship of food chain length (FCL)-adjusted logTMF] and length-adjusted [THg]. The dashed line indicates the observed trend (nonsignificant, $p = 0.09$). The first or first two letters of the lake name are used and codes are given in Table 2. TMF = trophic biomagnification factors; THg = total mercury.

Trophic magnification factors alone are not sufficient to predict Hg concentrations in char (no significant correlation between the average THg and TMF), as the results from this and our accompanying study revealed [6]. While TMFs are indicative of the rate of biomagnification (slope of the regression), the extent of Hg uptake is ultimately determined by FCL. In addition, because TMFs are derived from the slopes of MeHg and TL regression of all organisms in the food web, they indicate the efficiency of Hg (MeHg) transfer through the food web, rather than the quantity of Hg delivered to a top predator. We conclude that TMFs are useful to describe efficiency in trophic uptake in lake food webs that are well characterized; however, TMF has to be applied in conjunction with a measure of food web structure, such as FCL.

The aforementioned observations led us to attempt to model the combined effect of TMFs and FCL on THg in char. Therefore, we multiplied the slope of the regression of biomagnification (log[TMF]) with FCL for each of the 18 lakes. The results of log[TMF] × FCL were then plotted against the length-adjusted THg concentrations (Fig. 6). Again, the predictive capacity of log[TMF] × FCL calculation was low (linear regression $r^2 = 0.12$; $p = 0.09$) for all 18 lakes. In addition a

GLM model was run (log[adjHg] = logTMF + FCL = logTMF × FCL, resulting in similarly low predictability ($r^2_{adj} = 0.001$) of Hg in char. This observation may also apply to other bioaccumulative contaminants [24,46], and we suggest it be considered in future bioaccumulation studies.

Hg species in water and sediments

Concentrations of THg in water ranged from 0.29 to 0.72 ng/L in North Lake and Lake F, respectively. Length-adjusted THg concentrations in Arctic char, which should reflect >90 % MeHg according to literature values for salmonid species [48], were not correlated with THg concentrations in surface water ($p > 0.05$) or those of surface sediments (Fig. 5A,B). Measurements of MeHg in water were available for six lakes, although they were typically near or below the detection limits of the analytical laboratories (∼0.01–0.02 ng/L), making valid comparisons impossible. In surface water, Hg is mainly present in its inorganic forms (92%–97% of THg), which may explain the poor relationship. In addition, surface water samples were obtained as a single annual sampling event per year, and may not accurately represent water Hg concentrations for the whole lake or season.

Concentrations of THg in sediments ranged from 6 to 156 ng/g (all dry wt) in Notgordie and Pingualuk lakes, respectively, averaging 27 ng/g overall ($n = 18$). Methylmercury available from eight lakes ranged from 0.10 ng/g in Teardrop and Char lakes to 0.24 ng/g (North Lake), thus representing only approximately 0.5 to 1.0 % of THg in sediment. Concentrations in sediment were expected to be correlated with fish Hg, because they represent the site of methylation and thus the entry point of bioavailable (organic) MeHg into food webs, as recently suggested by Chételat et al. [49]. However, sediment THg (log[THg]) and MeHg (log[MeHg]) were not predictive of THg in fish (linear regression $p > 0.05$).

THg in char and environmental parameters

Relationships of length-adjusted THg concentrations in Arctic char with several known predictors of geographic trends of THg in fish revealed only a few of the predictors to be explanatory of the present study data. Latitude appears to have no effect on Hg concentrations in landlocked char (linear regression $p > 0.05$, Fig. 5C). Nor was there any correlation between THg in char and longitude ($p > 0.05$, Fig. 5E). Catchment area and lake area did not predict THg concentrations in char (both linear regression $p > 0.05$). However, catchment-to-lake area ratios (CA/LA) did show a positive relation (linear regression $r^2 = 0.27$; $p = 0.01$) with THg in char, with greater CA/LA ratios leading to higher THg concentrations in char (Fig. 5F; Table 3). The CA/LA ratios were highest for Amituk and North lakes, which also had higher Hg concentrations in char. Lake Pingualuk, a meteor impact crater lake, has a minimal catchment area, and low THg concentrations. Multiple linear regression analysis using CA/LA ($n = 17$) and FCL as independent variables revealed the relationship log[length-adjTHg] = −1.039 + (0.00585 × CA/LA) + (0.0662 × FCL), and weakly predictive of length-adjusted THg ($r^2 = 0.24$; $p = 0.058$). These results indicate that input of Hg from the surrounding land into lakes may have an effect on concentrations in char, which previous studies of lower trophic organisms [49] and Hg in sediments in Arctic lakes have also suggested [14]. The influence of CA/LA ratios on sediments have been previously described for mid-latitude lakes [50]. Muir et al. [17] suggest that increased aeolian inputs and/or greater erosion due to higher snowfall over the past 50 years could have affected sediment Hg concentrations in the central Canadian Arctic archipelago. Linkage of Hg deposition data and MeHg in a southern fish species (largemouth bass, *Micropterus salmoides* L.) has been established by Hammerschmidt and Fitzgerald [51], in multiple fish species in eastern North America [52], and in recent experimental studies in northwestern Ontario, Canada [53]. Moreover, the results of the present study highlights a possible linkage to climate vulnerability; melting permafrost or changes in precipitation under predicted climate scenarios [54] could increase Hg inputs from catchments to lakes. Predicted dryer conditions within the watershed due to warmer summers [55] could lead to drying out of wetlands and increased oxidative processes in normally wetted soil. Lakes of similar size within one watershed would respond similarly to change, as supported by our results from Ellesmere Island, Nunavut. Consequently, small lakes with large catchment areas may be most vulnerable to increased inputs due to climate induced changes in precipitation and temperature. Catchment-or-lake areas are indicative of contributions of Hg to sediments, however, to estimate the contribution of Hg from the atmosphere, catchment-specific precipitation needs to be taken in account.

Table 3. Relationships of log[length-adjTHg] with location and selected environmental factors[a]

	Linear regression parameters			
log[L-adj-THg] vs	slope	intercept	r^2_{adj}	p
Latitude	-	-	0.041	0.17
Longitude	-	-	-	0.36
Lake area (LA)	-	-	0.114	0.09
Catchment area (CA)	-	-	-	0.90
CA-LA ratio	0.2643	−0.9795	0.269	0.01
DOC	-	-	-	0.47
Chlorophyll *a*	-	-	-	0.69

[a] Hyphens (-) indicate value not determined; DOC = dissolved organic carbon.

Underlying geology can affect Hg concentrations in biota, as shown for Arctic fish by Lockhart et al. [1] and for ringed seals from Canada and Greenland [56]. However, the geology on Cornwallis Island and Ellesmere Island is similar; it is composed of tertiary sediment over carbonate bedrock [57] and all lakes on these islands can be considered to be on sedimentary bedrock [1]. Kent Peninsula area lakes may be influenced by metamorphic bedrock due to the nearby continental shelf/mainland. This could hold true for Baffin Island and the North Slope of the Yukon (Herbert Lake) as well, because underlying geology may be an alternative explanation for the high THg concentrations in Radar Lake.

CONCLUSION

Mercury concentrations from 27 Arctic char populations from a large geographical area are summarized here; the first comprehensive investigation of its kind for the circumpolar region. The present study has expanded the knowledge from previous studies on Hg concentrations in landlocked char

[1,7,58] not only by increasing number of lakes, but including information on food webs. We demonstrate intralake variability in THg concentrations due to difference in size, age, and trophic position of individual char within a lake, which concurs with the literature on other freshwater fish. A combination of the above factors in conjunction with biotic lake characteristics (such as TMFs and FCL combined) appear to influence THg concentrations in Arctic char of a given lake. Spatial comparison of a length-adjusted subset of samples indicated significant interlake variability of THg, however, catchment-to-lake-area ratios were related to length-adjusted THg concentrations in fish. This relationship highlights a possible linkage of Hg concentrations in fish and climate change because increased catchment inputs of THg or MeHg may result in greater Hg concentrations in fish.

Acknowledgement—The authors thank Pilipoosie Iqaluk and Günter Köck for assistance with field collections from 2005 to 2007. Polar Continental Shelf Project managers and employees provided logistical support (Project nos. 204-05, 661-06, 651-07) and contributed to safety and comfort during our field work. Milton Shears and co-workers collected all samples from Labrador. Parks Canada and staff aided greatly in sampling within Quttinirpaaq National Park on Ellesmere Island. Rick Wastle performed the aging of char otoliths at the Freshwater Institute, Winnipeg, MB (Canada). John Chételat and Marc Amyot provided MeHg data for surface water of six lakes. We thank Jessica Epp and Xiaowa Wang (EC Burlington) for assisting in THg analysis and determination of lake and catchment areas. This research was conducted under the terms of the Nunavut Research Institute (License: 0203206R-M), Department of Fisheries and Oceans (License: S-06/07-1015-NU), and the Ministère des Resources Naturelles et de la Faune du Québec (License: 9053.28). All work from 2005 to 2007 followed the of the University of Guelph animal care protocols (06R083). The Northern Ecosystem Initiative, Northern Contaminants Program, NCE Arctic Net, Canadian IPY Project – CVC on Chars in the Arctic, and Austrian Academy of Science (DOC-Stipend for lead author) provided funding for this research. The Royal Swedish Academy of Sciences provided funding for the Tundra Northwest 99 expedition. Two anonymous reviewers have improved the quality of this work through their comments.

REFERENCES

1. Lockhart WL, Stern G, Low G, Hendzel M, Boila G, Roach P, Evans MS, Billeck BN, DeLaronde J, Friesen S, Kidd KA, Atkins S, Muir DCG, Stoddart M, Stephens GR, Stephenson S, Harbicht S, Snowshoe N, Grey BJ, Thompson S, DeGraff N. 2005. A history of total mercury in edible muscle of fish from lakes in northern Canada. *Sci Total Environ* 351–352:427–463.
2. Munthe J, Bodaly RA, Branfireun BA, Driscoll CT, Gilmour CC, Harris R, Horvat M, Lucotte M, Malm O. 2007. Recovery of mercury-contaminated fisheries. *Ambio* 36:33–44.
3. Mergler D, Anderson HA, Hing Man Chan L, Mahaffey KR, Murray M, Sakamoto M, Stern AH. 2007. Methylmercury exposure and health effects in humans: A worldwide concern. *Ambio* 36:3–11.
4. Pacyna EG, Pacyna JM, Steenhuisenc F, Wilson S. 2006. Global anthropogenic mercury emission inventory for 2000. *Atmos Environ* 40:4048–4063.
5. Lindberg S, Bullock R, Ebinghaus R, Engstrom DR, Feng X, Fitzgerald WF, Pirrone N, Prestbo E, Seigneur C. 2007. A synthesis of progress and uncertainties in attributing the sources of mercury in deposition. *Ambio* 36:19–32.
6. Gantner N, Power M, Iqaluk D, Meili M, Borg H, Sundbom M, Solomon KR, Lawson G, Muir DCG. 2010. Mercury concentrations in landlocked arctic char (*Salvelinus alpinus*) from the Canadian High Arctic: Part I. Insights from trophic relationships in 18 lakes. *Environ Toxicol Chem* (this issue).
7. Muir DC, Wang X, Bright D, Lockhart L, Köck G. 2005. Spatial and temporal trends of mercury and other metals in landlocked char from lakes in the Canadian arctic archipelago. *Sci Total Environ* 351–352:464–478.
8. Evans MS, Lockhart L, Doetzel L, Low G, Muir D, Kidd KA, Stephens GR, Delaronde J. 2005. Elevated mercury concentrations in fish in lakes in the Mackenzie River Basin: The role of physical, chemical, and biological factors. *Sci Total Environ* 351–352:479–500.
9. Jewett SC, Duffy LK. 2007. Mercury in fishes of Alaska, with emphasis on subsistence species. *Sci Total Environ* 387:3–27.
10. Gantner N, Power M, Babaluk JA, Reist JD, Koeck G, Lockhart WL, Solomon KR, Muir DC. 2009. Temporal trends of mercury, cesium, potassium, selenium, and thallium in arctic char (*Salvelinus alpinus*) from Lake Hazen (Nunavut): Effects of trophic position, size and age. *Environ Toxicol Chem* 28:254–263.
11. Outridge PM, Sanei H, Stern G, Hamilton PB, Goodarzi F. 2007. Evidence for control of mercury accumulation rates in Canadian high arctic lake sediments by variations of aquatic primary productivity. *Environ Sci Technol* 41:5259–5265.
12. Schindler DW. 2001. The cumulative effects of climate warming and other human stresses on Canadian freshwaters in the new millennium. *Can J Fish Aquat Sci* 58:18–29.
13. Macdonald R, Harner T, Fyfe J, Loeng H, Weingartner T. 2003. The influence of global change on contaminant pathways to, within, and from the arctic. AMAP Assessment Report. Arctic Monitoring Assessment Program, Oslo, Norway.
14. Fitzgerald WF, Engstrom DR, Lamborg C, Tseng CM, Balcom PH, Hammerschmidt CR. 2005. Modern and historic atmospheric mercury fluxes in northern Alaska: Global sources and arctic depletion. *Environ Sci Technol* 39:557–568.
15. Steffen A, Douglas M, Amyot M, Ariya PA, Aspmo K, Berg T, Bottenheim J, Brooks S, Cobbett FD, Dastoor A, Dommergue A, Ebinghaus R, Ferrari C, Gardfeldt K, Goodsite M, Lean D, Poulain AJ, Scherz C, Skov H, Sommar J, Temme C. 2008. A synthesis of atmospheric mercury depletion event chemistry in the atmosphere and snow. *Atmos Chem Phys* 8:1445–1482.
16. Temme C, Blanchard P, Steffen A, Banic C, Beauchamp S, Poissant L, Tordon R, Wiens B. 2007. Trend, seasonal and multivariate analysis study of total gaseous mercury data from the Canadian atmospheric mercury measurement network (CAMNet). *Atmos Environ* 41:5423–5441.
17. Muir DC, Wang X, Yang F, Jackson TA, Evans M, Douglas M, Köck G, Pienitz R, Smol JP, Vincent W, Lamoureux S. 2009. Spatial trends and historical inputs of mercury and lead in eastern and northern Canada inferred from lake sediment cores. *Environ Sci Technol* 43:4802–4809.
18. Hall BD, Bodaly RA, Fudge RJ, Rudd JWM, Rosenberg DM. 1997. Food as the dominant pathway of methylmercury uptake by fish. *Water Air Soil Pollut* 100:13–24.
19. MacCrimmon HR, Wren CD, Gots BL. 1983. Mercury uptake by lake trout, *Salvelinus namaycush*, relative to age, growth, and diet in Tadanac Lake with comparative data from PreCambian Shield lakes. *Can J Fish Aquat Sci* 40:114–120.
20. Vander Zanden MJ, Rasmussen JB. 2001. Variation in $\delta^{15}N$ and $\delta^{13}C$ trophic fractionation: Implications for aquatic food web studies. *Limnol Oceanogr* 46:2061–2066.
21. Power M, Klein M, Guiguer KRRA, Kwan MKH. 2002. Mercury accumulation in the fish community of a sub-arctic lake in relation to trophic position and carbon sources. *J Appl Ecol* 39:819–830.
22. Guiguer KRRA, Reist JD, Power M, Babaluk JA. 2002. Using stable isotopes to confirm the trophic ecology of arctic charr morphotypes from Lake Hazen, Nunavut, Canada. *J Fish Biol* 60:348–362.
23. Rodgers DW. 1996. Methylmercury accumulation by reservior fish: Bioenergetic and trophic effects. In Miranda LE, DeVries DR, eds Multidimensional Approaches to Reservoir Fisheries Management. Symposium 16. American Fisheries Society, Bethesda, MD. pp 107-118.
24. Guildford S, Muir D, Houde M, Evans MS, Kidd KA, Drouillard K, Wang X, Anderson R, Bronte C, DeVault D, Haffner GD, Payne J, Kling H. 2008. Influence of geographic factors and possible implications of climate warming for PCB concentrations in lake trout (*Salvelinus namaycush*). *Environ Sci Technol* 42:8239–8244.
25. Kidd KA. 1998. Use of stable isotope ratios in freshwater and marine biomagnification studies. In Rose J, ed, Environmental Toxicology: Current Developments. Gordon and Breach Science, London, UK. pp 359–378.
26. Hammerschmidt CR, Fitzgerald WF, Lamborg C, Balcom PH, Tseng CM. 2006. Biogeochemical cycling of methylmercury in lakes and tundra watersheds of arctic Alaska. *Environ Sci Technol* 40:1204–1211.
27. Simonin HA, Loukmas JJ, Skinner LC, Roy KM. 2008. Lake variability: Key factors controlling mercury concentrations in New York State fish. *Environ Pollut* 154:107–115.
28. Scott WB, Crossman EJ. 1973. *Freshwater Fishes of Canada*. Fisheries Research Board of Canada, Ottawa, ON.

29. Power M, Reist J, Dempson B. 2008. Fish in high-latitude arctic lakes. In Vincent W, Laybourn-Parry J, eds Polar Lakes and Rivers. Oxford University Press, Oxford, UK. pp 249-268.
30. Johnson L, Burns B. 1984. *Biology of the Arctic Charr: Proceedings*, International Symposium on Arctic charr. University of Manitoba Press, Winnipeg, MB, Canada.
31. Hammar J. 1989. Freshwater ecosystems of polar regions: Vulnerable resources. *Ambio* 1:6–22.
32. Parker HH, Johnson L. 1991. Population structure, ecological segregation and reproduction in non-anadromous arctic charr, *Salvelinus alpinus* (L.), in four unexploited lakes in the Canadian high arctic. *J Fish Biol* 38:123–147.
33. Borg H, Holm K, Meili M. 2001. Distribution of trace elements in arctic lakes-some preliminary results from the Tundra Northwest expedition. Arctic Monitoring and Assessment Programme, Oslo, Norway.
34. Riget F, Asmund G, Aastrup P. 2000. Mercury in arctic char (*Salvelinus alpinus*) populations from Greenland. *Sci Total Environ* 245:161–172.
35. Babaluk JA, Wissink RH, Troke BG, Clark DA, Johnson JD. 2001. Summer movements of radio-tagged arctic charr (*Salvelinus alpinus*) in Lake Hazen, Nunavut, Canada. *Arctic* 54:418–424.
36. Keatley B, Douglas M, Smol JP. 2007. Limnological characteristics of a high arctic oasis and comparisons across northern Ellesmere Island. *Arctic* 60:294–308.
37. Smith RI. 2002. Diatom-based holocene paleoenvironmental records from continental sites on northeastern Ellesmere Island, High Arctic, Canada. *J Paleolimnol* 27:9–28.
38. Hobson KA, Welch HE. 1995. Cannibalism and trophic structure in a high arctic lake: Insight from stable-isotope analysis. *Can J Fish Aquat Sci* 52:1195–1201.
39. Muir DC, Köck G, Gantner N. 2006. Temporal trends of persistent organic pollutants and mercury in landlocked char in the high arctic. In Smith SL, Stow J, eds, Synopsis of Research Conducted Under the 2005–2006, Northern Contaminants Program. Indian and Northern Affairs Canada, Ottawa, ON. pp 155–161.
40. Johnson L. 1994. Long-term experiments on the stability of two fish populations in previously unexploited arctic lakes. *Can J Fish Aquat Sci* 51:209–225.
41. Vanriel P, Johnson L. 1995. Action principles as determinants of ecosystem structure: The autonomous lake as a reference system. *Ecology* 76:1741–1757.
42. Bouchard M. 1989. *L'Historie naturelle du Cratère du Nouveau Québec*. Universite de Montreal, Montreal, QC, Canada.
43. Bruce WJ, Spencer KD, Arsenault E. 1979. Mercury content data for Labrador fishes, 1977–78. Report 142. Department of Fisheries and Oceans, St John's, NL, Canada.
44. U.S., Environmental Protection Agency. 2007. Method 7473: Mercury in Solids and Solutions by Thermal Decomposition, Amalgamation, and Atomic Absorption Spectrophotometry. Washington, DC.
45. Systat Software. 2004. *SigmaStat 3.1*. San Jose, CA, USA.
46. Houde M, Muir D, Kidd KA, Guildford S, Drouillard K, Evans MS, Wang X, Whittle MD, Haffner D, Kling H. 2008. Influence of lake characteristics on the biomagnification of 16 persistent organic pollutants in lake trout food webs. *Environ Toxicol Chem* 27:2169–2178.
47. Semkin RG, Mierleb G, Neureuther RJ. 2005. Hydrochemistry and mercury cycling in a high arctic watershed. *Sci Total Environ* 342:199–221.
48. Kelly BC, Ikonomou MG, Higgs DA, Oacks J, Dubetz C. 2008. Mercury and other trace elements in farmed and wild salmon from British Columbia, Canada. *Environ Toxicol Chem* 27:1361–1370.
49. Chételat J, Amyot M, Coultier L, Poulain AJ. 2008. Metamorphosis in chironomids, more than mercury supply, controls methylmercury transfer to fish in high arctic lakes. *Environ Sci Technol* 42:9110–9115.
50. Lorey P, Driscoll CT. 1999. Historical trends of mercury deposition in Adirondack lakes. *Environ Sci Technol* 33:718–722.
51. Hammerschmidt CR, Fitzgerald WF. 2006. Methylmercury in freshwater fish linked to atmospheric mercury deposition. *Environ Sci Technol* 40:7764–7770.
52. Kamman M, Burgess NM, Driscoll CT, Simonin HA, Estabrook R, Hutcheson M, Major A, Scheuhammer AM. 2005. Mercury in freshwater fish of northeast North America - a geographic perspective based on fish tissue monitoring databases. *Ecotoxicology* 14:163–180.
53. Harris RC, Rudd JW, Amyot M, Babiarz CL, Beaty KG, Blanchfield PJ, Bodaly RA, Branfireun BA, Gilmour CC, Graydon JA, Heyes A, Hintelmann H, Hurley JP, Kelly C, Krabbenhoft DP, Lindberg S, Mason PS, Paterson MJ, Podermski CL, Robinson A, Sandilands KA, Southworth GR, St Louis V, Tate MT. 2007. Whole-ecosystem study shows rapid fish-mercury response to changes in mercury deposition. *Proc Natl Acad Sci U S A* 104:16586–16591.
54. Prowse TD, Wrona FJ, Reist J, Gibson JJ, Hobbie J, Le'vesque LMJ, Vincent W. 2006. Climate change effects on hydroecology of arctic freshwater ecosystems. *Ambio* 35:347–358.
55. Smol JP, Douglas MSV. 2007. Crossing the final ecological threshold in high arctic ponds. *Proc Natl Acad Sci U S A* 104:12395–12397.
56. Riget F, Muir DC, Kwan M, Savinova T, Nyman M, Woshner V, O'Hara T. 2005. Circumpolar pattern of mercury and cadmium in ringed seals. *Sci Total Environ* 351–352:312–322.
57. Natural Resources Canada. 1997. Geological map of Canada. Ottawa, ON.
58. Köck G, Babaluk J, Berger B, Bright D, Doblander C, Flannigan M, Kalra Y, Loseto L, Miesbauer H, Muir D, Niederstätter H, Reist J, Telmer K. 2004. Fish from Sensitive Ecosystems as Bioindicators of Global Climate Change - ''High-Arctic 1997-2003 ''. Veröffentlichungen der Universität Innsbruck, Innsbruck, Austria.