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COMPARISON OF MERCURY CONCENTRATIONS IN LANDLOCKED, RESIDENT, AND SEA-RUN FISH (*SALVELINUS* SPP.) FROM NUNAVUT, CANADA

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Abstract—Mercury concentrations ([Hg]) in Arctic food fish often exceed guidelines for human subsistence consumption. Previous research on two food fish species, Arctic char (*Salvelinus alpinus*) and lake trout (*Salvelinus namaycush*), indicates that anadromous fish have lower [Hg] than nonanadromous fish, but there have been no intraregional comparisons. Also, no comparisons of [Hg] among anadromous (sea-run), resident (marine access but do not migrate), and landlocked (no marine access) life history types of Arctic char and lake trout have been published. Using intraregional data from 10 lakes in the West Kitikmeot area of Nunavut, Canada, we found that [Hg] varied significantly among species and life history types. Differences among species–life history types were best explained by age-at-size and C:N ratios (indicator of lipid); [Hg] was significantly and negatively related to both. At a standardized fork length of 500 mm, lake trout had significantly higher [Hg] (mean 0.17 μg/g wet wt) than Arctic char (0.09 μg/g). Anadromous and resident Arctic char had significantly lower [Hg] (each 0.04 μg/g) than landlocked Arctic char (0.19 μg/g). Anadromous lake trout had significantly lower [Hg] (0.12 μg/g) than resident lake trout (0.18 μg/g), but no significant difference in [Hg] was seen between landlocked lake trout (0.21 μg/g) and other life history types. Our results are relevant to human health assessments and consumption guidance and will inform models of Hg accumulation in Arctic fish. Environ. Toxicol. Chem. 2011;30:1459–1467. © 2011 SETAC

Keywords—Lake trout Arctic char Mercury Life history Arctic food fish

INTRODUCTION

Mercury (Hg) is a neurotoxicant and contaminant of concern for both ecological and human health reasons [1,2]. The organic form of Hg, methylmercury (MeHg), is toxic and can bioaccumulate through aquatic and marine food webs to reach high concentrations in predatory fish [3–5]. These concentrations can exceed guidelines for human consumption, and much research has been devoted to elucidating factors that predict Hg concentrations ([Hg]) in fish [3,6–8]. This is particularly important in areas where human exposure to fish-derived Hg is potentially high, such as in Arctic communities with subsistence fisheries.

Consuming fish presents both risks and benefits to human health. Globally, fish are the dominant source of Hg to human populations ([9]; http://www.inchem.org/documents/ehc/ehc/ehc101.htm). In the Canadian Arctic, where a large diversity of traditionally harvested foods are eaten, tissues and organs from caribou and marine mammals represent the largest proportional contribution of Hg to human diets ([10]; http://www.collectionscanada.gc.ca/webarchives/20071206061307/http://www.ainc-inac.gc.ca/ncp/pub/helttoc_e.html). Fish such as Arctic char (*Salvelinus alpinus* L.) and lake trout (*Salvelinus namaycush* W.) represent a food source that is relatively lower in [Hg] compared with some other traditionally harvested foods, but [Hg] in these fish still often exceeds guidelines for commercial sale (0.5 μg/g wet wt) and subsistence consumption (0.2 μg/g wet wt) [4,5,7,11]. Despite these figures, consumption of Arctic char and lake trout is usually encouraged in Arctic communities because of the provision of essential nutrients and fatty acids, and the economic, cultural, and spiritual benefits of sharing traditionally harvested foods [2,10]. Effectively assessing and communicating both risks and benefits of fish consumption in the Canadian Arctic is obviously an ongoing and challenging task and is contingent on a thorough understanding of factors that influence fish [Hg].

Mercury concentrations in fish vary with factors that affect Hg inputs to lakes, rates of Hg methylation, and bioaccumulation processes. Atmospheric deposition of anthropogenically derived Hg is thought to be the most important source of Hg to most remote lakes in the Canadian Arctic [12,13]. Methylation of Hg tends to increase with decreasing lake area and pH, and increase with increasing primary productivity, lake temperature, and concentrations of dissolved organic carbon (DOC) [14]. Bioaccumulation and biomagnification of Hg in fish is influenced by fish age, size [6,15], trophic position (as indicated by $\delta^{15}N$) [3], and growth rate [15]. Although older, larger fish that feed at a high trophic position tend to have higher [Hg], fish [Hg] usually decreases with faster growth rate and higher body condition [6,16,17]. Fish with higher lipid content also tend to have lower [Hg] [14]. Growth rate, condition, trophic position, and lipid content can vary with fish species and life history [18–20] and may explain differences in [Hg] that have been observed among species–life history types of Arctic char and lake trout [4,5,21], but further research is necessary.

Arctic char are known to have a very plastic life history [18], and in the Canadian Arctic lake trout have more plasticity in their life history than was previously thought [19]. Populations of Arctic char and lake trout can be landlocked or partially anadromous (sea-run) in Canadian Arctic lakes [18,19].

All Supplemental Data may be found in the online version of this article.

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Partially anadromous populations of Arctic char and lake trout are composed of sympatric resident and anadromous individuals [18,19] that are genetically indistinct [22]. Therefore, there are three main life history types: anadromous, resident (access to marine environments but stay in freshwater), and landlocked (no access to marine environments).

Our knowledge of [Hg] variation among the three life history types of Arctic char and lake trout is fragmented and incomplete. Lake trout generally have higher [Hg] than Arctic char in the Canadian Arctic [4,5,20]. Anadromous fish have been reported to have lower [Hg] than freshwater-only fish, but this appears to differ between species and depend on whether the freshwater-only fish are landlocked or resident [4,5,20]. In a recent study, Swanson and Kidd [20] found that [Hg] did not differ between sympatric anadromous and resident Arctic char, but that [Hg] was significantly lower in anadromous lake trout than in sympatric resident lake trout. Comparisons with landlocked lake trout and Arctic char were not possible in this earlier study. Previous research has shown that landlocked Arctic char have higher [Hg] than anadromous Arctic char [4,5,21]; however, these comparisons have often been confounded by large spatial scales. Also, [Hg] has not been previously compared between landlocked and resident Arctic char.

Many factors have been implicated to explain observed differences in [Hg] among species–life history types of Arctic char and lake trout. Evans et al. [4] suggested that higher [Hg] in lake trout relative to Arctic char may be explained by slower growth rates in lake trout or warmer temperatures in lakes that support lake trout; warmer temperatures lead to faster rates of Hg methylation [14]. Consistent with the growth explanation of Evans et al. [4], Swanson and Kidd [20] found in a study of six coastal Arctic lake that differences in age-at-size explained much of the difference in [Hg] between Arctic char and lake trout. Evans et al. [4] also suggested that slower growth rates may explain why landlocked Arctic char have higher [Hg] than anadromous Arctic char. Alternatively, these authors suggested that higher [Hg] in landlocked Arctic char may result from higher incidence of cannibalism in landlocked Arctic char, higher temperatures and concentrations of dissolved organic carbon in lakes that support landlocked Arctic char, or higher [Hg] in freshwater prey items relative to marine prey items. Swanson and Kidd [20] found that lower [Hg] in anadromous lake trout relative to resident lake trout reflected faster growth rates and higher C:N (indicator of lipid [23]). These authors also found no difference in [Hg] between marine and freshwater prey items.

To address the fragmented and sometimes confounded state of knowledge on differences in [Hg] between species and life history types of Arctic char and lake trout in the Canadian Arctic, we combined data from Swanson et al. [20] and Gantner et al. [7,8] to create a dataset that includes [Hg] and biological information for landlocked, resident, and anadromous Arctic char and lake trout. We restricted the analysis to fish captured in the West Kitikmeot region of Nunavut, Canada, and made intraregional comparisons of [Hg] between species and among all three life history types. To explain observed patterns and contribute to development of predictive models, we related [Hg] to individual-level covariates ($\delta^{15}N$, $\delta^{13}C$, age, length, condition, C:N ratios) and compared significant covariates among species–life history types. We also compared ecosystem-level covariates of Hg methylation and Hg availability (lake size, chlorophyll *a* concentrations, MeHg concentrations in zooplankton, and %MeHg in zooplankton) between lakes with and without access to the marine environment.

METHODS

Site selection

Ten lakes in the West Kitikmeot region of Nunavut were sampled for Arctic char, lake trout, and zooplankton in the open water seasons of 2006 to 2008 (Fig. 1). Four of these lakes allow fish passage to the marine environment via outflows, whereas the remaining six are isolated from the marine environment by either impassable barriers (e.g., waterfalls) or small outflows. Four lakes contained landlocked Arctic char (Notgordie, Gavia, Little Nauyuk, and Keyhole; no access to the sea), two lakes contained landlocked lake trout (Patch and Doris; no access to the sea), three lakes contained partially anadromous populations (i.e., both anadromous and resident individuals) of Arctic char and lake trout (Roberts, Hovaktok, and Nauyuk; access to the sea), and one lake contained a partially anadromous population of lake trout but no Arctic char (Glenn; access to the sea) (Table 1). All lakes are located within a radius of approximately 175 km and are within approximately 150 km of Cambridge Bay, Nunavut (69° 9′N, 105° 23′W; Fig. 1). At least four of the 10 lakes (Keyhole, Nauyuk, Hovaktok, Little Nauyuk) are used for subsistence fish harvest in winter by residents of Cambridge Bay (A. Buchan, Hope Bay Mining, Nunavut, Canada, personal communication), but this harvest has not been quantified.

Sample collection and aging

Arctic char and lake trout were collected using a Smith-Root Type 12 backpack electrofisher, sinking gill nets (mesh sizes ranging from 1.9 to 8.9 cm stretched mesh), and ice fishing. Local Inuit aided in fish collections and fish processing. All fish were measured (nearest mm) and weighed (nearest g) in the field (see Supplemental Data for summary statistics) and dissected for dorsal muscle tissue and sagittal otoliths. Muscle tissue was frozen immediately after processing, and otoliths were cleaned and dried.

Fish captured in lakes without access to the sea were classified as landlocked. Fish captured in lakes with access to the sea were differentiated into anadromous and resident life history types in a previous study using otolith microchemistry [19]. Ages were determined for each fish, using the break-and-burn technique [24]. Under reflected light, one year was defined as an opaque zone (summer growth) followed by a translucent hyaline zone (winter growth).

Bulk zooplankton samples were collected from each lake and from the marine environment of Melville Sound with multiple surface tows. A 40- to 200-μm mesh net with a 0.5-m diameter opening was used. Samples were frozen in Nasco Whirl-Pak® bags within 1 d of collection.

Mercury and stable isotope analyses

Fish muscle tissues and bulk zooplankton samples were freeze-dried and homogenized. All samples were analyzed for stable isotope ratios of carbon (C; $\delta^{13}C$) and nitrogen (N; $\delta^{15}N$). Fish samples were analyzed for concentrations of total Hg, and bulk zooplankton samples from lakes were analyzed for concentrations of methyl Hg. Unfortunately, low sample masses of marine zooplankton precluded Hg analysis.

Total Hg concentrations were determined in fish tissues with Milestone DMA-80 Direct Mercury Analyzers (see Gantner et al. [7] and Swanson and Kidd [20] for complete methods) at the National Water Research Institute (NWRI, Burlington, ON, Canada), and the University of New Brunswick (Saint John, NB, Canada). Quality assurance/quality control procedures and

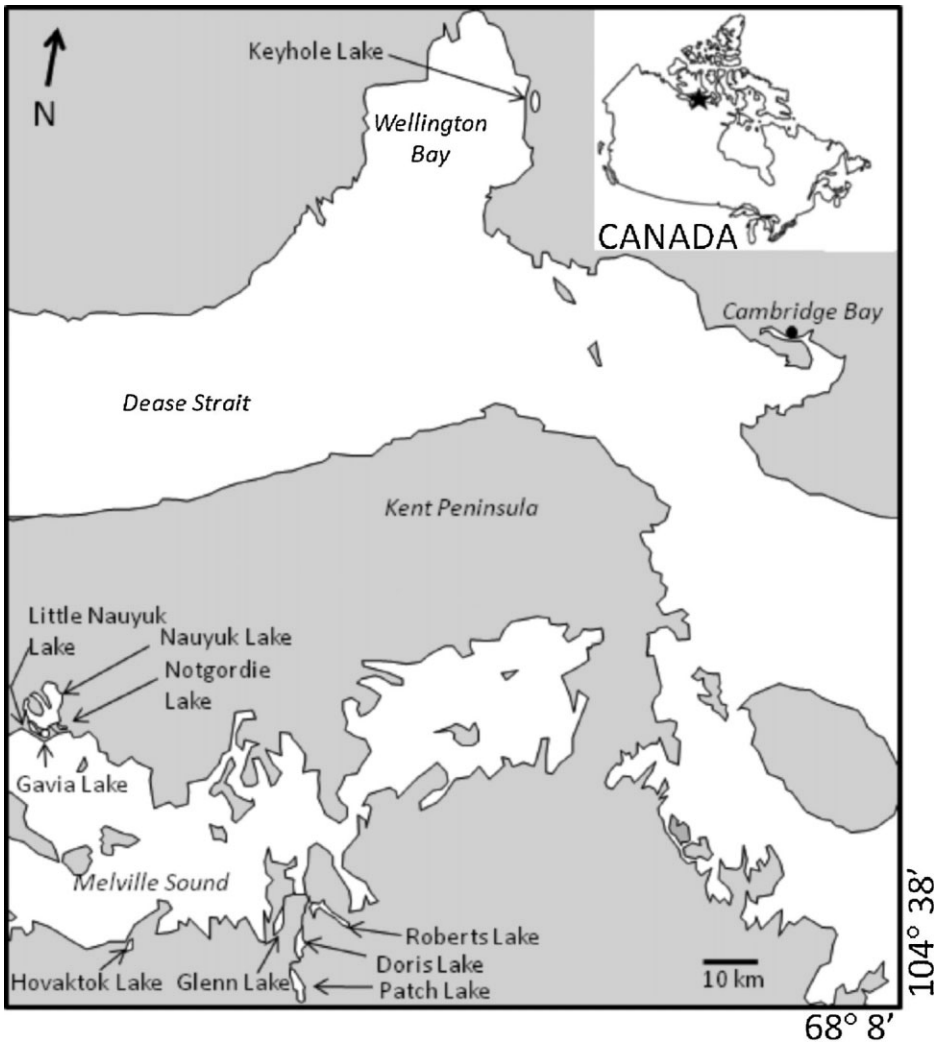

Fig. 1. Map of the study lakes. Lakes were sampled during the open water seasons of 2006 to 2008. Landlocked Arctic char were captured in Little Nauyuk, Notgordie, Gavia, and Keyhole lakes, landlocked lake trout were captured in Doris and Patch lakes, anadromous and resident Arctic char were captured in Nauyuk, Hovaktok, and Roberts lakes, and anadromous and resident lake trout were captured in Nauyuk, Hovaktok, Roberts, and Glenn lakes.

results are presented in the Supplemental Data section. All total Hg concentrations for fish are reported in μg/g wet weight.

Methylmercury concentrations [MeHg] in zooplankton samples collected from landlocked Arctic char lakes (Keyhole, Gavia, Little Nauyuk, Notgordie) were analyzed by acid leaching and solvent extraction followed by propylation with purge and trap, thermal desorption, and gas chromatography–cold vapor atomic fluorescence spectroscopy detection at NWRI (see Gantner et al. [8] for details). Methylmercury concentrations in zooplankton collected from Doris and Patch lakes and from all lakes with access to the sea were determined by Hg speciation analyses at Quicksilver Scientific (Lafayette, CO, USA) using Hg-Thiourea complex liquid chromatography–cold vapor atomic fluoresence spectrometry [25]. The quality assurance/quality control information for both laboratories is available in the Supplemental Data section. All MeHg concentrations for zooplankton are presented in ng/g dry weight.

Table 1. Fish species and life-history types captured in each of the 10 study lakes

Lake	Marine access?	Species present	Life histories present[a]
Doris	No	Lake trout	L
Patch	No	Lake trout	L
Keyhole	No	Arctic char	L
Little Nauyuk	No	Arctic char	L
Notgordie	No	Arctic char	L
Gavia	No	Arctic char	L
Glenn	Yes	Lake trout	A and R
Hovaktok	Yes	Arctic char Lake trout	A and R (both species)
Nauyuk	Yes	Arctic char Lake trout	A and R (both species)
Roberts	Yes	Arctic char Lake trout	A and R (both species)

[a] L, landlocked; R, resident; A, anadromous.

Stable C and N isotope analyses were performed at the Environmental Isotope Laboratory (University of Waterloo, Waterloo, ON, Canada) and the Stable Isotopes in Nature Laboratory (University of New Brunswick, Fredericton, NB, Canada). Details are provided in Gantner et al. [7] and Swanson et al. [19]. Briefly, samples were analyzed using continuous-flow isotope-ratio mass spectrometers connected to elemental analyzers. Carbon and nitrogen were analyzed simultaneously with the elemental analyzer and converted to molar ratios (C:N).

Stable C and N isotope ratios are expressed as parts per mil (‰) delta values ($\delta^{13}C$, $\delta^{15}N$) from Pee-Dee Belemnite and N_2 gas, respectively. Delta values were calculated using the standard formula (Eqn. 1)

$$\delta^{15}\text{N or } \delta^{13}\text{C} = [(R_{\text{sample}} - R_{\text{standard}})/R_{\text{standard}}] \cdot 1,000 \quad (1)$$

where $R = {}^{15}N/{}^{14}N$ or ${}^{13}C/{}^{12}C$. Quality assurance/quality control data are presented in the Supplemental Data section.

Data analysis

To correct for among-lake differences in baseline $\delta^{15}N$ [26,27], we used a modified version of the model proposed by Cabana and Rasmussen [26]. Zooplankton was the only invertebrate taxa that could be captured in all lakes and in the marine environment of Melville Sound; thus, zooplankton was chosen to represent the baseline. We assumed that resident and landlocked fish were composed entirely of freshwater-derived nutrients, and calculated adjusted $\delta^{15}N$ ratios accordingly (Eqn. 2)

$$\text{adjusted } \delta^{15}\text{N } (\delta^{15}\text{N}_{\text{adj}}) = \delta^{15}\text{N}_{\text{fish}} - \delta^{15}\text{N}_{\text{lake zooplankton}} \quad (2)$$

Results from isotope mixing models show that anadromous Arctic char from these lakes are composed of approximately 93% marine-derived nutrients, whereas anadromous lake trout are composed of approximately 67% marine-derived nutrients (Swanson et al., unpublished data). Adjusted $\delta^{15}N$ for anadromous fish were calculated using these estimates (Eqns. 3,4).

$$\delta^{15}\text{N}_{\text{adj}} \text{ (Arctic char)} = \delta^{15}\text{N}_{\text{fish}} - (0.93 \cdot \delta^{15}\text{N}_{\text{marine zooplankton}} + 0.07 \cdot \delta^{15}\text{N}_{\text{lake zooplankton}}) \quad (3)$$

$$\delta^{15}\text{N}_{\text{adj}} \text{ (lake trout)} = \delta^{15}\text{N}_{\text{fish}} - (0.67 * \delta^{15}\text{N}_{\text{marine zooplankton}} + 0.33 \cdot \delta^{15}\text{N}_{\text{lake zooplankton}}) \quad (4)$$

Variation in lipid content among fish has the potential to confound analyses of $\delta^{13}C$ because lipids are relatively depleted in the heavier isotope [23]. Because fish $\delta^{13}C$ ratios were not significantly related to C:N ratios, however (linear regression, $t = 0.87$, $p = 0.39$, df (degrees of freedom) = 1,271), $\delta^{13}C$ ratios were left unadjusted.

Fish condition was estimated using Fulton's condition factor [28] (Eqn. 5).

$$\text{Condition} = W \times 100/L^3 \quad (5)$$

where W = weight and L = length. All statistical analyses were performed with SAS version 9.1.3 [29]. Variables were log-transformed as required, and homogeneity of variance was assessed by examining residual plots after each analysis. Testing of residuals was achieved with Kolmogorov-Smirnov and Shapiro-Wilk statistics. Alpha was set at 0.05.

Total [Hg] was compared among species and life history types with a mixed model that included species, life history, and fork length as fixed effects and lake as a random effect. Lake was included as a random variable because we sampled only a subset of lakes of interest and because some study lakes contained two fish life history types whereas others contained only one fish life history type. Least-squares means (LSM) were calculated at a standardized fork length of 500 mm (this length was within the captured size range for all life history types; no extrapolation was necessary) and compared among species and life history types with post-hoc Tukey's tests.

The best predictive covariates of [Hg] were determined with a stepwise multiple regression that related [Hg] to fork length, age, $\delta^{13}C$, $\delta^{15}N_{adj,}$ C:N ratio, and condition. Data from both species and all life history types were included in this regression. We then investigated which covariates best explained differences in [Hg] among species and life history types. Because [Hg] was compared among species and life history types at a standardized fork length of 500 mm, variation attributable to fork length was removed by computing residuals of the [Hg]–fork length relationship. Residuals were then related to age, $\delta^{13}C$, $\delta^{15}N_{adj,}$ C:N ratio, and condition in a stepwise multiple regression. Significant covariates were compared among species and life history types with mixed models. If significant differences were found, LSMs for covariates were calculated at a fork length of 500 mm and compared among species and life history types with post-hoc Tukey's tests.

Our study lakes could be placed into one of two categories, marine access (Glenn, Hovaktok, Nauyuk, Roberts) or landlocked (Gavia, Notgordie, Little Nauyuk, Keyhole, Doris, Patch). To determine whether differences in [Hg] among life history types was related to [MeHg] at the base of the food chain or to lake characteristics that may affect Hg methylation, we used a t test to compare [MeHg] in zooplankton, %MeHg (of total Hg) in zooplankton, lake surface area (data from Ganter et al. [8] and Miramar Hope Bay [30]; ftp://ftp.nirb.ca/02-REVIEWS/COMPLETED%20REVIEWS/02MN134-DORIS%20NORTH%20GOLD%20MINE%202004/118%20Final%20EIS/), and concentrations of chlorophyll *a* (data taken from Gantner et al. [8] and Miramar Hope Bay [30]) between lakes with and without marine access.

RESULTS

Differences in [Hg] among life history types and species

Mercury concentrations in all Arctic char analyzed ranged from 0.01 to 0.51 μg/g, and overall arithmetic mean [Hg] (±SD) in Arctic char (not corrected for size) was 0.07 ± 0.08 μg/g. Mercury concentrations varied significantly among life history types (mixed model, $F = 4.76$, $p < 0.05$, $df = 2,116$). At a standardized fork length of 500 mm, resident and anadromous Arctic char had significantly lower [Hg] (each 0.04 μg/g) than landlocked Arctic char (0.19 μg/g) (Tukey's test, $p < 0.05$; Fig. 2). No significant difference was seen in [Hg] between anadromous and resident Arctic char (Tukey's test, $p > 0.05$; Fig. 2). Although the interaction between life history type and fork length was significant (mixed model, $F = 6.84$, $p < 0.05$, $df = 2,116$), the pattern of pairwise differences in [Hg] was generally consistent across the range of captured body sizes (Supplemental Data, Fig. S1).

Mercury concentrations in all lake trout analyzed ranged from 0.013 to 1.40 μg/g, and overall arithmetic mean [Hg]

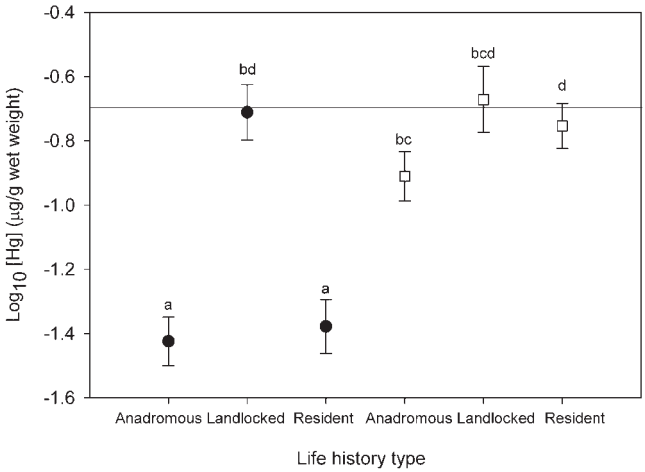

Fig. 2. The LSM $\log_{10}$[Hg] ± standard error for each species (Arctic char, black circles; lake trout, white squares)–life history type at a standardized size of 500 mm fork length. The reference line represents the Canadian subsistence consumption guideline (0.2 µg/g). Letters indicate pairwise differences between all species–life history types. Landlocked Arctic char had significantly higher [Hg] than resident or anadromous Arctic char (Tukey's test, $p < 0.05$). Resident lake trout had significantly higher [Hg] than anadromous lake trout, but no significant difference was seen between landlocked lake trout and resident lake trout. Within life history types, Arctic char had lower [Hg] than lake trout.

(±SD, not corrected for size) in lake trout was 0.23 ± 0.23 µg/g. Mercury concentrations differed significantly among life history types of lake trout (mixed model, $F = 9.75$, $p < 0.05$, $df = 2,155$). At a standardized fork length of 500 mm, anadromous lake trout had significantly lower [Hg] (0.12 µg/g) than resident lake trout (0.18 µg/g) (Fig. 2; Tukey's test, $p < 0.05$). Unlike Arctic char, no significant differences in [Hg] were seen between landlocked lake trout (0.21 µg/g) and either anadromous or resident lake trout (Fig. 2; Tukey's test, $p > 0.05$). Differences in [Hg] among lake trout life history types were consistent across the range of captured fish sizes; the interaction between life history type and fork length was not significant for lake trout (mixed model, $F = 1.08$, $p > 0.05$, $df = 2,155$) (Supplemental Data, Fig. S2).

With all life history types included in a mixed model, lake trout had significantly higher [Hg] than Arctic char ($F = 55.58$, $p < 0.05$, $df = 1,273$; Fig. 2). At a standardized fork length of 500 mm, LSM [Hg] was 0.09 µg/g for Arctic char and 0.17 µg/g for lake trout. A significant interaction was seen between species and life history type (mixed model, $F = 7.96$, $p < 0.05$, $df = 2,273$), however, which is evident in Figure 2. Whereas anadromous and resident Arctic char had significantly lower [Hg] than all life history types of lake trout (Tukey's test, $p < 0.05$; Fig. 2), landlocked Arctic char had very similar [Hg] to lake trout (Tukey's test, $p > 0.05$; Fig. 2).

Covariates of [Hg]

Both fish age and C:N were significantly related to [Hg]. All other covariates, including δ^{15}N, δ^{13}C, condition, and length, were not significantly related to [Hg] (stepwise multiple regression, $F < 3.02$, $p > 0.05$, $df = 5,248$). The relationship between [Hg] and age was positive (Fig. 3A) and significant ($F = 590$, $p < 0.05$, $df = 2,248$) and with all data in a stepwise multiple regression, age explained 70% of the variation in [Hg]. The relationship between [Hg] and C:N was negative (Fig. 3B) and significant (stepwise multiple regression, $F = 49.13$, $p < 0.0001$, $df = 2,248$), but C:N explained only 3% of the variation in [Hg].

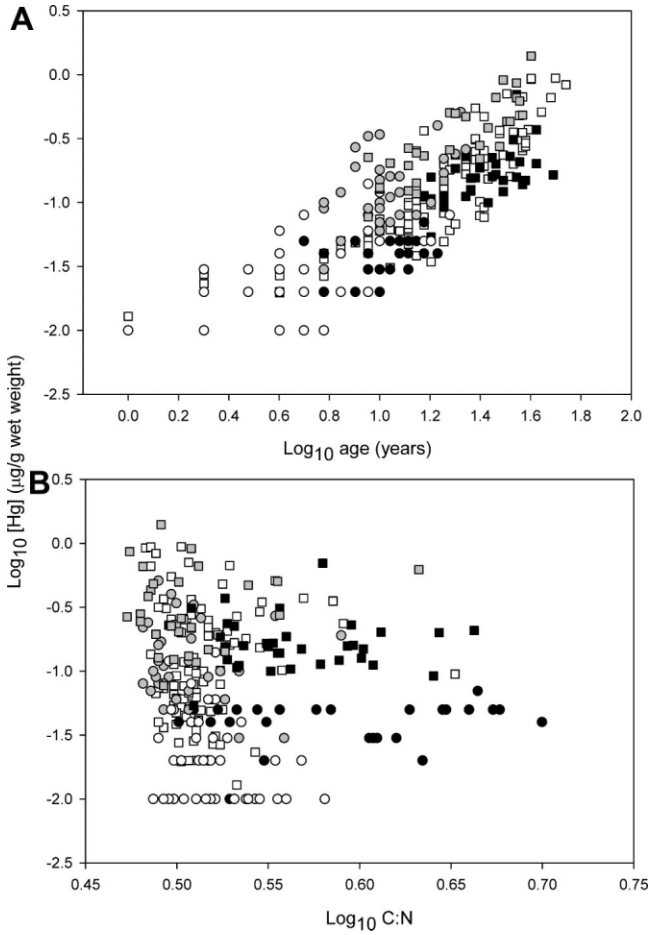

Fig. 3. Scatterplots of $\log_{10}$[Hg] $-\log_{10}$age (**A**) and $\log_{10}$[Hg] $-\log_{10}$C:N (**B**). Circles represent Arctic char, squares represent lake trout, black represents anadromous fishes, gray indicates landlocked fishes, and white indicates resident fishes. With all data in a stepwise multiple regression, the relationship between $\log_{10}$[Hg] and $\log_{10}$ age was significant ($F = 590$, $p < 0.0001$, degrees of freedom = 2,248) and positive, and the relationship between [Hg] and C:N significant ($F = 49.13$, $p < 0.0001$, $df = 3,248$) and negative.

Age and C:N appeared to explain the pattern of [Hg] observed in the six species–life history types. Because comparisons of [Hg] were performed at a standardized fork length of 500 mm, we removed variation attributable to fork length before investigating which covariates may explain [Hg] differences. A stepwise multiple regression performed on residuals of the [Hg]–fork length relationship showed that age and C:N were the only significant covariates ($F > 54.3$, $p < 0.05$, $df > 3,248$). Age and C:N each explained 18% of variation, and, similar to earlier analyses, [Hg] increased with age and decreased with C:N. Both age and C:N differed significantly among species and life history types (mixed model, $F > 5.10$, $p < 0.05$, $df > 1,250$). Arctic char were significantly younger (12 years) and had higher C:N (3.47) than lake trout (20 years and 3.34, respectively) at 500 mm fork length (Tukey's tests, $p < 0.05$). Within species, no significant differences were seen in age-at-size (Tukey's tests, $p > 0.05$; Fig. 4A), but in both species landlocked and resident fish had lower C:N than anadromous fish (Tukey's tests, $p < 0.05$, Fig. 4B).

Differences in LSM age and C:N among species and life history types (Fig. 4A, B) did not exactly match those observed for [Hg] (Fig. 2). That is, species–life history types with significantly higher [Hg] did not always have significantly older age-at-size and significantly lower C:N. Most patterns of LSM

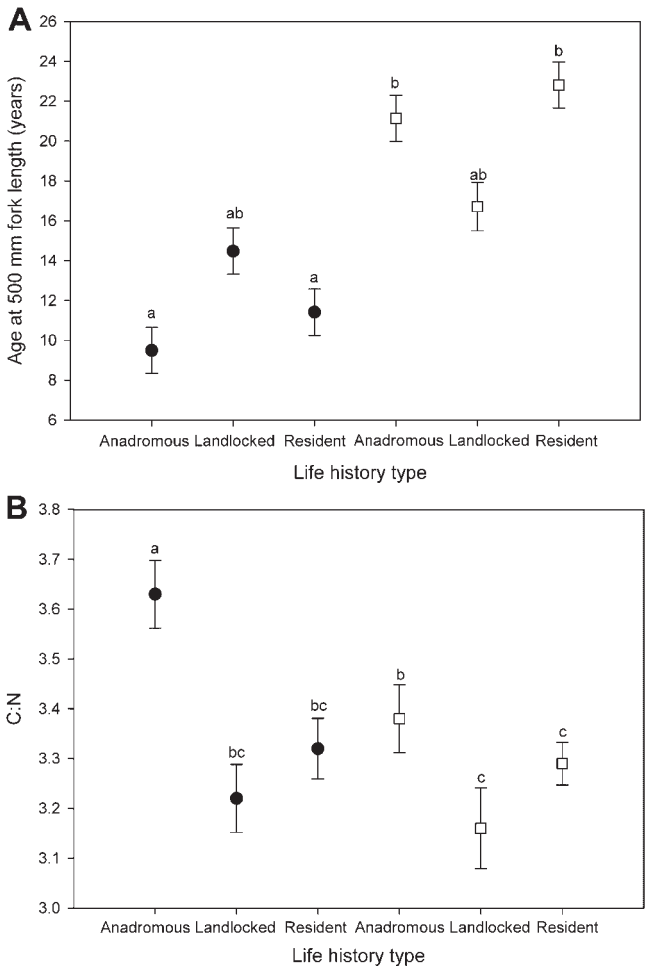

Fig. 4. The LSmean age (**A**) and C:N (**B**) ± standard error for each species (Arctic char, black circles; lake trout, white squares)–life history type at a standardized size of 500 mm fork length. Letters indicate pairwise differences between all species–life history types. Within species, age-at-size did not differ significantly between life history types (Tukey's test, $p > 0.05$). Arctic char were significantly younger than lake trout at 500 mm fork length, however (Tukey's test, $p < 0.05$). Anadromous Arctic char had significantly higher C:N than all other species–life history types, and anadromous lake trout had significantly higher C:N than landlocked or resident lake trout (Tukey's test, $p < 0.05$).

age and C:N were consistent with those of [Hg], however. For example, anadromous and resident Arctic char had the lowest [Hg] and also the youngest age at 500 mm fork length (Figs. 2, 4A). Anadromous Arctic char also had the highest C:N, which may contribute to relatively lower [Hg]. Arctic char had significantly lower [Hg] (see earlier discussion) and were significantly younger (12 years) than lake trout (20 years) at 500 mm fork length (Tukey's test, $p < 0.05$). Also, landlocked Arctic char and lake trout, which had generally high [Hg], had low C:N (Figs. 2, 4A, 4B).

Whole-lake variables and [MeHg] at the base of the food chain did not appear to explain any of the differences in [Hg] among species-life history types. Two possible covariates of Hg methylation, lake surface area and concentrations of chlorophyll *a,* did not differ significantly between lakes with and without access to the marine environment (*t* test, $t \leq 1.12$, $p \geq 0.34$, $df \geq 3.15$) (Table 2). Similarly, [MeHg] in zooplankton and percent MeHg in zooplankton (of total Hg) did not differ between lake types (landlocked vs marine access) (*t* test, $t < 0.87$, $p > 0.41$, $df = 8$) (Table 2).

Delta$^{15}N_{adj}$ is commonly a good predictor of [Hg], but we found that [Hg] was not significantly related to $\delta^{15}N_{adj}$. We compared $\delta^{15}N_{adj}$ among species–life history types to further investigate the lack of relationship. Delta$^{15}N_{adj}$ differed significantly among life history types (mixed model, $F = 14.3$, $p < 0.05$, $df = 2{,}260$), but did not differ significantly between species (mixed model, $p > 0.05$, $df = 1{,}260$). At 500 mm fork length, $\delta^{15}N_{adj}$ was highest in anadromous fish (~10‰) and significantly lower in resident and landlocked fish (~4–5‰; Tukey's test, $p < 0.05$, Supplemental Data, Fig. S3). This trend is the opposite of what would be expected if [Hg] increased with $\delta^{15}N_{adj}$.

Exceedances of [Hg] guidelines

Of the 290 fish analyzed in the current study, 75 (26%) exceeded the Canadian [Hg] guideline for subsistence consumption (0.2 μg/g Hg wet wt). This guideline is consistent with that of the World Health Organization. No anadromous or resident Arctic char exceeded these guidelines; all eight Arctic char exceedances were landlocked fish. Of the 67 lake trout with [Hg] greater than 0.2 μg/g, six were anadromous, 22 were landlocked, and 33 were resident. Arctic char with [Hg] greater than 0.2 μg/g ranged from 400 to 620 mm fork length, whereas lake trout with [Hg] greater than 0.2 μg/g ranged from 350 to 880 mm fork length.

Table 2. Concentrations of chlorophyll *a* and methylmercury (MeHg) in zooplankton in each lake, as well as lake area and %MeHg in zooplankton[a]

	Lake	Area (km^2)	Chlorophyll *a* (μg/L)	Mean [MeHg] in zooplankton (ng/g dry wt)	SD[b] [MeHg] in zooplankton	% MeHg in zooplankton
No marine access	Doris	3.5[c]	8.1[c]	16.2	0.28	58.5
	Gavia	0.2[d]	0.3[d]	3.00	NA[e]	13.6
	Keyhole	0.4[d]	1.1[d]	56.0	NA	42.0
	Little Nauyuk	0.5[d]	0.1[d]	4.00	2.83	11.3
	Notgordie	0.1[d]	0.6[d]	35.0	35.0	39.0
	Patch	5.8[c]	0.9[c]	1.42	2.12	30.0
Marine access	Glenn	2.2[c]	1.8[c]	7.05	0.35	30.0
	Hovaktok	1.7[c]	NA	21.5	1.49	39.5
	Nauyuk	27[c]	1.1[c]	3.75	0.21	18.0
	Roberts	3.8[c]	4.0[c]	4.05	0.21	50.5

[a] No significant differences were seen in any of these variables between lakes with and without marine access.
[b] Standard deviation.
[c] Data taken from Miramar Hope Bay Ltd. 2006.
[d] Data taken from Gyselman and Mohr [36].
[e] Not available.

DISCUSSION

Comparisons among species and life histories

This is the first study to make intraregional comparisons of [Hg] and covariates among anadromous, resident, and landlocked life history types of Arctic char and lake trout. Mercury concentrations were highest in lake trout and landlocked Arctic char and lowest in resident and anadromous Arctic char. We found that landlocked Arctic char had significantly higher [Hg] than resident Arctic char. In contrast, landlocked and resident lake trout had similar [Hg]. Consistent with previous studies, we also found that anadromous and resident Arctic char had significantly lower [Hg] than landlocked Arctic char [4,5,21], anadromous lake trout had significantly lower [Hg] than resident lake trout [20], and Arctic char had significantly lower [Hg] than lake trout [4,5,20].

High [Hg] in landlocked Arctic char did not appear to reflect higher Hg methylation or higher [Hg] at the base of the food chain. Despite having different life histories, anadromous and resident Arctic char had similar, lower [Hg] compared with landlocked Arctic char. Our analyses indicated that lake-scale effects, such as increased Hg methylation or increased [Hg] at the base of the food chain, did not seem to contribute to these differences. Comparisons of lake surface area, chlorophyll *a*, [MeHg] in zooplankton, and %MeHg in zooplankton between lakes with and without access to the marine environment yielded no significant differences. Because our analysis included only two possible covariates of Hg methylation, future studies should ideally include a greater number of lakes and comparisons of temperature, pH, and dissolved organic carbon. Percent wetland in the watersheds also should be assessed, because this can affect [MeHg] in lakes [31].

Fish age and C:N were the best predictors of [Hg] and best explained differences in LSM [Hg] among species–life history types. Before the effect of fork length was removed, fish age and C:N were the only significant covariates of [Hg], and age was a much better predictor than C:N. In a previous study that compared lake trout [Hg] between the Mackenzie River basin and more southerly locations, age predicted lake trout [Hg] better than length [32]. Age also partially explained interregional differences in [Hg]. Similarly, Swanson and Kidd [20] found that age explained 72% of variation in fish [Hg], and age explained many of the differences in [Hg] among species–life history types of Arctic char and lake trout.

Fork length, $\delta^{15}N$, $\delta^{13}C$, and condition were not significantly related to [Hg]. Many previous studies have found significant positive relationships between [Hg] and $\delta^{15}N$ [3,6–8]. For example, in a recent investigation of 27 landlocked Arctic char populations, Gantner et al. [7] reported that [Hg] was significantly related to $\delta^{15}N$ in 71% of studied populations. In contrast, we found that [Hg] was not significantly related to $\delta^{15}N_{adj}$, largely because anadromous fish had high $\delta^{15}N_{adj}$ but low [Hg]. De Freitas et al. [33] and Huckabee et al. [34] asserted that growth and longevity can be as important in explaining fish [Hg] as biomagnification from prey, and this appears to be true in our study.

Age-at-size is a proxy for growth rate, and negative relationships between fish [Hg] and growth rate are well documented. Fish with faster growth rates experience lower cumulative Hg uptake, because less time is required to reach a given size [35]. Also, fish with faster growth rates tend to have higher growth efficiency, and at a given food and contaminant intake, fish that grow more efficiently have lower metabolic costs and produce more flesh to dilute their Hg burden [15].

Although C:N was a weaker predictor of [Hg] than age before the effect of fork length was removed, the two variables explained approximately equal amounts of variation after the effect of fork length was removed. We found a negative relationship between [Hg] and C:N; life history types with higher C:N tended to have lower [Hg]. This is consistent with previous research showing that Hg binds to proteins rather than lipid, and that fish with higher lipid have lower [Hg] [14,20]. The range in C:N that we observed represented a 60-fold range in lipid content in muscle tissue, from approximately 0.2% to approximately 12% (H. Swanson, unpublished data).

Some of the pairwise differences in [Hg] that we observed were best explained by age-at-size, whereas others were best explained by C:N or a combination of the two variables. Consistent with previous comparisons [4,5,20], we found that Arctic char had significantly lower [Hg] than lake trout. Although Arctic char were both significantly younger and had significantly higher C:N at the standardized size than lake trout, qualitative analyses suggested that age-at-size may better explain this interspecies difference. Not all pairwise differences among species–life history types were significant, but whereas all life history types of Arctic char were younger-at-size than lake trout, only anadromous Arctic char had higher C:N than lake trout. Evans et al. [4] speculated that slower growth rates or warmer lake temperatures (stimulating Hg methylation) may lead to higher [Hg] in lake trout relative to Arctic char. Unlike most previous investigations, in which study lakes with lake trout were located further south than those with Arctic char, our study included four lakes with sympatric populations of Arctic char and lake trout. Thus, warmer lake temperatures do not explain higher [Hg] in lake trout in our study; slower growth rates and lower lipid are the best explanations.

Higher [Hg] in landlocked Arctic char relative to other life history types appeared to result from a combination of older age-at-size and lower C:N, whereas higher [Hg] in resident lake trout relative to anadromous lake trout appeared to be primarily driven by lower C:N. Although we found that anadromous lake trout were younger than resident lake trout at 500 mm fork length, the difference was not significant. Anadromous lake trout had significantly higher C:N than resident lake trout, however. This may result from reduced foraging costs, better food quality, or greater food abundance for anadromous lake trout, but further study is required. Whatever the cause of higher C:N in anadromous lake trout, higher lipid content appears to result in lower [Hg].

Mercury concentrations in landlocked lake trout require further study. Only two lakes were sampled for landlocked lake trout, and this may have affected results. Despite the fact that landlocked lake trout had significantly lower C:N than anadromous lake trout, no significant differences were seen in [Hg]. This appeared to result in part from higher uncertainty in the LSM estimate for landlocked lake trout [Hg]. It also may reflect the fact that landlocked lake trout were growing more quickly (although the difference was not significant) than both anadromous and resident lake trout; this warrants further investigation.

Application to human health

Arctic char and lake trout can represent up to 35% of human Hg intake in the Canadian Arctic, and Arctic char and lake trout are the second most frequently consumed traditionally harvested food (after caribou) in most Inuit regions of Canada [10]. Our results suggest that in areas of the Canadian Arctic where consumption of fish is a major route of Hg exposure for

humans, human Hg exposure may be reduced by targeting anadromous fish when possible and targeting Arctic char when possible. A thorough dietary survey that determines absolute and relative harvest levels of life history types is necessary to achieve full application of our results to human consumption. Results of the present study demonstrate that intraspecific differences in life history should be taken into account when developing region- and area-specific fish consumption guidelines.

CONCLUSIONS

We found that [Hg] varied among species and life history types of Arctic char and lake trout. Significant differences in [Hg] among species–life history types did not appear to be related to trophic level and were best explained by differences in age-at-size (i.e., growth rate) and C:N ratios. Although we focused on two species found in the Canadian Arctic, we suggest that [Hg] may vary similarly within any species that displays plasticity in life history strategy, especially when this plasticity results in differences in growth rate or proximate composition. We further suggest that when modeling [Hg] in slow-growing Arctic populations of fish, age is likely a much more useful predictor variable than length.

SUPPLEMENTAL DATA

Material and Methods—additional QC/QA.

Table S1. Descriptive statistics.

Figure S1. Log_{10} [Hg] vs. log_{10} fork length for anadromous, landlocked, and resident Arctic char. (93 KB DOC)

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