

Mercury Concentrations in Arctic Food Fishes Reflect the Presence of Anadromous Arctic Charr (*Salvelinus alpinus*), Species, and Life History

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Single-spawning (semelparous) anadromous fishes are known to transport contaminants from marine to freshwater habitats, but little research has been conducted on contaminant biotransport by multiple-spawning (iteroparous) anadromous fishes. We examined the effect of iteroparous, anadromous Arctic charr (*Salvelinus alpinus*) on mercury concentrations ([Hg]) in freshwater biota and compared [Hg] between species and life history types of Arctic charr and lake trout (*Salvelinus namaycush*). Data from six lakes and one coastal marine area in the Arctic territory of Nunavut, Canada, indicated that 1) lake trout had significantly lower [Hg] in lakes where anadromous Arctic charr were present; 2) [Hg] was significantly lower in recently discovered anadromous lake trout than in resident lake trout; and 3) regardless of life history, Arctic charr had significantly lower [Hg] than lake trout. These differences were explained by fish condition, age-at-size, and C:N. Biomagnification of Hg, measured as $\log_{10}[\text{Hg}] - \delta^{15}\text{N}$ slopes, did not differ between lakes with and without anadromous Arctic charr but was significantly higher in freshwater food webs (~ 0.2) than in the marine food web (0.08). Some biomagnification estimates were affected by correction for fish age and size. In contrast to semelparous anadromous species, biotransport of Hg by anadromous Arctic charr appears to be offset by increased growth of freshwater fishes.

Introduction

Mercury (Hg) is a neurotoxicant and contaminant of concern for both ecological and human health reasons. The organic form of Hg, methyl mercury (MeHg), is toxic and can bioaccumulate through aquatic and marine food webs to reach high concentrations in predatory fishes (e.g. refs 1–3). Much research has been devoted to elucidating factors that predict Hg concentrations ([Hg]) in fishes. This is particularly important in areas where fish [Hg] exceed Canadian guidelines for commercial sale (0.5 $\mu\text{g}/\text{g}$ wet weight (ww)) and subsistence consumption (0.2 $\mu\text{g}/\text{g}$ ww) (2, 3) and where human exposure to fish-derived Hg is high, such as in Arctic communities with subsistence fisheries for Arctic charr (*Salvelinus alpinus*) and lake trout (*Salvelinus namaycush*).

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Most Hg ($\geq 95\%$) in fish is in the form of methylmercury (e.g. ref 4); thus, concentrations of total Hg ([THg]) and methyl Hg ([MeHg]) in fish are often reported interchangeably.

Atmospheric deposition of anthropogenically derived Hg is the most important source of Hg to most Arctic lakes (5). Mercury may also be transferred between ecosystems by migratory animals (i.e., biotransport), however, and anadromous populations of Arctic charr have been identified as a potential biotransport vector for contaminants in the Canadian Arctic (6). Unlike other anadromous fishes that transport contaminants from marine environments to freshwater lakes (e.g., *Oncorhynchus* spp. (7)), Arctic charr are iteroparous and do not die immediately after spawning. This may limit their efficiency as a biotransport vector (6), but further research is required.

In addition to loading, many biological and life history characteristics affect [Hg] in fish. Because MeHg bioaccumulates and biomagnifies, concentrations in fish tend to increase with fish age, size, and $\delta^{15}\text{N}$ (indicator of trophic position) (e.g. refs 8 and 9). While older, larger fish tend to have higher [Hg], fish [Hg] usually (but not always) decrease with faster growth and greater body condition (e.g. refs 10 and 11). This is called growth dilution and often means that fish with younger age-at-size and higher condition have lower [Hg]. Because anadromous fishes often grow faster than resident (freshwater-only) fishes, the former often have relatively lower [Hg] (e.g. ref 2).

Perhaps due to low carrying capacities in Arctic lakes, Arctic charr and lake trout do not usually occur in sympatry if the Arctic charr population is landlocked (12). They do occur in sympatry, however, if the Arctic charr population is partially anadromous (i.e., some individuals are anadromous whereas others are freshwater residents (12)). In an earlier study that compared ecology of lakes with and without partially anadromous Arctic charr, we found that resident lake trout had significantly higher C:N (indicator of lipid (13)) and condition factors in lakes where anadromous Arctic charr were present (14). Thus, there are two possible competing effects of anadromous Arctic charr on resident lake trout [Hg] in Arctic lakes: 1) increased Hg loading due to biotransport and 2) increased growth.

Predictions of fish [Hg] often rely on models of biomagnification through food webs. This can be evaluated by calculating regression slopes between $\log[\text{Hg}]$ and $\delta^{15}\text{N}$. Slopes are usually significant and positive when applied to fish communities or whole food webs (e.g. refs 1 and 15) and are surprisingly consistent among tropical, temperate, Arctic, marine, and freshwater systems (usually ~ 0.2) (1, 16, 17). Slope calculations do not often account for organism size or age, however, and because these variables affect both [Hg] and $\delta^{15}\text{N}$ (9, 18, 19), previous comparisons of $\log_{10}[\text{Hg}] - \delta^{15}\text{N}$ slopes among systems may be confounded by differences in fish size and/or age.

Using six lake food webs and one coastal marine food web in the Arctic territory of Nunavut, Canada, we compared 1) [Hg] in resident lake trout and forage fishes between lakes with and without partially anadromous Arctic charr; 2) [Hg] in Arctic charr and lake trout between species and life history types (anadromous versus resident); 3) slopes of $\log_{10}[\text{Hg}]$ vs $\delta^{15}\text{N}$ using different (or no) standardization techniques for fish age and/or size; and 4) [Hg] in traditional food fishes to human consumption guidelines. Results are condensed into practical considerations that could decrease fish-derived Hg intake in subsistence fishers.

Methods

Sample Collection. During summers 2006–2008, Arctic charr, lake trout, forage fishes (cisco (*Coregonus artedii*), lake whitefish (*Coregonus clupeaformis*), ninespine stickleback (*Pungitius pungitius*), fourhorn sculpin (*Myoxocephalus quadricornis*), Pacific herring (*Clupea pallasii*), and capelin (*Malloctus villosus*)), benthic invertebrates (isopods (*Saduria entomon*), opossum shrimp (*Mysis relicta*), amphipods (*Gammarus lacustris* and *Hyallela azteca*), chironomids (Chironomidae)), and zooplankton were collected from six lakes and one coastal marine ecosystem in the West Kitikmeot region of Nunavut, Canada (~68°N and 107°W; Figure S1 and Table S1). Sample sizes are listed in Table S2. All lakes contained lake trout, forage fishes, and similar invertebrate taxa. The study was designed such that three lakes contained partially anadromous populations of Arctic charr (Roberts, Hovaktok, and Nauyuk), whereas three lakes contained no Arctic charr (Doris, Patch, Glenn; Figure S1 and Table S2). Midway through the study, however, we discovered anadromous lake trout in four of the study lakes (Roberts, Hovaktok, Nauyuk, and Glenn) (20). Interpretation of some analyses (e.g., testing for effects of anadromous fishes on [Hg] in freshwater biota) were therefore altered to include assessment of potential effects of anadromous lake trout. Anadromous and resident Arctic charr and lake trout were differentiated in a previous study using results from otolith microchemistry and stable carbon (C), nitrogen (N), and sulfur (S) isotope analysis (20).

Following capture, fish were measured for fork length (nearest mm) and weighed (nearest g) in the field, and all fishes except ninespine stickleback and fourhorn sculpin were dissected for skinless dorsal muscle tissue and sagittal otoliths. Analyses conducted on both muscle and whole bodies of ninespine stickleback and fourhorn sculpin revealed that [Hg] did not differ between tissue types (mixed model, $F < 3.03$, $P > 0.09$, $df > 1, 10$). Thus, whole bodies were used for the majority of analyses for these two species, and data from both tissue types were pooled. All tissue samples were frozen immediately after processing, and otoliths were cleaned and dried. Benthic invertebrate samples were kept in clean lake water for 12–24 h to purge gut contents before being sorted to major taxon and frozen whole. Bulk zooplankton samples were frozen within 12 h of collection.

Laboratory Analyses. Fish tissues and whole invertebrates were freeze-dried, ground to a fine powder, and subsampled for stable isotope and Hg analyses. When single invertebrates did not provide enough mass for analysis, composite samples of many individuals were used. Quality assurance/quality control procedures for analytical laboratories are described in the Supporting Information.

Stable C and N isotope analysis for fish and invertebrates were performed at the Stable Isotope in Nature Laboratory at the University of New Brunswick, Fredericton, NB (see ref 14 for details). Carbon and nitrogen content were converted to molar ratios (C:N), and stable C and N isotope ratios are expressed as parts per mil (‰) delta values ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$) relative to international standards (Pee-Dee Belemnite (C) and N_2 gas (N)).

Fish age determinations for lake trout and Arctic charr are described in ref 20. Total Hg concentrations were determined in dry fish samples with a direct mercury analyzer at the University of New Brunswick, Saint John, NB. MeHg concentrations were determined in dry invertebrate samples with Hg speciation analysis at Quicksilver Scientific, Lafayette, Colorado (21). Two to four samples each of amphipods, chironomids, isopods, and mysids (each lake and Melville Sound) as well as zooplankton (each lake) were analyzed for [MeHg]. Because accurate wet weight could not be determined for invertebrates in the field, invertebrate [MeHg] were analyzed and reported as $\mu\text{g/g}$ dry weight. Using a mean

moisture content of 80% (H. Swanson, unpublished data), fish Hg concentrations were converted to wet weight for most statistical analyses. All reported fish Hg concentrations represent total Hg (wet weight), except for calculations of Hg biomagnification through whole food webs (see below).

Data Analysis. Statistical analyses were performed with SAS version 9.1.3 (22). Fork length was used as a covariate in many statistical analyses. Whenever size-adjusted least-squared means (LSMEANS option (22)) were calculated, we used 650 mm fork length as the covariate level of comparison for lake trout and Arctic charr, 400 mm for lake whitefish, 270 mm for cisco, 100 mm for fourhorn sculpin, capelin, and Pacific herring, and 50 mm for ninespine stickleback. These sizes were within the size range captured in each lake; no extrapolation was necessary. Homogeneity of variance was assessed by examination of residuals plots and variables were transformed (\log_{10}) as required. Alpha was set at 0.05.

Condition was calculated for each fish as

$$K = W \times 100 / L^3 \quad (1)$$

where W = wet weight (g), and L = fork length (cm) (23).

Variation in lipid content among fishes may confound analyses of $\delta^{13}\text{C}$ because lipids are relatively depleted in the heavier isotope (13). We found a significant negative relationship between fish $\delta^{13}\text{C}$ and C:N (linear regression, $t = -7.87$, $P < 0.0001$, $df = 1,433$). Delta ^{13}C values were therefore adjusted for lipid content using C:N ($\delta^{13}\text{C}_{\text{adj}}$; see refs 13 and 14).

When $\delta^{15}\text{N}$ is compared among lakes, it is necessary to correct for differences at the base of the food web (e.g. ref 24). Delta ^{15}N ratios were adjusted for among-system differences in baseline ($\delta^{15}\text{N}_{\text{adj}}$) by subtracting system-specific $\delta^{15}\text{N}$ ratios for mysids (see the Supporting Information).

To assess whether the presence of anadromous Arctic charr affected [THg] in fish and [MeHg] in invertebrates, species- and taxa-specific [Hg] in resident lake trout, cisco, lake whitefish, ninespine stickleback, mysids, isopods, chironomids, and zooplankton were compared among lakes with ANCOVAs (fish; fork length covariate) and ANOVAs (invertebrates). Posthoc Tukey's tests were performed, and pairwise differences were evaluated in the context of whether anadromous Arctic charr were present (Hovaktok, Nauyuk, Roberts) or absent (Doris, Glenn, Patch). Pairwise differences were also interpreted in the context of whether any anadromous fishes were present (i.e., including anadromous lake trout). If pairwise differences indicated an effect of anadromous Arctic charr and/or anadromous lake trout, LSmeans were calculated at species-specific standardized sizes and compared between lake types (anadromous fish present or absent) with a t test. If the t test was significant for a fish species, [Hg] was related to $\delta^{15}\text{N}_{\text{adj}}$, $\delta^{13}\text{C}_{\text{adj}}$, C:N, age, and condition with stepwise multiple regression (maximum R^2 selection). Variance inflation factors and condition indices indicated low collinearity among independent variables (22). Significant covariates were compared among lakes with an ANCOVA or ANOVA, and pairwise differences were evaluated in the context of presence/absence of anadromous Arctic charr and/or anadromous lake trout. If there appeared to be an effect of anadromous fish, LSmeans of the covariates were then calculated and compared between lake types with a t test. This was done to determine what covariates may be associated with differences in [Hg] between lakes with and without anadromous Arctic charr and/or lake trout.

Mercury concentrations were compared between species and life history types of Arctic charr and lake trout with a mixed model; species, life history type, and fork length were fixed effects, whereas lake was a random effect. LSmeans were determined for each species-life history type at a standardized size and compared with a posthoc Tukey's test.

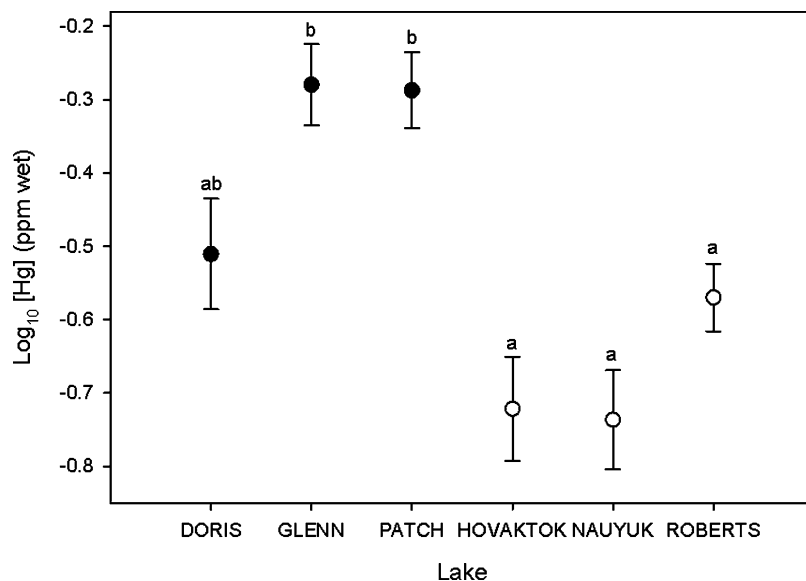


FIGURE 1. LSmean \pm SE [Hg] in lake trout (at 650 mm). Black symbols indicate lakes without anadromous Arctic charr, whereas white symbols indicate lakes with anadromous Arctic charr. Anadromous lake trout were present in Glenn, Hovaktok, Nauyuk, and Roberts lakes. [Hg] in resident lake trout differed significantly among lakes (ANCOVA, $F = 4.19$, $P = 0.0016$, $df = 5,115$), and both pairwise differences (indicated by letters; Tukey's test, $P < 0.05$) and a t test ($t = 3.42$, $P = 0.027$, $df = 4$) indicated that resident lake trout had significantly lower [Hg] in lakes with anadromous Arctic charr.

To determine what may be driving differences between species and life history types, [Hg] was related to age, C:N, $\delta^{15}\text{N}_{\text{adj}}$, $\delta^{13}\text{C}_{\text{adj}}$, and condition with stepwise multiple regression (maximum R^2 selection). When a variable was significant in the multiple regression, it was compared between species and life history types with a mixed model (as described above).

To determine if differences in [Hg] between life history types of Arctic charr and lake trout could be explained by differences in [Hg] of prey items, [Hg] in forage fishes and invertebrates were compared between marine and freshwater environments with a mixed model. For forage fishes, fork length and environment (marine or freshwater) were fixed effects in a mixed model, and species was a random effect. For invertebrates, species-specific [MeHg] for mysids, amphipods, and isopods were compared between marine and freshwater environments with t tests.

Biomagnification of Hg was assessed for each food web (defined as fish and invertebrates) by regressing dry [MeHg] (\log_{10} -transformed) against $\delta^{15}\text{N}$ with simple linear regression. Fish [THg] was converted to [MeHg] by multiplying by 0.95; this was the average proportion of MeHg in our fish samples (H. Swanson, unpublished data). For lakes, regressions included data from all invertebrate taxa, forage fishes, and resident Arctic charr and lake trout. The regression for Melville Sound (marine environment) included data from the three invertebrate taxa captured, capelin, Pacific herring, and all anadromous Arctic charr and lake trout; anadromous fish were primarily composed of marine-derived nutrients, regardless of whether capture occurred in freshwater or saltwater (H. Swanson, unpublished data).

Regressions were performed three ways. The first method did not account for effects of fish size or age on either [MeHg] or $\delta^{15}\text{N}$. For each lake, raw [MeHg] was regressed against raw $\delta^{15}\text{N}$ ratios with simple linear regression. The second method adjusted for fish size using LSmeans. LSmean [MeHg] and LSmean $\delta^{15}\text{N}$ ratios were generated at species-specific standardized sizes in each lake with ANCOVAs. Mean [MeHg] and mean $\delta^{15}\text{N}$ were calculated for each invertebrate taxa in each lake. LSmean and mean [MeHg] were then regressed against LSmean and mean $\delta^{15}\text{N}$ ratios with simple linear regression. The third method adjusted for fish age as well as size. Fish [MeHg] and $\delta^{15}\text{N}$ were regressed against fork length and age with multiple regressions performed for each lake.

Residual values of [MeHg] and $\delta^{15}\text{N}$ for each fish, along with raw values of [MeHg] and $\delta^{15}\text{N}$ for each invertebrate sample, were used in lake-specific linear regressions of [MeHg] against $\delta^{15}\text{N}$. Within each estimation method, regression slopes were compared among lakes with an ANCOVA. Slopes were also qualitatively compared among estimation methods.

Results and Discussion

Comparisons of [Hg] between Lakes with and without Anadromous Fishes. Previous studies have compared contaminant concentrations in freshwater fishes between lakes with and without semelparous, anadromous Pacific salmon species and have found higher contaminant concentrations in lakes where anadromous Pacific salmon are present (e.g. ref 7). In contrast, we found that resident lake trout had lower [Hg] in lakes where anadromous Arctic charr were present. Among-lake comparisons of resident lake trout [Hg] were significant (ANCOVA, $F = 4.19$, $P = 0.0016$, $df = 5,115$). The pattern of pairwise differences was consistent with a charr-induced effect (Tukey's test, $P < 0.05$, Figure 1), and a t test on lake-specific LSmeans indicated that resident lake trout had significantly lower [Hg] in lakes where anadromous Arctic charr were present ($t = 3.42$, $P = 0.027$, $df = 4$).

Resident lake trout [Hg] was significantly related to age (positive; $R^2_{\text{partial}} = 49\%$), $\delta^{15}\text{N}_{\text{adj}}$ (positive; $R^2_{\text{partial}} = 34\%$), $\delta^{13}\text{C}_{\text{adj}}$ (negative; $R^2_{\text{partial}} = 7\%$), and condition (negative; $R^2_{\text{partial}} = 4\%$) (stepwise multiple regression, $F \geq 4.90$, $P \leq 0.029$, $df = 4,121$). Age explained the most variation in resident lake trout [Hg] and varied significantly among lakes (ANCOVA, $F = 9.69$, $P < 0.0001$, $df = 1,121$), but LSmean age-at-size did not show a pattern with respect to the presence/absence of Arctic charr and, therefore, did not explain why lake trout [Hg] was lower in lakes with Arctic charr. LSmean age (at 650 mm) of lake trout in Doris, Glenn, and Patch lakes (charr absent) was 24, 37, and 26 years, respectively, whereas LSmean age of lake trout in Hovaktok, Nauyuk, and Roberts lakes (charr present) was 36, 38, and 28 years, respectively (SE was 1.05–1.07 years).

Resident lake trout were in significantly better condition in lakes where anadromous Arctic charr were present (14), and this likely explained their significantly lower [Hg]. Earlier comparisons of $\delta^{15}\text{N}_{\text{adj}}$, $\delta^{13}\text{C}_{\text{adj}}$, and condition among the study lakes showed that condition differed significantly between lakes with and without Arctic charr, whereas their trophic

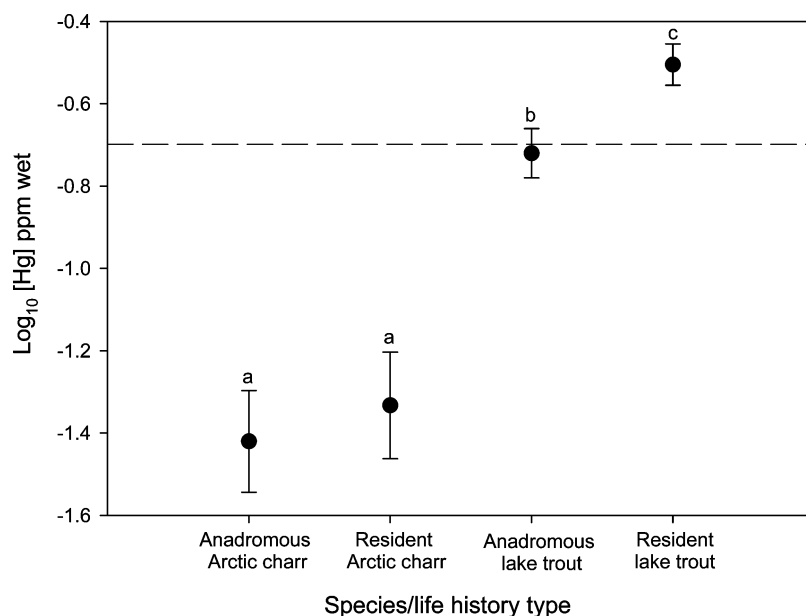


FIGURE 2. LSmean [Hg] \pm SE for each species-life history type at a fork length of 650 mm. Letters indicate pairwise differences. Regardless of life history type, Arctic charr had significantly lower [Hg] than lake trout (Tukey's test, $P < 0.05$). Anadromous fishes had lower [Hg] than resident fishes, but the difference was only significant for lake trout (Tukey's test, $P < 0.05$). The reference line indicates the Canadian subsistence consumption guideline for [Hg] in fish ($0.2 \mu\text{g/g}$).

position and carbon source ($\delta^{15}\text{N}_{\text{adj}}$ and $\delta^{13}\text{C}_{\text{adj}}$) did not (14). Resident lake trout were in significantly better condition (i.e., significantly heavier at a given length) in lakes with Arctic charr, perhaps because these lakes had greater prey availability or higher lake productivity (14). In agreement with previous studies, we found that the relationship between lake trout [Hg] and condition was negative (10, 25).

If resident lake trout were in significantly better condition in lakes with Arctic charr because of higher primary productivity (further research required, see ref 14), it is possible that biomass dilution of Hg was occurring (26). In more productive systems, the amount of bioavailable Hg in the water column is distributed among a greater number of algal cells. This may result in lower [Hg] throughout the food web (26). Biomass dilution did not appear to explain differences in resident lake trout [Hg] between lakes with and without Arctic charr. Although we did not determine [MeHg] in algae, [Hg] in forage fishes and [MeHg] in invertebrates did not differ significantly between lakes with and without Arctic charr. Mercury concentrations in cisco, ninespine stickleback, and lake whitefish differed significantly among lakes (ANCOVA, $F \geq 2.76$, $P \leq 0.04$, $\text{df} \geq 4,38$), but pairwise differences (Tukey's test, $P < 0.05$) did not indicate a charr-induced effect (Table S3). Similarly, [MeHg] in mysids and zooplankton differed significantly among lakes (ANOVA, $F \geq 184.18$, $P \leq 0.0001$, $\text{df} \geq 5,6$), but pairwise differences did not indicate a charr-induced effect (Table S3). [MeHg] in chironomids and isopods did not differ significantly among lakes (ANOVA, $F \leq 0.83$, $P \geq 0.57$, $\text{df} \geq 5,7$) (Table S3).

When resident lake trout [Hg] was compared among lakes with an ANCOVA, the interaction between lake and fork length was significant (ANCOVA, $F = 4.74$, $P = 0.0006$, $\text{df} = 5,115$). A scatterplot of raw [Hg] vs fork length showed that resident lake trout had relatively lower [Hg] in lakes with anadromous Arctic charr at fork lengths $\geq \sim 245$ mm ($\log_{10}245 = 2.8$; Figure S2). Because condition best explained why [Hg] in resident lake trout was lower in lakes with Arctic charr, there may be a change in lake trout growth at ~ 245 mm that results in lower Hg bioaccumulation in resident lake trout from lakes with Arctic charr. This requires further research.

Although the study design (comparison of three lakes with anadromous Arctic charr to three lakes without anadromous

Arctic charr) was confounded by the presence of anadromous lake trout, there appeared to be an independent effect of anadromous Arctic charr on resident lake trout [Hg]; Hg concentrations in resident lake trout from Glenn Lake (anadromous Arctic charr absent, anadromous lake trout present) were similar to those observed in Doris and Patch lakes and were significantly higher than those observed in Hovaktok, Nauyuk, or Roberts lakes (Figure 1). Also, patterns of pairwise differences in [THg] of forage fishes and [MeHg] of invertebrates did not indicate an effect of anadromous lake trout (Table S3).

Comparisons of [Hg] between Species and Life History Types of Arctic Charr and Lake Trout. This was the first study to examine [Hg] in recently discovered anadromous lake trout and the first study to compare [Hg] between sympatric resident and anadromous Arctic charr. Mercury concentrations varied significantly between species and life history types of Arctic charr and lake trout (mixed model, $F > 15.95$, $P < 0.0001$, $\text{df} = 1,244$; Figure 2). Regardless of life history type, Arctic charr had significantly lower [Hg] (at a standardized size) than lake trout (Figure 2). This difference between lake trout and Arctic charr was consistent with results of previous studies, and Hg concentrations of all Arctic charr and lake trout were within the range of previously reported values (2, 3). Within species, anadromous fishes generally had lower [Hg] (at a standardized size) than resident fishes, but the difference was only significant for lake trout (Tukey's test, $P < 0.05$; Figure 2).

Consistent with the suggestion of Evans et al. (2), significant differences in [Hg] between species and life history types of Arctic charr and lake trout were best explained by fish age. Age explained 72% of the variation in [Hg] whereas C:N, $\delta^{13}\text{C}_{\text{adj}}$, and $\delta^{15}\text{N}_{\text{adj}}$ explained 5, 1.5, and 1% of remaining variation, respectively. Age also differed significantly between species and life history types (mixed model, $F > 10.86$, $P < 0.011$, $\text{df} = 1,263$), and pairwise differences in age-at-size were consistent with patterns observed in [Hg] (Tukey's test, $P < 0.05$; Table 1). That is, at a standardized size, lake trout were significantly older than Arctic charr, and resident lake trout were significantly older than anadromous lake trout.

In contrast with previous studies (2, 3), anadromous Arctic charr did not have significantly lower [Hg] than resident Arctic

TABLE 1. LSmean (and SE) Age, $\delta^{15}\text{N}_{\text{adj}}$, $\delta^{13}\text{C}_{\text{adj}}$, and C:N for Anadromous and Resident Arctic Charr and Lake Trout

		LSmean ^a log age (years)	SE	LSmean age (years)	LSmean $\delta^{15}\text{N}_{\text{adj}}$ (‰)	SE	LSmean $\delta^{13}\text{C}_{\text{adj}}$ (‰)	SE	C:N ^b	SE
Anadromous fishes										
	Anadromous Arctic charr	1 (a)	0.08	10	10.36 (a)	0.34	-23.29 (a)	0.60	3.93 (a)	0.06
	Anadromous lake trout	1.41 (b)	0.08	26	7.97 (b)	0.51	-22.38 (b)	0.60		
Resident fishes										
	Resident Arctic charr	1.11 (c)	0.10	13	10.82 (a)	0.33	-25.32 (c)	0.65	3.56 (b)	0.06
	Resident lake trout	1.48 (d)	0.07	30	8.77 (b)	0.29	-26.31 (c)	0.56		

^a LSmeans were calculated at a fork length of 650 mm (\log_{10} fork length = 2.81), and letters indicate pairwise differences (Tukey's test). ^b LSmeans are not shown for each species-life history pair because C:N did not differ significantly between species (Tukey, $P > 0.05$).

charr. Previous comparisons of [Hg] between landlocked and anadromous Arctic charr may have been confounded by differences in region of fish capture (2, 3), but it is also possible that landlocked (no access to the sea) Arctic charr have significantly higher [Hg] than either resident (access to the sea, but do not migrate) or anadromous individuals. Future research should compare [Hg] and covariates among landlocked, resident, and anadromous Arctic charr (preferably within a region) to determine how [Hg] differs among all three life history types.

Our finding of significantly lower [Hg] in anadromous lake trout relative to resident lake trout has implications for human health. As shown in Figure 2, anadromous lake trout [Hg] were similar to the Canadian subsistence consumption guideline, but resident lake trout [Hg] were above this guideline. Significantly lower [Hg] in anadromous lake trout relative to resident lake trout was not due to significantly lower [Hg] in marine prey items; [Hg] did not differ significantly between marine and freshwater environments for either forage fishes (mixed model, $F = 1.91$, $P = 0.17$, $df = 1, 194$; Figure S3) or invertebrates ([MeHg], t tests, $1.33 \leq t \leq 0.80$, $P \geq 0.23$, $df \geq 3$). Sample sizes for forage fish were unbalanced, however (Figure S3), and differences may be significant with a larger sample size and size range of marine forage fishes. Although anadromous lake trout have been confirmed in only 4 lakes, they likely extend across the mainland Arctic coast of North America, and anadromous lake trout can comprise ~14–42% of the total lake trout population in lakes where they exist (20). This should be a consideration when examining past and future [Hg] data in lake trout from coastal Arctic lakes and when developing guidance for human consumption of lake trout in the Arctic.

Although age-at-size appeared to best explain the difference in [Hg] between anadromous and resident lake trout, this difference may also be related to C:N. C:N was significantly and negatively related to [Hg] in Arctic charr and lake trout (multiple regression, $F = 61.08$, $P < 0.0001$) and explained 5% of the variation in [Hg]. There was no difference in C:N between species (mixed model, $F = 0.19$, $P = 0.66$, $df = 1, 239$), but anadromous fishes had significantly higher C:N than resident fishes (mixed model, $F = 29.34$, $P < 0.0001$, $df = 1, 239$; Table 1). Higher C:N in anadromous fishes could lead to lower [Hg] because Hg binds to cysteine in proteins (e.g. ref 27). Fish with more lipid relative to protein (higher C:N) (13) should therefore have lower [Hg].

The effect of life history on fish [Hg] did not vary with fork length (life history*fork length interaction was not significant, mixed model, $P > 0.05$). Differences in [Hg] between Arctic charr and lake trout (both life history types included) did vary with fork length (significant species*fork length interaction), however (mixed model, $F = 97.5$, $P < 0.0001$, $df = 1, 244$). Similar to the interaction analysis presented above, examination of raw [Hg]-fork length plots showed that the interspecies difference was likely only significant at fork

lengths $> \sim 245$ mm ($\log_{10}245 = 2.8$; Figure S4). In this analysis, most of the variation in [Hg] was explained by age (72%) and C:N (5%); thus, it seems likely that at a fork length of ~ 245 mm, there is a change in growth and/or lipid content that leads to greater Hg bioaccumulation in lake trout relative to Arctic charr.

Comparison of [MeHg]- $\delta^{15}\text{N}$ Slopes. Previous calculations of $\log_{10}[\text{Hg}]-\delta^{15}\text{N}$ slopes for freshwater and marine food webs have usually used raw data. Estimates have varied between ~ 0.13 and 0.48 (1, 15–17, 28) but are most often close to 0.2. We used three different data sets (raw data, size-adjusted means, residuals from size and age) to calculate $\log_{10}[\text{Hg}]-\delta^{15}\text{N}$ slopes. Using raw data (best comparison with previous studies), we found that freshwater $\log_{10}[\text{Hg}]-\delta^{15}\text{N}$ slopes were relatively close to 0.2 and varied between 0.16 and 0.26 (Table 2). Slopes varied significantly among lakes (ANCOVA, $F = 5.98$, $P < 0.0001$, $df = 5, 382$), but pairwise differences did not indicate an effect of anadromous Arctic charr and/or anadromous lake trout (Tukey's test, $P < 0.05$; Table 2). Therefore, differences in Hg biomagnification between lakes with and without anadromous Arctic charr did not help explain why resident lake trout had lower [Hg] in lakes where anadromous Arctic charr were present.

The $\log_{10}[\text{Hg}]-\delta^{15}\text{N}$ slope for Melville Sound was 0.08 (raw data; Table 2). This was significantly lower than any of the slopes calculated for freshwater food webs (ANCOVA, $F = 7.97$, $P < 0.0001$, $df = 6, 485$; Table 2) and was also lower than previous estimates for Arctic marine food webs (0.2 (15); 0.223 (28); 0.15 (converted from natural log slope) (16)). The food web we analyzed contained only fishes and invertebrates, whereas other studies have often included sea birds and/or marine mammals. Reanalysis of previous data would reveal if slope estimates are affected by inclusion of higher trophic-level organisms such as marine mammals and indicate whether our $\log_{10}[\text{Hg}]-\delta^{15}\text{N}$ slope for Melville Sound is indeed lower than those calculated for the Northwater Polynya (28), Lancaster Sound (15), and West Greenland (16). Regardless, the relatively low $\log_{10}[\text{MeHg}]-\delta^{15}\text{N}$ slope in Melville Sound may help explain why anadromous lake trout had significantly lower [Hg] than resident lake trout. In addition to younger age-at-size and higher C:N, it is possible that reduced biomagnification of Hg through the marine food web resulted in lower [Hg] in anadromous lake trout relative to resident lake trout.

Adjusting for fish age and/or size had variable, lake-specific effects on calculations of $\log_{10}[\text{MeHg}]-\delta^{15}\text{N}$ slopes (Table 2). The effect of fish age and/or size adjustment may have been inconsistent among lakes because the strength of [Hg]-age and [Hg]-size relationships differed among lakes. The difference between unadjusted slopes (raw data) and size-adjusted slopes (LSmean data) was largest for Nauyuk Lake, where the slope increased from 0.18 (no covariate adjustment) to 0.24 (size-adjusted LSmeans for fish) (Table 2). Further analysis revealed that neither [MeHg] nor $\delta^{15}\text{N}$

TABLE 2. Statistics for Linear Regressions of log₁₀[MeHg] vs δ¹⁵N for Each Lake and Melville Sound

lake ^a	raw log ₁₀ [MeHg] vs raw δ ¹⁵ N				LSmean log ₁₀ [MeHg] vs LSmean δ ¹⁵ N (standardized fish sizes) ^b			residuals of log ₁₀ [MeHg] vs residuals of δ ¹⁵ N (fish size and age variation removed) ^c			
	slope	pairwise diffs	P	N	slope	P	N	slope	pairwise diffs	P	N
Doris	0.20	abe	<0.0001	52	0.22	0.0483	8	0.19	a	<0.0001	51
Glenn	0.23	bde	<0.0001	82	0.25	0.0007	9	0.23	ad	<0.0001	79
Patch	0.25	bd	<0.0001	62	0.23	0.0054	8	0.23	ad	<0.0001	62
<i>Hovaktok</i>	0.16	a	<0.0001	41	0.16	0.0281	7	0.11	bc	<0.0001	41
<i>Nauyuk</i>	0.18	ae	<0.0001	64	0.24	0.0034	9	0.17	ab	<0.0001	57
<i>Roberts</i>	0.26	d	<0.0001	96	0.23	0.0288	9	0.24	d	<0.0001	91
Melville Sound (marine)	0.08	d	<0.0001	105	0.09	0.0270	7	0.10	c	<0.0001	96

^a Italicized lakes contained anadromous Arctic charr. These lakes, and Glenn lake, also contained anadromous lake trout.

^b LSmeans were calculated at species-specific standardized sizes. ^c Variation due to fork length and age removed.

were significantly related to fish size in Nauyuk Lake (linear regression, $-1.77 \geq t \leq -0.43$, $P > 0.09$, $df \geq 2,33$) but that both relationships were slightly negative. Removal of this variation likely led to the increase in slope.

In Nauyuk and Hovaktok lakes, adjusting for fish age as well as size had a considerable effect on log₁₀[MeHg]-δ¹⁵N slopes (Table 2). In both lakes, slopes were approximately 30% lower when variation due to age was removed. This decrease was apparent even when size- and age-adjusted slopes were compared to size only adjusted slopes (Table 2). For both Nauyuk and Hovaktok lakes, age was a much stronger covariate for [MeHg] (multiple regression, $t \geq 2.76$, $P \leq 0.005$, $df \geq 2,19$) than size (multiple regression, $-1.47 \leq t \leq -0.83$, $P \geq 0.16$, $df \geq 2,19$). The variable response of log₁₀[MeHg]-δ¹⁵N slopes to standardization for fish age and/or size indicates that future studies should consider effects of organism size and age on both [Hg] and δ¹⁵N and adjust for these covariates when necessary. We demonstrated two possible methods for size and age adjustment, but other methods are available and should be explored. Our results also indicate that unadjusted estimates of [Hg]-δ¹⁵N slopes may reflect bioaccumulation rather than biomagnification if effects of covariates have not been considered.

Exceedances of [Hg] Guidelines. Of 313 lake trout, Arctic charr, and lake whitefish (common traditional food fish species), 17 exceeded the Canadian Hg guideline for commercial sale. All exceedances were lake trout. These fish ranged from 498–880 mm fork length and represented Roberts, Glenn, Hovaktok, and Patch lakes. Sixteen of the 17 fish were resident; only one anadromous lake trout exceeded the commercial sale guideline.

There were 70 exceedances of the Canadian Hg guideline for subsistence consumption, representing approximately 22% of the total food fish catch. None of these exceedances were Arctic charr. Fifty-six of the exceedances were resident lake trout, 12 were anadromous lake trout, and two were lake whitefish. The lake whitefish were from Roberts Lake, whereas lake trout exceedances were found in all systems. This comparison again illustrates the importance of differentiating anadromous lake trout from resident lake trout when developing guidance for human consumption of fish. Fishes that exceeded the subsistence consumption guideline ranged in size from 348–880 mm for lake trout and 531–546 mm for lake whitefish.

Balancing risks and benefits of consuming traditional food fishes in the Canadian Arctic is a complex issue, both for scientists and northerners. One of the goals of this study was to condense results into practical considerations that can be used when northerners choose a site and species to fish. Our results indicate that northerners may reduce their Hg intake from fish consumption by 1) fishing for resident lake trout in lakes that contain both Arctic charr and lake trout; 2)

choosing Arctic charr before lake trout; 3) choosing anadromous (i.e., catch them at sea) fish before resident fish; and 4) choosing smaller lake trout (preferably less than 350 mm and definitely less than 500 mm) and lake whitefish. Our results also indicate that the mass of Hg biotransported by anadromous Arctic charr to freshwater lakes is either negligible or that the effect of increased Hg loading on freshwater fish [Hg] is counteracted by increased fish growth rates. To further quantify how anadromous fishes may affect [Hg] in Arctic freshwater lakes, net mass of imported, biotransported Hg should be estimated for lakes that contain anadromous fishes. These estimates could then be used in comparisons of Hg mass budgets between Arctic lakes that do and do not contain anadromous fishes.

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Supporting Information Available

Laboratory QA/QC, corrections for baseline δ¹⁵N, sample sizes, location, and limnological data for each lake and figures to support statistical analyses. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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