Critical Review

Screening-level risk assessment of methylmercury for non-anadromous Arctic char (*Salvelinus alpinus*)

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Running Head: Risk of methylmercury for non-anadromous Arctic char

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Abstract: Non-anadromous forms of Arctic char (*Salvelinus alpinus*), those that are restricted to lakes and rivers, typically have higher mercury concentrations than anadromous forms that migrate to and from the sea. Using tissue burden data from the literature and our own analyses, we performed a screening-level risk assessment of methylmercury for non-anadromous Arctic char. Our assessment included 1569 fish distributed across 83 sites. Site-specific mean total mercury concentrations in non-anadromous Arctic char muscle varied considerably from 0.01 to 1.13 μg/g wet weight (ww), with 21% (17 of 83 sites) meeting or exceeding a threshold-effect level in fish of 0.33 μg/g ww, and 13% (11 of 83 sites) meeting or exceeding a threshold-effect level in fish of 0.5 μg/g ww. Of the sites in exceedance of the 0.33 μg/g threshold, seven were located in Greenland and ten in Canada (Labrador, Nunavut, Yukon). All but one of these sites were located in interfrost or permafrost biomes. Maximum total mercury concentrations exceeded 0.33 μg/g ww at 53% of sites (40 of the 75 sites with available maximum mercury values), and exceeded 0.5 μg/g ww at 27% (20 of 75 sites). Collectively, these results indicate that certain populations of non-anadromous Arctic char located mainly in interfrost and permafrost regions may be at risk for methylmercury toxicity. This approach provides a simple statistical assessment of methylmercury risk to non-anadromous Arctic char, and does not indicate actual effects. We highlight the need for studies that evaluate the potential toxic effects of methylmercury in non-anadromous Arctic char, as well as those that aid in the development of a methylmercury toxic-effect threshold specific to this species of fish. This article is protected by copyright. All rights reserved

INTRODUCTION

Human activities, including mining and fossil-fuel combustion, release and transport mercury (Hg) in the environment (UNEP 2013). Gaseous elemental mercury (Hg^0) emitted from these sources has a long atmospheric residence time (up to 1 year) allowing it to be distributed globally, and thus even unpopulated regions are subject to Hg contamination (Driscoll et al. 2013). Methylmercury (MeHg), produced by microbes, bioaccumulates and biomagnifies in aquatic food webs (Driscoll et al. 2013). As a result, fish worldwide are contaminated with Hg, and MeHg comprises the majority of the total Hg (THg) present in fish (Bloom 1992; Grieb et al. 1990).

A growing body of evidence indicates that MeHg concentrations regularly found in fish may negatively impact their health. Methylmercury causes oxidative stress, and results in altered gene expression, biochemistry, and histology in wild fish as well as fish exposed in the laboratory (Sandheinrich and Wiener 2011). Laboratory exposures have demonstrated that environmentally relevant concentrations of dietary MeHg negatively impact reproduction. For example, in fathead minnows (*Pimephales promelas*) exposed to dietary MeHg at environmentally relevant concentrations, circulating sex hormones were suppressed in both sexes leading to decreased fecundity in females and altered reproductive behavior in males (Drevnick and Sandheinrich 2003; Drevnick et al. 2006; Sandheinrich and Miller 2006). The combined effects of these laboratory exposures on females and males resulted in reproductive failure (Hammerschmidt et al. 2002). Field studies have also reported altered hormone levels in fish with elevated Hg concentrations, albeit in environments grossly polluted with other contaminants (Adams et al. 1999; Friedmann et al. 2002; Hontela et al. 1995; Webb et al. 2006).

Analyses of the available data for MeHg toxicity in fish indicate that effects are likely to occur at concentrations in edible muscle equal to or above 0.33 and 0.5 µg/g wet weight (ww) (equivalent whole-body concentrations are 0.2 µg/g ww (Beckvar et al. 2005), or 0.3 µg/g ww (Dillon et al. 2010; Sandheinrich and Wiener 2011), respectively). Comparing muscle THg concentrations found in wild fish to a threshold-effect level (TEL) is a simple statistical approach for assessing the potential for toxic effects. Sandheinrich et al. (2011) used this approach to estimate that 43% of sexually mature female walleye (*Sander vitreus*) populations in the Great Lakes region of North America are at risk of MeHg toxicity. A similar analysis for yellow perch (*Perca flavescens*) demonstrated that 6.5% of the 691 populations sampled exceeded the Hg TEL of 0.2 µg/g ww (whole-body) in the Great Lakes region (Wiener et al. 2012).

Arctic char (*Salvelinus alpinus*, Linnaeus, 1758) is a species of fish in the family Salmonidae that is found within water bodies of Arctic, subarctic, boreal, and temperate climate regions. The species' preference for cold water is consistent with a distribution across the northern regions of Canada, Greenland, Europe, and Asia between approximately 42°N to 83°N latitude (Klemetsen et al. 2003). Within alpine lakes and freshwaters north of ~75° N Arctic char is often the only species of fish present, and the species is both ecologically and culturally important. Though all Arctic char use freshwater habitats for reproduction and as nurseries for their young, anadromous forms make annual migrations to the sea after spending several years in freshwater. There are also landlocked (those lacking access to the sea) and resident populations that comprise non-anadromous forms (Power et al. 2008).

Anadromous Arctic char have relatively low THg concentrations in muscle tissue; an analysis by Evans et al. (2015) reported that across Canada the mean THg concentration in anadromous Arctic char muscle (n>1000) was 0.05 ± 0.02 µg/g ww. Consequently, anadromous

Arctic char have been promoted as a nutritious food source by public health authorities. In contrast, Hg concentrations are higher in non-anadromous (landlocked and resident) Arctic char (Gantner et al. 2010; Lockhart et al. 2005; Swanson et al. 2011a; van der Velden et al. 2015; van der Velden et al. 2013b). These observed differences are likely a result of higher THg and MeHg levels at the base of lacustrine relative to marine food webs (van der Velden et al. 2013a). Regardless of life-history type, Hg contamination of Arctic char is of concern from a human health perspective given that many Indigenous communities rely on Arctic char as a part of their traditional diets and typically augment stores in late winter from lake-based fisheries (Boivin and Power 1990). Additionally, the potential deleterious effects of Hg to Arctic char are not well characterized.

Given the ecological and cultural importance of Arctic char, we performed a screening-level risk assessment of MeHg for non-anadromous Arctic char (NAAC), similar to efforts to assess the risk of MeHg for Great Lakes fish (Sandheinrich et al. 2011). Our assessment was carried out by comparing published MeHg toxicity thresholds and the available Hg data for populations of NAAC. Here, we focus on NAAC given the assumption that their generally higher Hg concentrations put them at greater risk for toxic effects than anadromous conspecifics.

METHODS

Literature review

We performed a state-of-the-art review (Grant and Booth 2009) to better understand levels of Hg contamination in wild NAAC. A literature search was performed with Web of Science, SCOPUS, and ProQuest Agricultural & Environmental Science Database. The first and last searches were performed on November 1st, 2017 and December 17th, 2017 respectively. The searches in each database were performed with the following terms:

Salvelinus alpinus OR *Arctic char* OR *Arctic charr* AND *Mercury* OR *methylmercury* OR *Hg*

OR *CH3Hg* OR *MeHg*

The following inclusion criteria were selected prior to performing the literature search.

1. Study discusses NAAC, and differentiates between anadromous and non-anadromous forms.

2. Study presents mean or geometric mean THg or MeHg concentrations in fish muscle.

3. Study presents data for a minimum of three fish.

4. Study provides geographic location of the samples.

The Web of Science search generated 68 results, while the SCOPUS and ProQuest searches generated 219 and 832 results respectively. In total, 21 studies were included from this search as they met the inclusion criteria. All of the studies reported THg in NAAC muscle, and only one study (Lescord et al. 2015) reported muscle THg and the percentage of MeHg in a subset of NAAC. From the selected studies, we gathered THg data (minima, maxima, arithmetic means, geometric means, and medians) for NAAC muscle, as well as arithmetic mean lengths, weights, and ages of the fish, when available. To standardize lengths, we calculated total lengths from fork lengths by multiplying fork lengths by 1.05 (Tallman et al. 2013). If the same site was sampled for multiple years we collected data for the most recent sampling period. We also documented the terrestrial biomes surrounding the water bodies from which NAAC were collected to better understand the populations that have been sampled for Hg. Terrestrial biomes were initially assigned by visual comparison of sampling locations with the Major Biomes map produced by the United States Department of Agriculture (USDA-NRCS 1999). Subsequently, a map with biomes (alpine, boreal, tundra interfrost, and tundra permafrost) and geographic

locations of the various NAAC populations was generated using ArcGIS 10.4 and several

shapefiles (Brown et al. 2017; Olson et al. 2001; Pecher et al. 2011). Note that we classified

lakes as alpine if their elevations were above 1500 m.

Sampling and analysis of Arctic char muscle

In addition to information obtained from the literature, we also present our original data for

NAAC sampled from eight lakes. Four of these lakes, Boomerang (73.94°N, -92.89°W),

Aqiatusuk (74.70°N, -94.23°W), Lake 10 (68.14°N, -106.40°W), and Lake 32 (68.15°N, -

106.52°W) are located in Nunavut, Canada. Arctic char from Boomerang and Aqiatusuk have

been sampled for THg as previously described in Muir et al. (2010) and (2005a). The remaining

four lakes, Fragasso (47.37°N, -71.31°W), Pemmican (47.64 °N, -70.77°W), Arthabaska (47.66

°N, -70.78°W), and des Îles (47.65 °N, -70.78°W) are located in Québec, Canada. Arctic char

from each of these sites were collected with gillnets. Fish were sacrificed, before lengths and

weights were recorded. Axial muscle was collected and frozen at -20 °C until analyzed for THg.

Otoliths of Arctic char from the eight lakes were collected to estimate age using either a thin-

section or break-burn method (Swanson et al. 2010). Ages for Arctic char collected from the

Quebec lakes were estimated by Mark Gautreau (Canadian Rivers Institute, University of New

Brunswick). Age estimates of Arctic char from Nunavut lakes were provided by John Babaluk

(Fisheries and Oceans Canada).

Muscle tissue samples were analyzed for THg by direct mercury analyzers (DMA-80,

Milestone Inc., Monroe, CT) according to USEPA method 7473 (EPA 2007) at Burlington's

National Laboratory for Environmental Testing (NLET; Boomerang and Aqiatusuk), INRS-ETE

(Fragasso, Pemmican, Arthabaska, and des Îles), and at the University of New Brunswick's

(UNB) Canadian Rivers Institute (Lakes 10 and Lake 32). Quality control data associated with

original THg data are presented in Supporting Information.

Risk of methylmercury exposure for non-anadromous Arctic char

To assess the potential for toxic effects of MeHg exposure, we compared THg concentrations in

NAAC muscle to the TELs developed by Beckvar et al. (2005), Dillon et al. (2010), and

Sandheinrich and Wiener (2011). Note that the majority of the THg in muscle of large Arctic

char (fork length > 180 mm) is present as MeHg (Lescord et al. 2015), which is similar to other

upper trophic level fish (Bloom 1992). Therefore, THg is an acceptable proxy for MeHg for the

purposes of risk assessment. Specifically, we compared means, maxima, and medians to the

TELs and noted whether they met or exceeded these values. Mean THg concentrations in muscle

were available for all of the populations included in the present study except for the sample of

Arctic char collected from Lake Tjulträsk, Sweden. For this sample, only a geometric mean THg

concentration was reported and, therefore for this sample only we compared the geometric mean

THg concentration to the TELs. For 61 of the 83 populations included in the present study, we

obtained data for each of the individuals in the populations allowing us to report medians and

maxima. In addition to the maximum THg concentrations obtained from the raw data, maximum

THg concentrations were published for 14 other Arctic char samples giving us a total of 75

maximum concentrations that could be compared to the TELs.

The TEL developed by Beckvar et al. (2005) (0.33 μg/g ww) is based on laboratory

studies that assessed sublethal endpoints including growth, reproduction, development, and

behavior. The TEL developed by Dillon et al. (2010) (0.5 μg/g ww) is based on laboratory

studies with endpoints most relevant to ecological risk assessment i.e., mortality, survival,

reproductive success, and lethal developmental abnormalities. The major difference between the

TELs developed by Beckvar et al. (2005) and Dillon et al. (2010) is that the former included growth and behavior in their model. Sandheinrich and Wiener (2011) reported a TEL equivalent to that of Dillon et al. (2010), however both laboratory and field studies were included in their assessment and the authors indicated that toxic effects at or above the TEL would include biochemical changes, cell and tissue damage, and reduced reproduction.

Statistical analyses

Data were tested for normality using Shapiro-Wilks tests and tested for outliers using Grubb's tests. If not normal, data were $\log_{10}$ transformed. Relationships among variables were first examined in bivariate scatterplots and tested by simple correlation (Pearson r). Simple regression models were only tested using the ordinary least-squares equation if the bivariate scatterplots indicated a possible relationship. If data remained non-normal, a non-parametric correlation was reported (Spearman r). For all statistical tests $\alpha = 0.05$ was used. All statistical analyses were performed with JMP Pro 13 Statistical Analysis Software (SAS Institute, Cary, NC, USA).

RESULTS

In the present study, we included data for NAAC from 83 sites comprising a total of 1569 individual fish. Of these 83 datasets, eight are original to the present study. Information about sampling sites, Arctic char biological data (mean lengths (mm), weights (g), ages (yr)), and THg (µg/g ww) descriptive statistics (minima, maxima, means, medians) are presented in Table 1. A map presenting locations and mean muscle THg concentrations of NAAC is provided in Figure 1. The sites ranged in latitude from ~42°N (French Alps) to 82°N (northern Ellesmere Island). Most of the sites were located in Canada (59 sites) followed by Greenland (10 sites), France (5 sites), Norway (3 sites), Austria (2 sites), Italy (2 sites), with one site in each of the Faroe Islands, Sweden, and Russia. The majority of sampling sites were located in the tundra

permafrost and interfrost biomes (n=65 combined), whereas fewer sites were located in alpine, boreal or temperate biomes (n=18 combined).

Site-specific mean THg concentrations in NAAC muscle varied considerably from 0.01 to 1.13 μg/g ww. The lowest mean THg concentration was reported for a sample of resident Arctic char from Roberts Lake, Nunavut (Swanson and Kidd 2010), and the highest mean THg concentration was reported for a sample of landlocked Arctic char collected from an unnamed lake in southwest Greenland (Rigét et al. 2010). Mean THg concentrations were equal to or above the TEL of 0.33 μg/g ww at 21% of the sites, whereas Arctic char from only 13% were equal to or above the TEL of 0.5 μg/g ww. Of the 83 Arctic char datasets included in this analysis, we obtained maximum THg concentrations for 75 of the datasets. Of these 75 maxima, 53% were equal to or above 0.33 μg/g ww and 27% were equal to or above 0.5 μg/g ww.

Mean THg concentrations in Arctic char from seven of the sites in Greenland and ten in Canada (Labrador, Nunavut, Yukon) exceeded the TEL of 0.33 μg/g ww. Mean THg concentrations in Arctic char from six of the sites in Greenland also met or exceeded the TEL of 0.5 μg/g ww, as did mean THg concentrations in Arctic char collected from five sites in Canada (Labrador, Nunavut, Yukon) (Barst et al. 2016; Bruce et al. 1979b; CACAR 2012; Gantner et al. 2010; Muir et al. 2005a; Rigét et al. 2000; Rigét et al. 2010). All of the Arctic char samples with mean THg concentrations exceeding at least one of the TELs (n=17) were located between ~54°N and 75°N, and most (n=16) were in regions of tundra interfrost and permafrost.

Though median THg concentrations were available for a smaller number of datasets (n=61) than mean concentrations, the percentages of sites exceeding the TELs were similar when using means or medians as a basis for comparison; 13 of 61 (21%) median THg concentrations in

Arctic char muscle exceeded 0.33 μg/g ww, and 9 out of 61 (15%) median THg concentrations exceeded 0.5 μg/g ww (Table 1 and Figure 2).

Across sites, THg concentrations in NAAC muscle were positively correlated with total lengths (r=0.53, p<0.0001; n=59 datasets) and ages (r=0.47, p<0.0001; n=44 datasets) (Figure 3). Mean total lengths (r=0.36, p=0.0008; 82 datasets) and mean ages (r=0.60, p<0.0001; 69 datasets) of NAAC increased significantly with latitude (Figure 4), however mean THg did not vary significantly with latitude (r=0.11, p=0.32; n=83 datasets).

DISCUSSION

In the present study, we compared mean (n=83 datasets), median (n=75 datasets), and maximum (n=61 datasets) THg concentrations in muscle of NAAC to available toxicity thresholds for MeHg. The screening-level approach we employed is a simple method to assess risk of MeHg for NAAC. Our study is unique in that we applied this type of screening-level assessment to NAAC, whereas in the past these types of assessments have been carried out for species distributed at lower latitudes (Sandheinrich et al. 2011; Wiener et al. 2012). Though screening-level assessments are relatively simple to conduct, there are limitations which should be noted. For example, while our assessment may suggest risk, it does not indicate actual effects and therefore it should be validated with laboratory and field studies with Arctic char. The major uncertainties associated with the use of TELs have been summarized by Dillon et al. (2010). Here we briefly discuss uncertainties in the context of NAAC.

Importantly, none of the laboratory exposure studies that serve as the basis for the TELs generated by Beckvar et al. (2005) and Dillon et al. (2010) used Arctic char as a study species. Similarly, no studies with Arctic char were incorporated into the estimate of Sandheinrich and Wiener (2011). This is relevant since a difference in sensitivities to MeHg across fish species is

plausible given that differences have been documented for species of birds (median lethal concentrations for MeHg dosed embryos ranged from 0.12 to 2.42 µg/g ww) (Heinz et al. 2009). Fathead minnow were the predominate species of fish used in the laboratory studies serving as the basis for the various TELs, and this species is known to be less sensitive to chemicals than rainbow trout (*Oncorhynchus mykiss*) (Dillon et al. 2010; Teather and Parrott 2006), a species in the same family as Arctic char. If this holds true for MeHg, the TELs may underestimate the toxicity of MeHg to Arctic char, and by extension more NAAC populations may be at risk than indicated by our assessment.

Data demonstrating the effects of MeHg to Arctic char are lacking. A study by de Oliveira Ribeiro et al. (2002) demonstrated that a single dietary dose of MeHg (0.26 ± 0.5 µg/g body weight) results in histological changes in Arctic char liver including necrosis and alterations to cytoplasmic organization, though it is unclear what the muscle THg (or MeHg) concentrations were in these fish (de Oliveira Ribeiro et al. 2002). Our previous work with wild NAAC collected from lakes spanning a Hg contamination gradient in Nunavut, Canada indicate that MeHg is not effectively detoxified in livers, which could explain tissue damage in the form of hepatic fibrosis in Arctic char with elevated MeHg concentrations (Barst et al. 2016). Hepatic fibrosis was more prevalent in Arctic char collected from Amituk Lake, where 73% of the Arctic char exceeded both the 0.3 and 0.5 µg/g ww TELs. Despite the elevated levels of Hg in Amituk Arctic char, it is difficult to attribute the observed hepatic tissue damage to MeHg exposure given the existence of confounding variables (for example other contaminants and parasites) (Barst et al. 2016). Neither of these studies included endpoints that relate to the TELs. We therefore highlight the need for future laboratory and field studies that continue to address the potential effects of MeHg to Arctic char. Ideally, both laboratory and field studies should report

axial muscle or whole-body THg concentrations to allow comparisons to be made to TELs. Furthermore, a laboratory-based dosing study with Arctic char is necessary to develop a species-specific TEL. Future studies should also aim to understand how contaminant mixtures affect wild Arctic char, given that previous research has reported varying levels of polychlorinated biphenyls (Cabrerizo et al. 2018; Dietz et al. 2018), organochlorine pesticides (Cabrerizo et al. 2018; Cleemann et al. 2000), and non-essential elements (Muir et al. 2005a) in the tissues of NAAC.

A limitation of the present study is the low sample sizes of Arctic char from certain sites. If we assume that the samples included in the present study are representative of their respective populations then our analysis suggests that 21% of the NAAC populations included in the present study are at risk for the toxic effects of MeHg. Additionally, 53% of the most contaminated individuals in their respective populations are also at risk. These potential effects include changes in growth, reproduction, development, and behavior, which are the sublethal endpoints included in the 0.33 μg/g ww TEL estimated by Beckvar et al. (2005).

There are also uncertainties associated with the extrapolation from laboratory to wild fish (Dillon et al. 2010). During laboratory exposures confounding variables are purposely limited in order to isolate contaminant effects, whereas in wild systems fish are exposed simultaneously to a myriad of other stressors (other contaminants, parasites, predation, temperature changes, etc.) that may act in a synergistic fashion with MeHg to cause deleterious effects. Thus, it is difficult to gauge how well the 0.3 and 0.5 μg/g ww TELs apply to wild NAAC.

Total lengths and ages were strong predictors of muscle THg across sites, indicating that the largest and oldest individuals have the highest THg concentrations. The positive relationships between latitude and both mean lengths and mean ages were consistent with the larger and older

NAAC being found in tundra regions. Our study was biased towards Arctic char datasets representing tundra interfrost and permafrost regions, with about 78% of the NAAC included in the study being located in these regions. The bias in the dataset included a much greater range in Arctic char total lengths and ages for these regions. For Arctic char collected from Heintzelman Lake, van der Velden et al. (2012) reported that fish age best explained differences in THg among individuals. The authors also suggested that both changes in life history and diet shifts altered THg levels of Arctic char, but size or length-at-age had little influence (van der Velden et al. 2012).

Other factors also affect THg concentrations in fish, however, including rates of atmospheric deposition and the ability of receiving landscapes and waterbodies to methylate deposited Hg (Munthe et al. 2007). Atmospheric deposition of Hg decreases with increasing latitude (Muir et al. 2009), which could explain relatively high THg concentrations for small, NAAC from the boreal zone, where deposition rates are elevated and wetlands are prominent features of the landscape. In the Canadian High Arctic, Gantner et al. (2010) explored how trophic position, food web length, and abiotic factors influenced THg concentrations in 24 populations of landlocked Arctic char. Size, $\delta^{15}N$ (trophic position), and age were good predictors of THg concentrations in landlocked Arctic char populations. Additionally, Gantner et al. (2010) reported that the catchment-to-lake area ratio was positively correlated with THg concentrations of landlocked Arctic char, indicating that both biotic and abiotic factors influence Arctic char Hg levels.

Considerable efforts have been made to understand the temporal trends of THg in Arctic populations of NAAC. Though results demonstrate no change in THg concentrations over time in certain populations, both decreasing and increasing THg concentrations have been reported for

others (AMAP 2011; Muir 2017; van der Velden et al. 2015). For several landlocked Arctic char populations on Cornwallis Island THg concentrations have declined slowly since 2005 in five of six lakes for which long-term trends are available (Muir 2017). These decreases are in step with decreasing atmospheric Hg concentrations, but could also be related to increased photodegradation of MeHg as a result of decreased ice cover, which has been demonstrated for Arctic marine ecosystems (Point et al. 2011). However, THg concentrations in landlocked Arctic char from a sixth lake on Cornwallis Island (Small Lake) have been increasing since 2008. Notably, MeHg in water and dissolved organic carbon were elevated in Small Lake relative to the other five lakes on Cornwallis Island (Muir 2017). In contrast, no significant trend was reported for Arctic char collected from Lake Abiskojaure, Sweden over a 25 year period (AMAP 2011). The 83 datasets included in our analysis represent NAAC sampled from a wide range of years (1977 – 2015). Given the discrepancy in temporal trends of THg in populations of NAAC it is possible that the THg concentrations included in our analysis are either representative, below, or above current THg levels depending on the population.

IMPLICATIONS

Permafrost soils store approximately double the amount of Hg than all other soils, the atmosphere, and oceans combined (Schuster et al. 2018). Permafrost is thawing as a result of climate change (Hinzman et al. 2005), which in turn can release Hg and organic matter to the environment (Creed et al. 2018). Dissolved organic matter (DOM) influences the fate of Hg in aquatic environments in a variety of ways including influencing inorganic Hg and MeHg transport in catchments (Ravichandran 2004), the methylation of inorganic Hg by bacteria (Graham et al. 2013), the incorporation of MeHg at the base of food webs (Tsui and Finlay 2011), and the movement of inorganic Hg and MeHg within waterbodies (Creed et al. 2018;

Ravichandran 2004). The impacts of Hg and DOM release from permafrost stores are not well understood, but could involve greater production of MeHg in adjacent water bodies and greater contamination of food webs. An even greater proportion of NAAC populations from tundra permafrost regions may be at risk for MeHg toxicity if Hg released from permafrost soils is methylated and incorporated into the food webs supporting these fish.

Arctic char play an essential role in subsistence, economic, and socio-cultural aspects of northern communities (Roux et al. 2011). In general, anadromous Arctic char are more frequently consumed than NAAC by Inuit (Lemire et al. 2015; Roux et al. 2011). A recent food recall survey reported that anadromous Arctic char was the second most consumed country food by Nunavik Inuit communities, and landlocked Arctic char was consumed at a much lower frequency (less than 1 to 2 %) (Lemire et al. 2015). While current exploitation of lacustrine Arctic char may be relatively low, future reliance on these populations may increase with potential decreases in anadromy as a result of climate change (Finstad and Hein 2012). We therefore, reinforce the need for continued monitoring of trends in Hg concentrations of Arctic char.

Data Availability – Please contact the corresponding author (bdbarst@alaska.edu) for any

requests for access to data.

Conflict of interest – The authors declare no conflict of interest.

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Figure 1. Geographic distribution of non-anadromous Arctic char (*Salvelinus alpinus*) samples included in the screening-level risk assessment (n=83). Populations of non-anadromous Arctic char were grouped by mean total mercury concentrations in muscle (green circles = below 0.33 µg/g wet weight; orange circles = 0.33 to 0.5 µg/g wet weight; red circles = above 0.5 µg/g wet weight). Groupings are based on threshold-effects levels 0.33 µg/g wet weight (Beckvar et al. 2005) and 0.5 µg/g wet weight (Dillon et al. 2010; Sandheinrich and Wiener 2011). Inset figures bordered by solid lines show their respective dashed line boxes in greater detail. Terrestrial biome demarcations (alpine, boreal, tundra interfrost, and tundra permafrost) were created with ArcGIS 10.4 and various shapefiles (Brown et al. 2017; Olson et al. 2001; Pecher et al. 2011).

Figure 2. Box and whisker plots representing total mercury concentrations in a subset (n=61) of non-anadromous Arctic char (*Salvelinus alpinus*) samples included in the screening-level risk assessment. Boxes signify the interquartile range (IQR), the whiskers define the distance to the furthest point within 1.5 X IQR, and the center line is the median. The solid line spanning the graph represents the mercury threshold-effect level reported by Beckvar et al. (2005) (0.33 µg/g wet weight) and the dashed line represents the threshold-effect level reported by Dillon et al. (2010) and Sandheinrich and Wiener (2011) (0.5 µg/g wet weight). Arctic char with muscle total mercury concentrations in the grey region are potentially at risk for toxic effects. The letters A, B, I, and P signify alpine, boreal, tundra interfrost, and tundra permafrost biome groupings, respectively.

Figure 3. Bivariate scatterplots of Arctic char log10 transformed muscle total Hg (µg/g wet weight) versus total lengths (mm) and ages (years). Data points represent individual non-anadromous Arctic char sampled from 59 and 44 water bodies for lengths and ages, respectively.

Figure 4. Bivariate scatterplots of latitude (degrees) versus mean ages (years) and mean total lengths (millimeters) of Arctic char. Data are from non-anadromous Arctic char sampled from 82 and 69 water bodies for lengths and ages, respectively.

Table 1. Details of sample collection sites and non-anadromous Arctic char included in the screening-level risk assessment. Summary total mercury (THg) data (µg/g wet weight in muscle) are also presented.

Waterbody	Biome	Location	Latitude	Longitude	Date Sampled (yr)	Sample Size	Mean total length (mm)	Mean weight (g)	Mean age (yr)	Minimum total Hg in muscle (µg/g ww)	Maximum total Hg in muscle (µg/g ww)	Mean total Hg in muscle (µg/g ww)	STDEV total Hg in muscle (µg/g ww)	Geometric mean total Hg in muscle (µg/g ww)	Median total Hg in muscle (µg/g ww)	Reference
Rotfelssee	Alpine	Austria	47.23	11.01	2007	24	202	—	—	0.02	0.10	0.05	0.02	0.05	0.05	Koinig et al. (2015)
Schwarzsee ob Sölden	Alpine	Austria	46.97	10.95	2011	8	212	59	—	0.03	0.07	0.05	0.02	0.05	0.06	Koinig et al. (2015)
Étang d'Aubé	Alpine	France	42.74	1.34	1996, 1997	25	290	268	3	0.04	0.08	0.06	0.01	—	0.05	Rognerud et al. (2002)
Poursollet	Alpine	France	45.05	5.90	2008	8	264	309	6	0.08	0.14	0.10	0.02	—	—	Marusczak et al. (2011)
Crop	Alpine	France	45.21	5.99	2008	27	166	92	5	0.10	0.35	0.17	0.07	—	—	Marusczak et al. (2011)
La Sagne	Alpine	France	45.22	6.08	2008	18	204	91	5	0.08	0.46	0.19	0.08	—	—	Marusczak et al. (2011)
Bramant	Alpine	France	45.20	6.18	2008	14	239	185	6	0.06	0.25	0.15	0.06	—	—	Marusczak et al. (2011)
Lago Lungo	Alpine	Italy	46.73	11.08	1993	5	245	123	12	0.01	0.06	0.04	0.02	0.04	0.05	Wathne et al. (1997)
Lago di Latte	Alpine	Italy	46.72	11.07	1993	5	173	39	13	0.03	0.07	0.05	0.02	0.05	0.06	Wathne et al. (1997)
Lake Norsjø (Fjærekilen)	Boreal	Norway	59.19	9.54	2014, 2015	77	145	38	9	0.07	1.13	0.24	0.21	—	—	Olk et al. (2016)
Rivière de la Trinité	Boreal	Québec, Canada	49.42	-67.32	2009, 2010	10	192	54	5	0.19	0.26	0.22	0.02	0.22	0.22	van der Velden et al. (2013a)
Arthabaska	Boreal	Québec, Canada	47.66	-70.78	2011	5	211	57	4	0.24	0.41	0.29	0.07	0.29	0.26	This study
des Îles	Boreal	Québec, Canada	47.65	-70.78	2011	5	208	59	4	0.08	0.10	0.09	0.01	0.09	0.10	This study
Pemmican	Boreal	Québec, Canada	47.64	-70.77	2011	5	203	55	4	0.22	0.27	0.25	0.02	0.03	0.26	This study
Fragasso	Boreal	Québec, Canada	47.37	-71.31	2011	5	202	55	3	0.05	0.07	0.05	0.01	0.05	0.05	This study

Nipishish Lake	Boreal	Labrador, Canada	54.17	-60.77	1977	7	316	330	8	0.28	0.46	0.35	0.07	—	—	Bruce et al. (1979a)
Lake Chibini	Boreal	Russia	67.69	33.44	1993	5	257	169	7	0.03	0.06	0.04	0.01	0.04	0.04	Wathne et al. (1997)
á Mýrunum	Temperate	Faroe Islands	62.18	-7.12	2012	20	280	194	7	0.17	0.36	0.26	0.10	—	—	Nielsen et al. (2014)
Ikarut Lake	Tundra interfrost	Labrador, Canada	58.13	-63.63	1978	10	439	560	14	0.07	0.42	0.19	0.12	—	—	Bruce et al. (1979a)
Komaktorvik Lake	Tundra interfrost	Labrador, Canada	59.15	-64.23	1978	6	471	750	15	0.06	0.25	0.11	0.07	—	—	Bruce et al. (1979a)
Mistastin Lake	Tundra interfrost	Labrador, Canada	55.90	-63.27	1978	4	523	1230	10	0.04	0.22	0.11	0.08	—	—	Bruce et al. (1979a)
Mistinippi Lake	Tundra interfrost	Labrador, Canada	54.78	-61.32	1978	13	426	640	11	0.13	0.96	0.50	0.24	—	—	Bruce et al. (1979a)
Saglek Lake	Tundra interfrost	Labrador, Canada	58.82	-63.35	1978	17	516	1090	19	0.11	0.34	0.19	0.07	—	—	Bruce et al. (1979a)
Shallow Lake	Tundra interfrost	Labrador, Canada	57.65	-63.23	1978	5	459	620	13	0.24	0.39	0.33	—	—	—	Bruce et al. (1979a)
Shapio Lake	Tundra interfrost	Labrador, Canada	54.97	-61.28	1977	26	402	—	9	0.08	1.75	0.45	—	—	—	Bruce et al. (1979a)
Tasialuk	Tundra interfrost	Labrador, Canada	56.74	-62.69	2007	10	430	777	—	0.08	0.17	0.13	0.02	0.13	0.13	Gantner et al. (2010)
Coady's Pond	Tundra interfrost	Labrador, Canada	56.64	-63.63	2007	10	366	452	—	0.07	0.12	0.10	0.02	0.10	0.10	Gantner et al. (2010)
Coady's Pond #2	Tundra interfrost	Labrador, Canada	56.64	-63.63	2007	20	316	478	6	0.06	0.21	0.12	0.04	0.11	0.12	van der Velden et al. (2013a)
Esker Lake	Tundra interfrost	Labrador, Canada	57.15	-62.88	2008	20	278	306	11	0.04	0.86	0.11	0.18	0.08	0.07	van der Velden et al. (2015)
Hebron Lake #2	Tundra interfrost	Labrador, Canada	57.97	-64.02	2008	25	357	—	12	—	—	0.19	0.17	—		van der Velden et al. (2015)
Upper Nakvak Lake	Tundra interfrost	Labrador, Canada	58.66	-63.32	2007	20	287	360	12	0.03	0.49	0.13	0.12	0.08	0.07	van der Velden et al. (2015)
Galggojavri	Tundra interfrost	Norway	69.35	21.06	2011	49	252	184	4	—	—	0.12	0.01	—	—	Kahilainen et al. (2016)
Tasiapik Lake	Tundra interfrost	Nunavik, Canada	58.52	-68.36	2009, 2010	31	296	659	6	0.06	0.26	0.11	0.05	0.10	0.10	van der Velden et al. (2013a)
Lake Tjulträsk	Tundra interfrost	Sweden	65.97	16.07	1995, 1996	20	—	—	—	—	—	—	—	0.13	—	AMAP (2005)

Avanersuaq	Tundra permafrost	Greenland, Denmark	77.42	-66.36	1994, 1995	26	412	492	14	0.04	0.50	0.23	0.14	0.19	0.21	Rigét et al. (2000)
Nuuk	Tundra permafrost	Greenland, Denmark	64.18	-51.70	1994, 1995	25	368	435	11	0.09	1.30	0.60	0.33	0.49	0.60	Rigét et al. (2000)
Lake A (Qaqortoq)	Tundra permafrost	Greenland, Denmark	60.99	-47.51	1994, 1995	50	392	558	12	0.07	3.73	0.80	0.60	0.66	0.60	Rigét et al. (2000)
Lake B (Qaqortoq)	Tundra permafrost	Greenland, Denmark	60.97	-47.56	1994, 1995	22	347	403	11	0.15	1.71	0.58	0.35	0.49	0.48	Rigét et al. (2000)
Lake C (Qaqortoq)	Tundra permafrost	Greenland, Denmark	60.96	-47.53	1994, 1995	25	350	354	12	0.22	1.32	0.64	0.30	0.58	0.54	Rigét et al. (2000)
Lake D Qqaqortoq)	Tundra permafrost	Greenland, Denmark	60.87	-47.58	1994, 1995	22	335	300	11	0.23	2.01	0.62	0.35	0.57	0.57	Rigét et al. (2000)
Tasiilaq (Ammassalik) Lake	Tundra permafrost	Greenland, Denmark	65.61	-37.64	1994, 1995	4	478	1169	19	0.32	0.48	0.40	0.07	0.40	0.40	Rigét et al. (2000)
Tasiilaq River	Tundra permafrost	Greenland, Denmark	65.61	-37.64	1994, 1995	21	235	—	11	0.05	0.19	0.12	0.16	0.26	—	Rigét et al. (2000)
unnamed lake (near fjord Isortoq)	Tundra permafrost	Greenland, Denmark	65.58	-51.90	2008	20	387	328	—	0.70	3.56	1.13	0.61	1.05	0.98	Rigét et al. (2010)
Lake Fergusson	Tundra permafrost	Greenland, Denmark	66.97	-50.64	2005	27	373	486	9	0.03	0.39	0.18	0.09	0.15	0.18	Wathne et al. (1997)
Pingualuk	Tundra permafrost	Nunavik, Canada	61.28	-73.66	2007	10	402	395	17	0.11	0.38	0.17	0.08	0.16	0.15	Gantner et al. (2010)
Laflamme	Tundra permafrost	Nunavik, Canada	61.32	-73.71	2007	7	543	1669	13	0.10	0.24	0.14	0.04	0.14	0.14	Gantner et al. (2010)
Second Portage	Tundra permafrost	Nunavut, Canada	65.01	-96.02	2008	15	354	—	—	—	—	0.08	—	—	—	CACAR (2012)
Angajurjualuk	Tundra permafrost	Nunavut, Canada	71.12	-79.00	2006, 2007	28	408	—	—	—	—	0.33	—	—	—	CACAR (2012)
Sheardown	Tundra permafrost	Nunavut, Canada	71.31	-79.31	2006, 2007	26	356	—	—	—	—	0.16	—	—	—	CACAR (2012)
9-Mile	Tundra permafrost	Nunavut, Canada	74.81	-95.20	2011, 2012	30	366	356	18	0.06	0.39	0.15	0.09	—	0.12	Barst et al. (2016)
Amituk	Tundra permafrost	Nunavut, Canada	75.05	-93.75	2011, 2012	26	375	407	15	0.14	2.08	0.84	0.47	0.69	0.83	Barst et al. (2016)
North	Tundra permafrost	Nunavut, Canada	74.78	-95.09	2011, 2012	28	393	439	17	0.09	0.70	0.26	0.15	0.22	0.21	Barst et al. (2016)
Small	Tundra permafrost	Nunavut, Canada	74.76	-95.06	2011, 2012	30	358	274	14	0.04	0.19	0.10	0.04	—	0.09	Barst et al. (2016)
Little Nauyuk	Tundra permafrost	Nunavut, Canada	68.35	-107.76	2006	10	363	327	12	0.07	0.22	0.13	0.05	0.12	0.12	Gantner et al. (2010)

	Habitat	Location			Year											Reference
Radar	Tundra permafrost	Nunavut, Canada	68.44	-66.86	1999	8	453	860	—	0.23	1.92	0.99	0.61	0.81	0.91	Gantner et al. (2010)
12 Mile	Tundra permafrost	Nunavut, Canada	74.82	-95.34	2006	10	351	515	17	0.10	0.20	0.14	0.04	0.13	0.14	Gantner et al. (2010)
Aqiatusuk	Tundra permafrost	Nunavut, Canada	74.70	-94.23	2011, 2016	15	481	1003	18	0.16	1.03	0.56	0.28	0.49	0.59	This study
Boomerang	Tundra permafrost	Nunavut, Canada	73.94	-92.89	2007, 2012	23	455	560	19	0.09	0.54	0.24	0.14	0.20	0.16	This study, Gantner et al. (2010)
Hazen	Tundra permafrost	Nunavut, Canada	81.83	-70.42	2006	10	346	337	19	0.04	0.29	0.10	0.08	0.08	0.07	Gantner et al. (2010)
Lake D	Tundra permafrost	Nunavut, Canada	82.11	-67.48	2007	7	349	498	19	0.04	0.26	0.11	0.09	0.08	0.07	Gantner et al. (2010)
Lake F	Tundra permafrost	Nunavut, Canada	81.81	-69.35	2007	8	275	344	15	0.07	0.67	0.17	0.20	0.13	0.12	Gantner et al. (2010)
Lake G	Tundra permafrost	Nunavut, Canada	81.82	-69.18	2007	10	299	184	20	0.15	0.26	0.19	0.05	0.19	0.18	Gantner et al. (2010)
Char Lake	Tundra permafrost	Nunavut, Canada	74.71	-94.88	2010, 2011	13	332	278	10	0.15	0.48	0.32	0.11	0.30	0.34	Lescord et al. (2015)
Meretta Lake	Tundra permafrost	Nunavut, Canada	74.68	-94.92	2010, 2011	30	397	563	5	0.11	0.36	0.19	0.06	0.18	0.17	Lescord et al. (2015)
Resolute	Tundra permafrost	Nunavut, Canada	74.69	-94.95	2010, 2011	30	421	609	18	0.06	0.33	0.17	0.07	0.16	0.16	Lescord et al. (2015)
Kangiqsujuaq	Tundra permafrost	Nunavut, Canada	61.60	-71.96	1998	7	500	—	—	—	—	0.14	—	—	—	Lescord et al. (2015)
Sapphire	Tundra permafrost	Nunavut, Canada	75.36	-89.49	2001	9	448	606	23	0.16	1.07	0.45	0.29	0.39	0.36	Muir et al. (2005b)
Hovaktok	Tundra permafrost	Nunavut, Canada	68.05	-107.20	2006-2008	7	468	1581	11	0.02	0.08	0.04	0.02	0.04	0.05	Swanson and Kidd (2010)
Nauyuk	Tundra permafrost	Nunavut, Canada	68.38	-107.68	2006-2008	18	274	575	6	0.02	0.16	0.06	0.04	0.05	0.04	Swanson and Kidd (2010)
Roberts	Tundra permafrost	Nunavut, Canada	68.16	-106.52	2006-2008	34	237	210	4	0.01	0.04	0.01	0.01	0.01	0.01	Swanson and Kidd (2010)
Gavia Faeces	Tundra permafrost	Nunavut, Canada	68.34	-107.73	2006	8	289	211	12	0.05	0.51	0.17	0.18	0.11	0.08	Swanson et al. (2011b)
Keyhole	Tundra permafrost	Nunavut, Canada	69.38	-106.24	2006	7	449	1167	9	0.03	0.27	0.13	0.10	0.09	0.10	Swanson et al. (2011b)
Notgordie	Tundra permafrost	Nunavut, Canada	68.35	-107.66	2006	5	464	1134	9	0.15	0.34	0.24	0.90	0.23	0.19	Swanson et al. (2011b)

Heintzelman	Tundra permafrost	Nunavut, Canada	81.70	-66.93	2001	119	216	101	12	0.03	0.32	0.11	0.05	0.10	0.11	van der Velden et al. (2012)
Crazy Lake	Tundra permafrost	Nunavut, Canada	63.87	-68.47	2010	47	185	—	7	—	—	0.11	0.16	—	0.06	van der Velden et al. (2013a)
Iqalugaarjuit Lake	Tundra permafrost	Nunavut, Canada	66.57	-66.71	2004	20	185	65	6	0.07	0.30	0.12	0.05	0.11	0.11	van der Velden et al. (2013a)
Unnamed Lake	Tundra permafrost	Nunavut, Canada	72.59	-77.97	2010	18	254	304	10	0.08	0.42	0.23	0.11	0.20	0.19	van der Velden et al. (2013a)
Herbert	Tundra permafrost	Yukon, Canada	69.42	-139.63	1999	8	328	206	—	0.21	2.34	0.69	0.74	0.48	0.35	Gantner et al. (2010)
West Lake	Tundra permafrost	Nunavut, Canada	74.89	-109.60	2008, 2009	26	410	448	—	0.05	0.44	0.17	0.09	0.15	0.16	This study
East Lake	Tundra permafrost	Nunavut, Canada	74.89	-109.54	2008, 2009	17	406	327	—	0.05	0.29	0.11	0.06	0.10	0.10	This study
Arresjøen	Tundra permafrost	Svalbard, Norway	79.67	10.80	1996, 1997	25	336	442	16	0.04	0.44	0.18	0.11	—	0.12	Rognerud et al. (2002)
Lake 10	Tundra permafrost	Nunavut, Canada	68.14	-106.40	2006-2008	7	398	724	9	0.03	0.08	0.05	0.02	0.05	0.05	This study
Lake 32	Tundra permafrost	Nunavut, Canada	68.15	-106.52	2006-2008	12	338	375	11	0.02	0.23	0.09	0.05	0.08	0.08	This study
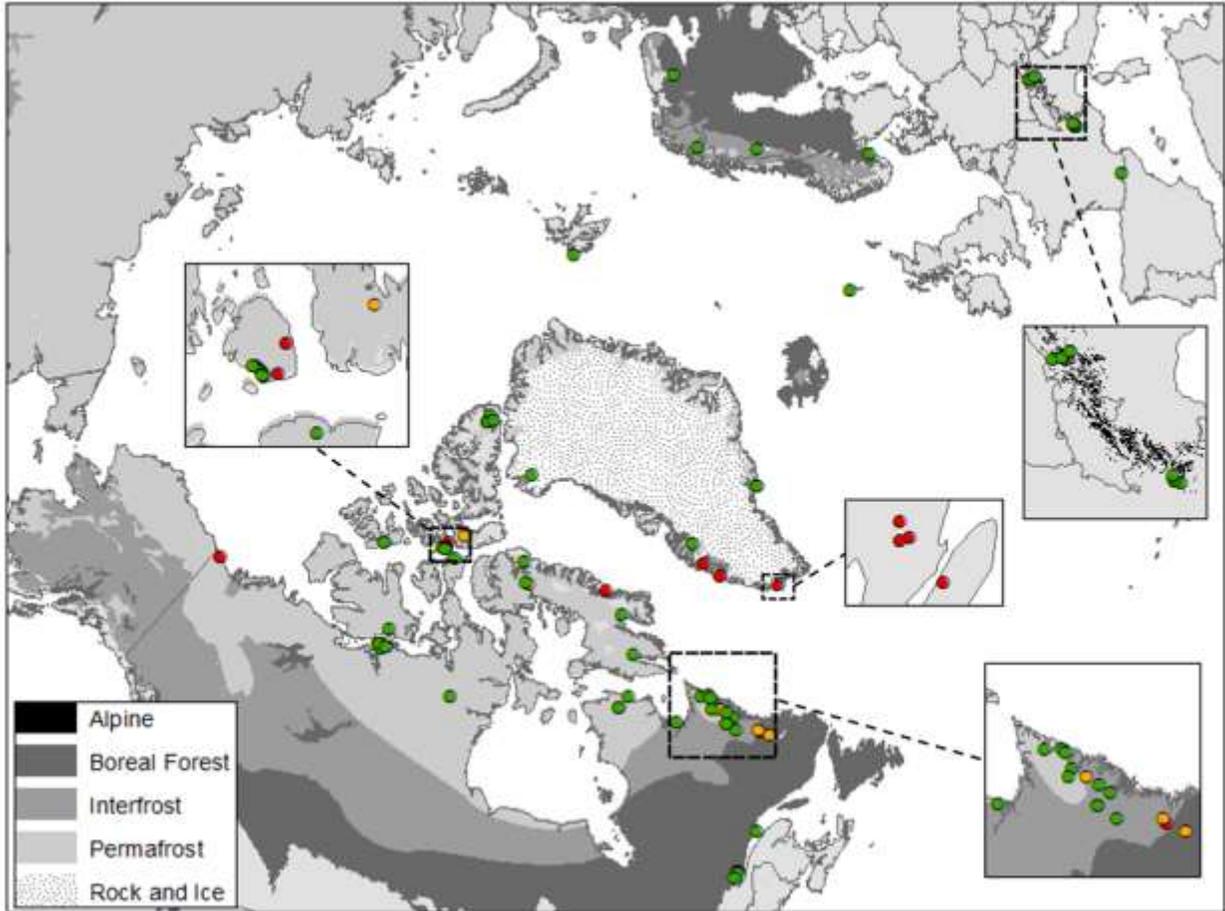
Figure 1
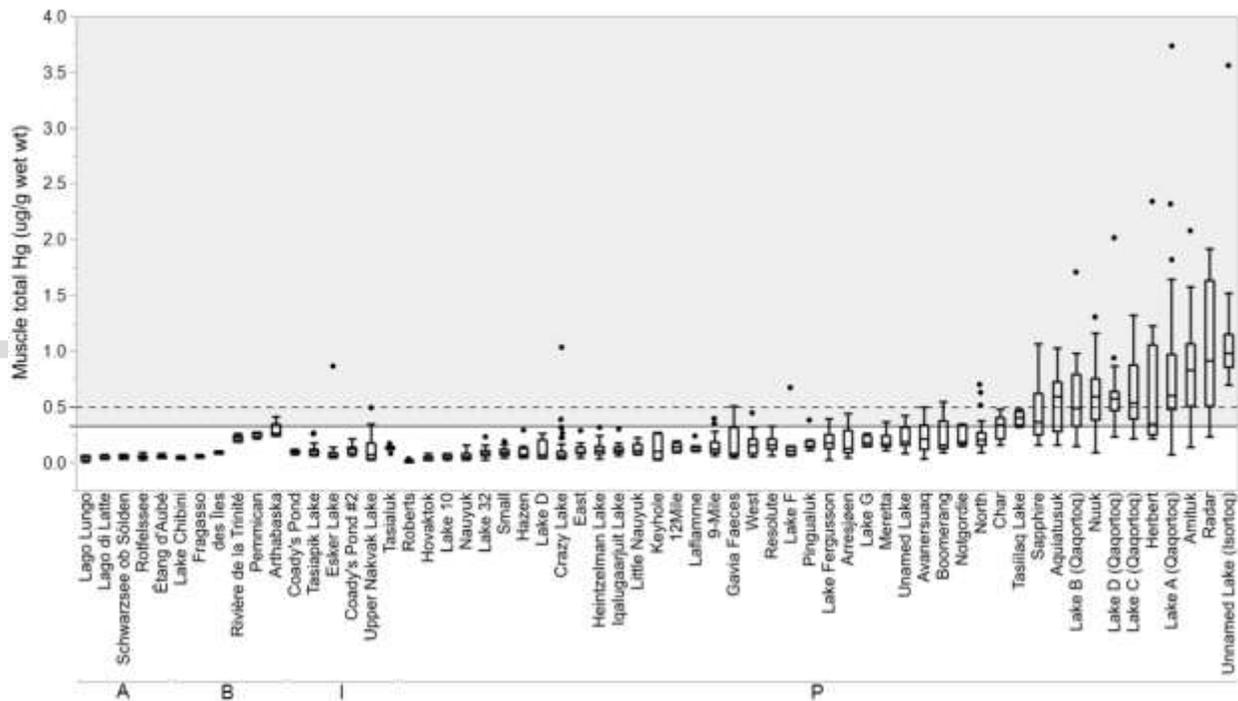
Figure 2

Figure 3

Figure 4