Factors affecting mercury concentrations in anadromous and non-anadromous Arctic charr (*Salvelinus alpinus*) from eastern Canada

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Author's declaration

I hereby declare that I am the sole author of this thesis. This is a true copy of the thesis,

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Abstract

Mercury concentrations in freshwater and marine biota are an ongoing concern, even in areas remote from local point sources, such as in the Canadian Arctic and sub-Arctic. Anadromous Arctic charr, which feed in the marine environment, have lower mercury concentrations than non-anadromous Arctic charr, which feed strictly in freshwater, but the two life-history forms have rarely been studied together, and the mechanisms driving the difference are unclear. Here, data from nine pairs of closely-located anadromous and non-anadromous Arctic charr populations were used to explore the impact of biological and life-history factors on individual total mercury concentration ([THg]) across a range of latitudes (49 – 81° N) in eastern Canada. From six of these sampling locations, additional samples of lower trophic level biota (i.e., algae, invertebrates, and forage fishes) were obtained in order to investigate patterns of total mercury (THg) and methylmercury (MeHg) biomagnification in the marine and lacustrine foodwebs supporting Arctic charr. Arctic charr mean [THg] ranged from 20 to 114 ng/g wet weight (ww) in anadromous populations, and was significantly higher in non-anadromous populations (all $p < 0.01$), ranging from 111 to 227 ng/g ww. Within-population variations in Arctic charr [THg] were best explained by fish age, and were also positively related to fork-length and $\delta^{15}N$-inferred trophic level. Across all sampling sites, the relationship between Arctic charr [THg] and fish age was significant and statistically similar in both life-history types, but only the non-anadromous fish demonstrated a significant relationship with trophic level. Fork-length and site latitude did not explain significant additional variation in Arctic charr [THg] across sampling

locations. Trophic magnification factors were 1.98 – 5.19 for THg and 3.02 – 6.69 for MeHg in lacustrine foodwebs, and 1.59 – 2.82 for THg and 2.72 – 5.70 for MeHg in marine foodwebs, and did not differ significantly between the two feeding habitats for either THg or MeHg. The biomagnification rate of MeHg exceeded that of THg in both habitats. Mercury concentrations at the base of the foodweb were higher in the lacustrine environment (estimated at 17 – 139 ng/g dw for THg and 5 – 42 ng/g dw for MeHg) than in the marine environment (8 – 39 ng/g dw for THg and 1 – 11 ng/g dw for MeHg). The proportion of mercury in the methylated form was related to trophic level, and the relationship was statistically similar in the lacustrine and marine habitats. There was no effect of site latitude on mercury concentrations in marine or lacustrine biota, thus the difference between feeding habitats was consistent across a range of latitudes (56 – 72°N) in eastern Canada. We conclude that a difference in prey mercury concentration, driven by differential mercury concentrations at of the base of the lacustrine and marine foodwebs, is important for explaining the difference in mercury concentration between anadromous and non-anadromous Arctic charr.

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Chapter 1

Background information

1.1 Arctic charr

      Arctic charr (*Salvelinus alpinus*) have a northern circumpolar distribution and are the only freshwater fish found in the high Arctic (Power et al. 2008). The natural distribution of Arctic charr is more widespread than any other salmonid fish, and natural populations are found in 16 countries (Maitland 1995). In North America, the latitudinal range of Arctic charr extends from approximately 43°N in Maine, USA to 82°N on Ellesmere Island, Nunavut (Johnson 1980; Power et al. 2008). Arctic charr exhibit both non-migratory lacustrine and anadromous life-history strategies throughout much of their range, although anadromy is considered to be a facultative trait, and declines toward both the northern and southern limits of the species' distribution (Doucett et al. 1999; Power et al. 2008). Anadromous individuals begin seaward migrations in May and June at three to eight years old, spend six to eight weeks feeding in the marine coastal environment, and return to freshwater to overwinter (Power et al. 2008).

      Arctic charr are long-lived fish, with individuals from some populations reaching 30 years or older (e.g., Guiguer et al. 2002), and one specimen from Aquiatusuk Lake, Cornwallis Island, Nunavut aged at 44+ years (Gantner et al. 2010a). Arctic charr display large variations in somatic growth rate and size at maturity (Johnson 1980; Klemetsen et al. 2003a; Parker and Johnson 1991), and often display bimodal size distributions or "standing waves" in length-frequency distributions (Johnson 1980; Power et al. 2008). Thus, Arctic charr of the same size can be of very different ages.

The somatic growth rate of individual Arctic charr is related to lake characteristics such as temperature (Larsson et al. 2005), altitude, depth, and microcrustacean density (Cavalli et al. 2002). Within populations, individual growth rate covaries with differences in morphotype (Reist et al. 1995), diet composition, and stomach fullness (Kahilainen and Lehtonen 2002). The somatic growth of Arctic charr tends to follow a sigmoidal pattern where life begins with a period of relatively slow growth, followed by rapid growth that may be associated with a change in diet to marine feeding (in anadromous individuals) or piscivory (Kahilainen and Lehtonen 2002). Anadromous Arctic charr generally have higher growth rates than lacustrine fish (Rikardsen et al. 2000).

The occurrence of more than one form of Arctic charr in a lake (polymorphism) has been well documented (Johnson 1980; Klemetsen et al. 2003a; Power et al. 2008), with up to four different Arctic charr morphotypes identified in a single lake (e.g., lake Thingvallavatn, Iceland; Malmquist et al. 1992; Sandlund et al. 1992). Different forms of Arctic charr can be differentiated by morphology, colouration, growth rate, size and age at maturity, life-history type, habitat use and diet, and time and place of spawning (Adams et al. 2003; Adams et al. 1998; Amundsen et al. 2008; Andersson 2003; Johnson 1980; Malmquist et al. 1992; Power et al. 2005; Power et al. 2008; Reist et al. 1995; Sandlund et al. 1992). Where multiple forms of Arctic charr coexist, there is often a small benthivorous and large pelagic planktivorous form, as well as a large piscivorous form (Power et al. 2008).

Arctic charr are opportunistic feeders, and can make use of a variety of food items where they are available. In the lacustrine environment, Arctic charr feed on benthic and pelagic invertebrates, including surface insects, chironomid larvae and

pupae, molluscs, amphipods, copepods, and cladocerans (Amundsen and Knudsen 2009; Cavalli and Chappaz 1996; Klemetsen et al. 2003a,b; Knudsen et al. 2007; Malmquist et al. 1992; Svenning et al. 2007). In many lakes, there is a shift to piscivory with larger size (e.g., Guiguer et al. 2002; Svenning et al. 2007), and cannibalistic feeding on smaller Arctic charr is not uncommon (Hammar 2000; Svenning and Borgstrom 1995). Arctic charr may feed almost exclusively on chironomids in high Arctic lakes (Johnson 1980). In the marine environment, the diet of anadromous Arctic charr includes fish (e.g., sand lance, *Ammodytes* spp.; capelin, *Mallotus villosus*; sculpins, *Myoxocephalus* spp.), amphipods, mysids, copepods, and krill (Dempson et al. 2002; Rikardsen et al. 2000). Non-anadromous Arctic charr feed throughout the year (Klemetsen et al. 2003a,b; Svenning et al. 2007). In contrast, anadromous Arctic charr obtain the majority of their nutrition during summer marine feeding forays, and the dietary contribution from lacustrine or riverine sources is minimal after the onset of anadromy (Johnson 1980; Rikardsen et al. 2003; Swanson et al. 2011a). The diet and habitat use of Arctic charr vary with life-history strategy (anadromous or non-anadromous), life-stage, time of year, and morphotype (Power et al. 2008).

Along with other northern fish species, Arctic charr are an important food resource for the Inuit people of northern Canada (Evans et al. 2005a). Consuming country food, including Arctic charr, provides nutritional, economic, and cultural benefits to northern people (Van Oostdam et al. 2005). In addition to their importance in subsistence fisheries, Arctic charr are also valuable for sport and commercial fishing (Evans et al. 2005a; Maitland 1995).

1.2 Mercury in the aquatic environment

Mercury (Hg) is a transition metal and has an atomic number equal to 80. Long-range transport of gaseous Hg in the atmosphere is responsible for conveying mercury around the globe, and to relatively pristine Arctic regions (AMAP/UNEP 2008; Munthe et al. 2011; Pacyna et al. 2010). Naturally, mercury is released to the atmosphere through volcanoes, geothermal vents, and rock weathering, as well as from previously contaminated soil and water (AMAP/UNEP 2008). Principle anthropogenic sources of mercury include coal combustion, waste incineration, mining, and metal smelting (Pacyna et al. 2006; Pacyna et al. 2010). Global mercury emissions have been relatively constant (between 1881 and 2235 tons/year) since 1980, with reduced European emissions balanced out by increasing emissions from Asia (Pacyna et al. 2006). By country, China is by far the largest emitter of mercury, followed by India and the United States (Pacyna et al. 2010). Mercury is primarily released to the atmosphere in the form of gaseous elemental mercury, Hg(0) (Pacyna et al. 2006), which can be oxidized to Hg(II) and then deposited to Arctic aquatic environments via precipitation or deposition onto ice and snow (Douglas et al. 2011; Munthe et al. 2011). Once deposited, Hg(II) can be converted to methylmercury (MeHg) by bacteria, primarily in anoxic water and sediments (Barkay and Poulain 2007; Morel et al. 1998; Ullrich et al. 2001).

In single-celled aquatic organisms, the uptake of mercury across the cell membrane is accomplished primarily via passive diffusion of the neutrally-charged chloride complexes $HgCl_2$ and CH_3HgCl (Mason et al. 1996). Thus the rate of mercury uptake is controlled by mercury speciation, which is ultimately determined by water

chemistry parameters such as pH, salinity, and the concentrations of chloride, sulphide, and total organic carbon and/or organic ligands (Lawson and Mason 1998; Morel et al. 1998; Ullrich et al. 2001; Watras et al. 1998). Biological factors such as zooplankton species composition (Chetelat and Amyot 2009; Pickhardt et al. 2005), phytoplankton cell size (Mason et al. 1996), algal concentration (Chen and Folt 2005; Pickhardt et al. 2005, 2002), and growth rate (Gorski et al. 2006; Karimi et al. 2007) are also important factors influencing [Hg] at the base of the foodweb. Furthermore, site-specific characteristics such as lake size and catchment area, lake productivity, underlying geology, surrounding land use, and temperature influence mercury concentrations in lacustrine biota (Chen et al. 2005; Evans et al. 2005b; Greenfield et al. 2001; Kamman et al. 2004; Lockhart et al. 2005; Marusczak et al. 2011; Rose et al. 1999; Schindler et al. 1995), as does atmospheric mercury deposition (Hammerschmidt and Fitzgerald 2006). Considering the abundance of confounding variables among sample sites, it is not surprising that mercury concentrations in water and sediments are often poorly correlated to fish mercury concentrations (Evans et al. 2005b; Gantner et al. 2010a; Kamman et al. 2004; Rose et al. 1999).

While direct uptake from water is an important source of Hg to organisms at the base of aquatic foodwebs (Morel et al. 1998), the vast majority of mercury accumulated by organisms at higher trophic levels (e.g., fish) is obtained through consumed food (Hall et al. 1997). Once it has been taken up by biota, mercury is well known to bioaccumulate in aquatic organisms and biomagnify in temperate and polar aquatic food webs (e.g., Cabana and Rasmussen 1994; Campbell et al. 2005; Gantner et al. 2010b; Jæger et al. 2009; Sharma et al. 2008; Wyn et al. 2009). van Leeuwen and Hermens

(1995) defined bioaccumulation as, "the net result of uptake, distribution and elimination of a substance due to all routes of exposure, i.e. exposure to air, water, soil/sediment and food," which includes bioconcentration, "the net result of uptake, distribution and elimination of a substance due to water-borne exposure of an organism" and biomagnification, "the accumulation and transfer of chemicals via the food web (e.g. algae-invertebrate-fish-mammal) due to ingestion, resulting in an increase of the internal concentration in organisms at the succeeding trophic levels." Due to biomagnification, mercury concentrations in higher trophic level biota are positively related to food chain length (Cabana and Rasmussen 1994; Cabana et al. 1994; Schindler et al. 1995).

Methylmercury is more efficiently transferred from prey to predator than inorganic mercury (Mason et al. 1995; Morel et al. 1998), thus the proportion of mercury in the methylated form increases with trophic level in aquatic foodwebs (Douglas et al. 2011). Methylmercury is also considered the most toxic form of Hg, causing reproductive deficiencies and behavioural changes in wildlife (Scheulhammer et al. 2007), and developmental neurotoxicity and cardiovascular effects in humans (Mergler et al. 2007).

1.3 Mercury in fish

High mercury concentrations in fish tissue are an ongoing concern in the Canadian Arctic, as the levels in desirable food species may approach or exceed recommended guidelines for human consumption (Evans et al. 2005a,b; Lockhart et al. 2005; Swanson et al. 2011b). The Health Canada guideline for mercury in commercially sold fish is 0.5 ppm (500 ng/g wet weight), and the recommended

guideline for sensitive consumers (i.e., women of childbearing age, children, and frequent fish consumers) is 0.2 ppm Hg (200 ng/g wet weight) (Health Canada 2007).

In addition to the site-specific abiotic and biotic factors mentioned in the previous section, individual biological characteristics influence fish total mercury concentration ([THg]). Mercury concentrations in non-anadromous Arctic charr have been positively related to fish age, size, and trophic level (Gantner et al. 2010a, 2009; Muir et al. 2005; Riget et al. 2000; Rognerud et al. 2002), but relationship strengths vary among populations and are usually not examined in anadromous Arctic charr (Riget et al. 2000 is an exception). Life-history strategy (i.e., feeding habitat) is also a key determinant of mercury concentration, with anadromous Arctic charr having lower measured [THg] than non-anadromous conspecifics (Evans et al. 2005a; Lockhart et al. 2005; Riget et al. 2000; Swanson et al. 2011b).

It has been suggested that by increasing somatic growth rates, [THg] in fish can be reduced without changing the mercury concentration in fish prey items (Lepak et al. 2009; Verta 1990). A negative relationship between total mercury concentration and somatic growth rate has been observed in fish species including: juvenile Atlantic salmon, *Salmo salar* (Ward et al. 2010), walleye, *Sander vitreus* (Simoneau et al. 2005), yellow perch, *Perca flavescens* (Essington and Houser 2003), and northern pike, *Esox lucius* (Sharma et al. 2008; Verta 1990). Similarly in Arctic charr, Hammar et al. (1993) found higher levels of PCB and DDE contaminants in a slow-growing dwarf form than in faster-growing "normal" charr. Therefore, the lower [THg] observed in anadromous, relative to non-anadromous, Arctic charr may be related to the higher rate of somatic growth evident in anadromous fish (Rikardsen et al. 2000). However, while somatic

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growth rate may influence fish [THg], it is considered less important than the mercury concentration in fish diet (Essington and Houser 2003; Henery et al. 2010; Lindqvist et al. 1991; Verta 1990; Ward et al. 2010), and the effects of growth rate changes on fish [THg] can be masked when increases in size are associated with a shift in diet (Lepak et al. 2009).

1.4 Applications of stable isotope analysis

Stable isotope analysis provides useful information for a wide variety of ecological studies. For example, mapping isotopes in soil or water can reveal patterns of nutrient cycling at the landscape level, sulphur and oxygen isotope signatures can be used to track animal migrations, and stable carbon and nitrogen isotopes can demonstrate complex foodweb relationships (Fry 2006).

The carbon and nitrogen isotope ratios of an organism reflect its average feeding pattern, as opposed to stomach content analysis which provides a snapshot of recently consumed prey items (Vinson and Budy 2011). In aquatic foodwebs, the nitrogen isotope ratio ($\delta^{15}N$) increases by approximately 3.4‰ between a consumer and its food source and provides a quantitative measure of trophic level (Minagawa and Wada 1984; Post 2002). In contrast, carbon stable isotope values ($\delta^{13}C$) increase by a small amount between trophic levels (< 1‰; Post 2002; Vander Zanden and Rasmussen 2001), but can differentiate between benthic and pelagic (Hecky and Hesslein 1995) or marine and freshwater (Doucett et al. 1999; Kim and Rochford 2008) carbon sources. Stable isotopes have been used to distinguish between Arctic charr morphotypes displaying trophic polymorphism (Adams et al. 2003; Guiguer et al. 2002; McCarthy et al. 2004;

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Power et al. 2005), and can differentiate between anadromous and non-anadromous individuals (Doucett et al. 1999). To compare consumer δ^{13}C and δ^{15}N between sites, stable isotope values must be corrected for differences in isotope signatures at the base of the foodweb, which can vary substantially between locations as a result of anthropogenic or biogeochemical influences (Post 2002).

Because they reflect the average assimilated diet of a consumer (Peterson and Fry 1987), δ^{13}C and δ^{15}N isotopes provide key information explaining mercury concentrations in upper trophic level consumers (e.g., fish) that obtain mercury primarily through consumed food (Hall et al. 1997). Mercury concentrations in fish tend to increase with increasing δ^{15}N or trophic level (Cabana and Rasmussen 1994; Kidd et al. 1995), and have been inversely related to δ^{13}C or reliance on benthic food sources (Kidd et al. 2003; Lavoie et al. 2010; Power et al. 2002). Stable isotopes have also been widely used to quantify the rate of mercury biomagnification in aquatic foodwebs, using δ^{15}N or δ^{15}N-inferred trophic level versus log [THg] or log [MeHg] (Borgå et al. 2012; Jardine et al. 2006).

1.5 Research objectives

Previous studies examining [THg] in Arctic charr have often been based on small sample sizes of large fish, and have usually focused on characterizing spatial and/or temporal trends among populations (e.g., Evans et al. 2005a; Gantner et al. 2010a; Lockhart et al. 2005; Muir et al. 2005; Riget et al. 2000; Swanson et al. 2011b). Less is known about the biological and life-history factors impacting individual [THg] within and among populations, particularly for anadromous Arctic charr. Furthermore, mercury

concentrations of anadromous and non-anadromous Arctic charr have been studied together at only a few locations: the coast of Labrador, Canada (Bruce and Spencer 1979), near Qaqortoq in southern Greenland (Riget et al. 2000), Kangiqsujuaq, northern Quebec, Canada (Lockhart et al. 2005), and the West Kitikmeot region, Nunavut, Canada (Swanson et al. 2011b). In view of the limited knowledge about large scale spatial differences in, and the causal mechanisms behind, known differences in [THg] between anadromous and non-anadromous Arctic charr, this research compares latitudinally-paired populations of anadromous and non-anadromous Arctic charr to address a number of critical hypotheses as outlined below.

In chapter 2, the impacts of biological factors influencing Arctic charr [THg] across a range of latitudes (49 – 81° N) in eastern Canada are explored using nine pairs of co-located anadromous and non-anadromous Arctic charr populations (Table 1.1). Specifically, the following hypotheses are tested:

1. Anadromous Arctic charr have lower [THg] than conspecifics from closely located or sympatric non-anadromous populations, and this difference is consistent across a range of latitudes in eastern Canada

2. Within-population differences in [THg] are positively related to individual age, fork-length, and δ^{15}N-inferred trophic level, and negatively related to average somatic growth rate and δ^{13}C-inferred benthic connection, with relationships being statistically similar in anadromous and non-anadromous populations

3. Among-population differences in [THg] are positively related to differences in age, fork-length, and trophic level, and negatively related to average somatic growth rate and site latitude

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In chapter 3, data from six spatially-paired marine and lacustrine foodwebs along a latitudinal gradient in eastern Canada (Table 1.1) are used to examine mercury accumulation patterns in feeding habitats important for Arctic charr. The specific hypotheses addressed are:

1. Biomagnification rates (quantified using the slope of mercury concentration versus δ^{15}N-inferred trophic level) are lower in marine foodwebs than in spatially-proximate lacustrine foodwebs

2. Mercury concentrations at the base of the foodweb are lower in the marine than in the lacustrine feeding habitats

3. The proportion of methylated mercury increases with trophic level, therefore biomagnification rates are higher for MeHg than THg, and the trend is similar in both feeding habitats used by Arctic charr

4. The differences between feeding habitats are consistent across a range of latitudes (56 – 72°N) in eastern sub-Arctic and Arctic Canada

1.6 Tables

Table 1.1. Location of sample collection sites for anadromous and non-anadromous Arctic charr.

Type	Sample site	Region	Latitude (N)	Longitude (W)	Sampling year(s)	n[b]
Anadromous						
	Rivière de la Trinité	Quebec	49°25'	67°18'	2010	10
	Fraser River, Nain Bay[a]	Labrador	56°37.072'	62°15.243'	2008	20
	Okak Bay[a]	Labrador	57°33.250'	62°05.914'	2008	20
	Saglek Bay (North Arm)[a]	Labrador	58°32'41.5"	63°27'30.6"	2007	22
	Nepihjee River, Dry Bay[a]	Ungava Bay, Quebec	58°32'	68°17'	2009, 2010	33
	Frobisher Bay, Iqaluit[a]	Baffin Is., Nunavut	63°41'22.5"	68°25'17.2"	2004, 2010	30
	Pangnirtung Fjord	Baffin Is., Nunavut	66°8'15.2"	65°45'11.2"	2004, 2009	40
	Salmon River, Pond Inlet[a]	Baffin Is., Nunavut	72°40'46.3"	78°3'11.2"	2005-08, 2010	70
	Heintzelman Lake	Ellesmere Is., Nunavut	81° 42'	66° 56'	2001	7
Non-anadromous						
	Rivière de la Trinité	Quebec	49°25'	67°18'	2009, 2010	10
	Coady's Pond #2, Nain Bay[a]	Labrador	56°38'32.5"	63°37'33.9"	2007	20
	Esker Lake, Okak Bay[a]	Labrador	57°9'14.8"	62°52'39.4"	2008	20
	Upper Nakvak Lake, Saglek Bay[a]	Labrador	58°39'46.1"	63°18'59.8"	2007	20
	Tasiapik Lake, Dry Bay[a]	Ungava Bay, Quebec	58°31'	68°21'	2009, 2010	31
	Crazy Lake, Iqaluit[a]	Baffin Is., Nunavut	63°52'13.4"	68°28'10.5"	2010	47
	Iqalugaarjuit Lake, Pangnirtung	Baffin Is., Nunavut	66°34'21.8"	66°42'48.5"	2004	19
	unnamed lake, Pond Inlet[a]	Baffin Is., Nunavut	72°35'40.0"	77°58'19.8"	2010	18
	Heintzelman Lake	Ellesmere Is., Nunavut	81°42'	66°56'	2001	112

[a]Additional samples of lower trophic level biota (i.e., algae, invertebrates, and forage fishes) were obtained in order to test hypotheses regarding mercury biomagnification in the marine and lacustrine foodwebs supporting Arctic charr.
[b]The total number of Arctic charr analyzed for [THg] from all sampling years.

Chapter 2

Comparing total mercury concentrations in anadromous and non-anadromous Arctic charr (*Salvelinus alpinus*) from eastern Canada

2.1 Introduction

Mercury concentrations in Arctic biota have increased by an order of magnitude since pre-industrial times (Dietz et al. 2009), and represent an ongoing threat to the health of Arctic ecosystems and inhabitants (Dietz et al. 2011; Stow et al. 2011). This is particularly concerning for species such as Arctic charr (*Salvelinus alpinus*), which are consumed regularly by northern people and are culturally and economically significant (Evans et al. 2005a). The primary route of exposure to mercury is through diet, thus contamination of traditional country foods has critical implications for human health (Stow et al. 2011; Van Oostdam et al. 2005).

Arctic charr have a holarctic distribution that extends farther north than that of any other freshwater fish species, and as far south as 43°N in North America (Power et al. 2008). Where access to the sea exists, anadromous Arctic charr migrate to nearshore coastal areas for summer feeding, and return to freshwater to overwinter. However, sympatric populations of anadromous and lake-resident Arctic charr are not uncommon (Klemetsen et al. 2003). The prevalence of anadromy is related to latitude, declining to the northern and southern extremes of the species distribution (Power et al. 2008).

Total mercury concentrations ([THg]) in Arctic charr are variable, both within and among populations (AMAP 2005; Evans et al. 2005a; Lockhart et al. 2005). In non-

anadromous Arctic charr, within-population [THg] has been positively related to fish age, size, and trophic level (Gantner et al. 2010a, 2009; Muir et al. 2005; Riget et al. 2000; Rognerud et al. 2002; Swanson et al. 2011a), but relationship strengths vary among populations and are usually not examined in anadromous Arctic charr (Riget et al. 2000 is an exception). Anadromous Arctic charr have higher somatic growth rates than non-anadromous conspecifics (Rikardsen et al. 2000), and high rates of somatic growth have been associated with low [THg] in some fish species (e.g., Doyon et al. 1998; Ward et al. 2010).

Anadromous Arctic charr generally have lower [THg] than non-anadromous conspecifics (AMAP 2005; Evans et al. 2005a; Lockhart et al. 2005). However, the two life-history forms have been studied together at only a few sites: the coast of Labrador, Canada (Bruce and Spencer 1979), near Qaqortoq in southern Greenland (Riget et al. 2000), Kangiqsujuaq, northern Quebec, Canada (Lockhart et al. 2005), and the West Kitikmeot region, Nunavut, Canada (Swanson et al. 2011a). No wide-scale spatial investigation of differential [THg] in closely-located populations of anadromous and non-anadromous Arctic charr of which we are aware has been conducted. Furthermore, the mechanisms driving the difference in mercury concentration between the two life-history forms are unknown, though they may be related to differing fish ages or somatic growth rates (Evans et al. 2005a,b; Swanson et al. 2011a), trophic levels (Kim and Rochford 2008), or exposure concentrations in the marine versus freshwater feeding habitats (Mason et al. 2006).

The vast majority of mercury in fish is obtained from ingested food (Hall et al. 1997). Thus stable isotope ratios of carbon (δ^{13}C) and nitrogen (δ^{15}N) are potentially

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useful in explaining differential mercury concentrations, as they reflect the average assimilated diet of a consumer (Peterson and Fry 1987). In aquatic foodwebs, $\delta^{15}N$ is enriched by an average of 3.4‰ in a consumer relative to its diet, and provides a quantitative measure of trophic level (Jardine et al. 2006; Minagawa and Wada 1984; Post 2002). In contrast, $\delta^{13}C$ changes by only a small amount (~0.4‰, Post 2002) between trophic levels, and can distinguish between benthic and pelagic sources of carbon in aquatic environments (Hecky and Hesslein 1995). Mercury concentrations in fish tend to increase with increasing $\delta^{15}N$ or trophic level (Cabana and Rasmussen 1994; Kidd et al. 1995), and have been inversely related to $\delta^{13}C$ or reliance on benthic food sources (Kidd et al. 2003; Lavoie et al. 2010; Power et al. 2002).

In view of the limited knowledge about the large scale spatial differences in [THg] between anadromous and non-anadromous Arctic char, we explore the impact of biological factors influencing Arctic charr [THg] across a range of latitudes (49 – 81° N) in eastern Canada. Specifically we test the following hypotheses: [1] anadromous Arctic charr have lower [THg] than conspecifics from closely located or sympatric non-anadromous populations, and that this difference is consistent across a range of latitudes in eastern Canada; [2] within-population differences in [THg] are positively related to individual age, fork-length, and $\delta^{15}N$-inferred trophic level, and negatively related to average somatic growth rate and $\delta^{13}C$-inferred benthic connection, with relationships being statistically similar in anadromous and non-anadromous populations; and [3] among-population differences in [THg] are positively related to differences in age, fork-length, and trophic level, and negatively related to average somatic growth rate and site latitude.

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2.2 Methods

2.2.1 Sample collection

Nine sampling locations were selected to represent a range of latitudes in eastern Canada from Rivière de la Trinité, Quebec (49° N) in the south to Heintzelman Lake, Ellesmere Island (81° N) in the north (Figure 2.1). At each location, matching anadromous and non-anadromous Arctic charr populations were sampled, either from near-by locales containing discrete life-history form populations, or from a single site where the two life-history forms co-occurred. Distances between paired anadromous and non-anadromous sampling sites (0 to 68 km) were minimized to remove possible confounding spatial factors (e.g., airborne depositional Hg gradients), with the proximity of sample sites being controlled by population occurrences and the realities of sampling logistics. Details on sampling sites are provided in Table 2.1, including the known fish assemblages for the lakes.

Arctic charr samples were collected between 2001 and 2010 from purpose designed field programs or in conjunction with Government of Canada monitoring programs, often with the aid of local Hunters and Trappers Organizations (HTOs). Sample sizes ranged from 7 to 112 individual fish from a particular location and for a given life-history type (Tables 2.3 and 2.4). All non-anadromous Arctic charr, except for those from Rivière de la Trinité, were captured from lakes using multi-mesh nylon multifilament sinking gillnets as described by Johnson (1983) set perpendicular from shore in open water. Anadromous and non-anadromous fish were captured during the spring (May) migration in the Rivière de la Trinité using a fyke trap, with previously non-

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anadromous fish being intercepted prior to reaching the marine environment. Anadromous Arctic charr were collected from the nearshore marine environment using 114, 127, and 140 mm mesh gillnets or angling, and were obtained from local subsistence fishers at Pond Inlet, Pangnirtung, and Iqaluit. Anadromous Arctic charr from Dry Bay were trapped while migrating upstream in a fyke net set in the Nepihjee River. Anadromous individuals were captured concurrently with non-anadromous fish in Heintzelman Lake and Iqalugaarjuit Lake (Pangnirtung).

Samples of primary consumers were collected from as many sampling sites as possible (7 of 9 marine and 7 of 9 freshwater sites) in order to correct for differences in the isotope baseline, which can vary considerably among sampling sites (Post 2002). Benthic macroinvertebrates were used (filter-feeding bivalves or other primary consumers, Table 2.1), as they provide a time-integrated measure of basal δ^{13}C and δ^{15}N values (Post 2002). Invertebrates were collected by hand or using forceps from rocks or pools in the littoral zone (snails, mussels, clams, barnacles, and gammarids) or by using dipnets and a kick-sweep method at 0.5 – 1 m depth (chironomid larvae). All invertebrate samples were frozen and transported to the laboratory for stable isotope analysis.

In some cases, whole and undigested invertebrates were opportunistically sampled from Arctic charr stomachs, which can provide a useful source of prey items for stable isotope analyses (Peterson 1999; Tieszen et al. 1983). To guard against potential biases imparted by digestion, only intact dietary items obtained directly from the stomach were used (Guelinckx et al. 2008), with the influence of digestion on stable isotope values of such prey items having been shown to be negligible in Arctic charr

dietary studies from Loch Ness (Grey et al. 2002). To further minimize the influence of digestive tract material on prey stable isotope values, all stomach-derived prey items were rinsed with deionized water prior to analysis.

2.2.2 Laboratory analyses

All collected Arctic charr were processed immediately after capture or were frozen and transported to the laboratory for processing. Each fish specimen was weighed whole (g) and measured for fork-length (mm). A sample of dorsal muscle tissue (≈ 5 g) was removed from the left side of each fish in the region dorsal to the lateral line and posterior to the dorsal fin. Muscle samples were kept frozen prior to use in stable isotope and mercury analyses.

Sagittal otoliths were removed from all Arctic charr, and fish ages were determined using standardized methods by Fisheries and Oceans Canada in Winnipeg, Canada, and cross validated with aging conducted at the University of Waterloo, Waterloo, Canada. Ageing methods included visual inspection of whole otoliths under reflected light, break and burn (Reist et al. 1995), or thin-section techniques. Otoliths to be sectioned were embedded in epoxy (EPO-THIN resin 20-8140-032 and hardener 20-8142-016, Buehler Ltd., Lake Bluff, Illinois) mixed according to the manufacturer's directions, and left to harden for one week. Embedded otoliths were cut through the nucleus perpendicular to the sulcus using a Buehler Isomet 1000 precision sectioning saw (Buehler Ltd., Lake Bluff, Illinois) outfitted with a Buehler diamond wafering blade (152 mm in diameter, 0.5 mm thick, No. 11-4246, series 15 HC), and a speed set to 575

rpm. The micrometer was advanced 1.0 mm and a 0.5 mm section was cut from the otolith, which was then examined under reflected light.

Arctic charr life-history form (anadromous or non-anadromous) was determined from sampling location in the case of landlocked fish, or anadromous Arctic charr caught at sea or while conducting upstream migrations in late summer. In the case of sympatric populations of anadromous and lake-resident Arctic charr (Rivière de la Trinité, Iqalugaarjuit Lake, unnamed lake near Pond Inlet, and Heintzelman Lake) life-history strategy was assessed using the stable sulphur isotope ratio (δ^{34}S) of dorsal muscle tissue (Doucett et al. 1999), and confirmed using scanning proton microprobe analysis of sagittal otoliths, using methods described in Loewen et al. (2009).

Total mercury concentrations of individual Arctic charr were quantified via thermal decomposition and atomic absorption spectroscopy following U.S. EPA method 7473 (U.S. Environmental Protection Agency 2007) using a Milestone Direct Mercury Analyzer, DMA-80 (Milestone S.r.l., Sorisole, Italy). A subsample of 0.1 – 0.2 g of frozen, non-homogenized dorsal muscle tissue was used for analysis, and the results are expressed as ng/g wet weight (ww). Certified reference materials and blanks were run at the beginning and end of each batch of 20 samples. The method detection limit, determined as 3x the standard deviation of the blanks, was 0.33 ng Hg. One sample was run in triplicate during each sample batch, and the mean relative standard deviation of the triplicates was 4.8%. The reference materials used and percent recoveries (mean percentage of certified value ± 1 standard deviation) were NIST 1566b (89.9 ± 5.4) and NIST 2976 (98.6 ± 6.4) from the National Institute of Standards and Technology (Standard Reference Materials Program, Gaithersburg, USA), as well as

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TORT-2 (100.8 ± 2.5) and DORM-1 (96.2 ± 8.3) from the National Research Council

Canada (Institute for National Measurement Standards, Ottawa, Canada).

Stable isotope analyses were conducted using dorsal muscle tissue for each

Arctic charr specimen, and whole bodies of invertebrates (bivalves, gastropods, and

barnacles were first removed from shells). Small invertebrates (e.g., chironomid larvae)

were pooled to obtain sufficient mass for a single sample (≈ 0.1 – 0.5 g ww), while

larger invertebrates were analyzed individually. Tissue samples were oven dried at

50°C for 48 hours, homogenized using a Retsch MM 301 ball mill grinder (Retsch

GmbH, Haan, Germany), and weighed into tin capsules using a high-precision analytical

balance (UMX2, Mettler-Toledo GmbH, Greifensee, Switzerland). Stable isotope

analyses were conducted at the Environmental Isotope Laboratory at the University of

Waterloo, Waterloo, Canada following standard protocol. Briefly, a Delta Plus

Continuous Flow Stable Isotope Ratio Mass Spectrometer (Thermo Finnigan, Bremen,

Germany) coupled to a Carlo Erba Elemental Analyzer (CHNS-O EA1108, Carlo Erba,

Milan, Italy) was used for simultaneous determination of δ^{13}C and δ^{15}N. Analysis of δ^{34}S

was completed using an Isochrom Continuous Flow Stable Isotope Ratio Mass

Spectrometer (GVInstruments, Micromass, Manchester, UK) coupled to a Costech

Elemental Analyzer (CNSO 4010, Costech Analytical Technologies Inc., Valencia,

USA). All stable isotope ratios are expressed using standard delta (δ) notation as parts-

per-thousand differences from recognized international standards, with a precision in

organic material of ± 0.2‰ for carbon, ± 0.3‰ for nitrogen, and ± 0.5‰ for sulphur.

2.2.3 Data analysis

To compare trophic positions of Arctic charr from different sampling sites, mean $\delta^{15}N$ values of primary consumers were used to assess baseline values ($\delta^{15}N_{base}$) for each site (Table 2.1). Arctic charr trophic level was calculated using site-specific $\delta^{15}N_{base}$ following:

Trophic level$_{Arctic\ charr}$ = 2 + ($\delta^{15}N_{Arctic\ charr} - \delta^{15}N_{base}$) / 3.4

While the trophic fractionation of $\delta^{15}N$ ($\Delta\delta^{15}N$) is variable (e.g., -2.1 to +5.4‰, McCutchan et al. 2003; -0.7 to +9.2‰, Vander Zanden and Rasmussen 2001; -1.16 to +5.89‰, Vanderklift and Ponsard 2003), the average value of 3.4‰ provides a reasonable approximation of $\Delta\delta^{15}N$ when averaged across multiple trophic levels, particularly when a primary consumer is used for the baseline value (Jardine et al. 2006; Post 2002; Vander Zanden and Rasmussen 2001). In order to evaluate the potential bias associated with using an estimated $\Delta\delta^{15}N$ value, all statistical analyses involving trophic level were also carried out using baseline-adjusted $\delta^{15}N$, calculated as:

Adjusted $\delta^{15}N_{Arctic\ charr}$ = $\delta^{15}N_{Arctic\ charr} - \delta^{15}N_{base}$

The use of baseline-adjusted $\delta^{15}N$ rather than trophic level produced no difference in the results of any statistical significance testing, presumably because the conversion to trophic level is a scalar transformation that does not affect the relative position of data points. Therefore, the results presented herein are based on calculations with trophic level.

A marine $\delta^{15}N_{base}$ was used to assess the trophic level of anadromous fish, because migratory Arctic charr feed primarily in the marine environment, and the dietary contribution from lacustrine or riverine sources is minimal after the onset of anadromy

(Johnson 1980; Rikardsen et al. 2003; Swanson et al. 2011b). Sampling was not

conducted in the Gulf of St. Lawrence, the marine feeding environment of the Rivière de

la Trinité anadromous Arctic charr, therefore a literature value was used for $\delta^{15}N_{base}$.

The $\delta^{15}N$ (7.2 ± 0.1‰) for blue mussel (*Mytilus edulis*) was selected from Lavoie et al.

(2010), a figure that agrees well with previously reported values of 7.4‰ and 7.3‰ for

blue mussel from the St. Lawrence Estuary and Gulf of St. Lawrence (Lesage et al.

2001). Trophic levels were not calculated for Arctic charr from one anadromous

population (Heintzelman Lake) and 2 non-anadromous populations (Iqalugaarjuit Lake,

Pangnirtung and Heintzelman Lake) due to lack of suitable baseline signature samples.

All statistical analyses were conducted using the R program for statistical

computing (R Development Core Team 2009) with Type I error set to $\alpha = 0.05$. Total

mercury concentration data were natural-log transformed prior to inclusion in analyses

in order to satisfy assumptions of normality. Normality was confirmed using a Shapiro-

Wilk test (Shapiro and Wilk 1965). Means of two groups were compared using t-tests

adjusted for variance equality or inequality (Zar 2010). Variances of two groups were

compared using an F-test when the data were normally distributed, and a Levene's test

using the median when the data were not normally distributed. Relationships between

two variables were assessed using Pearson's correlation coefficients and simple linear

regression. Partial correlation analysis was used to evaluate the significance of a single

term while other factors were held constant (e.g., Zar 2010).

The effect of fork-length, given a constant age, was used to explore the

significance of average somatic growth rate (i.e., individuals with a faster average

growth rate are larger at a given age than slower-growing conspecifics) within and

among Arctic charr populations. To further visualize the effect of average somatic growth rate on mercury concentrations among populations, the age versus ln [THg] and ln age versus ln fork-length relationships for each population were used to estimate population-specific ln [THg] and ln fork-length at a standardized age. Age 9 was selected, as it was within the ranges of most anadromous and non-anadromous populations, so no extrapolation was required. The exception was the Rivière de la Trinité populations, for which age-standardized ln [THg] and fork-length were not possible owing to the limited age ranges (anadromous = 4 – 6 years, non-anadromous = 5 – 6 years) and fork-length ranges (anadromous = 239 – 339 mm, non-anadromous = 181 – 202 mm) that did not overlap with many of the other populations. Plots of age versus length for all populations are provided in Appendix 1 (Figure S1).

A general linear model was conducted to explain individual ln [THg] using fish age, fork-length, and trophic level (covariates), as well as life-history type and sampling location (factors). In order to evaluate the effects of biological and life-history factors on fish mercury concentration, regardless of sampling site, linear mixed models were employed. Mixed models were conducted within each life-history type to explain individual ln [THg] using fish age, trophic level, and site latitude. A final model was conducted using Arctic charr from both life-history types, and described individual ln [THg] as a function of fish age and trophic level (inner covariates), life-history type (fixed effect factor), and site latitude (outer covariate). Interaction terms were included for age x type, trophic level x type, and latitude x type to determine whether the relationships differed between the anadromous and non-anadromous life-history types. Sample site was included as a random-effect factor in the mixed models to allow for variations in the

slopes of ln [THg] versus age and trophic level, as well as the model intercept, among sampling sites.

2.3 Results

Individual anadromous Arctic charr had [THg] ranging from 9 to 217 ng/g ww in muscle, while the concentrations in non-anadromous fish ranged from 28 to 1033 ng/g ww. Only 2 of the 549 individuals analyzed had [THg] greater than the Health Canada limit for mercury concentration in commercially sold fish (500 ng/g ww), and both were non-anadromous. The [THg] of 40 individuals exceeded the Health Canada guideline for frequent fish consumers (200 ng/g ww); of these, 38 fish were non-anadromous and 2 fish were anadromous. These guidelines are the same as those recommended by the World Health Organization for internationally marketed fish (500 ng/g ww) and for fish consumed by sensitive groups (i.e., women of childbearing age, children, and frequent fish consumers; 200 ng/g ww). Similarly, all anadromous and most non-anadromous Arctic charr had muscle mercury concentrations lower than the U.S. Food and Drug Administration guideline (1000 ng/g ww) and more conservative U.S. Environmental Protection Agency guideline (300 ng/g ww) for mercury concentrations in fish.

Location-specific mean [THg] ranged from 20 to 114 ng/g ww in anadromous populations, and from 111 to 227 ng/g ww in non-anadromous populations (Table 2.2, Figure 2.2). The non-anadromous populations had mean [THg] 1.8 to 6.3 times higher than their paired anadromous counterparts, and there was a significant difference in ln [THg] for each pair (two-sample t-tests; all $p < 0.01$; Table 2.2). There was no

significant relationship between latitude and the difference in mean [THg] between the two life-history types (p = 0.88).

The anadromous Arctic charr were significantly larger than non-anadromous conspecifics at all locations except Nain Bay (two-sample t-test; p = 0.17), but there was no consistent difference in mean age between the two life-history types (Table 2.2; see also Appendix 1 Figure S1). At most locations, the anadromous fish occupied a higher mean trophic level than the non-anadromous conspecifics where a significant difference existed, although the opposite was true at Dry Bay (two-sample t-test; p < 0.001; Table 2.2).

Within-population ln [THg] data were positively related to both fish age and fork-length, and the relationships tended to be stronger in the non-anadromous populations (Figures 2.3 and 2.4; Table 2.3). Fish age was significantly related to ln [THg] in 6 of 9 anadromous and 7 of 9 non-anadromous populations (Figure 2.3; Table 2.3). Significant positive relationships existed between ln [THg] and fork-length in 3 of 9 anadromous and 7 of 9 non-anadromous populations (Figure 2.4; Table 2.3). Partial correlation analysis revealed that fish age was a better predictor of within-population ln [THg] than fork-length when controlling for the effect of other factors, a pattern that held true for both life-history types (Table 2.4). Significant relationships between ln [THg] and $\delta^{15}N$ or $\delta^{13}C$ occurred in approximately half of the studied populations, with the magnitude and even direction of the relationships varying among populations (Table 2.3; see also Figure 2.5 and Appendix 1 Figures S2, S3, and S4). Within a given sampling location, anadromous Arctic charr usually had lower [THg] than non-anadromous conspecifics at a similar age, fork-length, or trophic level (Figures 2.3, 2.4,

and 2.5), thus the difference in mercury concentration between the two life-history types could not be attributed to differences in age, size, or trophic level.

Little evidence of a negative effect of average somatic growth rate on Arctic charr [THg] was found, either within or among populations. The within-population relationships between ln [THg] and fork-length after controlling for the effects of age and $\delta^{15}N$ (a measure of average somatic growth rate) were significant and negative in only 3 of 18 populations (Table 2.4). When individual Arctic charr were pooled across sampling locations, there was no significant correlation between ln [THg] and fork-length (after controlling for the effects of age and trophic level) within either life-history type (Table 2.5). A negative correlation between ln [THg] and fork-length, when controlling for other factors, occurred only when all Arctic charr from both life-history types were combined. While there was a wide range in age-specific ln fork-length among populations of both life-history types, there was no relationship to age-specific ln [THg] among the anadromous or non-anadromous populations (Figure 2.6).

The general linear model evaluating the relative importance of biological, life-history, and location characteristics in explaining ln [THg] found that the effect of fork-length was not significant (F = 0.96, p = 0.33), therefore it was removed from further analyses. Subsequent analysis of ln [THg] indicated significant effects of fish age, trophic level, life-history type, and sampling location (R^2 = 0.74, adjusted R^2 = 0.73, $F_{10,390}$ = 110.5, p < 0.001). To estimate the importance of site-to-site variability in predicting fish [THg], the model was also conducted without including sampling location as a predictor variable, and the percentage of explained variation was substantially reduced (R^2 = 0.54, adjusted R^2 = 0.54, $F_{3,397}$ = 155.3, p < 0.001).

Results of the linear mixed models indicated a significant positive effect of fish

age on individual ln [THg] for both life-history types, but a significant positive effect of

trophic level was only apparent in the non-anadromous Arctic charr (Table 2.6). Site

latitude did not significantly impact ln [THg] in either life-history type. The more complex

model including fish from both life-history types (n = 401 observations from 15 sites)

demonstrated that the age effect was statistically similar between the anadromous and

non-anadromous fish (i.e., no age x type interaction, p = 0.30). When both life-history

types were considered, there was a negative effect of site latitude (p < 0.01) that did not

differ significantly between the anadromous and non-anadromous fish (p = 0.08).

However, inspection of Figure 2.2 suggests that the apparent decline in [THg] with

latitude may rely on a single high mercury and low latitude location, Rivière de la Trinité.

Indeed, when the linear mixed model was conducted without the fish from Rivière de la

Trinité (n = 381 observations from 13 sites), there was no longer a significant effect of

latitude (p = 0.16).

2.4 Discussion

Evidence was found to support the hypothesis that anadromous Arctic charr have

lower [THg] than non-anadromous conspecifics, and that the difference is independent

of latitude (49 – 81° N) in eastern Canada. Within-population [THg] was most

consistently related to fish age in both anadromous and non-anadromous Arctic charr,

although significant positive relationships with fork-length and δ^{15}N or trophic level also

occurred. Differences in δ^{13}C were poorly related to [THg], particularly in non-

anadromous Arctic charr. Fish [THg] was poorly related to fork-length, when controlling

for the effects of fish age and $\delta^{15}N$ or trophic level, within and among populations. Among-population differences in individual [THg] were positively related to fish age in both life history types, but only the non-anadromous Arctic charr demonstrated a significant increase in [THg] with trophic level. Individual fork-length and site latitude did not explain significant additional variation in fish [THg] across sampling locations.

The mean mercury concentrations observed in the anadromous populations (20 to 64 ng/g except Rivière de la Trinité = 114 ng/g) are consistent with previously reported values of mean [THg] in migratory Arctic charr (e.g., 10 – 130 ng/g, Bruce and Spencer 1979; 30 – 80 ng/g, Evans and Muir 2010; 30 – 70 ng/g, Evans et al. 2005a; 40 – 50 ng/g, Riget et al. 2000; 40 ng/g, Swanson et al. 2011a). The mean [THg] observed in non-anadromous populations (111 to 227 ng/g) were also within the range of previously reported concentrations in non-migratory Arctic charr (e.g., 110 – 500 ng/g, Bruce and Spencer 1979; 50 – 1760 ng/g, Evans et al. 2005a; 70 – 1310 ng/g, Gantner et al. 2010a; 97 – 185 ng/g, Marusczak et al. 2011; 147 – 1520 ng/g, Muir et al. 2005; 30 – 940 ng/g, Muir et al. 2009a; 120 – 801 ng/g, Riget et al. 2000; 55 – 179 ng/g, Rognerud et al. 2002; 190 ng/g, Swanson et al. 2011a).

As expected, mercury concentrations were lower among anadromous Arctic charr than in closely located non-migratory conspecifics across the full range of latitudes represented in the present study. The difference in [THg] between life-history types existed whether the non-anadromous fish were landlocked, or co-occurred with sympatric anadromous individuals. In contrast, Swanson et al. (2011a) did not detect a significant difference in mercury concentration between anadromous and lake-resident Arctic charr, and estimated mercury concentrations of 40 ng/g at a standardized length

of 500 mm for both life-history types. Here the non-anadromous fish had mean [THg] 1.8 to 6.3 times higher than paired anadromous conspecifics, whereas others have reported concentrations 3.5 to 15 times higher in non-anadromous compared to anadromous Arctic charr (Bruce and Spencer 1979; Lockhart et al. 2005; Riget et al. 2000; Swanson et al. 2011a).

Lower mercury concentrations have also been detected in anadromous lake trout, *Salvelinus namaycush* (Swanson et al. 2011a), Atlantic salmon, *Salmo salar* (Bruce and Spencer 1979), and brown trout, *Salmo trutta* (Kim and Rochford 2008) than in non-migratory conspecifics. In sockeye salmon, *Oncorhynchus nerka*, [THg] was higher in freshwater-reared smolts than in adults returning from the sea to spawn (Baker et al. 2009). In fact, there is a general tendency for marine or estuarine fish to have lower mercury concentrations than nearby freshwater fish (Baker et al. 2009; Holsbeek et al. 1997; Jewett and Duffy 2007; Kütter et al. 2009; Mason et al. 2006), although the opposite trend was found in a survey of 10 freshwater species and 10 marine species from a fish market in Hong Kong (Cheung et al. 2008).

The lower mercury concentrations in marine relative to freshwater fish may be due to differential incorporation of Hg at the base of the marine and freshwater foodwebs (Mason et al. 1995). Total mercury concentrations measured in freshwater are generally higher than those in seawater (Leopold et al. 2010; Ullrich et al. 2001; and references therein), and in the Canadian Arctic and sub-Arctic specifically, [THg] observed in lakes (e.g., 0.30 – 1.39 ng/L, Chetelat and Amyot 2009; 1.8 ± 0.8 ng/L, Evans et al. 2005b; 0.29 – 0.72 ng/L, Gantner et al. 2010a; 0.56 ± 0.09 to 1.44 ± 0.55 ng/L, Loseto et al. 2004; 2.66 ± 2.70 ng/L in ponds and 1.11 ± 0.48 ng/L in lakes, St.

Louis et al. 2005) were usually higher than concentrations in marine water (0.14 – 0.24 ng/L, St. Louis et al. 2007; 0.40 ± 0.47 ng/L, Kirk et al. 2008). Furthermore, it is the methylated form of mercury that biomagnifies in aquatic foodwebs (Watras et al. 1998), and the proportion of methylated Hg is often higher in freshwater (≈30%) than in seawater (typically <5%) (Leopold et al. 2010; Ullrich et al. 2001; and references therein), although high percentages of methylated Hg (30 – 40%) have been measured in Arctic marine waters at depth or under sea ice (Kirk et al. 2008; St. Louis et al. 2007).

An alternative explanation for higher [THg] in freshwater fish may be a higher rate of mercury biomagnification (quantified as the slope of δ^{15}N versus log [Hg]) through freshwater foodwebs (Swanson and Kidd 2010). However, while biomagnification rates vary among lakes (Gantner et al. 2010b; Kidd et al. 1995; Wyn et al. 2009), similar rates have been found across diverse aquatic foodwebs, including freshwater and marine ecosystems (e.g., Campbell et al. 2005; Chumchal et al. 2011; Riget et al. 2007; and references therein). Furthermore, there is evidence that varying biomagnification rates may not predict fish mercury concentrations among lakes (Gantner et al. 2010a; Wyn et al. 2009).

Fish age was the most consistent predictor of [THg] within both anadromous and non-anadromous Arctic charr, although significant relationships between age and [THg] did not always occur. In some cases, the lack of a relationship may be related to the small sample sizes obtained (n ≤ 10 for anadromous Arctic charr from Heintzelman Lake and both life-history types from Rivière de la Trinité). Our findings agree with previous studies that have found age to be a better predictor of [THg] than size in Arctic charr (Rognerud et al. 2002; Swanson and Kidd 2010; Swanson et al. 2011a) and other

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northern fish species (Evans et al. 2005b). In contrast, size was a better predictor of

mercury concentration in non-anadromous Lake Hazen Arctic charr, where larger

individuals were piscivorous and smaller fish were insectivorous (Gantner et al. 2009),

thus ontogenetic shifts in diet with increasing size can obscure the age versus [THg]

relationship. Riget et al. (2000) also found that fish size better predicted [THg] than age

in non-anadromous Arctic charr populations, but did not investigate differences in

trophic position. Relationships between [THg] and fork-length were observed in many

of the Arctic charr populations examined here, but were not consistent among

populations, particularly after the effects of age and trophic level or δ^{15}N were removed.

Similarly, the relationships between [THg] and δ^{15}N or δ^{13}C were variable among

populations. Therefore, the relationships between [THg] and fish size or trophic position

appeared to be population-specific, and influenced by foodweb and/or abiotic

differences among sampling sites. In contrast, the trend for higher [THg] with increasing

fish age was relatively system-independent.

In most cases the relationships between fish [THg] and δ^{15}N were positive and

consistent with the expected increase in mercury concentration with trophic level (e.g.,

Cabana and Rasmussen 1994; Kidd et al. 1995). However, negative relationships

between [THg] and δ^{15}N and δ^{13}C were observed in two of the anadromous populations.

The negative trends occurred in areas with extensive estuaries (Rivière de la

Trinité/Gulf of St. Lawrence and Dry Bay), and may reflect a gradient in the usage of

freshwater, estuarine, and marine feeding habitats within the studied Arctic charr

populations. In estuarine environments, there is a trend for higher δ^{15}N and δ^{13}C values

and lower [THg] as freshwater influence declines (Attrill et al. 2009; Cossa and

Rondeau 1985; Fry and Chumchal 2012). There were no negative relationships

between ln [THg] and $\delta^{15}N$ or $\delta^{13}C$ in any of the non-anadromous populations. Most

non-anadromous populations showed no relationship between mercury concentration

and $\delta^{13}C$, which may reflect the strong reliance on benthic prey (particularly

chironomids) in Arctic lakes, and little variation in prey carbon source among individual

Arctic charr (Gantner et al. 2010b).

While a growth dilution effect cannot be entirely ruled out, it is unlikely that the

difference in Arctic charr [THg] between the life-history types is caused by differential

somatic growth rates. A negative relationship between ln [THg] and fork-length, after

controlling for the effects of age and trophic level (a measure of average somatic growth

rate), occurred only when Arctic charr from both life-history types were combined. The

lack of a relationship within either the anadromous or non-anadromous fish suggests

that the apparent dilution effect between life-history types is due to the shift in feeding

habitat that drives both a reduction in tissue mercury concentration (e.g., due to a lower

exposure concentration, Mason et al. 2006) and increase in somatic growth rate

(Rikardsen et al. 2000) in anadromous relative to non-anadromous Arctic charr. It has

been previously demonstrated that any effects of differential growth rates can be

masked when increases in size are associated with a shift in diet (Lepak et al. 2009).

Similarly, the anadromous populations had higher age-specific length (i.e., average

somatic growth rate) than all but two of the non-anadromous populations, but even the

non-anadromous populations with high growth rates (at Coady's Pond #2, Nain Bay and

Tasiapik Lake, Dry Bay) did not experience a reduction in mean [THg] relative to the

other non-anadromous populations (Figure 2.6). Thus, this work supports the body of

literature concluding that somatic growth rate is less important than fish diet and/or feeding habitat in predicting fish mercury concentrations (e.g., Essington and Houser 2003; Henery et al. 2010; Lepak et al. 2009; Verta 1990). The enhanced growth rates of the non-anadromous Arctic charr from Coady's Pond #2 and Tasiapik Lake may be explained by the small size of the two lakes (≤ 1 km^2) and the presence of lake trout, which is known to increase the somatic growth rate of sympatric Arctic charr (Fraser and Power 1989).

Across all populations, approximately 54% of the variation in individual [THg] could be predicted using only fish biological factors (age, trophic level, and life-history type). The percentage of explained variation increased to 74% when sampling location was also included in the model, demonstrating the importance of individual sample site characteristics in determining fish [THg]. Fish mercury concentrations have been related to physical parameters (e.g., temperature, lake size, catchment area, underlying geology), chemical factors (e.g., concentration of dissolved organic carbon, pH, alkalinity), and biological considerations such as zooplankton density (Chen and Folt 2005; Chen et al. 2005; Essington and Houser 2003; Greenfield et al. 2001; Pickhardt et al. 2002; Schindler et al. 1995; Watras et al. 1998). At the base of the foodweb, the species composition (Chetelat and Amyot 2009) and concentration of mercury in biota (Chumchal et al. 2008) may also critically influence [THg] in fish.

Considering the abundance of confounding variables among sample sites, it is not surprising that a latitudinal trend in fish [THg] was not apparent once the Rivière de la Trinité populations were removed from the analysis, despite trends of mercury in lake sediments that indicate a decrease in Hg deposition from south to north (Muir et al.

2009b; Schindler et al. 1995). The high mercury concentrations observed in the Rivière de la Trinité populations likely reflect greater industrialization and Hg pollution in the St. Lawrence River Basin (e.g., Cossa and Gobeil 2000, and references therein), compared to the more northern locales.

2.4.1 Summary

The present study is, to our knowledge, the first wide-scale spatial investigation of mercury concentrations in co-located populations of anadromous and non-anadromous Arctic charr. Mercury concentrations were usually low; less than 1% of all individuals measured exceeded the Health Canada limit for mercury in commercially sold fish (500 ng/g), while 0.8% of anadromous and 12.8% of non-anadromous Arctic charr exceeded the more conservative guideline for frequent fish consumers (200 ng/g). The difference in mercury concentration between anadromous and non-anadromous Arctic charr was independent of site latitude, and could not be explained by differential fish ages, fork-lengths, or trophic levels. Therefore, it is reasonable to expect that factors relating to the marine versus lacustrine feeding environments, such as Hg concentrations in biota at the base of the foodweb (Chumchal et al. 2008; Wyn et al. 2009) or rates of biomagnification (Swanson and Kidd 2010) are responsible for the difference in Arctic charr [THg] with life-history type. A key finding of the present study is that mercury concentrations are determined by fish age rather than fork-length in Arctic charr, a species characterized by a long life-span and highly variable length-at-age (Johnson 1980; Klemetsen et al. 2003). Therefore, we recommend that fish [THg]

be corrected using fish age when comparing concentrations across populations,

particularly when examining long-lived species.

2.5 Tables and Figures

Table 2.1. Details on sample collection sites for anadromous and non-anadromous Arctic charr. Abbreviations for lacustrine fish community members are: AC-A = anadromous Arctic charr (*Salvelinus alpinus*); AC-N = non-anadromous Arctic charr; AE = American eel (*Anguilla rostrata*); AS = Atlantic salmon (*Salmo salar*); LC = lake chub (*Couesius plumbeus*); LT = lake trout (*Salvelinus namaycush*); 3SS = threespine stickleback (*Gasterosteus aculeatus*); BT = brook trout (*Salvelinus fontinalis*); LS = longnose sucker (*Catostomus catostomus*); RW = round whitefish (*Prosopium cylindraceum*); 9SS = ninespine stickleback (*Pungitius pungitius*). Location abbreviations are defined in Figure 2.1.

Type	Sampling site (location abbreviation)	Lake area (km^2)	$\delta^{15}N_{base}$ (n)	Base taxon	Lacustrine fish community
Anadromous					
	Rivière de la Trinité[a] (TR)	-	7.2 ± 0.1 (21)[b]	Mussel	-
	Fraser River, Nain Bay (NB)	-	9.0 ± 0.5 (5)	Mussel	-
	Okak Bay (OB)	-	8.8 ± 0.8 (5)	Mussel	-
	Saglek Bay (SB)	-	7.9 ± 0.5 (5)	Mussel	-
	Nepihjee River, Dry Bay (DB)	-	8.5 ± 0.3 (5)	Mussel	-
	Frobisher Bay, Iqaluit (IQ)	-	8.7 ± 0.2 (5)	Barnacle	-
	Iqalugaarjuit Lake, Pangnirtung Fjord (PG)	-	10.5 ± 0.4 (5)	Clam	-
	Salmon River, Pond Inlet (PI)	-	8.6 ± 0.9 (10)	Gammarid	-
	Heintzelman Lake (HZ)	-	NA	NA	-
Non-anadromous					
	Rivière de la Trinité[a] (TR)	0.9	3.5 ± 0.3 (5)	Mussel	AC-A, AC-N, AE, AS
	Coady's Pond #2 (NB)	1.0	2.8 ± 0.6 (3)	Clam	AC-N, LC, LT, 3SS
	Esker Lake (OB)	33.2	5.2 ± 0.6 (4)	Chironomid	AC-N, LC, LT, BT, LS, RW
	Upper Nakvak Lake (SB)	5.9	5.1 ± 0.9 (3)	Chironomid	AC-N
	Tasiapik Lake (DB)	0.3	4.2 ± 0.5 (18)	Snail	AC-N, LT, 3SS
	Crazy Lake (IQ)	4.4	4.0 ± 0.6 (4)	Chironomid	AC-N

Table 2.1. (Continued)

Type	Sampling site (location abbreviation)	Lake area (km^2)	$\delta^{15}N_{base}$ (n)	Base taxon	Lacustrine fish community
	Iqalugaarjuit Lake (PG)	1.4	NA	NA	AC-A, AC-N, 3SS
	unnamed lake (PI)	0.2	2.8 ± 0.4 (4)	Chironomid	AC-A, AC-N, 9SS
	Heintzelman Lake (HZ)	10.2	NA	NA	AC-A, AC-N

[a]Arctic charr of both life-history types overwinter in Lac Davidson. Marine feeding of anadromous individuals occurs in the Gulf of St. Lawrence, and non-anadromous fish were caught prior to making their first marine feeding foray (e.g., Doucett et al. 1999). The fish community and lake area are given for Lac Davidson.
[b]Collected in the Gulf of St. Lawrence, from Lavoie et al. (2010)

Table 2.2. Summary data for Arctic charr from the paired sampling sites. Sampling locations are listed in order of increasing latitude, using abbreviations defined in Figure 2.1. Means ± 1 standard deviation are given for the anadromous (A) and non-anadromous (N) populations at each location. Significant differences between the two life-history types at a given location are indicated as: * = $p < 0.05$, ** = $p < 0.01$, *** = $p < 0.001$.

Location	Age (years)		Fork-length (mm)		Trophic level		THg concentration (ng/g ww)	
	A	N	A	N	A	N	A	N
TR	4.8 ± 0.8	5.3 ± 0.5	275 ± 39	192 ± 7***	3.4 ± 0.2	3.6 ± 0.1	114 ± 54	220 ± 25***
NB	7.3 ± 2.0	5.6 ± 3.0*	363 ± 88	316 ± 124	3.7 ± 0.1	3.2 ± 0.1***	22 ± 8	120 ± 42***
OB	7.9 ± 2.4	10.7 ± 5.4*	430 ± 98	278 ± 106***	3.4 ± 0.3	2.8 ± 0.3***	47 ± 27	111 ± 179**
SB	8.2 ± 3.6	11.7 ± 6.7	390 ± 94	287 ± 166*	3.8 ± 0.2	3.3 ± 0.3***	20 ± 11	126 ± 124***
DB	5.6 ± 1.5	5.9 ± 3.6	416 ± 108	296 ± 165**	3.6 ± 0.2	4.1 ± 0.2***	38 ± 16	111 ± 50***
IQ	10.6 ± 2.3	7.3 ± 5.2***	517 ± 48	176 ± 100***	3.7 ± 0.3	3.3 ± 0.3***	35 ± 9	112 ± 160***
PG	12.1 ± 1.8	6.5 ± 2.5***	588 ± 72	188 ± 30***	3.1 ± 0.2	NA	50 ± 21	118 ± 53***
PI	13.9 ± 2.8	10.3 ± 8.0	644 ± 92	254 ± 151***	3.7 ± 0.2	3.8 ± 0.2	60 ± 21	227 ± 109***
HZ	9.4 ± 2.8	12.5 ± 5.1	284 ± 64	212 ± 63**	NA	NA	64 ± 16	117 ± 51**

Table 2.3. Pearson's correlation coefficients for within-population ln [THg] versus age, fork-length, δ^{15}N, or δ^{13}C. Sampling locations are ordered by increasing latitude, using abbreviations defined in Figure 2.1. Significance of the correlation is indicated as: * = p < 0.05, ** = p < 0.01, *** = p < 0.001.

Type	Location	n	ln [THg] age	ln [THg] fork-length	ln [THg] δ^{15}N	ln [THg] δ^{13}C
Anadromous						
	TR	10	-0.30	-0.44	-0.84**	-0.69*
	NB	20	0.60**	0.28	0.25	0.44
	OB	20	0.80***	0.26	0.35	0.59**
	SB	22	0.89***	0.75***	0.73***	0.69***
	DB	33	0.20	0.00	-0.35*	-0.36*
	IQ	30	0.58***	0.44*	-0.07	0.06
	PG	40	0.52**	-0.25	0.49**	0.34*
	PI	70	0.47***	0.46***	0.36**	0.39**
	HZ	7	0.65	-0.75	-0.07	0.01
Non-anadromous						
	TR	10	0.10	-0.16	0.54	-0.23
	NB	20	0.69***	0.68***	0.34	0.55*
	OB	20	0.74***	0.52*	0.63**	-0.11
	SB	20	0.95***	0.88***	0.72***	0.15
	DB	31	0.25	0.22	0.10	-0.07
	IQ	47	0.84***	0.93***	0.86***	-0.09
	PG	19	0.68**	0.57*	0.29	0.17
	PI	18	0.60**	0.50*	0.43	0.17
	HZ	112	0.70***	0.60***	0.40***	0.09

Table 2.4. Pearson's partial correlation coefficients for within-population ln [THg] versus age or fork-length when controlling for other factors. Sampling locations are ordered by increasing latitude, using abbreviations defined in Figure 2.1. Significance of the correlation is indicated as: * = p < 0.05, ** = p < 0.01, *** = p < 0.001.

Type	Location	n	ln [THg] age, given fork-length	ln [THg] fork-length, given age	ln [THg] age, given fork-length and $\delta^{15}N$	ln [THg] fork-length, given age and $\delta^{15}N$
Anadromous						
	TR	10	-0.22	-0.40	-0.24	-0.32
	NB	20	0.61**	-0.29	0.60**	-0.32
	OB	20	0.90***	-0.69***	0.89***	-0.70***
	SB	22	0.72***	0.04	0.60**	0.04
	DB	33	0.39*	-0.34*	0.19	0.02
	IQ	30	0.44*	0.11	0.46**	0.32
	PG	40	0.52***	-0.26	0.49**	-0.35*
	PI	70	0.19	0.21	0.25*	0.14
	HZ	7	0.54	-0.67	0.60	-0.58
Non-anadromous						
	TR	10	0.12	-0.17	0.02	-0.10
	NB	20	0.19	0.10	0.04	0.25
	OB	20	0.77***	-0.59**	0.72***	-0.60**
	SB	20	0.74***	-0.05	0.74***	-0.07
	DB	31	0.19	-0.10	0.19	-0.10
	IQ	47	0.13	0.75***	0.19	0.57***
	PG	19	0.47*	0.04	0.50*	0.05
	PI	18	0.40	-0.15	0.47*	-0.30
	HZ	112	0.55***	0.36***	0.36***	0.52***

Table 2.5. Pearson's partial correlation coefficients between ln [THg] and age or fork-length when controlling for other factors. Arctic charr of a given life-history type were pooled across all sampling locations. Significance of the correlation is indicated as: * = $p < 0.05$, ** = $p < 0.01$, *** = $p < 0.001$.

	Anadromous	Non-anadromous	Both life-history types
n (n with trophic level data)	252 (244)	297 (166)	549 (410)
ln [THg] age, given fork-length	0.33***	0.37***	0.55***
ln [THg] fork-length, given age	-0.08	0.30***	-0.49***
ln [THg] age, given fork-length and trophic level	0.28***	0.39***	0.53***
ln [THg] fork-length, given age and trophic level	-0.01	0.06	-0.50***

Table 2.6. Results for fixed effect factors from the linear mixed models to explain ln [THg] using individual age, trophic level, and site latitude within the two life-history types, given the random effect of sampling site. Type = anadromous (A) or non-anadromous (N).

Type	n (sample sites)	Source	Coefficient estimate	Standard error	F-value	p
A	236 (8)	Intercept	5.51	1.87	422.59	< 0.001
		Age	0.09	0.02	19.61	< 0.001
		Trophic level	0.02	0.10	0.11	0.75
		Latitude	-0.05	0.03	2.27	0.13
N	165 (7)	Intercept	2.74	1.48	1163.12	< 0.001
		Age	0.06	0.01	34.75	< 0.001
		Trophic level	0.70	0.13	26.69	< 0.001
		Latitude	-0.02	0.02	0.40	0.56
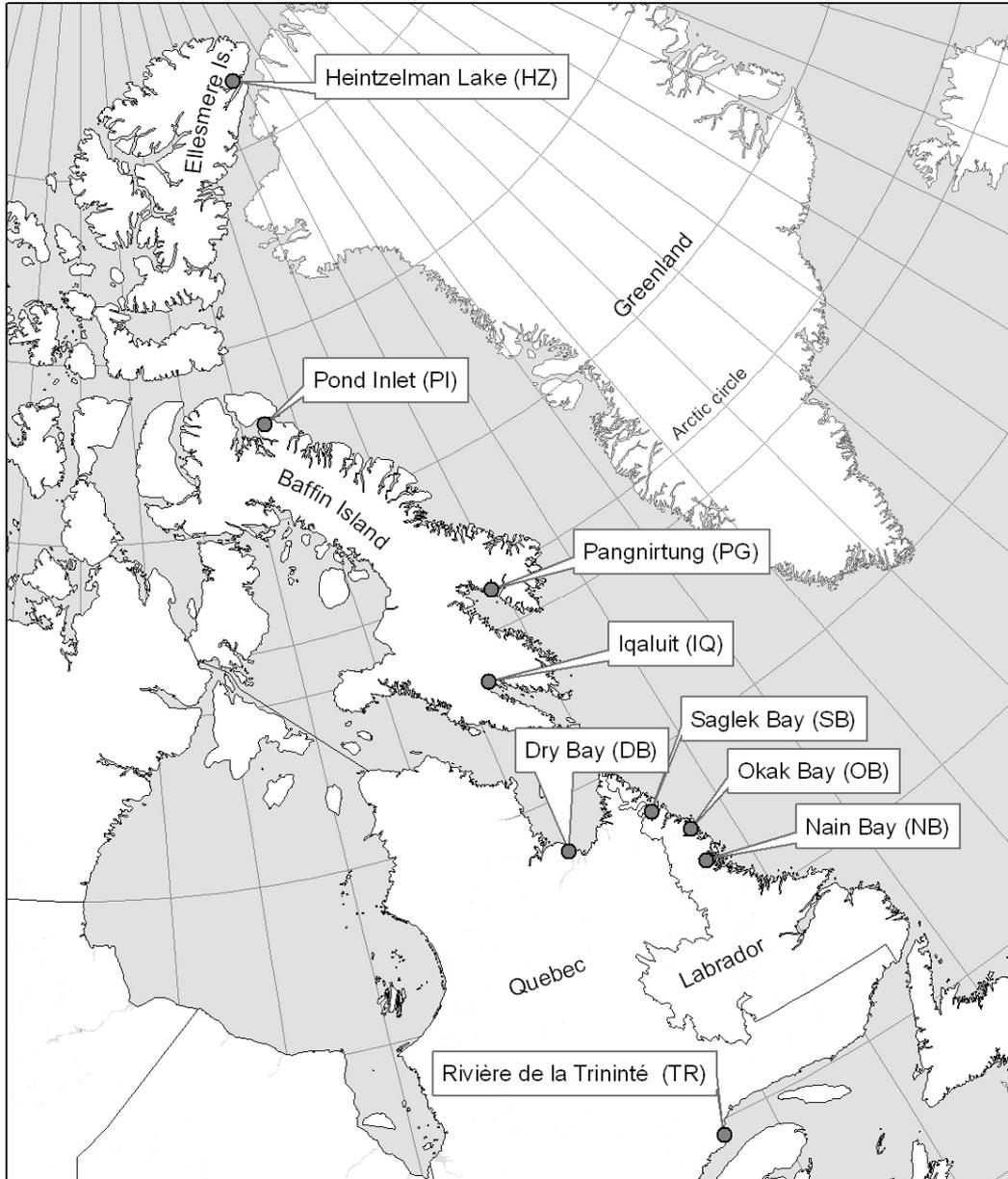
Figure 2.1. Map of north eastern Canada indicating the nine locations of Arctic charr collection. Location abbreviations used in Tables 2.1 – 2.4 are given in parentheses.
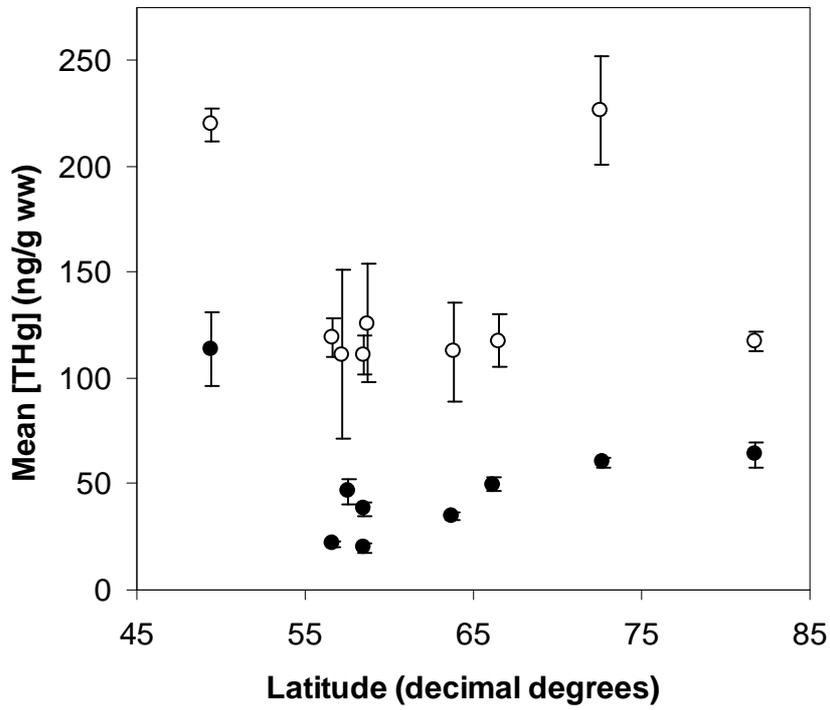
Figure 2.2. Latitude (decimal degrees) versus mean [THg] (ng/g ww) for Arctic charr from anadromous (solid circles) and non-anadromous (open circles) populations. Error bars represent one standard error of the mean.
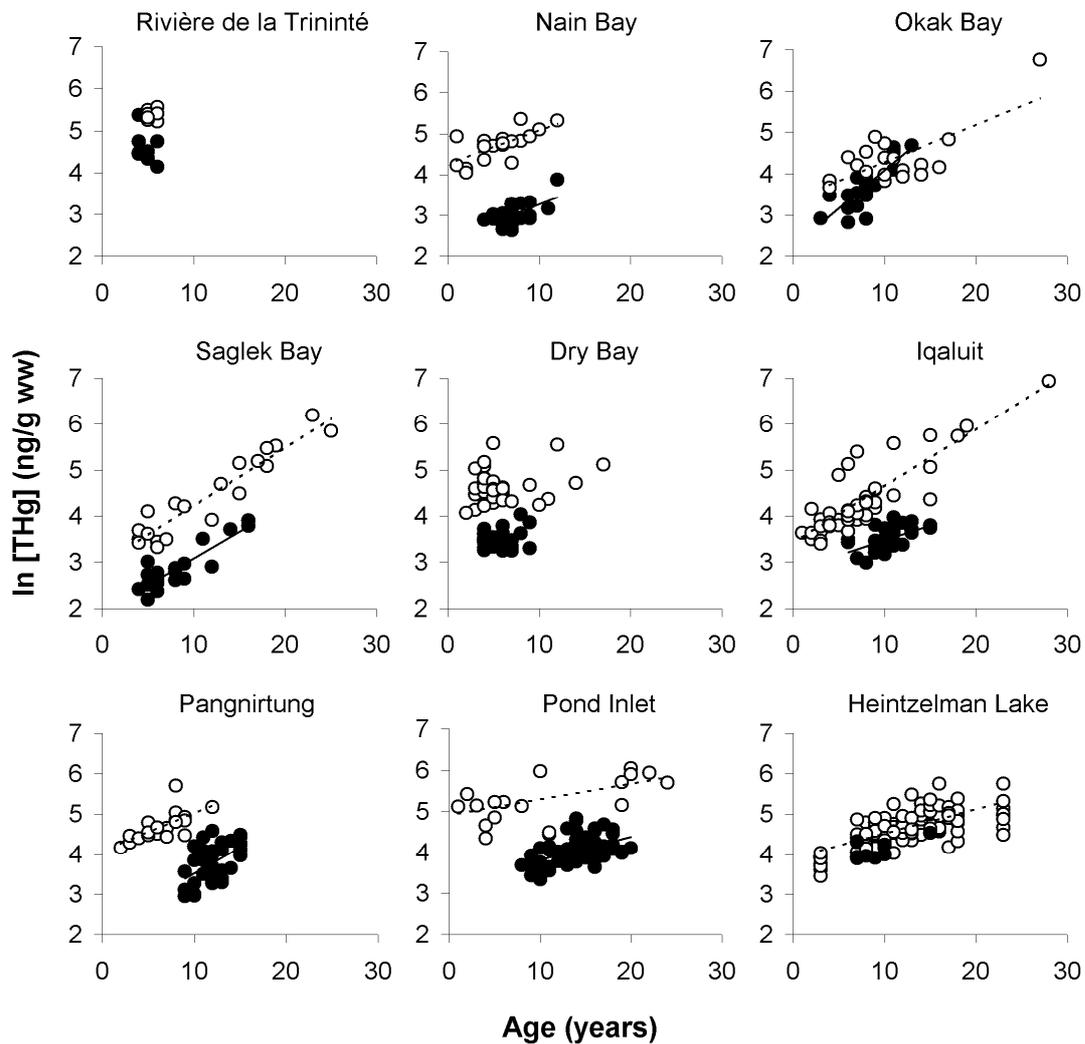
Figure 2.3. Age (years) versus ln [THg] (ng/g ww) for Arctic charr from 9 paired sampling sites. Sampling locations are ordered by increasing latitude (left to right then top to bottom). Data points represent individual fish from anadromous populations (closed circles) and non-anadromous populations (open circles). Lines indicate significant linear regressions in anadromous (solid line) and non-anadromous (dashed line) populations.
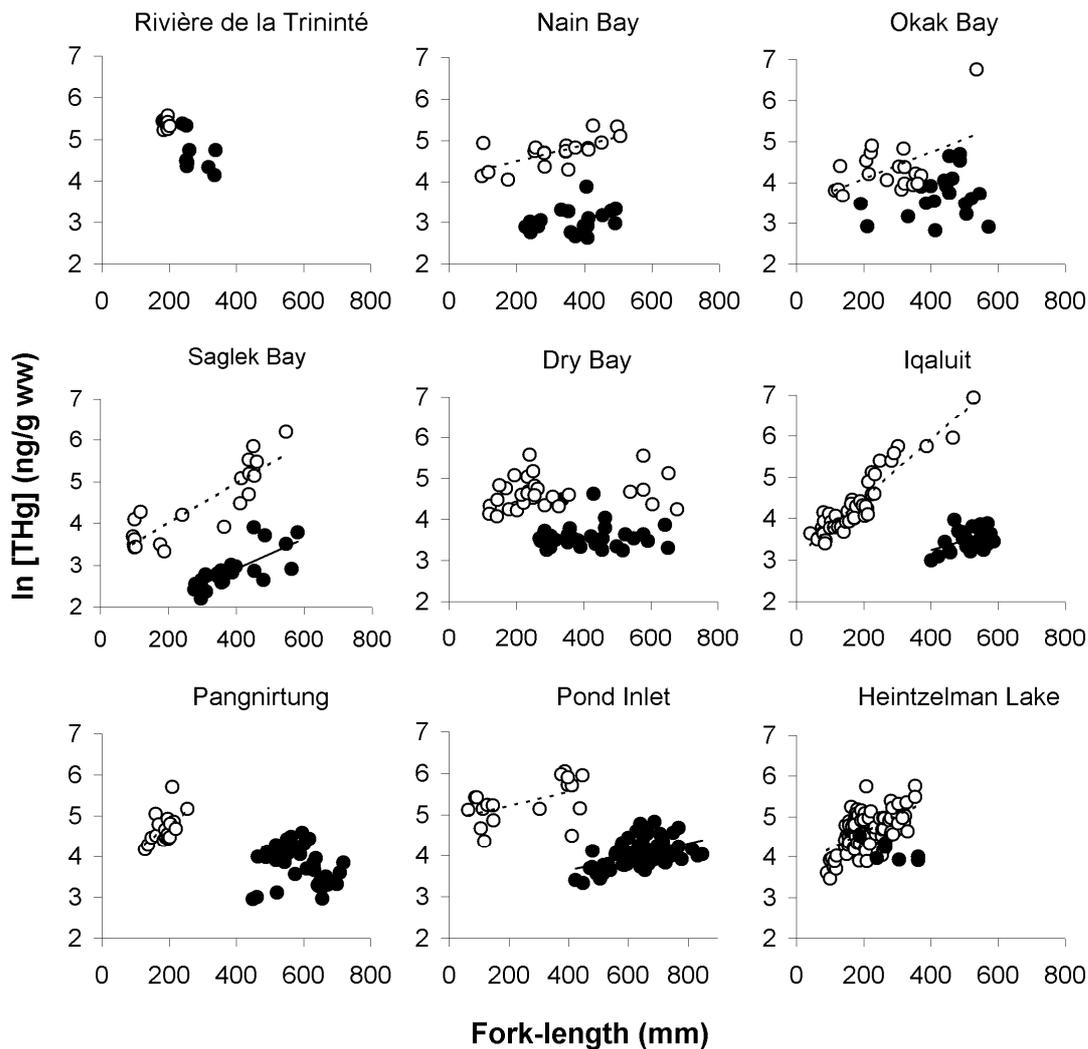
Figure 2.4. Fork-length (mm) versus ln [THg] (ng/g ww) for Arctic charr from 9 paired sampling sites. Sampling locations are ordered by increasing latitude (left to right then top to bottom). Data points represent individual fish from anadromous populations (closed circles) and non-anadromous populations (open circles). Lines indicate significant linear regressions in anadromous (solid line) and non-anadromous (dashed line) populations.
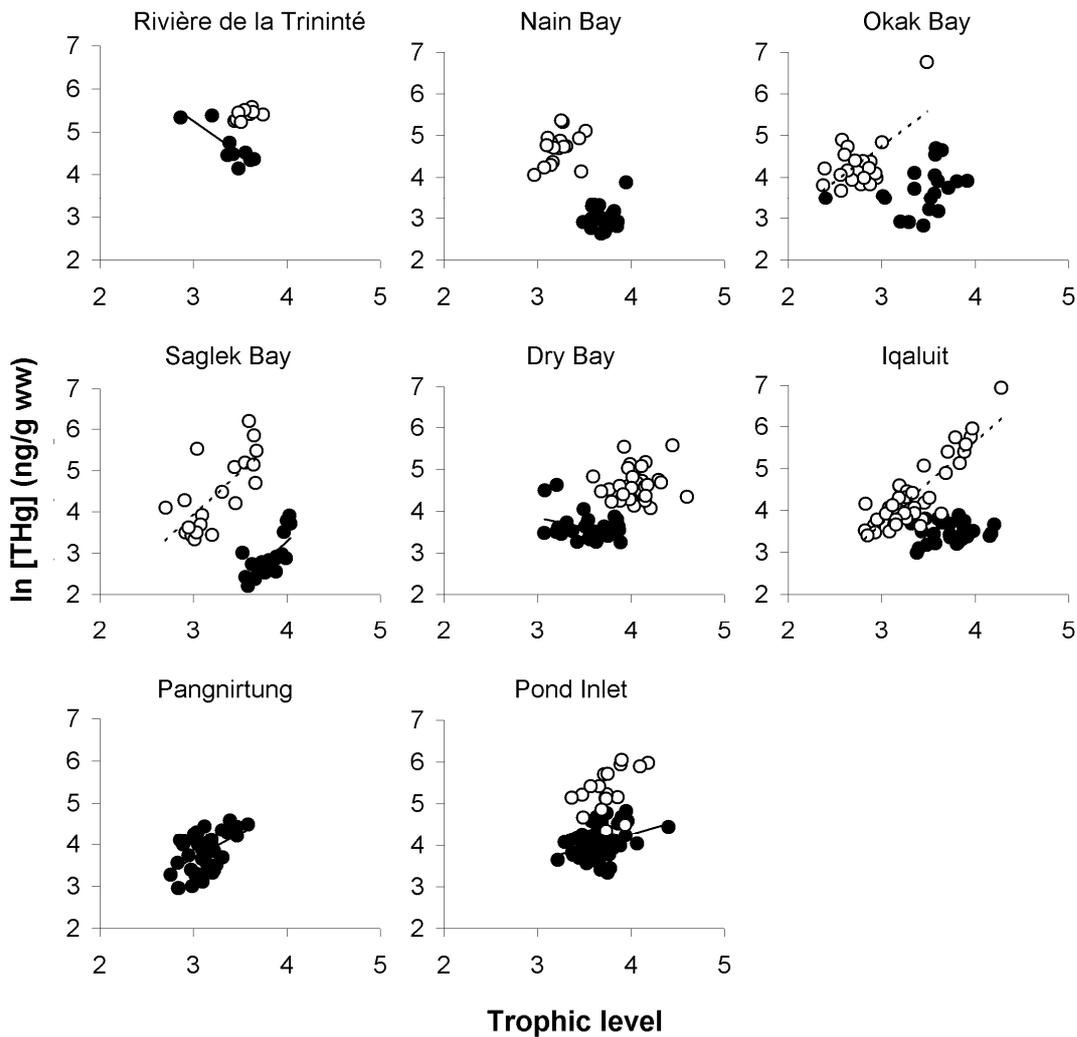
Figure 2.5. Trophic level versus ln [THg] (ng/g ww) for Arctic charr from 8 sampling

locations. Locations are ordered by increasing latitude (left to right then top to bottom).

Data points represent individual fish from anadromous populations (closed circles) and

non-anadromous populations (open circles). Lines indicate significant linear

regressions in anadromous (solid line) and non-anadromous (dashed line) populations.

Trophic level could not be calculated in three populations due to lack of suitable $\delta^{15}N_{base}$

values (Table 2.1).
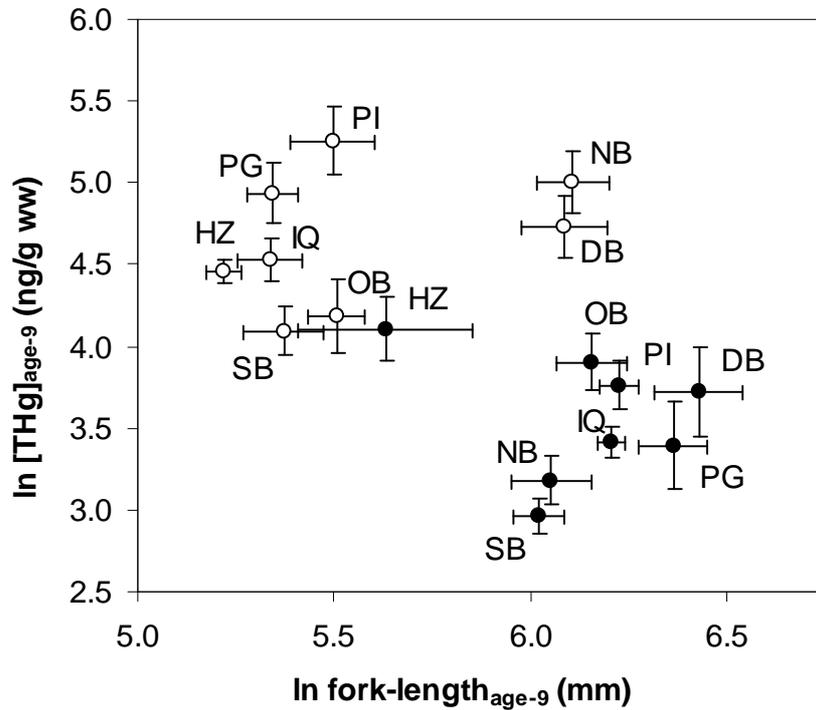
Figure 2.6. Age-adjusted ln fork-length (mm) versus age-adjusted ln [THg] (ng/g ww) for Arctic charr from the anadromous (closed circles) and non-anadromous (open circles) populations. All [THg] and fork-lengths were adjusted to a standard age of 9 years; error bars represent 95% confidence intervals of the estimates. Location abbreviations are defined in Figure 2.1. Age-standardized ln [THg] and fork-length were not calculated for the Rivière de la Trinité populations (see text).

Chapter 3

Basal mercury concentrations and biomagnification rates in freshwater and marine foodwebs: effects on Arctic charr (*Salvelinus alpinus*) from eastern Canada

3.1 Introduction

Mercury contamination presents a continuing threat to the health of Arctic ecosystems and inhabitants (Dietz et al. 2011; Stow et al. 2011). Mercury is transported to the Arctic and other remote regions primarily via long-range atmospheric transport (Munthe et al. 2011), and originates from both natural (e.g., volcanic activity) and anthropogenic (e.g., mining and metal production, fossil fuel burning) sources (AMAP/UNEP 2008; Munthe et al. 2011; Pacyna et al. 2010). Atmospheric mercury enters Arctic aquatic environments through precipitation or deposition onto ice and snow (Douglas et al. 2011; Munthe et al. 2011). Additional sources of mercury to the Arctic Ocean include inflows from the Atlantic and Pacific Oceans, coastal erosion, and inputs from rivers (Munthe et al. 2011), while watershed inputs such as snowmelt runoff and erosion of thawed soils are important for lakes (Douglas et al. 2011). Once in the aquatic environment, inorganic mercury can be transformed into methylmercury (MeHg), primarily by sulphate-reducing bacteria in anoxic water and sediments (Barkay and Poulain 2007; Morel et al. 1998; Ullrich et al. 2001). Mercury, particularly MeHg, is well known to bioaccumulate in organisms and biomagnifiy in aquatic foodwebs (Douglas et al. 2011; Mason et al. 1995; Morel et al. 1998; Watras et al. 1998).

Methylmercury is considered the most toxic form of mercury, causing reproductive deficiencies and behavioural changes in wildlife (Scheulhammer et al.

2007), and developmental neurotoxicity and cardiovascular effects in humans (Mergler

et al. 2007). The proportion of mercury in the methylated form (%MeHg) increases with

trophic level in aquatic foodwebs (Douglas et al. 2011), from <1 – 12.2% in lacustrine

primary producers (Bowles et al. 2001; Gantner et al. 2010a) to ≈10 – 80% in marine

and lacustrine zooplankton (Back et al. 2003; Campbell et al. 2005; Gantner et al.

2010a; Lavoie et al. 2010; Loseto et al. 2008; Swanson et al. 2011a) and to ≈80 – 100%

(often >90%) in muscle tissue of most marine and freshwater fish (Bloom 1992;

Chumchal et al. 2011; Gantner et al. 2010a; Jæger et al. 2009; Lasorsa and Allen-Gil

1995; Loseto et al. 2008; Riget et al. 2007; Wyn et al. 2009). Thus, the foodweb

biomagnification rate of MeHg is often higher than that of total Hg (e.g., Lavoie et al.

2010; Riget et al. 2007).

      While direct uptake from water is an important source of Hg to organisms at the

base of aquatic foodwebs (Morel et al. 1998), the vast majority of mercury in organisms

at higher trophic levels (e.g., fish) is obtained through consumed food (Hall et al. 1997).

Therefore, stable isotope ratio methods commonly used in ecological studies to

characterize trophic relationships (e.g., Guiguer et al. 2002; Grey et al. 2002; Swanson

et al. 2003) are potentially useful for examining the trophic transfer of mercury, as they

reflect the long-term assimilated diet of a consumer (Peterson and Fry 1987). Stable

isotope methods have been used to quantify the rate of biomagnification across trophic

levels (Borgå et al. 2012; Jardine et al. 2006), with previous studies having found that

mercury is positively related to δ^{15}N-inferred trophic level, and that the observed

biomagnification rate (the slope of the relationship between mercury concentration and

δ^{15}N or trophic level) is relatively consistent among diverse aquatic foodwebs (e.g.,

Campbell et al. 2005; Chumchal et al. 2011; Riget et al. 2007; and references therein).

However, Swanson and Kidd (2010) found that MeHg biomagnification rates in

freshwater foodwebs were significantly higher than in a nearby marine foodweb. Thus,

differences in mercury concentrations between common taxa functioning in different

foodwebs are likely related to differing biomagnification rates and/or mercury

concentrations at the base of the foodweb (Chumchal et al. 2008; Mason et al. 2006;

Wyn et al. 2009).

Within the Arctic, one key species known to function in a diverse range of aquatic

habitats and foodwebs (Power et al. 2008) is Arctic charr (*Salvelinus alpinus*). Arctic

charr have been internationally used as a sentinel species to evaluate spatial and

temporal trends in mercury contamination (AMAP 2005; Douglas et al. 2011), are an

important food resource for northern people (Evans et al. 2005a; Van Oostdam et al.

2005), and are vulnerable to changing climate (Reist et al. 2006). Arctic charr are found

throughout the Arctic, and have a distribution that extends farther north than that of any

other freshwater fish (Johnson 1980; Power et al. 2008). Where access to the sea

exists, anadromous Arctic charr migrate to the nearshore marine environment for brief

periods of intensive summer feeding, and return to freshwater to spawn and overwinter

(Johnson 1980). Anadromy is not obligate, and populations of landlocked and lake-

resident (i.e., have access to the sea but do not migrate) Arctic charr are common

(Johnson 1980; Klemetsen et al. 2003), with the prevalence of anadromy declining

toward the northern and southern limits of the species' distribution (Power et al. 2008).

Differences in Arctic charr life-history strategy have been related to total mercury

concentration ([THg]), with lower measured [THg] in anadromous than non-anadromous

fish (Bruce and Spencer 1979; Lockhart et al. 2005; Riget et al. 2000; Swanson et al. 2011a). Within-population differences in [THg] of anadromous and non-anadromous Arctic charr have been positively related to fish age, size, and trophic level (Gantner et al. 2010b, 2009; Muir et al. 2005; Riget et al. 2000; Rognerud et al. 2002; see also Chapter 2), although the significance and strength of the associations vary markedly by population.

Here we use data from six spatially-paired marine and lacustrine foodwebs along a latitudinal gradient in eastern Canada to examine mercury accumulation patterns in feeding habitats important for Arctic charr. Specifically, we test the following hypotheses: [1] biomagnification rates (quantified using the slope of mercury concentration versus δ^{15}N-inferred trophic level) are lower in marine foodwebs than in spatially-proximate lacustrine foodwebs; [2] mercury concentrations at the base of the foodweb are lower in the marine than in the lacustrine feeding habitats; [3] the proportion of methylated mercury increases with trophic level, therefore biomagnification rates are higher for MeHg than THg, and the trend is similar in both feeding habitats used by Arctic charr; and [4] the differences between the lacustrine and marine feeding habitats are consistent across a range of latitudes (56 – 72°N) in eastern sub-Arctic and Arctic Canada.

3.2 Methods

3.2.1 Sample collection

Six sampling locales were chosen from a range of latitudes in the eastern Canadian Arctic and sub-Arctic (Figure 3.1) on the basis of accessibility and the

presence of both anadromous and non-anadromous Arctic charr. At each locale,

spatially proximate nearshore marine and lacustrine foodwebs were sampled. Arctic

charr (anadromous or non-anadromous) were obtained from each foodweb, and

represented the top trophic level at each sample site (i.e., higher trophic level organisms

were not targeted in the present study). Anadromous Arctic charr were included as part

of the marine foodweb, because migratory Arctic charr feed primarily in, and were

typically sampled from, the marine environment. Furthermore, the dietary contribution

from lacustrine or riverine prey resources is minimal after the onset of anadromy

(Johnson 1980; Rikardsen et al. 2003; Swanson et al. 2011b). In instances where

putative anadromous fish were obtained late in the season from freshwater,

anadromous status was confirmed using sulphur stable isotopes following methods

outlined in Doucett et al. (1999). Sampling was conducted in conjunction with Fisheries

and Oceans Canada and the Northern Contaminants Program, in cooperation with local

Inuit Hunters and Trappers Organizations (HTOs).

In the lacustrine environment, Arctic charr were captured using multi-mesh nylon

multifilament sinking gillnets (Johnson 1983) set perpendicular from shore for a period

of ≥4 hours, usually overnight. Juvenile Arctic charr, threespine stickleback

(*Gasterosteus aculeatus*), and ninespine stickleback (*Pungitius pungitius*) were

obtained using small, un-baited fyke nets set near shore or with dipnets. Benthic

invertebrates (e.g., chironomids, trichoptera larvae, amphipods, gastropods) were

collected from sediments using dipnets and a kick-sweep method at 0.5 – 1 m depth, or

were removed from rocks and pools in the littoral zone. Zooplankton were sampled by

surface tows using a 65 or 125 µm zooplankton net. Filamentous algae and/or

periphyton were collected by hand from the water surface or from rocks in the littoral zone. Additional samples including terrestrial insects, benthic invertebrates, and stickleback were opportunistically sampled from Arctic charr stomach contents.

Anadromous Arctic charr were either captured in the nearshore marine environment using a combination of angling and 114 to 140 mm mesh gillnets, or were purchased from local subsistence fishers who used similar sampling gear (at Iqaluit and Pond Inlet). Anadromous individuals from Dry Bay were collected while migrating upriver in a counting fence fyke trap set in the Nepihjee River fishway. Additional fish species captured from marine sampling sites included: blennies (*Blennioidei* sp.), capelin (*Mallotus villosus*), gunnels (*Pholis fasciata*, *P. gunnellus*), ninespine stickleback, polar cod (*Boreogadus saida*), sandlance (*Ammodytes* sp.), sculpin (*Myoxocephalus* sp.), and threespine stickleback. Forage fish were collected along the shore using dipnets, or were opportunistically sampled from Arctic charr stomachs. Invertebrates including mussels (*Mytilus edulis*), barnacles (*Balanus* sp.), and gastropods (*Littorina* sp.) were removed from rocks in the intertidal zone. Gammarid amphipods were collected from pools in the intertidal zone. Arctic charr stomachs yielded additional invertebrate samples (e.g., hyperiid amphipods, mysids). Zooplankton were obtained by surface tows using a 65 or 125 µm zooplankton net. Marine algae including *Fucus* sp., kelp, filamentous algae, and others (e.g., *Ulva* sp.) were collected from rocky intertidal areas, or from fragments washed up on shore.

Previous studies have identified fish stomach contents as a useful source of prey items for stable isotope (Grey et al. 2002; Peterson 1999; Tieszen et al. 1983) and mercury analyses (e.g., Lepak et al. 2009; Ward et al. 2010). To minimize the impact of

digestion on prey samples obtained from fish stomachs, only intact dietary items obtained directly from the esophagus or upper stomach with no evidence of digestion were used (Guelinckx et al. 2008). All samples from stomachs were rinsed in deionized water prior to analysis to ensure minimal influence of digestive tract materials on completed analyses.

3.2.2 Sample processing and analysis

All samples of forage fish, invertebrates, and algae were frozen in the field and transported to the laboratory. Arctic charr were either processed in the field immediately after capture, or were frozen whole and transported to the laboratory. Fork-length (mm) and whole weight (g) were measured for each Arctic charr specimen, and sagittal otoliths were removed for fish aging. Further details regarding the Arctic charr populations used in this study can be found in Chapter 2.

Tissue types used for stable isotope and mercury analyses were as follows: for Arctic charr and larger forage fish, dorsal muscle tissue was excised from the left side of the fish posterior to the dorsal fin and dorsal to the lateral line. For small forage fish (stickleback and sculpin < 5 cm total length), the head, fins, and viscera were removed and the remainder of the body (primarily muscle tissue) was used for analysis. Whole bodies of invertebrates were used (snails, mussels, and barnacles were first removed from shells). Algae samples were rinsed in deionized water and cleaned of debris prior to analysis.

All stable isotope analyses were conducted at the Environmental Isotope Laboratory at the University of Waterloo, Waterloo, Canada, following methods

described in Chapter 2. Small invertebrates (e.g., chironomid larvae, cladocera) were pooled to obtain sufficient mass for analysis ($\approx$0.1 – 0.5 g wet weight), while larger invertebrates and fish were measured individually. Stable isotope ratios of carbon (δ^{13}C) and nitrogen (δ^{15}N) were quantified in a total of 847 samples. Sample sizes for each taxon are given for lacustrine and marine sampling sites in Tables 3.1 and 3.2, respectively.

Total Hg concentrations were measured on a Milestone Direct Mercury Analyzer, DMA-80 (Milestone S.r.l., Sorisole, Italy) via thermal decomposition and atomic absorption spectroscopy as described in U.S. EPA method 7473 (U.S. Environmental Protection Agency 2007). Total mercury concentrations were measured using 0.1 – 0.2 g of frozen, non-homogenized tissue for individual fish. Invertebrates were pooled to obtain samples of $\approx$1 – 2 g wet weight. All algae and invertebrate samples were lyophilized and then homogenized using an acid-washed glass mortar and pestle. A sample of 0.05 – 0.1 g of dried tissue was used for THg analysis. Blanks and standard reference materials were run at the beginning and end of each batch of 20 – 30 samples, and one sample per batch was run in triplicate. The standard reference materials used and percent recoveries (mean percentage of certified value ± standard deviation) were: NIST 2976 (96.7 ± 5.1) from the National Institute of Standards and Technology (Standard Reference Materials Program, Gaithersburg, USA), DORM-3 (108.9 ± 12.2), and DORM-1 (99.7 ± 6.2) from the National Research Council Canada (Institute for National Measurement Standards, Ottawa, Canada). The method detection limit (3x the standard deviation of blanks) was 0.34 ng Hg, and the mean relative standard deviation of triplicates was 4.4%. Total mercury concentrations were

directly measured in a total of 554 samples, and were calculated as MeHg + Hg (II) in a further 125 samples from mercury speciation analysis. Sample sizes for lacustrine and marine taxa are given in Tables 3.1 and 3.2, respectively.

All samples for mercury speciation analysis were freeze dried and then homogenized using an acid-washed glass mortar and pestle. Arctic charr were analyzed individually, while invertebrates and forage fish were pooled to obtain the required mass (≈1 – 2 g wet weight) for analysis. Samples were weighed (±0.1 mg) before and after lyophilization and the percent moisture was calculated for each sample to permit interconversion of wet weight (ww) and dry weight (dw) concentrations of THg and MeHg (Tables 3.1 and 3.2). Concentrations of MeHg and inorganic Hg (II) were measured at Quicksilver Scientific (Lafayette, CO, USA). Quality control measures included matrix spikes and certified reference materials. The limit of detection (LOD) and limit of quantification (LOQ) were LOD = 0.20 ng/g, LOQ = 0.50 ng/g Hg for both MeHg and Hg (II) in algae and invertebrate samples, and LOD = 1.33 ng/g, LOQ = 3.33 ng/g Hg for both MeHg and Hg (II) in fish samples. Where Hg (II) concentrations were below the limit of quantification or limit of detection (n = 16 samples), half of the LOQ/LOD value was used in order to calculate total mercury concentration. No samples had MeHg concentrations below the LOQ or LOD values. MeHg concentration was estimated for 554 samples (most were Arctic charr, n = 341) that had measured [THg] values, using the relevant mean %MeHg for the taxon (e.g., non-anadromous Arctic charr = 99.0 ± 0.7%). In the case where %MeHg was not measured for a taxon (e.g., marine calanoids), the %MeHg from a similar taxon was used (e.g., marine mixed zooplankton = 7.5 ± 4.3%). Mercury speciation analysis was conducted on a total of

125 samples. Sample sizes for lacustrine and marine taxa are given, respectively, in Tables 3.1 and 3.2.

3.2.3 Data analysis

  To compare the trophic positions of organisms from different sampling sites, mean $\delta^{15}N$ values of primary consumers were used to assess baseline values ($\delta^{15}N_{base}$) for each site (Tables 3.1 and 3.2). Trophic level (TL) was calculated using site-specific $\delta^{15}N_{base}$ following (Post 2002):

  Trophic level $_{sample}$ = 2 + ($\delta^{15}N_{sample} - \delta^{15}N_{base}$) / 3.4

  While the trophic fractionation of $\delta^{15}N$ is variable (e.g., -2.1 to +5.4‰, McCutchan et al. 2003; -0.7 to +9.2‰, Vander Zanden and Rasmussen 2001; -1.16 to +5.89‰, Vanderklift and Ponsard 2003), the average value of 3.4‰ provides a reasonable approximation of trophic fractionation when averaged across multiple trophic levels (Post 2002), particularly when a primary consumer is used for the baseline value (Vander Zanden and Rasmussen 2001).

  For each marine and lacustrine foodweb, the trophic magnification factor (TMF) of THg and MeHg was calculated following Borgå et al. (2012) and Jardine et al. (2006) as:

  $TMF = 10^{b}$

where the exponent b is the estimated slope coefficient obtained from the regression:
$\log_{10}$ [Hg] = a + b*(TL – 1). All trophic level data were subtracted by one in order to set the intercept of the regression line to the first trophic level (i.e., the presumed base of

the foodweb). The basal mercury concentration of each foodweb was estimated as 10^a where a represents the estimated intercept of the relationship.

Because invertebrate specimens were too small to conduct mercury and stable isotope analyses on the same individual, means of pooled composite samples were used for lower trophic level organisms. Individual values were used for fish following recommendations by Borgå et al. (2012) and to remain consistent with other studies that used individual values for fish (e.g., Gantner et al. 2010a; Swanson and Kidd 2010). In order to facilitate comparisons with studies that used different statistical approaches (e.g., Campbell et al. 2005; Riget et al. 2007; and references therein), results for the slope of mercury concentration versus $\delta^{15}N$ are also provided, and were calculated using wet weight as well as dry weight mercury concentrations. To evaluate the potential bias associated with using individual values for fish and mean composite values for invertebrates, slopes of $\delta^{15}N$ versus log [THg] and log [MeHg] were also calculated using mean values for all taxa.

To determine whether the biomagnification rate of MeHg exceeded that of THg, a general linear model (ANCOVA) was used to assess the statistical significance of a difference in the slopes of the trophic level versus log [THg] and trophic level versus log [MeHg] relationships for all sampling sites. Linear mixed models were further used to explain log [THg] or [MeHg] as a function of trophic level, habitat type (lacustrine or marine), and the interaction between trophic level and habitat type, with sampling site included as a random-effect factor. The effect of trophic level on the percentage of mercury in the methylated form was evaluated using the non-linear logistic model of growth towards an asymptote:

Percent MeHg = $100 / (1 + e^{(\beta 1 + \beta 2*TL)})$

where the asymptote was set to 100%, β1 represents the %MeHg when trophic level

equals zero (i.e., the estimated intercept of the relationship), and β2 reflects the rate at

which %MeHg approaches the asymptote as a function of trophic level.

All statistical analyses were conducted using the R program for statistical

computing (R Development Core Team 2009) with Type I error set to $\alpha = 0.05$. Prior to

inclusion in regression analysis, THg and MeHg concentrations were $\log_{10}$ transformed

to ensure an approximately normal data distribution. Compliance with all required

statistical model assumptions was verified using diagnostic plots (e.g., normal Q-Q and

fitted values versus residuals).

3.3 Results

Concentrations of total mercury in individual samples spanned 4 orders of

magnitude, from 4 ng/g dw in *Fucus* sp. from Nain Bay to 4293 ng/g dw in a single non-

anadromous Arctic charr from Crazy Lake. Mercury concentrations were lowest in

algae and zooplankton, and increased with trophic level to maximum values in Arctic

charr and other fish species (Tables 3.1 and 3.2). Measured [MeHg] ranged from 0.4

ng/g dw in *Fucus* sp. from Nain Bay and kelp from Saglek Bay to 1028 ng/g dw in

ninespine stickleback from an unnamed lake near Pond Inlet. The percentage of

methylated mercury ranged from a low of 3% in Kelp from Saglek Bay to >99%

observed in individual Arctic charr, polar cod, sandlance, and sculpin from a variety of

locations. Percent MeHg also increased with trophic level. Minimal values were

observed in algae and zooplankton, and most fish species had >90% MeHg (Tables 3.1

and 3.2). The measured %MeHg in all Arctic charr exceeded 95%, with means ±

standard deviation of 99 ± 0.7% in non-anadromous (n = 10 fish from 2 sites) and 98 ±

1.3% in anadromous (n = 10 fish from 2 sites) individuals.

Where taxa were collected in both feeding habitats, lake-dwelling organisms had

higher mercury concentrations than those captured in the marine environment (when

data were pooled across all sampling locations, not necessarily from matched sampling

sites). The trend was evident in Arctic charr (pooled mean [THg] ± standard deviation:

lacustrine = 546 ± 548; marine = 182 ± 96 ng/g dw), algae (lacustrine = 91 ± 74; marine

= 53 ± 15 ng/g dw), gammarid amphipods (lacustrine = 76 ± 12; marine = 39 ± 18 ng/g

dw), zooplankton (lacustrine = 286 ± 294; marine = 20 ± 16 ng/g dw), snails (lacustrine

= 84; marine = 45 ± 0.05 ng/g dw), ninespine stickleback (lacustrine = 915 ± 260;

marine = 206 ± 116 ng/g dw), and threespine stickleback (lacustrine = 474 ± 216;

marine = 179 ± 80 ng/g dw). At each sampling location, non-anadromous Arctic charr

had significantly higher mean [THg] than anadromous conspecifics (two-sample t-tests;

all $p < 0.01$).

Log-transformed [THg] and [MeHg] were both significantly positively related to

trophic level in each lacustrine and marine foodweb examined (Tables 3.3 and 3.4,

Figures 3.2 and 3.3). Trophic magnification factors (TMFs) averaged 3.18 in lacustrine

and 2.25 in marine foodwebs for THg (dw), and were not significantly different between

the two feeding habitats (Welch two sample t-test; t = 1.8, d.f. = 7.1, p = 0.11). For

methylmercury (dw), TMFs averaged 4.70 in lacustrine and 3.98 in marine foodwebs,

and did not differ significantly between the two feeding habitats (Welch two sample t-

test; t = 0.9, d.f. = 9.9, p = 0.40). Slopes of log [Hg] versus $\delta^{15}N$ averaged 0.14 in

lacustrine and 0.10 in marine foodwebs for THg (dw), and 0.19 in lacustrine and 0.17 in

marine foodwebs for MeHg (dw). When calculated using wet weight concentrations,

slopes of log [Hg] versus $\delta^{15}N$ increased to average 0.18 and 0.12 for THg in lacustrine

and marine foodwebs, and 0.23 and 0.19 for MeHg in lacustrine and marine foodwebs.

When slopes of log [Hg] versus $\delta^{15}N$ were calculated using mean values for all taxa,

there was no consistent difference in lacustrine foodwebs (mean slope = 0.14) and a

slight increase in marine foodwebs (mean slope = 0.11) for THg (dw), while slopes for

MeHg (dw) increased to average 0.21 in lacustrine and 0.22 in marine foodwebs.

Estimated basal [THg] values ranged from 17 to 139 ng/g dw in lacustrine sites and

from 8 to 39 ng/g dw in marine sites. Estimated basal [MeHg] values ranged from 5 to

42 ng/g dw in lacustrine and from 1 to 11 ng/g dw in marine sites.

When the data from all lacustrine sites were considered together, the slope of the

trophic level versus log [MeHg] (dw) relationship (linear regression slope = 0.48, 95%

confidence interval = 0.41 – 0.55) was significantly greater (ANCOVA; p = 0.009) than

the slope of the relationship with [THg] (dw) (slope = 0.35, 95% confidence interval =

0.29 – 0.41). The same pattern occurred when the data from all marine sites were

considered; the slope of the relationship between trophic level and [MeHg] (dw) (slope =

0.59, 95% confidence interval = 0.54 – 0.64) was significantly greater (ANCOVA; p <

0.001) than the slope of trophic level versus [THg] (dw) (slope = 0.35, 95% confidence

interval = 0.30 – 0.39).

The linear mixed models used to explain log [THg] or log [MeHg] as a function of

trophic level and habitat type indicated that trophic level was an important determinant

of taxon [THg] or [MeHg], and that mercury concentrations at the base of the foodweb

were higher in the lacustrine than marine habitats for both total and methylmercury (Table 3.5, Figures 3.4 and 3.5). There was no significant interaction between habitat type and trophic level for either THg or MeHg, signifying that there was no consistent difference in the rates of [THg] and [MeHg] biomagnification between the two habitat types. Linear mixed model analysis conducted including site latitude as an additional covariate indicated no effect of site latitude on log [THg] ($F_{1, 512}$ = 1.20, p = 0.27), or log [MeHg] ($F_{1, 512}$ = 0.13, p = 0.72), therefore the latitude term was removed from both models. For both [THg] and [MeHg], models that included the random effects of sample site on the intercept and slope provided a significantly better fit to the data than models without the random effects (likelihood ratio tests; all p < 0.001), indicating that the slope and intercept of the log [THg] or [MeHg] versus trophic level relationships varied significantly among sampling sites (see also Figures 3.4 and 3.5). The estimated [THg] at the base of the foodweb was 37 ng/g dw for lacustrine and 17 ng/g dw for marine sites, with TMFs of 2.92 and 2.19 in the lacustrine and marine environments, respectively. The estimated [MeHg] at the base of the foodweb was 14 ng/g dw for lacustrine and 4 ng/g dw for marine sites, with TMFs of 4.35 and 3.82 in the lacustrine and marine environments, respectively.

Non-linear logistic models indicated that the percentage of mercury as methylmercury was significantly related to trophic level in both marine and lacustrine taxa (Figure 3.6). When a model containing separate β1 and β2 parameters for marine and lacustrine taxa was compared with the common-parameter model (i.e., data from both habitat types considered together), the separate parameter model did not provide a significantly better fit to the data (nested model ANOVA; F = 0.90, p = 0.41). Therefore,

there was insufficient evidence to detect a difference in the trophic level versus percent MeHg relationships between the two habitat types, and the common-parameter model was accepted (Table 3.6).

3.4 Discussion

No evidence was found to support the hypothesis that biomagnification rates in lacustrine foodwebs are consistently higher than those in spatially proximate marine foodwebs located in the eastern Canadian Arctic and sub-Arctic. At a given trophic level, lacustrine biota had higher [THg] and [MeHg] than marine foodweb items, and the hypothesis that mercury concentrations at the base of the foodweb are higher in the lacustrine environment was supported. Both lacustrine and marine taxa demonstrated a non-linear trend of increasing percent MeHg with trophic level, and biomagnification rates of MeHg exceeded that of THg in both habitat types. There was no effect of site latitude on mercury concentrations in marine or lacustrine biota, thus the difference between feeding habitats was consistent across a range of latitudes (56 – 72°N) in eastern Canada.

The mean mercury concentrations observed in anadromous and non-anadromous Arctic charr are consistent with previously reported values (e.g., Evans et al. 2005a; Riget et al. 2000; Swanson et al. 2011a; see Chapter 2 for a more detailed comparison). Observed [THg] and [MeHg] in other lacustrine biota were similar to or higher than previously reported values for lakes in the Canadian Arctic and sub-Arctic (Gantner et al. 2010a; Power et al. 2002; Swanson and Kidd 2010). Observed total and methylmercury concentrations in marine biota were usually similar to or lower than

previously reported values in Arctic marine foodwebs (Atwell et al. 1998; Campbell et al. 2005; Loseto et al. 2008; Nfon et al. 2009; Riget et al. 2007).

In the present study, mercury biomagnification rates in the lacustrine foodwebs did not significantly exceed rates in the marine foodwebs. The finding is consistent with previous indications that the slope of the [Hg] versus δ^{15}N or trophic level relationship is relatively consistent among diverse aquatic habitats (Campbell et al. 2005; Chumchal et al. 2011; Riget et al. 2007). In contrast, Swanson and Kidd (2010) found that MeHg biomagnification rates in freshwater foodwebs (slopes of log [MeHg] versus δ^{15}N = 0.16 – 0.26) were significantly higher than in a nearby marine foodweb (slope = 0.08). In the lacustrine feeding habitat, slopes of δ^{15}N versus [Hg] calculated here were 0.09 – 0.21 for THg and 0.14 – 0.24 for MeHg, and are comparable to previously reported values for lakes in Canada including (for total Hg) 0.192 for Stewart Lake, northern Quebec (Power et al. 2002), 0.17 – 0.29 for lakes with five to eight fish species in north-western Ontario (Kidd et al. 1995), and (for MeHg) 0.16 – 0.26 for the West Kitikmeot region, Nunavut (Swanson and Kidd 2010), and 0.18 – 0.23 for lakes in Nova Scotia (Wyn et al. 2009). Observed lacustrine trophic magnification factors (TMFs) were 1.98 – 5.19 (THg) and 3.02 – 6.69 (MeHg), which are on the low end of the range of TMFs calculated for a series of lakes across the Canadian Arctic (3.6 – 64.3 for MeHg in muscle tissue/whole homogenates of fish and invertebrates; Gantner et al. 2010a).

In the marine feeding environment, calculated slopes of δ^{15}N versus [Hg] were 0.06 – 0.13 for THg and 0.13 – 0.22 for MeHg, which were usually lower than previously reported slopes for northern marine foodwebs including 0.232 – 0.255 for THg and 0.254 – 0.311 for MeHg in the Beaufort Sea (Loseto et al. 2008), 0.197 for THg and

0.223 for MeHg in the Northwater Polynya, Baffin Bay (Campbell et al. 2005), 0.183 for THg and 0.339 for MeHg in western Greenland (using the natural logarithm of [Hg]; Riget et al. 2007), 0.134 for THg and 0.201 for MeHg in the Gulf of St. Lawrence, Canada (Lavoie et al. 2010), 0.2 for THg in Lancaster Sound, Nunavut (Atwell et al. 1998), and 0.08 for MeHg in Melville Sound, Nunavut (Swanson and Kidd 2010). However, it should be noted that the previously mentioned studies (with the exception of Swanson and Kidd 2010) included birds and/or mammals in the examined foodwebs. The inclusion of homeotherms is known to increase the contaminant biomagnification rate in the foodweb (Fisk et al. 2001; Hop et al. 2002), and it is not surprising that these studies found higher biomagnification rates than those that examined only invertebrates and fish (this study and Swanson and Kidd 2010). Similarly, the TMFs calculated for our marine foodwebs (1.59 – 2.82 for THg) were lower than that calculated using seabirds and fish from Svalbard (4.87 for THg in muscle, Jaeger et al. 2009).

Foodweb biomagnification rates calculated using wet weight mercury concentrations exceed those calculated using dry weight concentrations, presumably because moisture content is variable, and is generally highest in invertebrates (Tables 3.1 and 3.2). Lavoie et al. (2010) also determined higher biomagnification rates for MeHg and THg using wet weight concentrations in the Gulf of St. Lawrence, as did Riget et al. (2007) in Davis Strait, west Greenland. When biomagnification rates were investigated using means for all species (as opposed to individual values for fish), the slopes of $\delta^{15}N$ versus [Hg] were relatively unaffected for total mercury, but increased for methylmercury. This is likely because of the high %MeHg observed in all fish species (usually >90%), and because of the high number of fish samples relative to non-fish

samples from our study sites. Because of the potential bias in the calculated

slopes/TMFs (particularly with MeHg) induced by using individual values for fish and

pooled composite values for invertebrates, it is recommended that the models

presented herein not be extrapolated to higher trophic levels beyond Arctic charr.

Pooling all values for Arctic charr would also be inappropriate, as the species frequently

occupies more than one trophic level. Cannibalism is common in Arctic charr (Johnson

1980; Klemetsen et al. 2003), and had the effect of lengthening the food chain in some

cases (e.g., Crazy Lake), a phenomenon also noted in Gantner et al. (2010a).

The similar biomagnification rates and difference in basal mercury concentrations

we observed between the two habitats suggest that mercury concentrations in upper

trophic level aquatic organisms are ultimately determined by mercury uptake rates at

the base of the foodweb (Chumchal et al. 2008; Loseto et al. 2008; Mason et al. 1995;

Wyn et al. 2009). Our conclusion, that basal [Hg] in the lacustrine environment exceeds

that in the marine environment, is corroborated by reported mercury concentrations in

northern lacustrine zooplankton (e.g., 2 – 26 ng/g ww THg, 50 – 660 ng/g dw THg and

25 – 483 ng/g dw MeHg, this study; 30 – 297 ng/g dw THg and 10 – 269 ng/g dw MeHg,

Chetelat and Amyot 2009; 60 ± 40 ng/g dw THg and 20 ± 30 ng/g dw MeHg, Gantner et

al. 2010a; 1 – 56 ng/g dw MeHg, Swanson et al. 2011a; 33 – 206 ng/g dw THg, Watras

et al. 1998; 90 – 230 ng/g dw MeHg, Wyn et al. 2009) that usually exceed

concentrations in northern marine zooplankton (e.g., 1 – 3 ng/g ww THg, 6 – 33 ng/g dw

THg and 1 – 3 ng/g dw MeHg, this study; 6 ± 5 ng/g ww THg, Nfon et al. 2009; 6 ± 2

ng/g ww THg, Campbell et al. 2005; 35 ± 5 ng/g dw THg and 10 ± 1 ng/g dw MeHg,

Loseto et al. 2008; 13 – 65 ng/g dw THg and 3 – 6 ng/g dw MeHg, Lavoie et al. 2010).

Higher basal [Hg] in freshwater foodwebs may be explained by enhanced bioavailability of mercury at low pH and low salinity, which are conditions typically found in freshwaters relative to seawater (Barkay et al. 1997; Morel et al. 1998; Ullrich et al. 2001). Furthermore, the uptake of MeHg by single-celled organisms is greater than that of inorganic Hg (Pickhardt and Fisher 2007), and mercury methylation is favoured by warm water temperature and relatively low sulphate concentration, as found in lakes relative to the marine environment (Ullrich et al. 2001). Accordingly, the proportion of methylated Hg is often higher in freshwater (≈30%) than in seawater (typically <5%) (Leopold et al. 2010; Ullrich et al. 2001; and references therein), although high percentages of methylated Hg (30 – 40%) have been measured in Arctic marine waters at depth or under sea ice (Kirk et al. 2008; St. Louis et al. 2007). Finally, total mercury concentrations measured in freshwater are generally higher than those in seawater (Leopold et al. 2010; Ullrich et al. 2001; and references therein), and in the Canadian Arctic and sub-Arctic specifically, [THg] observed in lakes (e.g., 0.30 – 1.39 ng/L, Chetelat and Amyot 2009; 1.8 ± 0.8 ng/L, Evans et al. 2005b; 0.29 – 0.72 ng/L, Gantner et al. 2010b; 0.56 ± 0.09 to 1.44 ± 0.55 ng/L, Loseto et al. 2004; 2.66 ± 2.70 ng/L in ponds and 1.11 ± 0.48 ng/L in lakes, St. Louis et al. 2005) were usually higher than concentrations reported for marine water (0.14 – 0.24 ng/L, St. Louis et al. 2007; 0.40 ± 0.47 ng/L, Kirk et al. 2008). The higher mercury concentrations in lakes relative to marine water may reflect watershed-related inputs of inorganic or methylmercury, such as surface runoff from melting snow or contributions from wetlands (Hammerschmidt et al. 2006; Loseto et al. 2004; Semkin et al. 2005).

The trend for the proportion of MeHg to increase with trophic level has been previously observed, although the present study is, to our knowledge, the first to explicitly model the relationship and test for differences among aquatic habitat types (lacustrine and marine). One novel finding of the present study is that the increase in %MeHg with trophic level was statistically similar in lacustrine and marine foodwebs. Percent MeHg increases with trophic level because MeHg is more efficiently transferred from prey to predator than total Hg (Mason et al. 1995; Morel et al. 1998), which also caused the biomagnification rate of MeHg to exceed that of THg in both feeding habitats. Our results suggest that the relative trophic transfer efficiencies of total and methylmercury are unaffected by the biotic and abiotic factors that differ between the lacustrine and marine feeding habitats (e.g., mercury concentrations in biota, water chemistry, species composition, etc.).

There was no effect of site latitude on [THg] or [MeHg] in marine or lacustrine biota, and the difference in [THg] of upper trophic level consumers (Arctic charr) between the two feeding habitats was independent of latitude (49 – 81° N) in eastern Canada (Chapter 2). This finding agrees with results presented in Gantner et al. (2010b), but contrasts with evidence from lake sediment cores suggesting a pattern of decreasing mercury deposition from south to north in North America (Muir et al. 2009b; Schindler et al. 1995). Furthermore, mercury methylation and uptake by biota increase with water temperature, productivity, and nutrient availability (Chetelat and Amyot 2009; St. Louis et al. 2005; Ullrich et al. 2001), which covary with latitude. Together these results suggest that large-scale abiotic factors such as temperature and mercury

deposition rates are not as important as site-specific chemical, physical, and/or biological factors in determining [Hg] in biota (Chetelat et al. 2008; Rose et al. 1999).

3.4.1 Summary

To our knowledge, the present study is the first wide-scale spatial investigation of mercury biomagnification in co-located pairs of lacustrine and marine foodwebs supporting a common predator (Arctic charr). We conclude that the difference in mercury concentration between anadromous and non-anadromous Arctic charr is not driven by differential biomagnification rates in lacustrine and marine foodwebs, a conclusion corroborated by evidence that differences in fish mercury concentrations among lakes may not be related to within-lake biomagnification rates (Wyn et al. 2009; Gantner et al. 2010b). Rather, a difference in prey mercury concentration, driven by differential mercury concentrations at of the base of the lacustrine and marine foodwebs, is important for explaining the difference in mercury concentration between anadromous and non-anadromous in Arctic charr. Indeed, results from the present study may be extended to explain the general tendency for marine or anadromous fish to have lower mercury concentrations than nearby freshwater fish. There was no effect of site latitude on mercury concentrations in marine or lacustrine biota, suggesting that individual site characteristics are more important than wide-scale regional differences in determining [Hg] in biota. Therefore, our results imply that spatial extrapolation (i.e., from site to site) will suffer from errors, and that more, not less, monitoring is needed.

3.5 Tables and Figures

Table 3.1. Summary of samples obtained from lacustrine sampling sites. Sample sites are ordered by increasing latitude, with location abbreviations, as defined in Figure 3.1, following in parentheses. Stable isotope and mercury data are presented as mean ± standard error when n ≥ 3 and as individual values when n < 3, trophic level and percent moisture (% H_2O) are given as means only. Within sites, taxa are ordered by increasing $\delta^{15}N$/trophic level. Sample sizes are provided in parentheses for stable isotope analysis, THg analysis, and MeHg analysis. NA = not analyzed.

Site	Taxon	$\delta^{13}C$ (‰)	$\delta^{15}N$ (‰)	Trophic level		[THg] (ng/g dw)		Percent MeHg		% H_2O
Coady's Pond #2 (NB)										
	Algae	-17.1 ± 0.5	0.7 ± 0.7	1.4	(3)	24	(1)	8	(1)	65
	Cladocera	-26.8 ± 0.4	1.2 ± 0.1	1.5	(3)	324 ± 59	(3)	NA		93
	Clams (Sphaeriidae)[a]	-24.3 ± 0.2	2.8 ± 0.4	2.0	(3)	NA		NA		NA
	Trichoptera	-22.4 ± 0.4	3.2 ± 0.1	2.1	(4)	115 ± 10	(3)	75, 76	(2)	83
	Diptera larvae	-24.4 ± 0.1	3.5 ± 0.2	2.2	(3)	388	(1)	77	(1)	88
	Coleoptera	-24.8 ± 0.9	4.6 ± 0.2	2.5	(6)	180	(1)	97	(1)	70
	Arctic charr	-21.1 ± 0.3	6.9 ± 0.1	3.2	(20)	503 ± 41	(20)	99 ± 0.3	(5)	76
	Threespine stickleback	-20.7 ± 0.6	7.0 ± 0.5	3.3	(3)	512, 1007	(2)	NA		NA
Esker Lake (OB)										
	Trichoptera	-24.4 ± 0.9	2.0 ± 0.3	1.1	(3)	16, 25	(2)	54, 57	(2)	65
	Chironimids[a]	-28.2 ± 0.4	5.2 ± 0.3	2.0	(4)	162, 184	(2)	82, 86	(2)	89
	Terrestrial insects[b]	-26.7, -26.2	3.9, 6.8	2.0	(2)	98 ± 15	(3)	63, 73	(2)	91
	Arctic charr	-24.0 ± 0.4	7.8 ± 0.2	2.8	(24)	555 ± 168	(24)	NA		NA
Upper Nakvak Lake (SB)										
	Terrestrial insects[b]	-22.1 ± 0.9	4.5 ± 0.2	1.8	(3)	103, 106	(2)	68, 70	(2)	92
	Chironimids[a]	-20.2 ± 0.7	5.1 ± 0.5	2.0	(3)	183, 248	(2)	63, 74	(2)	89
	Arctic charr	-19.2 ± 0.3	9.2 ± 0.2	3.2	(22)	492 ± 114	(22)	NA		NA

Table 3.1. (Continued)

Site / Taxon	$\delta^{13}C$ (‰)	$\delta^{15}N$ (‰)	Trophic level		[THg] (ng/g dw)		Percent MeHg		% H_2O
Tasiapik Lake (DB)									
Algae	-16.0 ± 0.5	2.2 ± 0.7	1.4	(18)	27 ± 2	(3)	13	(1)	89
Gastropods[a]	-22.4 ± 0.5	4.2 ± 0.1	2.0	(18)	85	(1)	NA		68
Gammarid amphipods	-21.1 ± 0.2	4.3 ± 0.2	2.0	(27)	76 ± 7	(3)	90, 91	(2)	87
Coleoptera	-25.7 ± 1.9	5.6 ± 0.4	2.4	(3)	200	(1)	97	(1)	81
Trichoptera	-23.3 ± 0.4	6.3 ± 0.1	2.6	(22)	85, 143	(2)	76, 87	(2)	80
Zooplankton	-26.0 ± 0.3	7.7 ± 0.5	3.0	(15)	47, 54	(2)	47	(1)	96
Cladocera	-29.6 ± 0.1	8.1 ± 1.3	3.1	(3)	211 ± 17	(3)	NA		96
Threespine stickleback	-24.9 ± 0.3	10.1 ± 0.1	3.7	(25)	364 ± 31	(7)	98, 99	(2)	75
Arctic charr	-23.9 ± 0.3	11.2 ± 0.1	4.1	(31)	462 ± 37	(31)	NA		NA
Crazy Lake (IQ)									
Algae	-29.4, -29.6	1.1, 1.5	1.2	(2)	31, 32	(2)	15	(1)	88
Zooplankton	-29.3, -29.5	3.4, 3.5	1.9	(2)	139, 156	(2)	22	(1)	95
Chironimids[a]	-17.8 ± 1.1	4.0 ± 0.3	2.0	(4)	232	(1)	61	(1)	89
Trichoptera	-17.3	5.0	2.3	(1)	60	(1)	59	(1)	76
Arctic charr	-19.3 ± 0.3	8.5 ± 0.2	3.3	(47)	467 ± 97	(47)	NA		NA
unnamed lake (PI)									
Algae	-23.1 ± 1.4	0.0 ± 0.8	1.2	(6)	166 ± 21	(6)	34	(1)	89
Chironomids[a]	-23.5 ± 0.2	2.8 ± 0.2	2.0	(4)	NA		NA		NA
Trichoptera	-29.6 ± 0.4	3.5 ± 0.2	2.2	(6)	156 ± 124	(4)	24, 32	(2)	78
Zooplankton	-31.0 ± 0.4	3.6 ± 0.5	2.2	(5)	642, 680	(2)	71	(1)	96
Ninespine stickleback	-27.5 ± 0.4	8.3 ± 0.2	3.6	(16)	959 ± 56	(22)	97 ± 1	(3)	78
Arctic charr	-25.8 ± 0.2	8.8 ± 0.2	3.8	(18)	1003 ± 108	(18)	99 ± 0.3	(5)	79

[a] Taxon used to assign $\delta^{15}N_{base}$
[b] Includes Diptera, Hymenoptera, and Hemiptera

Table 3.2. Summary of samples obtained from marine sampling sites. Sample sites are ordered by increasing latitude, with location abbreviations, as defined in Figure 3.1, following in parentheses. Stable isotope and mercury data are presented as mean ± standard error when n ≥ 3 and as individual values when n < 3, trophic level and percent moisture (% H_2O) are given as means only. Within sites, taxa are ordered by increasing $\delta^{15}N$/trophic level. Sample sizes are provided in parentheses for stable isotope analysis, THg analysis, and MeHg analysis. NA = not analyzed.

Site	Taxon	$\delta^{13}C$ (‰)	$\delta^{15}N$ (‰)	Trophic level		[THg] (ng/g dw)		Percent MeHg		% H_2O
Nain Bay (NB)										
	Algae (*Fucus* sp.)	-15.9 ± 0.5	6.4 ± 0.4	1.2	(4)	7 ± 1	(4)	10	(1)	70
	Mysids	-18.2 ± 0.1	8.6 ± 0.2	1.9	(3)	16	(1)	79	(1)	65
	Mussels[a]	-20.8 ± 0.1	9.0 ± 0.2	2.0	(5)	83	(1)	42	(1)	87
	Gammarid amphipods	-16.8 ± 0.1	9.5 ± 0.2	2.2	(4)	16, 17	(2)	78, 83	(2)	70
	Hyperiid amphipods	-20.6 ± 0.3	9.7 ± 0.2	2.2	(3)	22	(1)	NA		83
	Sculpin	-20.2 ± 0.2	10.9 ± 0.2	2.6	(5)	27 ± 2	(14)	84 ± 4	(3)	80
	Sandlance	-21.3 ± 0.3	12.2 ± 0.1	2.9	(5)	28 ± 3	(6)	97	(1)	74
	Blennies	-19.3 ± 0.6	12.9 ± 0.2	3.2	(5)	136 ± 8	(5)	NA		93
	Capelin	-20.6 ± 0.1	13.3 ± 0.2	3.3	(4)	67 ± 11	(5)	99	(1)	77
	Arctic charr	-19.1 ± 0.2	14.8 ± 0.1	3.7	(20)	89 ± 7	(20)	98 ± 1	(5)	75
Okak Bay (OB)										
	Algae (*Fucus* sp.)	-14.9 ± 0.4	3.8 ± 0.3	0.5	(3)	8 ± 1	(3)	19	(1)	74
	Gammarid amphipods	-17.5 ± 0.5	8.1 ± 0.7	1.8	(5)	46 ± 17	(3)	81 ± 9	(3)	84
	Mysids	-16.5 ± 0.0	8.3 ± 0.2	1.9	(3)	40, 221	(2)	85, 98	(2)	86
	Mussels[a]	-19.5 ± 0.4	8.8 ± 0.3	2.0	(5)	121, 134	(2)	54, 57	(2)	88
	Sculpin	-19.9 ± 0.1	11.2 ± 0.2	2.7	(5)	37 ± 3	(6)	88	(1)	79
	Sandlance	-19.8 ± 0.5	11.6 ± 0.1	2.8	(5)	57 ± 15	(6)	94 ± 4	(3)	72
	Threespine stickleback	-16.5 ± 0.7	12.8 ± 0.7	3.2	(5)	179 ± 33	(6)	95	(1)	NA
	Arctic charr	-18.6 ± 0.2	13.7 ± 0.3	3.4	(20)	193 ± 25	(20)	NA		NA
Saglek Bay (SB)										
	Algae (*Fucus* sp.)	-10.6, -10.4	4.0, 4.6	1.0	(2)	10	(1)	10	(1)	68
	Algae (filamentous)	-20.1, -16.2	4.4, 5.1	1.1	(2)	34	(1)	13	(1)	88

Table 3.2. (Continued)

Site	Taxon	δ^{13}C (‰)	δ^{15}N (‰)	Trophic level		[THg] (ng/g dw)		Percent MeHg		% H$_2$O
	Algae (kelp)	-17.0, -16.3	5.3, 5.4	1.3	(2)	17, 17	(2)	3	(1)	81
	Gammarid amphipods	-17.3 ± 0.2	7.7 ± 0.1	2.0	(3)	41 ± 2	(3)	69 ± 6	(3)	81
	Hyperiid amphipods	-22.8 ± 0.3	7.9 ± 0.5	2.0	(3)	27, 30	(2)	NA		88
	Mussels[a]	-19.9 ± 0.3	7.9 ± 0.2	2.0	(5)	70	(1)	61	(1)	77
	Mysids	-17.6 ± 0.9	8.6 ± 0.2	2.2	(4)	38	(1)	82	(1)	88
	Isopods	-19.0, -15.6	10.1, 11.6	2.9	(2)	21	(1)	NA		71
	Gunnels	-20.0 ± 0.1	11.3 ± 0.2	3.0	(5)	35 ± 4	(5)	NA		75
	Sculpin	-18.9 ± 0.2	11.8 ± 0.3	3.2	(5)	134 ± 55	(12)	94 ± 3	(3)	76
	Ninespine stickleback	-16.5 ± 1.0	12.2 ± 0.3	3.3	(5)	206 ± 44	(7)	NA		NA
	Decapod	-15.1	13.3	3.6	(1)	29	(1)	NA		75
	Arctic charr	-19.9 ± 0.1	14.0 ± 0.1	3.8	(22)	83 ± 10	(22)	NA		NA
Dry Bay (DB)										
	Algae (*Fucus* sp.)	-17.7 ± 0.3	4.6 ± 0.1	0.9	(5)	22, 27	(2)	30	(1)	80
	Algae (kelp)	-18.1 ± 0.2	4.7 ± 0.4	0.9	(5)	31, 37	(2)	17	(1)	92
	Algae (filamentous)	-14.5 ± 0.1	5.5 ± 0.2	1.1	(3)	51, 57	(2)	30	(1)	93
	Gammarid amphipods	-17.2 ± 0.2	8.2 ± 0.2	1.9	(6)	49 ± 13	(3)	68 ± 17	(3)	84
	Mussels[a]	-20.5 ± 0.2	8.5 ± 0.1	2.0	(5)	176 ± 22	(6)	54, 60	(2)	77
	Plankton	-20.3, -20.2	7.6, 10.5	2.2	(2)	31, 35	(2)	11	(1)	90
	Sandlance	-19.6 ± 0.2	11.8 ± 0.3	3.0	(3)	50 ± 7	(7)	95	(1)	73
	Sculpin	-17.5	12.7	3.2	(1)	211	(1)	NA		NA
	Arctic charr	-19.7 ± 0.1	13.8 ± 0.1	3.6	(33)	158 ± 12	(33)	NA		NA
Iqaluit (IQ)										
	Plankton	-22.5 ± 0.2	7.0 ± 0.3	1.5	(7)	5, 7	(2)	5	(1)	NA
	Algae (*Fucus* sp.)	-16.6 ± 0.6	7.1 ± 0.4	1.5	(6)	7 ± 1	(3)	22	(1)	81
	Algae (kelp)	-17.7 ± 0.9	7.8 ± 0.3	1.7	(3)	9 ± 0.1	(3)	5	(1)	87
	Barnacles[a]	-21.6 ± 0.2	8.7 ± 0.1	2.0	(5)	18 ± 1	(3)	41, 42	(2)	78
	Decapod	-22.5, -21.3	7.9, 10.9	2.2	(2)	54	(1)	NA		86
	Gastropods	-15.8 ± 0.1	9.8 ± 0.2	2.3	(5)	45, 45	(2)	NA		82
	Hyperiid amphipods	-21.6 ± 0.3	10.1 ± 0.2	2.4	(3)	45, 51	(2)	NA		86
	Gammarid amphipods	-17.4 ± 0.1	10.3 ± 0.2	2.5	(10)	30 ± 0.4	(5)	78 ± 1	(3)	82
	Mysids	-21.4 ± 0.2	11.0 ± 0.1	2.7	(3)	67	(1)	92	(1)	90
	Polar cod	-19.9 ± 0.1	14.5 ± 0.1	3.7	(20)	114 ± 6	(23)	99 ± 0.1	(3)	81

Table 3.2. (Continued)

Site	Taxon	$\delta^{13}C$ (‰)	$\delta^{15}N$ (‰)	Trophic level		[THg] (ng/g dw)		Percent MeHg		% H_2O
	Arctic charr	-19.0 ± 0.2	14.5 ± 0.2	3.7	(30)	144 ± 7	(30)	NA		NA
Pond Inlet (PI)										
	Algae (*Fucus* sp.)	-19.4 ± 1.2	6.0 ± 0.2	1.2	(3)	15	(1)	7	(1)	79
	Algae (other)	-19.5 ± 0.4	6.7 ± 0.4	1.4	(6)	10, 13	(2)	NA		85
	Algae (kelp)	-18.9 ± 1.3	7.1 ± 0.2	1.6	(3)	16	(1)	4	(1)	85
	Algae (filamentous)	-21.8, -21.4	7.2, 8.2	1.7	(2)	69	(1)	38	(1)	91
	Calanoids	-26.3 ± 0.2	8.2 ± 0.2	1.9	(3)	154	(1)	NA		93
	Gammarid amphipods[a]	-21.4 ± 0.4	8.6 ± 0.3	2.0	(10)	41 ± 6	(4)	66 ± 10	(3)	81
	Mysids	-24.0 ± 0.3	9.4 ± 0.1	2.2	(3)	50, 53	(2)	91, 91	(2)	91
	Hyperiid amphipods	-24.4 ± 0.6	9.5 ± 0.6	2.3	(3)	58 ± 1	(3)	NA		90
	Sandlance	-21.8 ± 0.1	12.5 ± 0.1	3.2	(25)	153 ± 15	(28)	98 ± 1	(3)	76
	Polar cod	-21.3 ± 0.1	13.6 ± 0.2	3.5	(16)	201 ± 20	(17)	98 ± 0.3	(3)	81
	Arctic charr	-21.1 ± 0.1	14.2 ± 0.1	3.7	(74)	247 ± 11	(74)	98 ± 0.3	(5)	74

[a]Taxon used to assign $\delta^{15}N_{base}$

Table 3.3. Relationships between log [THg] (ng/g) and $\delta^{15}N$ or trophic level within each marine and lacustrine foodweb. Sample sites are ordered by increasing latitude, with location abbreviations following in parentheses. All regressions were significant (all $p < 0.001$).

	$\log_{10}$ [THg] (ng/g) versus $\delta^{15}N$				$\log_{10}$ [THg] (ng/g dw) versus trophic level			
Sample site (location)	Slope (dw)	R^2	Slope (ww)	R^2	Intercept[a]	Slope	10^{slope} (TMF)	R^2
Lacustrine foodwebs								
Coady's Pond #2 (NB)	0.13	0.55	0.15	0.79	1.74	0.43	2.68	0.55
Esker Lake (OB)	0.21	0.60	0.22	0.61	1.30	0.72	5.19	0.60
Upper Nakvak Lake (SB)	0.15	0.42	0.21	0.62	1.39	0.52	3.33	0.43
Tasiapik Lake (DB)	0.11	0.67	0.14	0.60	1.48	0.37	2.37	0.70
Crazy Lake (IQ)	0.16	0.62	0.20	0.79	1.24	0.55	3.55	0.64
unnamed lake (PI)	0.09	0.50	0.14	0.75	2.14	0.30	1.98	0.50
Marine foodwebs								
Nain Bay (NB)	0.11	0.53	0.11	0.73	0.95	0.37	2.35	0.53
Okak Bay (OB)	0.11	0.50	0.12	0.61	1.31	0.37	2.33	0.50
Saglek Bay (SB)	0.06	0.26	0.07	0.36	1.36	0.20	1.59	0.26
Dry Bay (DB)	0.06	0.44	0.10	0.60	1.59	0.22	1.65	0.44
Iqaluit (IQ)	0.13	0.75	0.16	0.76	0.90	0.44	2.77	0.75
Pond Inlet (PI)	0.13	0.67	0.16	0.77	1.16	0.45	2.82	0.67

[a]At trophic level = 1

Table 3.4. Relationships between log [MeHg] (ng/g) and $\delta^{15}N$ or trophic level within each marine and lacustrine foodweb. Sample sites are ordered by increasing latitude, with location abbreviations following in parentheses. All regressions were significant (all $p < 0.001$).

Sample site (location)	$\log_{10}$ [MeHg] (ng/g) versus $\delta^{15}N$				$\log_{10}$ [MeHg] (ng/g dw) versus trophic level			
	Slope (dw)	R^2	Slope (ww)	R^2	Intercept[a]	Slope	10^{slope} (TMF)	R^2
Lacustrine foodwebs								
Coady's Pond #2 (NB)	0.22	0.64	0.24	0.81	1.06	0.73	5.42	0.64
Esker Lake (OB)	0.24	0.66	0.26	0.66	1.10	0.83	6.69	0.66
Upper Nakvak Lake (SB)	0.18	0.51	0.23	0.68	1.22	0.60	3.94	0.51
Tasiapik Lake (DB)	0.15	0.68	0.19	0.61	1.02	0.52	3.32	0.68
Crazy Lake (IQ)	0.22	0.80	0.26	0.86	0.72	0.76	5.79	0.81
unnamed lake (PI)	0.14	0.66	0.19	0.84	1.63	0.48	3.02	0.66
Marine foodwebs								
Nain Bay (NB)	0.16	0.65	0.17	0.81	0.47	0.56	3.63	0.66
Okak Bay (OB)	0.15	0.62	0.16	0.70	0.98	0.50	3.19	0.62
Saglek Bay (SB)	0.14	0.53	0.16	0.60	0.59	0.49	3.09	0.53
Dry Bay (DB)	0.13	0.63	0.16	0.68	1.03	0.43	2.72	0.63
Iqaluit (IQ)	0.22	0.73	0.24	0.74	0.07	0.75	5.57	0.73
Pond Inlet (PI)	0.22	0.79	0.25	0.82	0.37	0.76	5.70	0.79

[a]At trophic level = 1

Table 3.5. Results for fixed effect factors from the linear mixed models used to explain log [THg] or log [MeHg] using trophic level, habitat type, and the interaction between trophic level and habitat type, given the random effect of sampling site (n = 12 sites). Habitat type = lacustrine (L) or marine (M).

Response	n	Source	Coefficient estimate	Standard error	F-value	p
log [THg] (ng/g dw)	527	Intercept[a]	L: 1.57 M: 1.22	L: 0.12 M: 0.17	1664.60	< 0.001
		Trophic level	L: 0.47 M: 0.34	L: 0.05 M: 0.07	128.50	< 0.001
		Habitat type			31.03	< 0.001
		Trophic level x habitat			3.18	0.075
log [MeHg] (ng/g dw)	527	Intercept[a]	L: 1.14 M: 0.59	L: 0.15 M: 0.20	1067.59	< 0.001
		Trophic level	L: 0.64 M: 0.58	L: 0.06 M: 0.08	224.18	< 0.001
		Habitat type			28.48	< 0.001
		Trophic level x habitat			0.46	0.497

[a] At trophic level = 1

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Table 3.6. Results for the non-linear logistic model explaining percent MeHg as a function of trophic level. The model was fitted using data from both the lacustrine and marine habitat types.

Parameter	Estimate	Standard error	t-value	p	Lower CI	Upper CI
β1	4.46	0.60	7.38	< 0.001	3.26	5.66
β2	-2.43	0.31	-7.86	< 0.001	-3.04	-1.81
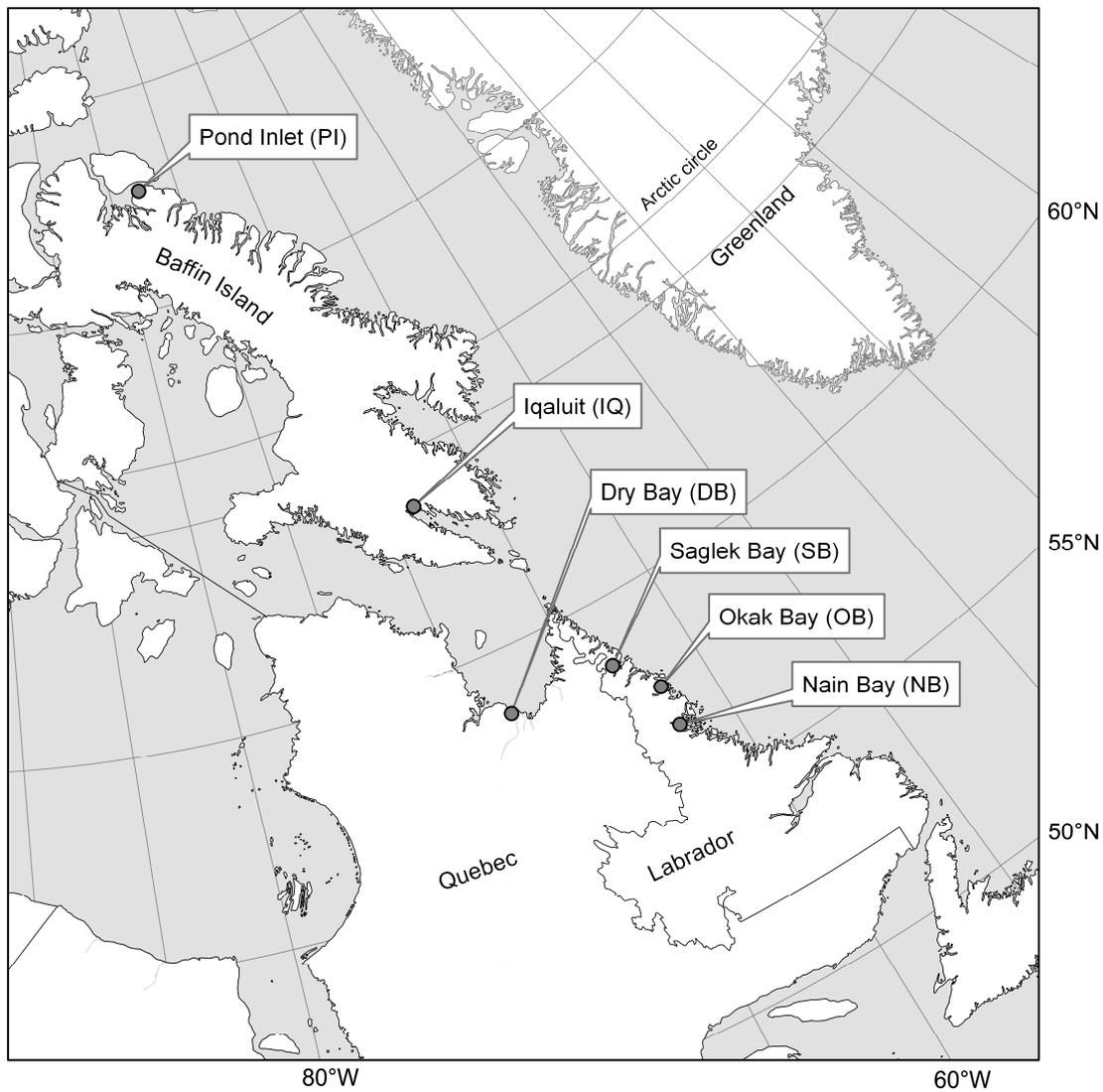
Figure 3.1. Map of north-eastern Canada indicating the six locales of marine and freshwater foodweb sampling. Location abbreviations are given in parentheses.
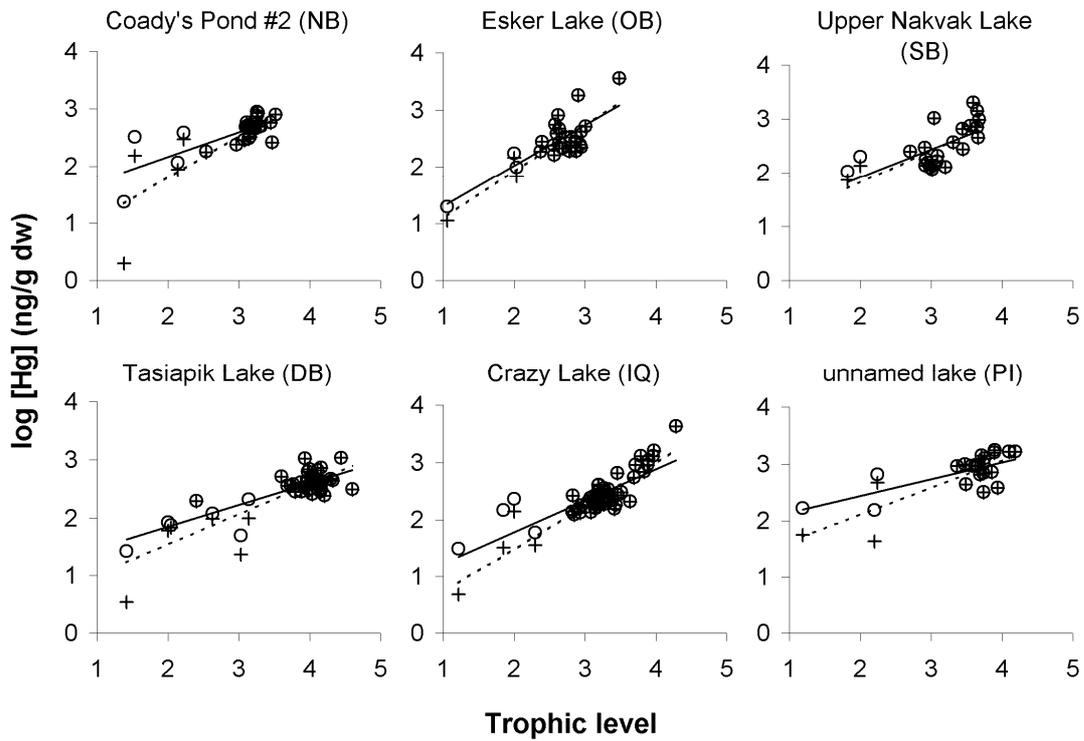
Figure 3.2. Trophic level versus log [Hg] (ng/g dw) for each lacustrine foodweb. Points represent [THg] (circles) and [MeHg] (+) of individual fish or invertebrate taxa. Significant linear regressions are indicated for [THg] (solid line) and [MeHg] (dashed line). Sample sites are ordered by increasing latitude, with location abbreviations following in parentheses.
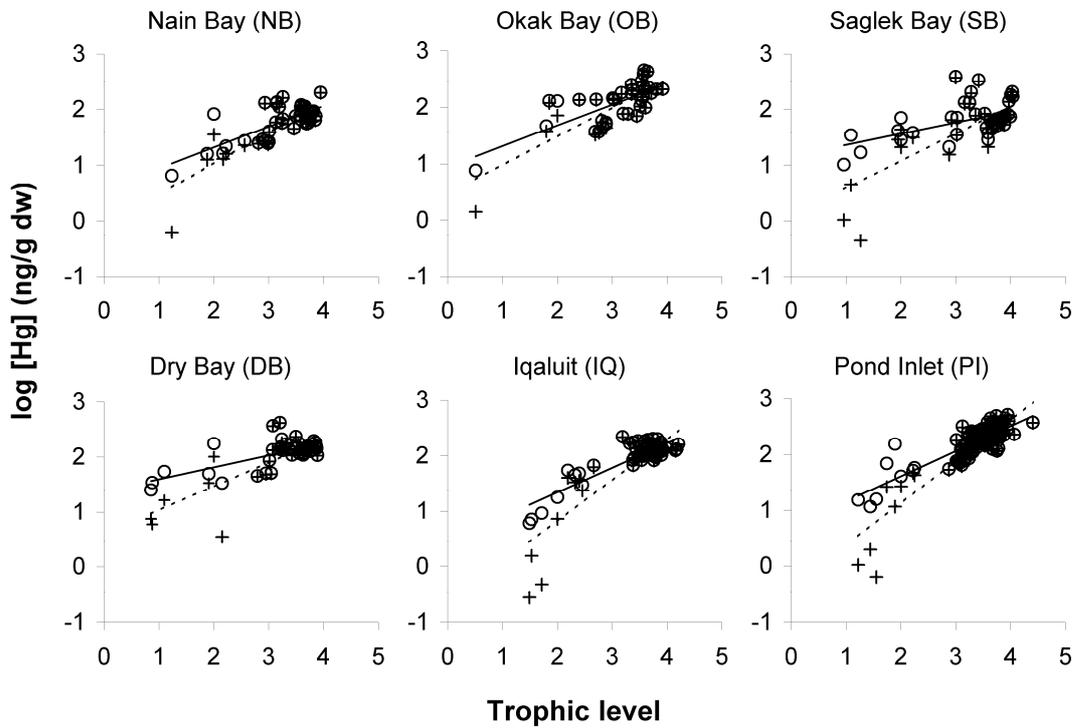
Figure 3.3. Trophic level versus log [Hg] (ng/g dw) for each marine foodweb. Points represent [THg] (circles) and [MeHg] (+) of individual fish or invertebrate taxa. Significant linear regressions are indicated for [THg] (solid line) and [MeHg] (dashed line). Sample sites are ordered by increasing latitude, with location abbreviations following in parentheses.
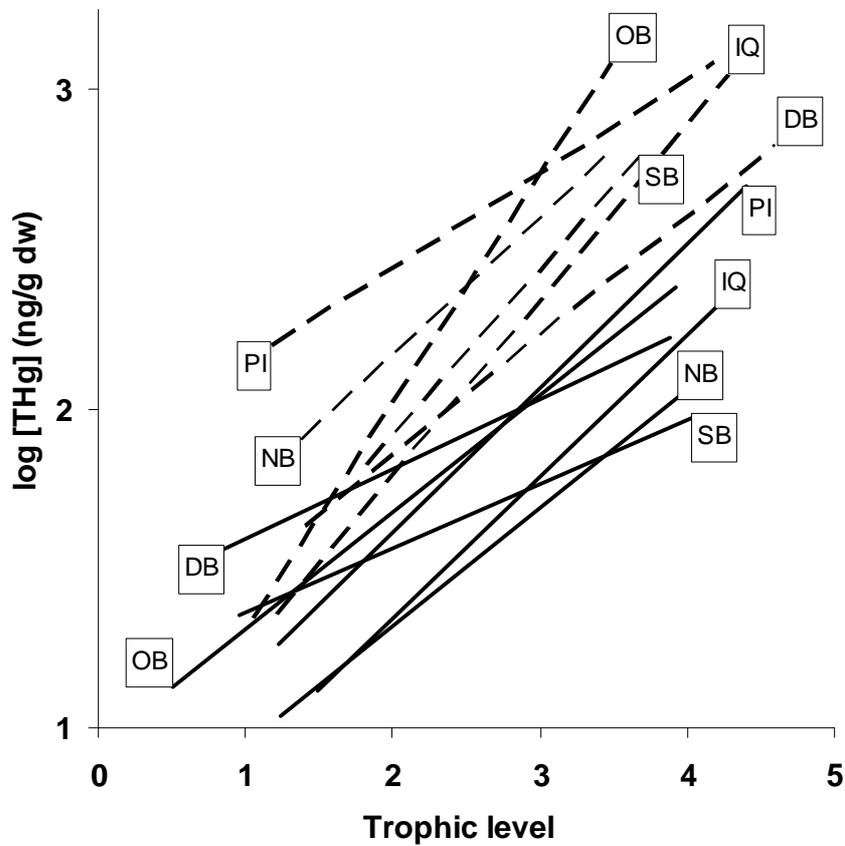
Figure 3.4. Trophic level versus log [THg] (ng/g dw) relationships for each of six lacustrine (dashed lines) and six marine (solid lines) foodwebs. Regression lines are identified using location abbreviations, located at the beginning or end of each line. To improve clarity of the display, individual data points are not plotted.
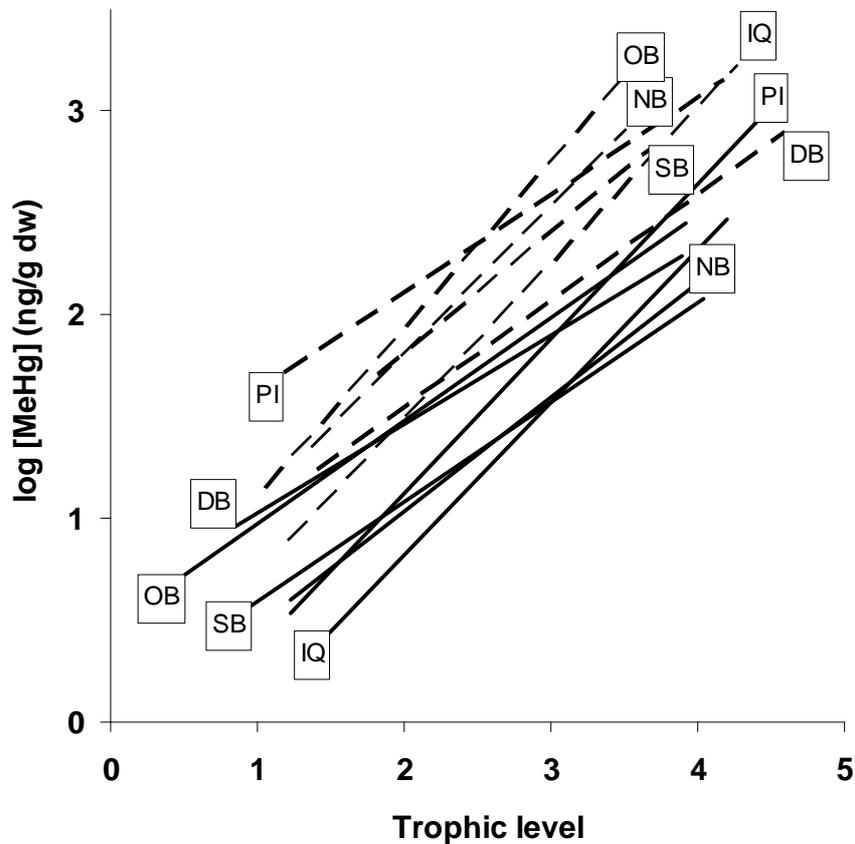
Figure 3.5. Trophic level versus log [MeHg] (ng/g dw) relationships for each of six lacustrine (dashed lines) and six marine (solid lines) foodwebs. Regression lines are identified using location abbreviations, located at the beginning or end of each line. To improve clarity of the display, individual data points are not plotted.
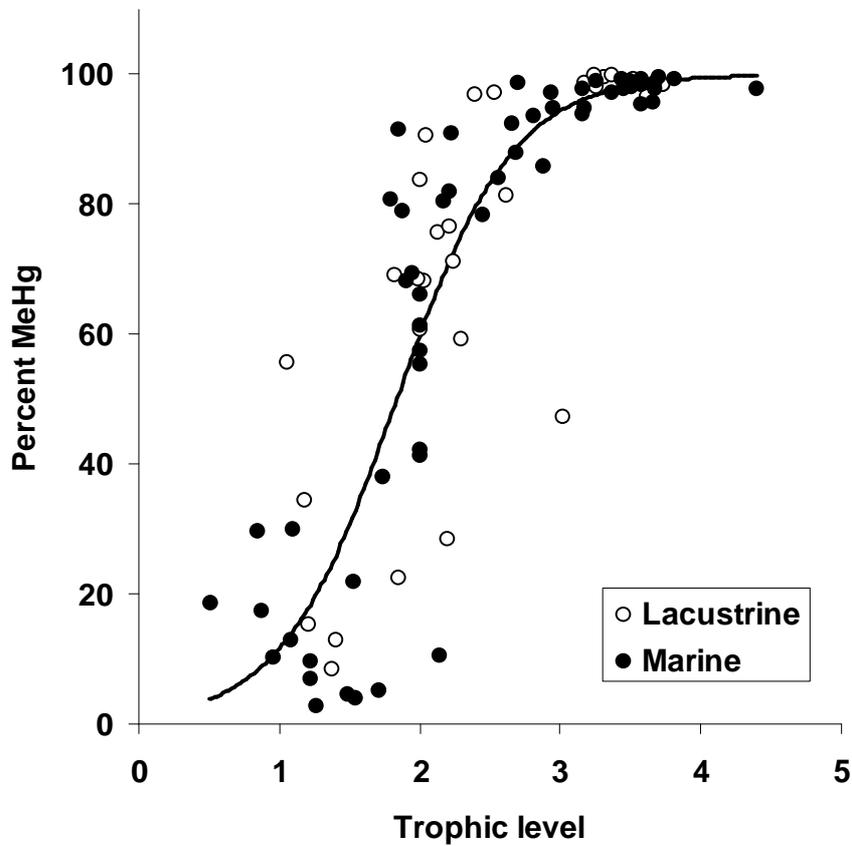
Figure 3.6. Trophic level versus percent MeHg for taxa from marine (solid circles) and lacustrine (open circles) foodwebs. The non-linear logistic model was estimated using data from both habitats.

Chapter 4

Conclusions and future research directions

That mercury in consumed fish poses a threat to human health has been known for over 50 years (McAlpine and Araki 1958), and mercury concentrations in fishes from the Canadian Arctic, and Arctic charr in particular, have been measured since the 1970s (Lockhart et al. 2005). Previous investigations of [THg] in Arctic charr have often been based on small sample sizes, and targeted large fish of potential interest to human consumers (e.g., Evans et al. 2005a; Evans and Muir 2010; Lockhart et al. 2005; Muir et al. 2009; Muir et al. 2005; Riget et al. 2000). Such studies provide valuable information concerning spatial and temporal variations in mercury concentrations, and are vital in predicting human health risks from fish consumption. Relatively less is known about the biological factors driving differences in [THg] among individual Arctic charr, although recent studies are beginning to fill that information gap (e.g., Gantner et al. 2010a,b, 2009; Swanson et al. 2011). Nevertheless, there remains a paucity of information explaining spatial patterns in, and mechanisms responsible for, differences in [THg] between anadromous and non-anadromous Arctic charr, which this M.Sc. project was designed to address.

This study represents the first wide-scale spatial comparison of mercury concentrations and associated biological variables (age, fork-length, and trophic position) between anadromous and non-anadromous Arctic charr. At each of nine sampling locations, anadromous Arctic charr had significantly lower mercury concentrations than spatially-proximate non-anadromous conspecifics, which is

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consistent with previous observations (Bruce and Spencer 1979; Lockhart et al. 2005; Riget et al. 2000; Swanson et al. 2011). However, anadromous Arctic charr also tended to be larger, and usually fed at a higher trophic level, than non-anadromous conspecifics, and both factors have been associated with higher [THg] in Arctic charr (e.g., Gantner et al. 2009). Thus, our results indicate that feeding habitat (lacustrine or marine) is more important than size or fork-length in determining fish mercury concentration. While the prevalence of anadromy in Arctic charr is related to latitude (Power et al. 2008), the difference in [THg] observed between the two life-history types was independent of latitude across 49 – 81° N in eastern Canada.

A key finding of the present study is that within-population [THg] is determined primarily by fish age in both anadromous and non-anadromous Arctic charr, a species characterized by a long life-span and highly variable length-at-age (Johnson 1980; Klemetsen et al. 2003). Individual trophic position was strongly related to fish [THg] in some populations, particularly where Arctic charr occupied more than one trophic level (e.g., due to cannibalism in large individuals from Crazy Lake). However, δ^{15}N-inferred trophic position did not consistently predict fish [THg], particularly in anadromous populations. Results presented herein suggest that individual fork-length is not directly related to fish mercury concentration. Rather, mercury concentrations were determined by age and, in some populations, trophic position, which both covary with fork-length. Similarly, differences in somatic growth rate may be correlated to differences in fish [THg], but the occurrence of a relationship does not imply causation, particularly where differences in somatic growth rate are associated with differences in diet (e.g., a switch to feeding in the marine environment).

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Thus, it is concluded that the accumulation of mercury in Arctic charr is driven primarily by age, but can be altered by a shift in diet (e.g., to piscivory), or habitat (e.g., anadromy). Accordingly, monitoring of Arctic charr for purposes of human health risk assessment should take into account the age distribution of consumed fish, particularly as climate change may significantly alter existing length-at-age relationships for many populations of Arctic charr. In addition, it is recommended that fish [THg] be corrected using fish age, rather than fork-length, when comparing concentrations across populations, particularly when examining long-lived species such as Arctic charr. Finally, in order to elucidate the mechanisms of mercury accumulation among individual fish, future studies should aim to obtain a reasonably large sample size including representatives from all size and age classes, morphotypes, and life-history variants (e.g., anadromous and non-anadromous) present in the population.

This study also marks the first wide-scale spatial investigation of mercury biomagnification in co-located pairs of lacustrine and marine foodwebs supporting Arctic charr. Biomagnification rates were quantified using slopes of δ^{15}N-inferred trophic level versus [THg] or [MeHg] in each of six spatially-paired foodwebs. While biomagnification rates varied among individual sampling sites, there was no consistent difference in the rate of total or methylmercury biomagnification between the marine and lacustrine feeding habitats. However, the mercury concentration at the base of the foodweb was higher in lacustrine than marine habitats, which may be driven by differences in water [THg], percent MeHg, or bioavailability of Hg to organisms at the base of the foodweb between the two habitat types. One novel finding of the present research is that the proportion of mercury in the methylated form increases with trophic level similarly in

lacustrine and marine foodwebs, suggesting that the relative trophic transfer efficiencies of total and methylmercury are independent of the physical, chemical, and biological differences that exist between the two habitat types.

While some general inferences can be made about [THg] in Arctic charr (e.g., anadromous individuals have lower [THg] than non-anadromous conspecifics), results presented herein suggest that it is not feasible to accurately predict fish mercury concentration among sampling sites using biological factors (e.g., age, fork-length, trophic level, life-history strategy) or wide-scale spatial information (e.g., site latitude). Rather, lake-specific characteristics influencing mercury input and cycling in the abiotic and biotic environment, and biological features such as species composition and food chain length, are critical factors for predicting differences in fish [THg] among lakes (Rose et al. 1999). The implication of this finding is that spatial extrapolation (i.e., site to site) will suffer from errors, and that more, not less, monitoring is needed.

While the results of this study offer insight into the spatial patterns and causal mechanisms driving the difference in [THg] between anadromous and non-anadromous Arctic charr, there are still many areas where additional research could be focused. Future research directions include:

1. An investigation of mercury concentrations comparing spatially-proximate landlocked, lake-resident, and anadromous Arctic charr. Swanson et al. (2011) found no difference in mercury concentration between anadromous and lake-resident fish, although both had a lower mean mercury concentration than landlocked Arctic charr. This contrasts with the results presented in Chapter 2, where anadromous fish had significantly lower [THg] than non-anadromous

conspecifics, regardless of whether the non-anadromous Arctic charr were landlocked, or were sympatric with anadromous fish. Future research would be warranted to resolve this discrepancy.

2. A study of mercury concentrations in an anadromous population of Arctic charr both before and after the onset of anadromy could be conducted. The goal here would be to investigate the reduction in tissue mercury concentration associated with the transition from feeding in the freshwater foodweb for the first several years of life, to feeding in the relatively mercury poor marine foodweb after the onset of anadromy.

3. Future studies should aim to characterize mercury concentrations and δ^{13}C values in benthic and pelagic organisms at the base of northern lacustrine and marine foodwebs, in order to compare basal mercury concentrations in the benthic versus pelagic food chains (as in Lavoie et al. 2010). In the present study, the effect of benthic connection (inferred using δ^{13}C) was investigated within, but not among, Arctic charr populations, due to a lack of benthic and pelagic baseline δ^{13}C in many of the studied foodwebs. Future research could investigate whether Arctic charr demonstrate a shift in benthic connection with anadromy (e.g., from primarily benthic feeding in the lacustrine environment to more pelagic foraging in the marine feeding habitat), that may be related to differences in [THg] (e.g., Kidd et al. 2003; Power et al. 2002).

4. Differences in Arctic charr [THg] among sample sites could be related to lake-specific chemical (e.g., pH, dissolved organic carbon, chloride concentration) and physical factors (e.g., lake size, catchment area, temperature), as well as THg

and MeHg concentrations in water. These relationships have been demonstrated in more southerly areas (e.g., Chen et al. 2005; Greenfield et al. 2001; Kamman et al. 2004; Marusczak et al. 2011; Rose et al. 1999; Schindler et al. 1995), but have been scarcely studied in Arctic lakes (although some lake-specific characteristics were examined in Evans et al. 2005b and Gantner et al. 2010a), and may help to explain differences in Arctic charr [THg] among lakes.

5. Laboratory studies are warranted in order to estimate physiological parameters such as the assimilation efficiency of Hg from water and ingested food, food consumption rates, and elimination rates of Hg (including loss to gonads during spawning) for Arctic charr reared at a range of tolerable temperatures. Such estimates have been produced for more southerly fish species (e.g., lake trout, *Salvelinus namaycush*, from Lake Ontario, Trudel and Rasmussen 2006; yellow perch, *Perca flavescens*, from the Ottawa River, Norstrom et al. 1976; walleye, *Sander vitreus*, from the Ottawa River, Trudel and Rasmussen 2001), and would allow bioenergetics modelling to predict changes in Arctic charr [THg] in response to changing parameters (e.g., prey [THg], somatic growth rate, temperature), as predicted by future climate warming scenarios.

6. Finally, sampling could be conducted from a wider geographic area to determine whether the trends seen here are consistent throughout the circumpolar range of Arctic charr (Johnson 1980). Mercury concentrations have been measured in Arctic charr from Greenland, Iceland, the Faroe Islands, northern Europe, and a single Russian site (AMAP 2005; Ford et al. 2005). However, studies that investigate the biological and life-history factors influencing Arctic charr [THg]

have predominantly used Canadian fish (e.g., Evans and Muir 2010; Gantner et al. 2010a,b, 2009; Muir et al. 2009; 2005; Swanson et al. 2011); an exception is Riget et al. (2000), where Arctic charr from Greenland were studied. The use of paired anadromous and non-anadromous Arctic charr from a wider geographic area would allow a more robust examination of spatial patterns in, and mechanisms influencing, differences in [THg] between the two life-history types.

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**Appendix 1**

Supporting figures
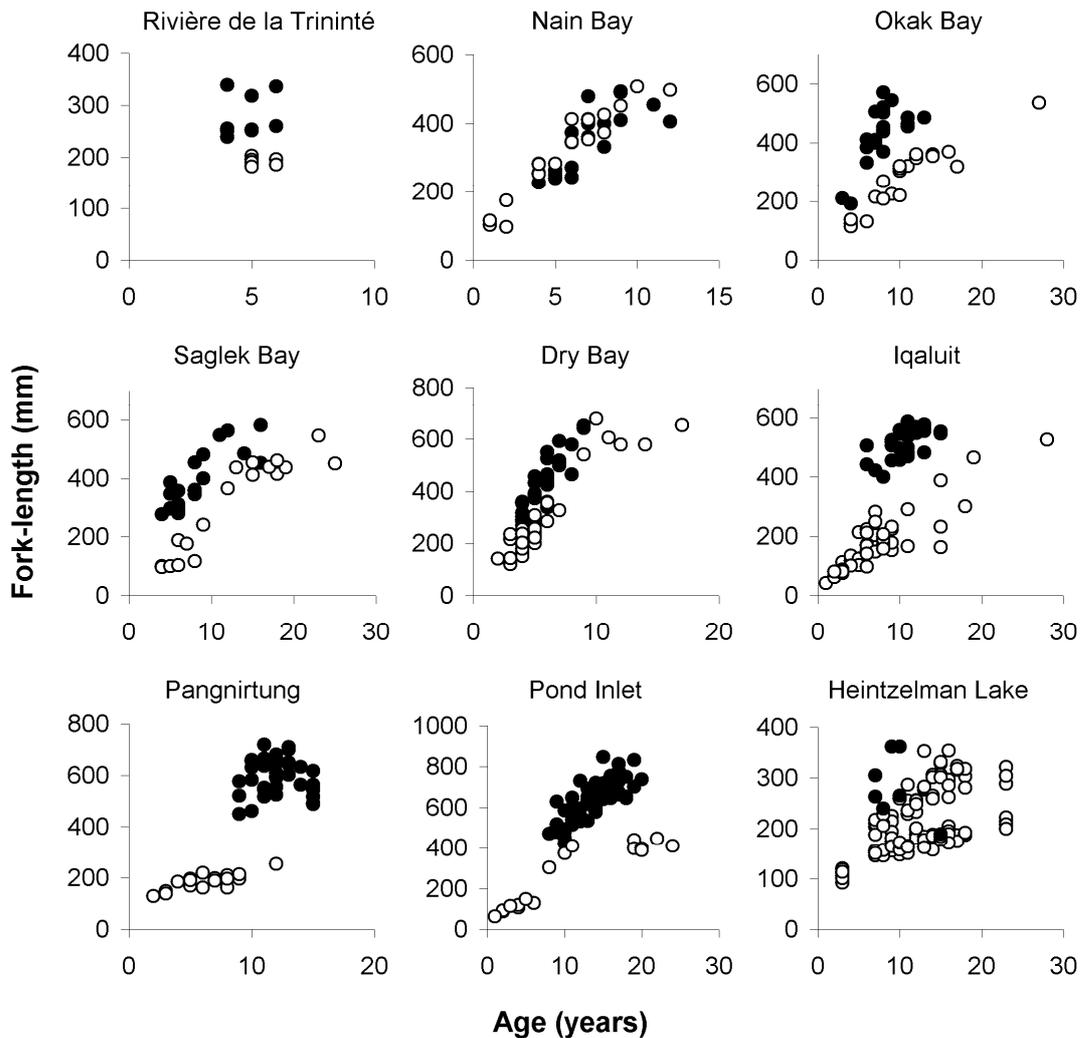
Figure S1. Age (years) versus fork-length (mm) for Arctic charr from 9 paired sampling sites. Sampling locations are ordered by increasing latitude (left to right then top to bottom). Data points represent individual fish from anadromous populations (closed circles) and non-anadromous populations (open circles).
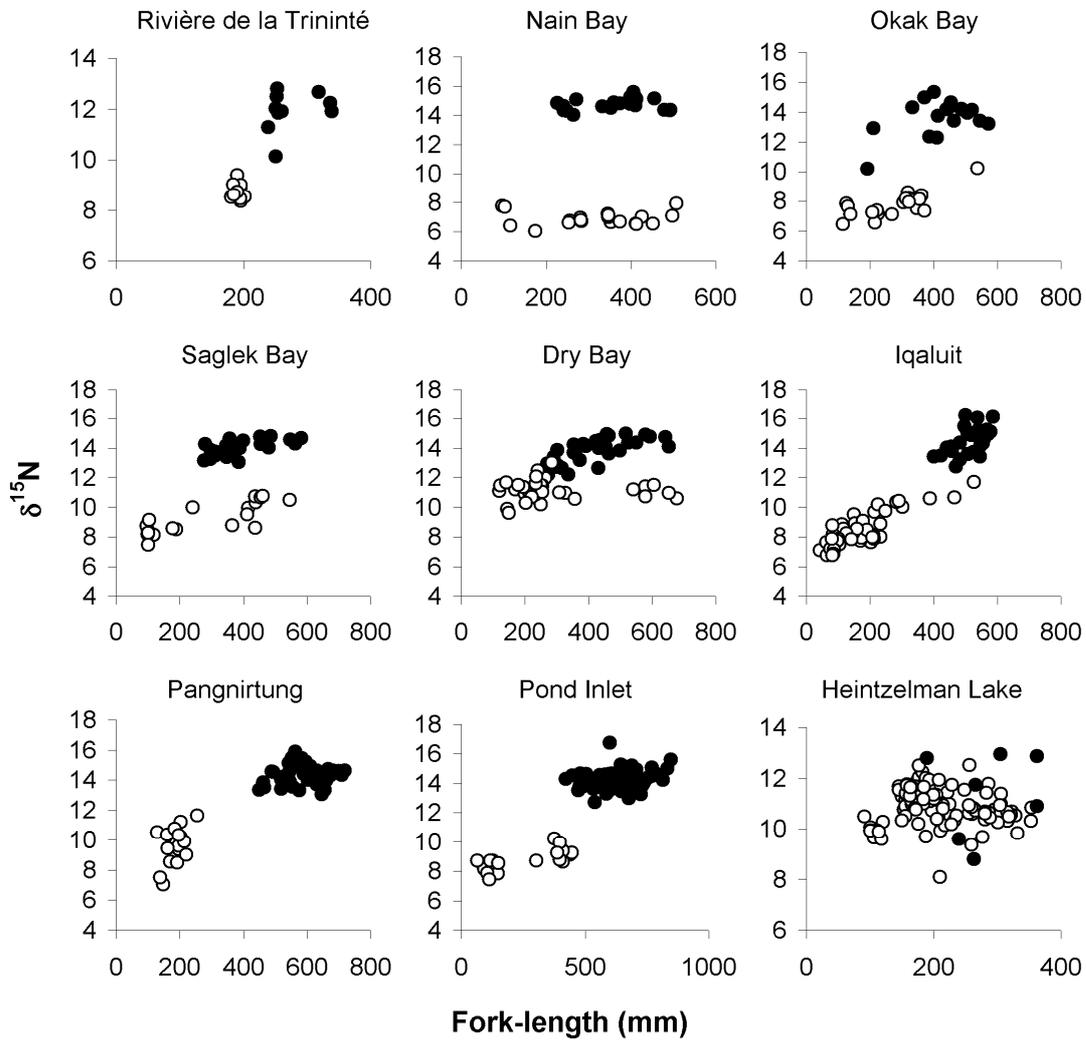
Figure S2. Fork-length (mm) versus $\delta^{15}N$ (‰) for Arctic charr from 9 paired sampling sites. Sampling locations are ordered by increasing latitude (left to right then top to bottom). Data points represent individual fish from anadromous populations (closed circles) and non-anadromous populations (open circles).
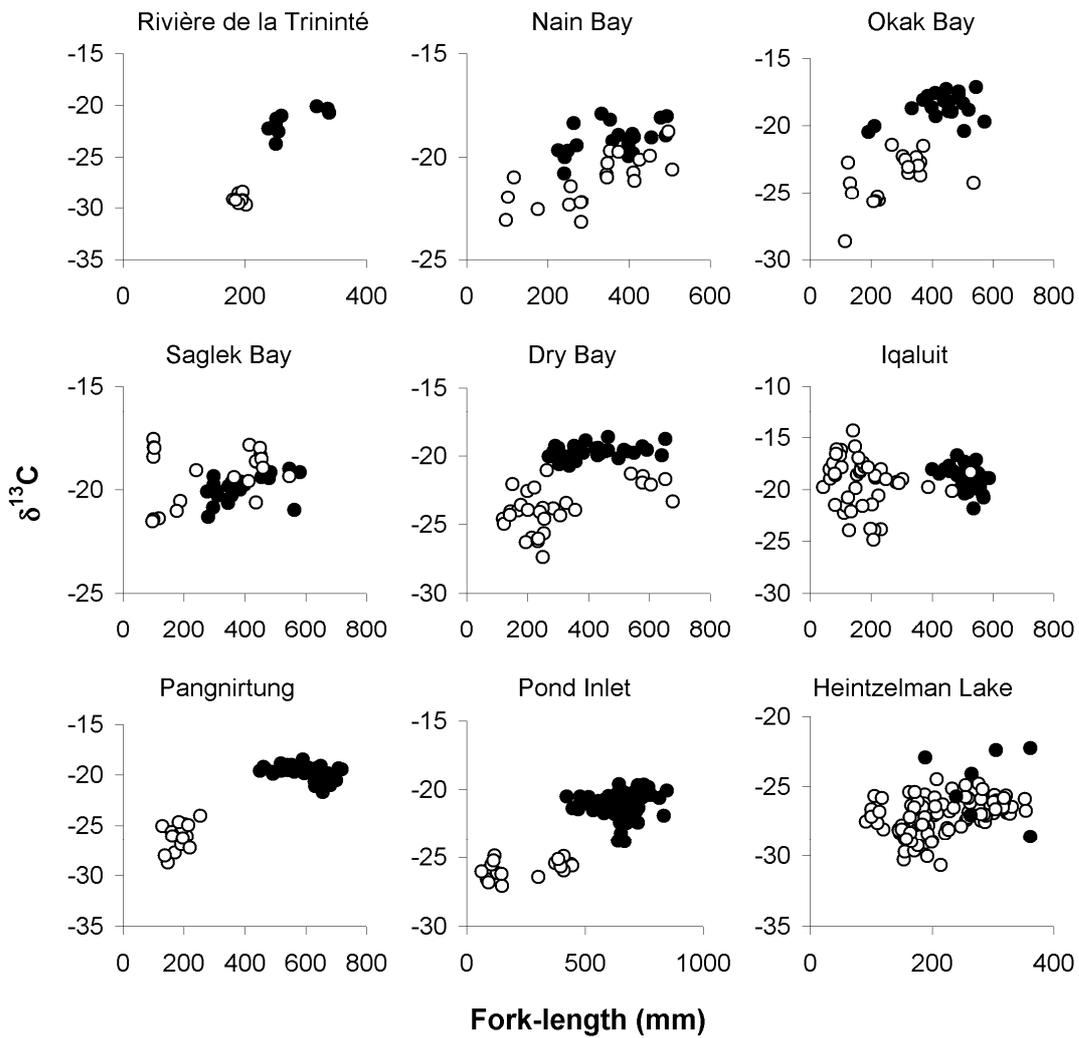
Figure S3. Fork-length (mm) versus $\delta^{13}C$ (‰) for Arctic charr from 9 paired sampling sites. Sampling locations are ordered by increasing latitude (left to right then top to bottom). Data points represent individual fish from anadromous populations (closed circles) and non-anadromous populations (open circles).

Figure S4. $\delta^{13}C$ (‰) versus ln [THg] (ng/g ww) for Arctic charr from 9 paired sampling sites. Sampling locations are ordered by increasing latitude (left to right then top to bottom). Data points represent individual fish from anadromous populations (closed circles) and non-anadromous populations (open circles).