Variations in Northern Dolly Varden (*Salvelinus malma malma*) total mercury concentrations from the northwestern Canadian Arctic

by

Lilian Tran

A thesis

presented to the University of Waterloo

in fulfilment of the

thesis requirement for the degree of

Master of Science

in

Biology

Waterloo, Ontario, Canada, 2014

Author's Declaration

I hereby declare that I am the sole author of this thesis. This is a true copy of the thesis, including any required final revisions, as accepted by my examiners.

I understand that my thesis may be made electronically available to the public.

Abstract

Mercury (Hg) is a ubiquitous contaminant with harmful impacts on the aquatic ecosystem as well as human health. Studies to date of the bioaccumulative effects of Hg on northern fishes have focussed on all but one of the key species consumed by humans. The exception is the Northern Dolly Varden (*Salvelinus malma malma*), prized as a food resource in the western Canadian Arctic. Northern Dolly Varden contaminant data were twinned with $\delta^{13}C$ and $\delta^{15}N$ stable isotope signature data as a means of relating observed variations in total mercury (THg) to feeding strategies. The objective of this thesis was to use archival tissue samples obtained from populations separated by space, time and life-history type to examine the spatial, temporal and life-history trends in Northern Dolly Varden THg concentrations as well as build a historical baseline against which future changes in contaminant loading may be assessed.

Historical THg spatial trends for anadromous Northern Dolly Varden were determined from 10 populations in the Yukon and Northwest Territories sampled across a range of latitudes (67-69° N) and longitudes (136-141° W) between the years 1988-1991. Unadjusted mean THg concentrations ranged from 15 to 254 ng/g wet weight (ww). Length-adjusted THg concentrations were significantly different among sites, but were not related to latitude or longitude. Within and among populations, THg was significantly related to fork-length, age, $\delta^{15}N$, and $\delta^{13}C$, with the variation in THg found among populations being best explained by size. The data serve as an important baseline against which future changes in THg levels in this important subsistence fishery may be compared to determine the significance of any observed trends.

To understand the importance of life-history type on THg, THg concentrations were measured in isolate, resident and anadromous Northern Dolly Varden from the Babbage River, Yukon Territory, Canada. Differences in mean THg concentrations were found starting at 22 ng/g ww in isolate, increasing to 56 ng/g ww in resident, and 108 ng/g ww in anadromous fish. The pattern is markedly different than that observed in other migratory charr species where anadromous fish typically have lower levels of THg. After adjusting THg to a standardized age and length, significant differences remained among the life-history types, with anadromous Northern Dolly Varden having the lowest THg concentrations. Trophic position was the most important factor in explaining observed differences among the individuals regardless of life-history types, with growth rate also contributing to explaining the variation among individuals. The contrast of higher absolute, but lower age and size adjusted THg levels in anadromous fish was explained by a combination of two counter-acting mechanisms, including: 1) a switch to feeding at higher trophic levels and the use of prey with higher THg concentrations in the marine environment that raises THg levels, and 2) somatic growth dilution that decreases THg as fish age and increase in size.

Temporal changes in Northern Dolly Varden THg concentrations were assessed between periods 1986-1988 to 2011-2013 from the Rat (67° 46’ 48”N and 136° 19’ 12”W) and the Firth (68° 40’ 12”N and 140° 55’ 12”W) rivers in the northwestern Canadian Arctic. In the Rat River, mean THg ranged from 79 ng/g ww in 1986-1988 to 109 ng/g ww in 2011-2013, while in the Firth River, THg ranged from 126 ng/g ww in 1986-1988 to 178 ng/g ww in 2011-2012. After factoring in size, $\delta^{13}C$ and $\delta^{15}N$, THg increased significantly over time. Relationships between log[THg] versus fork-length and log[THg] versus $\delta^{13}C$ have remained constant over time in the Rat River, but not in the Firth River, while relationships between log[THg] versus $\delta^{15}N$ have

remained constant in the Firth River, but not in the Rat River. Although increases in temperature coincide with increases in THg, the effects are not direct.

Acknowledgements

I would first like to thank my supervisor, Dr. Michael Power, for offering me a great project that has introduced me into fisheries biology. I am appreciative for the guidance, patience, and positive encouragement of Dr. Power and Dr. Jim Reist that they have provided over these past years, as well as for their help towards my future endeavors.

I also would like to thank the past and present members of the Power Lab for all their help and for creating such a positive work environment to come to everyday. I have been so fortunate to work with such a great group of people. Thanks for the de-stressing get-togethers and thank you Shannon van der Velden for introducing me to everything mercury and fish related and for your help with Doris, the DMA. A special thanks goes to Heidi Swanson for her help on statistics and for taking the time to meet with me one on one to go through my data. I must also thank Robert Fudge, Steven Sandstrom, Vic Gillman, Colin Gallagher, and Jim Johnson for their help in obtaining, managing and archiving the Northern Dolly Varden muscle tissue samples and acknowledge Jessica Ives, Nilo Sinnatamby, and Wendy Michaud for their statistical advice. I am also grateful for the advice and support from people I have met through conferences, particularly the members of DFO Winnipeg and my committee member, Dr. George Dixon. I also cannot forget my mom, dad and, Diana for their help, support, and understanding.

Lastly, I would like to thank Dr. Michael Power and Dr. Jim Reist for the incredible opportunities to go to the Arctic, experience the culture and to examine Northern Dolly Varden in person. Tracey Loewen and Dave Boguski, you were amazing field work buddies. Thank you for teaching me everything, taking care of me, and making my first field season a great one. The memorable experience would not have been complete without the help and generosity of Jordan

McLeod and Dennis Arey and their families as well as all the families at Shingle Point. Thank you for inviting me into your homes for food and tea and allowing me to experience the Inuvialuit culture that so few people ever get to see. It was an incredible experience and one that I will never forget.

Financial support for this project was provided by the Fisheries Joint Management Committee (FJMC), an NSERC ArcticNet and Discovery Grant to MP, University of Waterloo Graduate Scholarship, Graduate Research Studentship, Science Graduate Experience Award, and the Silverhill Graduate Research Grant.

Table of Contents

Author's Declaration ii

Abstract iii

Acknowledgements vi

List of Figures xi

List of Tables xii

Chapter 1: General Introduction 1

Mercury 2

Dolly Varden (*Salvelinus malma*) 3

Mercury in Dolly Varden 4

Stable Isotopes 5

Total mercury analyses 6

Study objectives 8

Chapter 2: Total mercury concentrations in anadromous Northern Dolly Varden from the northwestern Canadian Arctic: A historical baseline study 10

Introduction 11

Methods 13

Sample collection 13

Stable isotope analyses 14

Total mercury analyses 15

Methylmercury analyses 16

Statistical analyses 17

Results 19

Discussion 22

Chapter 3: Life-history dependent variation of total mercury concentrations in Northern Dolly Varden from the Babbage River, Yukon Territory, Canada 38

Introduction 39
Methods 42
Sample Collection 42
Stable isotope analyses 43
Total mercury analyses 44
Methylmercury analyses 45
Statistical analyses 45
Results 48
Discussion 50
Chapter 4: Comparison of contemporary and historical total mercury concentrations in Northern Dolly Varden from the northwestern Canadian Arctic 63
Introduction 64
Methods 66
Sample Collection 66
Stable isotope analyses 67
Total mercury analyses 67
Methylmercury analyses 68
Meteorological data 69
Statistical analyses 69
Results 70
Firth River 71
Rat River 72
Temperature 73
Discussion 73
Chapter 5: Conclusions and Future Directions 84
Summary 85

Study significance 86
Future research 88
References 90

List of Figures

Figure 1.1: Direct mercury analyzer program……9

Figure 2.1: Map of sampling sites……34

Figure 2.2: Log[THg] (ng/g ww) versus fork-length (mm) for each site……..35

Figure 2.3: Mean length-adjusted THg (ng/g ww)……36

Figure 2.4: Length-adjusted log[THg] (ng/g ww) versus age (years) for each site……...37

Figure 3.1: Map of sampling sites……59

Figure 3.2: Log[THg] (ng/g ww) versus life-history type……60

Figure 3.3: THg and stable isotope bi-plots for isolate, resident, and anadromous Northern Dolly Varden……61

Figure 3.4: Log[THg] (ng/g ww) versus $\delta^{15}N$ (‰), age (years) and growth rate (mm/year) for anadromous Northern Dolly Varden……..62

Figure 4.1: Map of sampling sites……..79

Figure 4.2: Log[THg] (ng/g ww) versus time period……80

Figure 4.3: Log[THg] (ng/g ww) versus fork-length (mm)……...81

Figure 4.4: Log[THg] (ng/g ww) versus $\delta^{13}C$ (‰) and $\delta^{15}N$ (‰)…….....82

Figure 4.5: Temperature (°C) versus year for Aklavik and Old Crow……..83

List of Tables

Table 2.1: Co-ordinates and sampling years of sites……...……..29

Table 2.2: Mean ± standard deviation and range of measured variables for each site…….….30

Table 2.3: Population-specific correlation coefficients…….31

Table 2.4: Population-specific partial correlation coefficients……..32

Table 2.5: Multiple regression model results explaining variation in unadjusted log[THg] (ng/g ww) as a function of biological and ecological variates……33

Table 3.1: Mean ± standard deviation and range of measured variables for each life-history type…….56

Table 3.2: Anadromous Northern Dolly Varden correlation coefficients…….57

Table 3.3: Anadromous Northern Dolly Varden partial correlation coefficients……..57

Table 3.4: Multiple regression model coefficients explaining variation in log[THg] (ng/g ww) as a function of biological and ecological variates……58

Table 4.1: Mean ± standard deviation and range of measured variables for each period in the Rat and Firth rivers……...77

Table 4.2: Two-sample t-test comparisons between periods for the Rat and Firth rivers…….78

Chapter 1: General Introduction

Mercury

Mercury (Hg) enters the atmosphere in its elemental form (Hg^0) from natural inputs, such as outgassing of the earth's crust, volcanic eruptions, and forest fires, and from anthropogenic sources, such as metal production, chlor-alkali plants, combustion of fossil fuels, and waste incineration (Schroeder and Munthe, 1998). From its elemental form, Hg^0 is eventually oxidized to form divalent inorganic Hg (Hg (II)) (Brosset, 1987; Schroeder and Munthe, 1998). Mercury has a residence time of 1-2 years and a low water solubility, which enables it to be transported over long distances to remote regions, such as the Arctic (Lindqvist and Rodhe, 1985; Schroeder and Munthe, 1998; Scott, 2001). In the Arctic, Hg (II) is deposited onto snow banks and enters lakes and rivers through snowmelt (Loseto et al., 2004), where it can then be methylated by anaerobic bacteria in the sediments to form toxic organic methylmercury (MeHg) (Jensen and Jernelov, 1969). Methylmercury is able to biomagnify up the food chain resulting in the measurably high levels of THg found in many top predators (Morel et al., 1998). Biomagnification can have harmful consequences for fish populations, causing decreased fertilization and reproductive success and altered behaviour in fish (Joensuu, 1971; Matta et al., 2001).

Methylmercury is obtained by fish through food sources and/or uptake across the gill surface (Health Canada, 2007). Once in the fish, Hg adheres to the muscle protein, where it can accumulate to levels greater than those of the surrounding water (Health Canada, 2007). Studies that have examined Hg levels in fish have found size effects (Barber et al., 1972; Jewett et al., 2003), age effects (Bache et al., 1971), both size and age effects (Scott and Armstrong, 1972; Jackson, 1991; Glass et al., 2000), as well as no age effects (Muir et al., 1999). Therefore, Hg

concentrations and the factors that control Hg concentrations can vary by species (Baker et al., 2009).

Consumption of fish is one of the main pathways of exposure for humans (Gupta et al., 2005). Once in the body, Hg can cause neurological impairment and have harmful effects on the nervous system, the kidneys and the cardiovascular system (Gupta et al., 2005). As a result, the maximum recommended limit for Hg allowed in commercially sold fish by the Canadian Food Inspection Agency is 0.5 ppm and 0.2 ppm for subsistence fish (Health Canada, 2008). The provisional tolerable daily intake for an adult is 0.4 µg MeHg per kg body weight for the protection against neurotoxicity effects (WHO, 2007), with lower levels recommended for pregnant women and children (WHO, 2004).

Dolly Varden (*Salvelinus malma*)

Dolly Varden is part of the charr species complex and consists of two distinct taxonomic forms, Northern Dolly Varden (*Salvelinus malma malma*) and Southern Dolly Varden (*Salvelinus malma lordi*) (Kowalchuk et al., 2010b). The northern form is distributed throughout western North America, ranging from Alaska to the Mackenzie River (Reist et al., 2002; Armstrong and Hermans, 2007) and has three life history types, anadromous, isolate, and resident (Kowalchuk et al., 2010a). The anadromous life history type is found in streams and rivers which drain towards the Beaufort Sea (Kowalchuk et al., 2010a). Annual anadromous migrations to the Beaufort Sea begin in late June when fish are between 3-5 years of age, with return over-wintering and spawning migrations occurring in late August (Gwich'in Renewable Resources Board, 2010). In contrast, both the isolate and resident life-history types spend their whole life in one river, are smaller in size and mature earlier than anadromous fish (Kowalchuk

et al., 2010a). Unlike the resident life history type, isolates are separated from the anadromous fish by migratory barriers (Kowalchuk et al., 2010a).

Mercury in Dolly Varden

Local native communities have fished Dolly Varden for subsistence purposes for generations, yet little is known about THg loadings in Dolly Varden or how THg concentrations in the fish may vary spatially or temporally by life-history type (Sandstrom et al., 2009). Many detailed studies have looked at THg contamination levels in other Arctic fish species in terms of spatial and temporal variability and the factors (physical and environmental) that influence concentrations in fish. The species studied include: Arctic charr (*Salvelinus alpinus*), northern pike (*Esox lucius*), broad whitefish (*Coregonus nasus*), walleye (*Stizostedion vitreum*), inconnu (*Stenodus leucichthys*) and lake trout (*Salvelinus namaycush*) (Stewart et al., 2003; Evans et al., 2005a; Lockhart et al., 2005; Muir et al., 2005). In contrast, Dolly Varden THg levels have only been studied incidentally, typically being reported as a single value in a list of studied fish (e.g., Deniseger et al., 1990; Lockhart et al., 2005). For example, Lockhart et al. (2005) studied spatial and temporal trends in THg in edible fish from northern Canada, but reported only a single value (1997, $n = 1$) for Dolly Varden from the Yukon Territory (0.004 μg/g). Although Deniseger et al. (1990) reported comparative values for Dolly Varden and changes in temporal accumulation trends, the data related specifically to mine contaminated sites in Vancouver Island, and therefore, are not pertinent for the Arctic.

An unpublished MSc thesis focusing on Alaskan Dolly Varden from the Aleutian Islands (Jeitner, 2009) noted THg concentrations did not correlate with fish size (length or weight) and that in a single study lake, smaller fish had higher mean THg concentration levels than larger

fish. As data were not accurately separated by life-history type, the direct influence of anadromy could not be accurately determined (Jeitner, 2009). There are no other studies of which I am aware, focusing on THg concentration levels in Dolly Varden from Canada, or elsewhere, and the effects of life-history, geography and time on observed variations in THg concentrations within and among populations of Dolly Varden.

Stable Isotopes

Species at the top of the food chain are known to have the highest Hg concentration levels, therefore, knowledge of prey and trophic level are essential to understanding patterns of THg accumulation in any species (Kidd et al., 1995). Analysis of gut contents as an approach to visualizing trophic interactions has been deemed unsuccessful in most instances (Kling et al., 1992). Gut contents may not be completely digested and/or absorbed and will only represent a short-term snapshot of the diet (Peterson and Fry, 1987; Kling et al., 1992). On the other hand, stable isotopes represent a temporally integrated view of diet based on tissue anabolism that has occurred over time and have been shown to be especially useful for inferring trophic relationships in slower growing Arctic fish (e.g., Hesslein et al., 1993). The stable isotope composition of nitrogen ($\delta^{15}N$) determines trophic level with the isotopically heavy nitrogen accumulating and increasing 3-5 ‰ at each trophic transfer (Peterson and Fry, 1987; Hesslein et al., 1993). Since THg concentrations in fish also often increase with increasing trophic level, or increasing $\delta^{15}N$, stable nitrogen isotopes may be used to trace Hg biomagnification through the aquatic food chain (Atwell et al., 1998). In contrast, carbon stable isotopes ($\delta^{13}C$) are best used to trace consumer carbon sources and the degree to which biota rely on pelagic or littoral primary production (Peterson and Fry, 1987; Kling et al., 1992; Hesslein et al., 1993; Power et al., 2002).

The reliability of $\delta^{13}C$ as a tracer of carbon sourcing relies on the fact that $\delta^{13}C$ composition barely changes with trophic transfer (Post, 2002).

The δ notation represents the stable isotope or isotopic composition of an element in permil units or parts per thousand (‰), where the isotope composition is the ratio of the heavy isotope to the light isotope of a particular element in a sample (Fry, 2006). For instance, the isotope composition of carbon ($\delta^{13}C$) measures the ^{13}C:^{12}C ratio (Kling et al., 1992), whereas the isotopic composition of nitrogen ($\delta^{15}N$) measures the ^{15}N:^{14}N ratio (Rolff et al., 1993). The δ value looks at the difference in the isotope composition of a sample relative to that of the standard reference material (Fry, 2006) and is expressed as:

$$\delta X\ (‰) = [(R_{sample}/R_{standard})\text{-}1] \times 1000$$

where X is the element and R is the isotope ratio of the sample or the standard (Peterson and Fry, 1987; Guiguer et al., 2002). An increase in the δ value indicates an increase in the amount of the heavy isotope, whereas a decrease indicates an increase in the amount of the light isotope (Peterson and Fry, 1987). All resulting measurements are expressed using standard delta notation as parts per thousand differences (‰) with respect to the international reference standards, carbonate rock from the Peedee Belemnite formation for $\delta^{13}C$ (Craig, 1957) and nitrogen gas in the atmosphere for $\delta^{15}N$ (Mariotti, 1983).

Total mercury analyses

Total mercury concentrations for a sub-sample (0.1-0.2g) of frozen, non-homogenized Northern Dolly Varden tissue were measured on a Milestone Direct Mercury Analyzer (DMA-80) using thermal decomposition followed by atomic absorption spectroscopy following U.S.

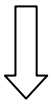
EPA method 7473 (U.S. Environmental Protection Agency, 2007). The DMA-80 is an automated machine and was run with a program set to run both the standard reference materials (SRMs) and samples as described in Figure 1.1. The program as described follows the method as used by Environment Canada's National Laboratory for Environmental Testing (NLET). The analytical protocol followed on a daily basis prior to commencing sample analyses consisted of running two blanks followed by 0.05-0.10g of each SRM, beginning with the SRM with the lowest certified value and ending with the SRM with the highest certified value. If the reported THg concentrations were in range for each of the SRMs, then two more blanks were run and Northern Dolly Varden tissue analyses were begun. To finish, two blanks, followed by the SRMs and two additional blanks were run, with SRM values checked against certified values to ensure continued optimal performance of the machine. When tested SRM values did not fall within certified range, the DMA was recalibrated following manufacturer specifications. In all analyses, a single sample was run in triplicate in each sample batch.

The DMA dries and decomposes the sample using temperatures as high as 650°C to release Hg from the sample. The released product is carried by flowing oxygen to the amalgamator which captures only Hg. The amalgamator is heated, releasing Hg vapour, which is carried by the flow of oxygen through absorbance cells located in front of a low pressure mercury lamp. The concentration of Hg is then obtained by measuring the UV absorption at a wavelength of 253.65 nm (Milestone, 2010) with results expressed as μg/kg wet weight.

Study objectives

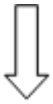
In view of the importance of Dolly Varden as a source of dietary protein (Daviglus et al., 2002), the increasing concerns for changes in THg accumulation rates in aquatic biota as a result of predicted climate change (Macdonald et al., 2005) and the fact that so little information exists on THg in Dolly Varden, the research described below aimed to improve the existing data and understanding of Hg levels in Dolly Varden. To accomplish the objective, existing archival tissue samples of Northern Dolly Varden were analyzed to determine THg concentration and stable isotope composition and the obtained data were used in spatial, life-history and temporal studies to better understand the factors that controlled THg in Northern Dolly Varden and describe the patterns of differences among populations. In the spatial study the objectives were to determine: [i] among-population level differences in THg; [ii] if significant latitudinal or longitudinal trends in THg existed; [iii] if THg levels within- and among-populations were positively correlated with the size and age of tested fish; and, [iv] if measured differences in THg levels within and among populations were correlated with corresponding stable isotope measures. In the life-history study the objectives were to determine if: [i] life-history types evidenced significantly different patterns of THg tissue concentrations; [ii] THg differences within and among life-history types were related to differences in feeding and trophic position; [iii] THg levels increase with size in all life-history types; and, [iv] THg and growth rate were correlated. And finally, in the temporal study the objectives were to determine for Northern Dolly Varden if: [i] significant temporal trends in THg existed; [ii] THg relationships that correlate with size, $\delta^{13}C$ and $\delta^{15}N$ have remained consistent through time; and, [iii] whether there was an association between changes in THg and mean annual temperature.

Drying Period (160°C)
1 min

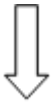


Decomposition Period 1 (650°C)
2 min

Decomposition Period 2 (650°C)
1 min



Purge Time
(time between the drying and decomposition periods are complete and the Hg measurement is about to begin)
240 sec



Amalgamation Time
(time required for the amalgamator to completely release the Hg collected in the absorption cell)
12 sec

Recording Time
30 sec

Total time: 8.7 min

Figure 1.1: Analytical program steps used by the DMA-80 to determine the THg concentrations in analyzed samples

Chapter 2: Total mercury concentrations in anadromous Northern Dolly Varden from the northwestern Canadian Arctic: A historical baseline study

This is an author-produced version of an article accepted for publication in Science of the Total Environment. The definitive publisher-authenticated version is as follows:

Tran, L., Reist, J.D. and Power, M. 2014. Total mercury concentrations in anadromous Northern Dolly Varden from the northwestern Canadian Arctic: A historical baseline study. *Science of the Total Environment.* Advance online publication. doi: 10.1016/j.scitotenv.2014.04.099

Introduction

Total mercury pollution from natural and anthropogenic sources has caused concern for the health of Arctic aquatic ecosystems (Braune et al., 1999), with long range atmospheric transport being the main pathway of exposure (Schroeder et al., 1998). After entering the aquatic ecosystems, mercury is methylated, resulting in toxic methylmercury (MeHg), which bioaccumulates and biomagnifies up the food chain (Booth and Zeller, 2005). Thus, fish consumption is a major exposure route of Hg for humans (Gupta et al., 2005). In the western Canadian Arctic, contamination levels in Dolly Varden (*Salvelinus malma*), a riverine charr, are of particular interest as this species is fished for subsistence purposes by the Inuvialuit and Gwich'in communities (Gallagher et al., 2011).

Dolly Varden occur in two distinct forms, southern and northern (Reist et al., 2002), with the northern form being distributed north of the Alaska Peninsula in the Bering, Chukchi, and Beaufort sea drainages east to the Mackenzie River (Reist et al., 2002). While there are many detailed THg studies of other subsistence fish species from the Canadian Arctic, THg studies of Dolly Varden are limited. For example, other subsistence fish species for which detailed THg studies have been completed include: Arctic charr (*Salvelinus alpinus*) (Muir et al., 2005; Gantner et al., 2010a,b; Gantner et al., 2012; van der Velden et al., 2013a,c), northern pike (*Esox lucius*) (Evans et al., 2005a), broad whitefish (*Coregonus nasus*) (Snowshoe and Stephenson, 2000; Evans and Lockhart, 2001; Evans et al., 2005a), walleye (*Sander vitreus*) (Evans and Lockhart, 2001; Evans et al., 2005a), burbot (*Lota lota*) (Snowshoe and Stephenson, 2000; Carrie et al., 2010), lake whitefish (*Coregonus clupeaformis*) (Bodaly et al., 1984), brook charr (*Salvelinus fontinalis*) (Braune et al., 1999), longnose sucker (*Catostomus catostomus*) (Lockhart

et al., 2005), Arctic grayling (*Thymallus arcticus*) (Lockhart et al., 2005), inconnu (*Stenodus leucichthys*) (Snowshoe and Stephenson, 2000; Lockhart et al., 2005), cisco (*Coregonus artedi*) (Lockhart et al., 2005), and lake charr (*Salvelinus namaycush*) (Power et al., 2002; Evans et al., 2005a). In contrast, Dolly Varden THg concentrations have been studied only incidentally, typically being reported as a single value in a list of studied fish (e.g., Deniseger et al., 1990; Lockhart et al., 2005).

Given the paucity of published mercury data on Dolly Varden, a key aim of this study was to create a spatially explicit baseline for the species to address the lack of information on mercury concentrations in Northern Dolly Varden and to facilitate establishing the significance of future rates of change in THg concentrations in Northern Dolly Varden that may be associated with industrial development and/or larger scale ecological changes (e.g., climate change). Using archival muscle tissue samples, the study further sought to explicitly test the following hypotheses concerning THg levels in Northern Dolly Varden: [i] as has been noted for other species of northern fish (Muir et al., 2005; Gantner et al., 2010a), there will be significant among-population level differences in THg; but, [ii] as has also been noted for other studied northern fish species (Gantner et al., 2010a), there will be no significant latitudinal or longitudinal trends in the spatial differences; [iii] measured THg levels within- and among-populations will be positively correlated with the size and age of tested fish (Bache et al., 1971; Grieb et al., 1990; Jewett et al., 2003); and [iv] measured differences in THg levels within- and among-populations will be correlated with carbon sources and trophic level, with THg being, respectively, negatively and positively correlated with $\delta^{13}C$ and $\delta^{15}N$ stable isotope measures (Power et al., 2002; Gantner et al., 2010a,b).

Methods

Sample collection

Dorsal muscle tissue samples of anadromous Dolly Varden were obtained from an archival collection maintained by the Department of Fisheries and Oceans Canada (DFO) in Winnipeg, Manitoba (Table 2.1) from sample sites covering roughly half the natural geographical range of the northern form of Dolly Varden within Canada. Approximately 5 g of muscle tissue was dissected with a clean scalpel from a standard area of each fish located posterior to the dorsal fin and above the lateral line (van der Velden et al., 2013c). Obtained tissue samples were subsequently split for use in THg and stable isotope analyses. Dolly Varden exhibit variant life-history types in this area: anadromous males and females and residual (non-migratory) males in rivers with access to the sea; and isolated populations upstream of impassable falls (Morrow, 1980). This study focused only on the anadromous life-history type.

All fish were sampled in the period July to October from rivers located throughout the northern Yukon and Northwest Territories in 1988, 1989, and 1991 (Fig. 2.1) and were classified as anadromous fish on the basis of capture point (e.g., in the marine environment) or size, morphology and coloration (Reist, personal communication) (Table 2.1). The whole otolith method was used to determine fish age (Sandstrom et al., 1997). The outer surface of the otolith was hand ground until the core/nucleus was visible. The otolith was then analyzed in a petri dish under a stereoscopic microscope with water or a clearing agent (e.g., oil of wintergreen) to elucidate the annuli. Sample sites were chosen because of the availability of a statistically sufficient sample size (e.g., Zar, 2010) and from as limited a range of years as possible so as to remove potential confounding temporal effects. Finally, samples were chosen so as to ensure that

appropriate biological data (e.g., fork-length (mm), weight (g), age (years), gonad weight (g), maturity, and sex) for statistical testing were available.

Stable isotope analyses

Portions of all muscle samples were used for the analysis of stable isotopes of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) following methods described in van der Velden et al. (2013c). Briefly, the samples were dried at 50 °C for 48 h and homogenated with a Retsch MM 301 ball mill grinder (F. Kurt Retsch GmbH Co., Haan, Germany) and weighed on a Mettler Ultra micro balance (UMX2, Mettler-Toledo GmbH, Greifensee, Switzerland). A range of 0.275–0.300mg of the homogenate was used for stable isotope analysis (SIA) completed with 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) at the Environmental Isotope Laboratory, University of Waterloo (Waterloo, Ontario). Machine analytical precision, respectively, for $\delta^{13}C$ and $\delta^{15}N$ was ±0.1‰ and ±0.2‰, and was determined by repeat analysis (duplicates run every $n = 11$). All resulting measurements were expressed using standard delta notation as parts per thousand differences (‰) with respect to the international reference standards, Vienna Peedee Belemnite for $\delta^{13}C$ (Coplen, 1994) and nitrogen gas in the atmosphere for $\delta^{15}N$ (Mariotti, 1983).

The comparison of organism stable isotope signatures from differing environments requires accounting for differences in the isotopic composition of primary producers from the food web within which organisms feed (Fry, 2006; Casey and Post, 2011). Selecting an appropriate baseline for marine feeding Dolly Varden is particularly challenging given variation in the factors which significantly influence baseline signatures in marine environments (e.g.,

temperature, depth and salinity (Casey and Post, 2011)), and the apparent movement of Dolly Varden among marine environments with differing physico-chemical characteristics (e.g., salinity, temperature, etc.). Dolly Varden enter the marine environment in spring (May–June) and move westward along the Beaufort coast. Within the marine environment, little is known about the nature and range of their movement, but they are assumed to feed in the highly productive nearshore zone (Sandstrom et al., 2009). Nevertheless, studies have reported anadromous Dolly Varden moving over extremely large distances and outside of the coastal zone (DeCicco, 1992, 1997). Furthermore, recent reports have suggested that Dolly Varden occur further offshore than previously thought (Cobb et al., 2008), a fact that would concur with the reported low catches of Dolly Varden in fish monitoring programs completed along the Beaufort coast (e.g., Jarvela and Thorsteinson, 1999). Given the complexity of possible Dolly Varden coastal and offshore marine movements, the lack of information about mean movement patterns for each of the studied populations, the known spatial variation in coastal baseline signatures caused by variations in depth, salinity, water temperature and water mass mixing (e.g., Jennings and Warr, 2003) and the implications for inferential error associated with incorrect baseline adjustment, no direct attempt was made to correct for among-population baseline differences. As Schell et al. (1998) have pointed out specifically for the Beaufort Sea, the assessment of trophic levels occupied by top predators (e.g., Dolly Varden) in the coastal zone must be done cautiously, even with accurate knowledge of the variation in primary producer isotope ratios in potential feeding habitats.

Total mercury analyses

Total mercury concentrations for a sub-sample (0.1–0.2 g) of non-homogenized muscle tissue were measured on a Milestone Direct Mercury Analyzer, DMA-80 (Milestone S.r.l., Sorisole, Italy) using thermal decomposition followed by atomic absorption spectroscopy following U.S. EPA method 7473 (U.S. Environmental Protection Agency, 2007), with results expressed as ng/g wet weight (ww). Standard reference materials (SRMs) were run at the beginning and end of every batch of 20 samples, with no less than 5 blanks run in each sample batch. The method detection limit determined as 3× the standard deviation of the blanks was 0.07 ng Hg (approximately 0.5 ng/g ww). The certified reference materials and percent recoveries (mean percentage of certified value ± 1 standard deviation) that were used and determined in this study include National Institute of Standards and Technology (NIST, Standard Reference Materials Program, Gaithersburg, MD, USA) 1566B, oyster tissue (95.3 ± 1.1%), NIST 2976, mussel tissue (103.1 ± 3.9%), NIST 2974a, freeze dried mussel tissue (100.9 ± 0.5%), and National Research Council of Canada (NRC) DORM-3, fish protein (110.0 ± 9.3%). In all analyses, a single sample was run in triplicate in each sample batch and the mean relative standard deviation of the triplicates was measured as 7.1%.

Methylmercury analyses

Methylmercury is the dominant form of mercury in organisms such as fish (Lindqvist and Rodhe, 1985). To determine whether the measurement of THg was an appropriate estimate of MeHg concentrations, random sub-samples (n = 20) were measured for MeHg concentrations. Muscle tissue samples were freeze dried for 48 h and then ground using an acid washed glass mortar and pestle and placed into acid washed vials. Samples were weighed (±0.1 mg) before and after lyophilization and the percent moisture was computed for each sample to facilitate

conversion of wet weight (ww) and dry weight (dw) concentrations of THg and MeHg. Concentrations of MeHg and inorganic Hg (II) were determined at Quicksilver Scientific (Lafayette, CO, USA) using method QS-LC-CVAF-001, matrix spikes and certified reference materials as quality control measures. The limit of detection (LOD) and limit of quantification (LOQ) were respectively, 2.0 ng/g and 5.0 ng/g for both MeHg and Hg (II) in analyzed tissue samples.

Statistical analyses

Given literature studies indicating the significance of fish size as an explanation for among-individual and among-population variation in THg (e.g., Muir et al., 2005; Gantner et al., 2010a, and van der Velden et al., 2013c), relationships between length and within- and among-site variation in THg were estimated for the Dolly Varden data using linear regression (e.g., van der Velden et al., 2013c). Length was found to account for a significant portion of the among-site variation in logTHg (65%) and was significantly positively correlated with logTHg (Fig. 2.2) for seven of the ten study sites (r^2 = 0.17 to 0.70). Accordingly, Dolly Varden mercury data were length-standardized following methods described in Fleming and Gross (1990):

(1) $$M_i = MO_i * (L/LO_i)^b$$

where M_i is the adjusted logTHg concentration for the i^{th} fish, MO_i is the observed logTHg concentration for the i^{th} fish, L is the mean length from all populations (427 mm), LO_i is the observed length of the i^{th} fish, and b is the common within-group slope of logTHg on log fork-length (Reist, 1986; Quinn et al., 1998). A test for homogeneity of slopes showed that the slopes

were significantly different ($p < 0.001$) and, therefore, the common slope model could not be used. As a result, population specific slopes were used to represent b. An analysis of variance (ANOVA) followed by Tukey's honestly significant difference post hoc test was then performed using length-adjusted logTHg to determine whether THg concentrations varied among populations.

All statistical analyses were performed using the statistical program SPSS (SPSS Inc., 2008) with Type I error set to $\alpha = 0.05$. Logarithmic transformation ($\log_{10}$) was completed on the THg concentration data prior to inclusion in statistical analyses to satisfy assumptions of normality and homogeneity of variance, with data being tested for normality using skewness and kurtosis tests and transformed where coefficients were >2 (Gantner et al., 2009). Relationships between log-transformed THg data and associated biological factors (e.g., length, age) were assessed using Pearson's correlation coefficients and simple linear regression (Zar, 2010). Partial correlation analysis was used to evaluate the significance of a single factor when other factors were held constant (e.g., Zar, 2010).

Data were also used to estimate multiple regression models of the form:

$$Y_i = a_0 + a_1X_{1i} + a_2X_{2i} + \ldots. + a_kX_{ki} + \varepsilon_i \quad (2)$$

where Y_i was the $\delta^{15}N$ or log-transformed THg value for the i^{th} fish, X_1 to X_k are the set of biological variates used to explain Yi and εi is a normally distributed error term with zero mean and unit variance (Draper and Smith, 1981). Variables selected for inclusion in models were chosen on the basis of forward selection stepwise regression (Draper and Smith, 1981) from among the set of biologically relevant variables reported in the literature as being correlated with $\delta^{15}N$, e.g., length, weight, age, $\delta^{13}C$ (Power et al., 2002; Jennings et al., 2008; Swanson et al., 2011; van der Velden et al., 2012) or logTHg, e.g., length, age, $\delta^{15}N$ and $\delta^{13}C$ (Rigét et al., 2000;

Power et al., 2002; Gantner et al., 2009, 2010a,b; van der Velden et al., 2012). Variable selection Selections>F-to-enter and F-to-remove criteria used were the conservative upper $\alpha = 0.05$ points of the F-distribution as recommended by Draper and Smith (1981). Sensitivity tests on variable selection were conducted by raising and lowering the F-to-enter criterion to the upper $\alpha = 0.01$ and 0.10 points, respectively, of the F-distribution and reversing the direction of selection (forward to backward). The AIC (Akaike information criterion) was also computed for the selected model and compared to AIC values computed for the set of all possible models that could be estimated from the set of considered biological variables.

As the magnitude of regression coefficients depends on the variates included in the regression and the units of measure (Dunn and Clark, 1987), standardized regression coefficients were calculated following Cox (1987) to judge the relative influence of each independent variable on $logTHg_i$. Standardized regression coefficients (β_j) measure the amount by which $logTHg_i$ changes in terms of units of its own standard deviation for a unit change in the standard deviation of the j^{th} independent variable, when all other independent variable values are held constant. The B_j coefficients, therefore, allow direct comparisons to be made between variables in terms of their relative importance for explaining variation in $logTHg_i$ and are useful for establishing which model variables have the greatest influence on measured concentrations of THg in Dolly Varden. Subsequent to estimation, all regression models were assessed for statistical adequacy by testing residuals for conformance to the underlying assumptions of linear estimation methods using standardized procedures (Dunn and Clark, 1987).

Results

A total of $n = 246$ samples from 10 locations were obtained from the DFO tissue archives for analysis. Individual muscle mercury concentrations ranged from 15 to 254 ng/g ww, with the average percentage of MeHg equaling 95 ± 3.4% (mean ± standard deviation) and ranging from 87 to 98% in the $n = 20$ samples assessed.

Mean values of THg in Northern Dolly Varden differed by a factor of 5.5, according to sample location, with concentrations ranging from a low of 29 ± 7 ng/g ww in Ptarmigan Bay to 161 ± 37 ng/g ww in the Firth River (Table 2.2). Fish sizes also varied by sample location, with the smallest (272 ± 40 mm) being found in Ptarmigan Bay and the largest (588 ± 67 mm) being found in the Firth River. When length-adjusted, the mean THg concentrations also demonstrated among-site variability, varying by a factor of just under 4. Among-site differences in mean age ($F_{(7,138)} = 17.893$, $p < 0.001$), weight ($F_{(9,233)} = 31.007$, $p < 0.001$), $\delta^{13}C$ ($F_{(9,236)} = 11.90$, $p < 0.001$), and non-baseline corrected $\delta^{15}N$ ($F_{(9,236)} = 19.70$, $p < 0.001$) were also evident as shown in Table 2.2. Variation in $\delta^{15}N$ was best explained by a multivariate regression model estimated by forward stepwise regression that did not include site (population), but did include length, $\delta^{13}C$, year of capture and age ($r^2 = 0.689$, all coefficients $p \leq 0.005$), thereby obviating the need for specific baseline adjustment of $\delta^{15}N$ by site.

Length-adjusted THg concentrations differed significantly among study sites ($F_{(9,232)} = 22.2$, $p < 0.001$),with the Ptarmigan Bay fish having the lowest concentrations (34.8± 8.5 ng/g ww) and the Firth River fish having the highest concentrations (128.2 ± 23.3 ng/g ww). Site means were not all significantly different from one another, with the ten study sites being grouped into six groups (Fig. 2.3) within which length-adjusted mean site THg concentrations did not differ significantly (Tukey's HSD; $p > 0.061$).

While significant differences in mean length-adjusted THg were evident among populations, no large-scale spatial patterns were evident in the data. Mean length-adjusted THg did not vary significantly with latitude (regression; $F_{(1,8)} = 3.8$, $p = 0.089$) or longitude (regression; $F_{(1,8)} = 0.5$, $p = 0.485$) across the range of latitudes (67°47′N to 67°35′N) and longitudes (136°10′W to 140°55′W) for which data were available.

The within-population relationships with fork-length were not significant in any of the 10 sample sites (Table 2.3), as a result of length standardization of the data. Within-population relationships with age were significant and positive in three of eight populations (Table 2.3; Fig. 2.4). When individual data were pooled across sampling locations, there was a significant positive relationship between non-length-adjusted THg and fork-length ($F_{(1,240)} = 437.0$, $p < 0.001$, $r^2 = 0.646$) and a significant positive but weak relationship between length-adjusted THg and age ($F_{(1,143)} = 15.2$, $p < 0.001$, $r^2 = 0.096$).

Within- and among-sampled populations, length-adjusted THg was related to sampled $\delta^{13}C$ and $\delta^{15}N$. Within-population relationships with $\delta^{13}C$ were significant and positive in two of 10 populations and negative in another population (Table 2.3). The within-population relationships with $\delta^{15}N$ were significant and positive in two of 10 populations (Table 2.3). When individual data were pooled across sampling locations, there were weak explanatory but significant positive relationships between length-adjusted THg and $\delta^{13}C$ ($F_{(1,240)} = 8.6$, $p = 0.004$, $r^2 = 0.034$) and $\delta^{15}N$ ($F_{(1,240)} = 52.2$, $p < 0.001$, $r^2 = 0.179$).

Partial correlation analysis of population-specific data indicated limited significant correlation with either fork-length or age when the other variable was controlled for, with three of eight populations showing significant correlation with either fork-length or age (Table 2.4). All partial correlations were positive and only the Pauline Cove data were correlated with both

fork-length and age when the other variable was controlled. When $\delta^{15}N$ was controlled for in addition to fork-length or age, only a single population was significantly correlated with either fork-length (Cache Creek) or age (Canoe River).

A multiple linear model using step-wise regression that included fork-length, age, site, $\delta^{15}N$ and $\delta^{13}C$ as candidate independent variables found that fork-length, age, and $\delta^{15}N$ contributed significantly to explaining 62% of the variation within the Northern Dolly Varden non-length-adjusted THg data set (all coefficients $p \leq 0.073$, $r^2 = 0.631$, $n = 145$) for which complete biological data were available (i.e., data for Babbage River and Ptarmigan Bay were not included owing to missing age data). Additional explanatory power was not provided by the inclusion of "site" as a variable in the final model ($p > 0.05$). Sensitivity analysis completed by raising and lowering the F-to-enter criterion to the upper $\alpha = 0.01$ and 0.10 points of the F-distribution and reversing the direction of selection (forward to backward) did not alter the estimated model. Furthermore, the estimated AIC for the selected model was lower than the AIC associated with any of the possible alternative models estimated using the same data. Standardized regression coefficients indicated that length was the most important determinant of THg, followed by age and $\delta^{15}N$ (Table 2.5), with length being 1.5× as important for explaining variation in THg as $\delta^{15}N$. Model residuals showed no evidence of serial correlation, heteroscedasticity or non-normality when tested at the $\alpha = 0.05$ level of significance.

Discussion

Analyzed archival samples indicated that length-adjusted THg differed spatially among populations, but that there were no geographically significant patterns in the data. Measured THg levels within and among populations were positively correlated with the size and age of tested

fish in some of the studied populations, with length generally being a better correlate with THg than age. There was also evidence to suggest that differences in THg levels within and among populations were influenced by differences in the trophic ecology of the studied populations. Within populations, the influence of $\delta^{15}N$ was limited. Among populations, $\delta^{15}N$ was an important explanatory factor.

For the purposes of comparing THg levels among populations, adjustments were made to control for the effects of biological variation (e.g., van der Velden et al., 2012). Fork-length was observed to have the most influence and was marginally more effective at predicting THg than age. In contrast, for closely related anadromous Arctic charr, Swanson and Kidd (2010) and van der Velden et al. (2013c) concluded that age was the better predictor of THg. Differences in growth rates between the anadromous charr species may explain the contrasting conclusions regarding the significance of length as an explanation of THg concentrations in Northern Dolly Varden. For example, size-at-age plots of anadromous Arctic charr reported in van der Velden et al. (2012) indicate that Arctic charr averaged just over 350 mm at age-10, whereas Northern Dolly Varden at age-10 average 504 mm. The comparative difference in tissue THg levels may arise as a result of somatic growth dilution when considering growth rates, food consumption rates and activity costs in Northern Dolly Varden, as has been observed for other northern fish species including lake whitefish, *Coregonus clupeaformis,* and Atlantic salmon, *Salmo salar* (Ward et al., 2010). Alternatively, differences among the species may be driven by differences in THg at the base of the food webs within which the species feed (van der Velden et al., 2013a).

The historical length-adjusted THg concentrations (means 35–128 ng/g ww) in anadromous Northern Dolly Varden described here were all below Health Canada's 0.5 ppm guideline for commercial sale of fish (Health Canada, 2008) and were similar to the levels

reported for contemporaneously sampled anadromous broad whitefish, lake whitefish, and landlocked Arctic charr from the Yukon Territory, YT, and Northwest Territories, NWT (Lockhart et al., 2005), a fact that would suggest that the health risks associated with Dolly Varden fish consumption were on par with those for other consumed fish species. For example, similar THg concentrations were found for anadromous broad whitefish from Campbell Lake, Kugaluk River, Lake 100, and Travaillant Lake, NWT (1988–1992), with recorded mean THg levels ranging from 20–70 ng/g ww (Lockhart et al., 2005). THg levels in lake whitefish from the Mackenzie Delta and Mayo Lake, YT and Burnt Lake, Colville Lake, Manuel Lake, and Lac Ste. Therese, NWT (1989–1993), ranged from 20–150 ng/g ww, and landlocked Arctic charr captured at Paulatuk, NWT, in 1984 registered mean THg concentrations of 40 ng/g ww (Lockhart et al., 2005).

Anadromous Northern Dolly Varden THg concentrations, however, were consistently lower than levels reported contemporaneously for other predatory fish species such as non-anadromous walleye, lake charr, and anadromous inconnu. Braune et al. (1999) and references therein determined mean THg levels in walleye to be 980 ng/g ww from Lac à Jacques, NWT in 1994 and 1340 ng/g ww from Lac Ste. Therese, NWT in 1992. Lockhart et al. (2005) found lake charr with mean levels of 310 ng/g ww in Colville Lake, NWT in 1993 and 210 ng/g ww in Yaya Lake, NWT in 1995, while Braune et al. (1999) and references therein reported levels ranging from 130 ng/g ww in Lac Belot, NWT in 1993 to 950 ng/g ww in Lac Ste. Therese, NWT in 1992. Inconnu were found to have concentrations of 170 ng/g ww from the Mackenzie Delta in 1992 and Yaya Lake, NWT in 1995 (Lockhart et al., 2005). Thus from a human health perspective, Northern Dolly Varden are a better consumption choice than many of the commonly used fish in the north.

Significant differences in THg among sites seen in this study can be attributed to a variety of factors. Physiological and ecological factors commonly influencing THg levels in fish include: size (Grieb et al., 1990; Berninger and Pennanen, 1995), trophic level (Cabana and Rasmussen, 1994), growth rate (Harris and Bodaly, 1998), age (Bache et al., 1971; Kim, 1995), and carbon source (Power et al., 2002). While it is presumed that measured concentrations are reflective of recent summer feeding/occupancy of marine environments, variations among populations may also be related to the marine coastal waters they occupy. This has been noted for freshwater systems where one study that looked at THg in fish from a similar location found that the variations in THg in fish among lakes from the Mackenzie River basin were due to dissolved organic carbon concentrations (Evans et al., 2005a). Other common abiotic factors influencing THg levels include temperature (Carrie et al., 2010) and high discharge rates (Leitch et al., 2007), both of which may contribute to spatial heterogeneity in coastal feeding environments (Schell et al., 1998).

The lack of latitudinal or longitudinal relationships with THg paralleled findings reported by Gantner et al. (2010a) who found significant spatial differences in non-anadromous Arctic charr population mean THg levels from across the Canadian Arctic, but no latitudinal or longitudinal trends. Van der Velden et al. (2013a,c) similarly reported no geographic trends in THg of anadromous and non-anadromous Arctic charr from the eastern Canadian Arctic.

As expected, significant correlations were found within and among populations for both size and age. A significant correlation between THg and fork-length and age was hypothesized as strong relationships between these variables are commonly observed in other northern fishes such as northern pike, Arctic grayling, walleye and lake charr (Jewett et al., 2003; Evans et al., 2005a). Positive THg–age and THg–fork-length trends can result from the continuous

accumulation and very slow excretion of Hg in fish (Bache et al., 1971; Van Walleghem et al., 2013) and the increased ability to feed on larger, more contaminated prey as fish age and grow (Stafford et al., 2004). Lockhart et al. (2005) similarly noted that size explained the variation in lake charr unadjusted THg levels among sites from throughout the Canadian Arctic. In contrast, van der Velden et al. (2013c) determined age, and not size, to be the variable that most influenced THg concentrations in anadromous Arctic charr. Swanson et al. (2011) and Swanson and Kidd (2010) also noted that age was one of the best predictors of among population THg differences in Arctic and lake charr.

It was hypothesized that THg would have an inverse relationship with $\delta^{13}C$, as seen in other studies (e.g., Power et al., 2002; Kidd et al., 2003, and Stern and Macdonald, 2005). Within populations, there was only a weak tendency for the relationship to hold and in many instances (9 of 10) the relationship was either reversed or non-significant. Among populations, Northern Dolly Varden appear to accumulate less THg when feeding in marine food webs with lower basal $\delta^{13}C$ values. The trend may result from the combination of high $\delta^{13}C$ and THg values prevalent to the west along the Alaskan coast and the migration pattern of Northern Dolly Varden. For example, an increase in copepod $\delta^{13}C$ (Schell et al., 1998) has been demonstrated when moving west along the Alaskan coast from the Yukon, the same direction of documented Dolly Varden migration (Kowalchuk et al., 2010a; references therein). In addition, Macdonald et al. (2005) suggested that the Arctic oscillation may play a significant role in the transfer of contaminants to the western Canadian Arctic and noted possible current transport of contaminants from the Chukchi Sea to the Beaufort Sea, with higher contaminant values observed in the Chukchi Sea.

As predicted, $\delta^{15}N$ was a factor in explaining the variation in length-adjusted THg among sites and within two populations. The positive relationship between THg and trophic level corroborates other studies of landlocked and anadromous populations of northern fishes (e.g., Cabana and Rasmussen, 1994; Kidd et al., 1995; Atwell et al., 1998; Power et al., 2002; van der Velden et al., 2013c) and is indicative of an accumulation of THg through the food web (Atwell et al., 1998). However, $\delta^{15}N$ was the third most important explanatory variable and, thus, did not account for the majority of the THg variation. Similarly, van der Velden et al. (2013c) observed significant correlations between THg and trophic level, but noted that trophic level did not explain the differences in THg concentrations between anadromous and non-anadromous Arctic charr. Overall, findings from the literature for charr species show that significant THg–$\delta^{15}N$ relationships are common in northern fish populations but that $\delta^{15}N$ is not always the best predictor of among or within population variation in THg (e.g., Power et al., 2002; Gantner et al., 2009; Swanson and Kidd, 2010).

Discrepancies among studies with respect to the importance of $\delta^{15}N$ may relate to issues associated with baseline adjustment which is typically recommended to account for variations in anthropogenic influences in freshwater for among-site comparisons (e.g., Cabana and Rasmussen, 1996 and Post, 2002). Here the heterogeneity of the nitrogen baseline along the Beaufort coast (e.g., Schell et al., 1998) presented problems for simple among-population baseline correction using the standard freshwater approaches (e.g., Casey and Post, 2011), or those employed for the study of anadromous Arctic charr (e.g., Swanson et al., 2010; van der Velden et al., 2012) where localized use of the nearshore environment could be reasonably assumed and locally-specific data for characterizing the baseline easily collected. If individual fish range as widely in the environment as telemetry studies are increasingly showing for

salmonid species as varied as Atlantic salmon (Holm et al., 2006; Reddin et al., 2011; Lacroix, 2013), Steelhead, *Oncorhynchus mykiss* (Johnson et al., 2010), Sockeye salmon, *Oncorhynchus nerka* (Cooke et al., 2008) and Dolly Varden (Bond and Quinn, 2013) and stable isotopes reflect the time-integrated effect of seasonal feeding (Fry, 2006), then individual Northern Dolly Varden $\delta^{15}N$ should represent a time-averaged index of seasonal feeding in multiple food webs with varying baselines. Differences in $\delta^{15}N$, therefore, may provide an appropriate index of differences in spatial and/or trophic feeding suitable for making direct comparisons among individuals.

In contrast, the traditional baseline correction that assumes locally specific movement for each population would erroneously adjust the signatures of individual Northern Dolly Varden and obscure understanding of the relationship between variables such as THg concentrations and $\delta^{15}N$. Indeed, recent telemetry studies with Dolly Varden have shown high variability within and among populations in the context of run timing and marine occupancy (Bond and Quinn, 2013), with behavior of individuals influenced by a combination of age, size, maturational state and the relative abundance of resources in marine and fresh waters. These factors create a complex array of individual migratory behaviors (Armstrong, 1984; Jonsson and Jonsson, 2011; Bond and Quinn, 2013). Such variation in marine behaviors by Dolly Varden further suggests interpreting $\delta^{15}N$ as an integrated index of varying reliance on the portfolio of locally differential baselines within which any single fish may interact. Together these all will influence and result in among-individual and among-population THg exposure and variation. Nevertheless, the variation in summer feeding, age, size, maturational state, and migratory behavior in Dolly Varden is an issue that requires further investigation.

Table 2.1: Sites for which anadromous Northern Dolly Varden samples were obtained for analysis. Site code, number of fish (*n*), geographic co-ordinates and year of capture are given below.

Site	Site code	*n*	Latitude	Longitude	Year of capture
Babbage River	BR	30	68°37'47"N	139°22'12"W	1991
Cache Creek	CC	30	68°31'11"N	136°13'47"W	1988
Rat River	RR	30	67°46'48"N	136°19'11"W	1988
Firth River	FR	14	68°40'12"N	140°55'11"W	1988
Ptarmigan Bay	PB	28	69°29'11"N	139°01'12"W	1988
Canoe River	CR	30	68°46'11"N	138°45'00"W	1988
Shingle Point	SP	16	68°58'48"N	137°31'12"W	1989
Pauline Cove	PC	30	69°34'47"N	138°52'12"W	1989
Thetis Bay	TB	8	69°32'59"N	139°01'48"W	1989
Big Fish River	BFR	30	68°27'00"N	136°11'60"W	1991

Table 2.2: Mean ± standard deviation and range of fork-length (mm), age (year), THg (ng/g wet weight), $\delta^{13}C$ (‰), and $\delta^{15}N$ (‰) for samples obtained from each site. Site codes correspond to those given in Table 2.1. Sites with biological measures not significantly different from one another at the $\alpha = 0.05$ level of significance are indicated with uppercase superscripts (e.g., A, B).

Site	Statistic	Fork-length (mm)	Age (yr)	THg (ng/g ww)	$\delta^{13}C$ (‰)	$\delta^{15}N$ (‰)
BR	Mean ± Stdev	497 ± 45^{AGH}	n/a	108 ± 39^{ABF}	-22.7 ± 0.6^{AC}	$+15.0 \pm 0.9^{ADF}$
	Range	393 – 612	n/a	30 – 181	-24.6 to -21.8	+13.0 to +16.5
CC	Mean ± Stdev	405 ± 27^{BCFI}	6.5 ± 1.6^{ABDE}	63 ± 14^{ABEF}	-22.2 ± 0.4^{BCFG}	$+14.0 \pm 0.4^{BCFGH}$
	Range	361 – 489	4 – 10	43 – 102	-23.0 to -21.5	+13.4 to +14.9
RR	Mean ± Stdev	438 ± 44^{BCGI}	7.4 ± 1.1^{ABEF}	83 ± 35^{ABF}	-23.3 ± 0.7^{ABCG}	$+13.7 \pm 0.7^{BCE}$
	Range	331 – 519	6 – 10	25 – 163	-25.4 to -21.8	+12.6 to +15.5
FR	Mean ± Stdev	588 ± 67^{DH}	11.8 ± 1.6^{C}	161 ± 37^{C}	-22.6 ± 0.8^{D}	$+15.0 \pm 0.6^{ADFGH}$
	Range	505 – 681	10 – 15	102 – 254	-24.3 to -20.8	+13.8 to +16.0
PB	Mean ± Stdev	272 ± 40^{E}	n/a	29 ± 7^{DF}	-23.1 ± 0.9^{EF}	$+13.1 \pm 0.6^{CE}$
	Range	205 – 363	n/a	16 – 45	-26.4 to -21.8	+11.7 to +14.1
CR	Mean ± Stdev	360 ± 24^{BF}	5.2 ± 0.8^{ADE}	45 ± 11	-22.7 ± 0.6^{BEF}	$+13.5 \pm 0.6^{BCE}$
	Range	305 – 401	4 – 6	$21 – 67^{ABEF}$	-23.9 to -21.5	+11.8 to +14.5
SP	Mean ± Stdev	439 ± 68^{BCGI}	6.6 ± 2.0^{ABDEF}	75 ± 48^{ABEF}	-22.9 ± 0.7^{BCFG}	$+14.4 \pm 0.7^{ABDFGH}$
	Range	354 – 623	4 – 10	27 – 167	-24.2 to -21.9	+13.3 to +15.9
PC	Mean ± Stdev	479 ± 97^{ACGH}	8.4 ± 3.0^{BEF}	80 ± 51^{BEF}	-23.3 ± 1.1^{BCG}	$+14.3 \pm 1.0^{BDFGH}$
	Range	292 – 627	3 – 15	15 – 212	-25.8 to -21.6	+12.1 to +16.0
TB	Mean ± Stdev	528 ± 91^{ADGH}	10.9 ± 2.9^{C}	93 ± 44^{ABDEF}	-22.5 ± 0.6^{ABCG}	$+14.9 \pm 0.7^{ADFGH}$
	Range	400 – 635	7 – 14	38 – 138	-23.3 to -21.6	+13.9 to +15.8
BFR	Mean ± Stdev	434 ± 32^{BCI}	7.0 ± 1.6^{ABEF}	64 ± 27^{ABEF}	-23.9 ± 0.6^{BCFG}	$+14.3 \pm 0.7^{BDFGH}$
	Range	364 – 498	4 – 10	29 – 140	-25.2 to -22.7	+12.9 to +15.8

Table 2.3: Population-specific correlation coefficients (*r*) for length-adjusted THg (ng/g ww) concentrations and $\delta^{15}N$ (‰), $\delta^{13}C$ (‰), fork-length (mm), and age (years).

	$\delta^{15}N$ (‰)	$\delta^{13}C$ (‰)	Fork-length (mm)	Age (years)
Babbage River	0.501**	0.195	-0.031	
Cache Creek	0.066	-0.506**	0.013	0.466*
Rat River	0.252	-0.302	-0.015	-0.114
Firth River	0.354	0.219	-0.016	0.616
Ptarmigan Bay	-0.004	0.418*	0.000	
Canoe River	0.253	0.048	-0.010	-0.287
Shingle Point	0.277	0.150	-0.017	0.596*
Pauline Cove	0.271	0.010	-0.014	0.449*
Thetis Bay	0.189	0.306	-0.046	0.271
Big Fish River	0.605***	0.535**	0.005	0.336

* Significant relationships at $p < 0.05$.
** Significant relationships at $p < 0.01$.
*** Significant relationships at $p < 0.001$.

Table 2.4: Sample partial correlation coefficients (*r*) for THg (ng/g ww) concentrations versus age or fork-length when controlling for other factors. Sampling site locations correspond to those defines in Table 2.1. No age data were available for Babbage River and Ptarmigan Bay.

Sampling Site	logTHg vs. age, given fork-length	logTHg vs. fork-length, given age	logTHg vs. age, given fork-length and $\delta^{15}N$	logTHg vs. fork-length, given age and $\delta^{15}N$
BR				
CC	0.596**	-0.328	0.638**	-0.344
RR	-0.112	0.529**	-0.113	0.385
FR	0.675	0.017	0.663	-0.007
PB				
CR	-0.297	0.580**	-0.297	0.544**
SP	0.620*	0.596	0.496	0.602
PC	0.566**	0.566**	0.441	0.287
TB	0.586	0.174	0.514	0.175
BFR	0.388	0.280	-0.043	0.135

* Significant relationships at $p < 0.05$.
** Significant relationships at $p < 0.01$.

Table 2.5: Estimated multiple regression model coefficients, significance values (p), and standardized regression coefficients (β_j) for the model, $r^2 = 0.615$, explaining variation in unadjusted log[THg] (ng/g ww) as a function of biological and ecological variates.

	Regression coefficient	Significance (p-value)	Standardized regression coefficient (β_j)
Fork-length (mm)	0.001	< 0.001	0.354
Age (years)	0.027	0.002	0.288
$\delta^{15}N$ (‰)	0.066	0.002	0.229

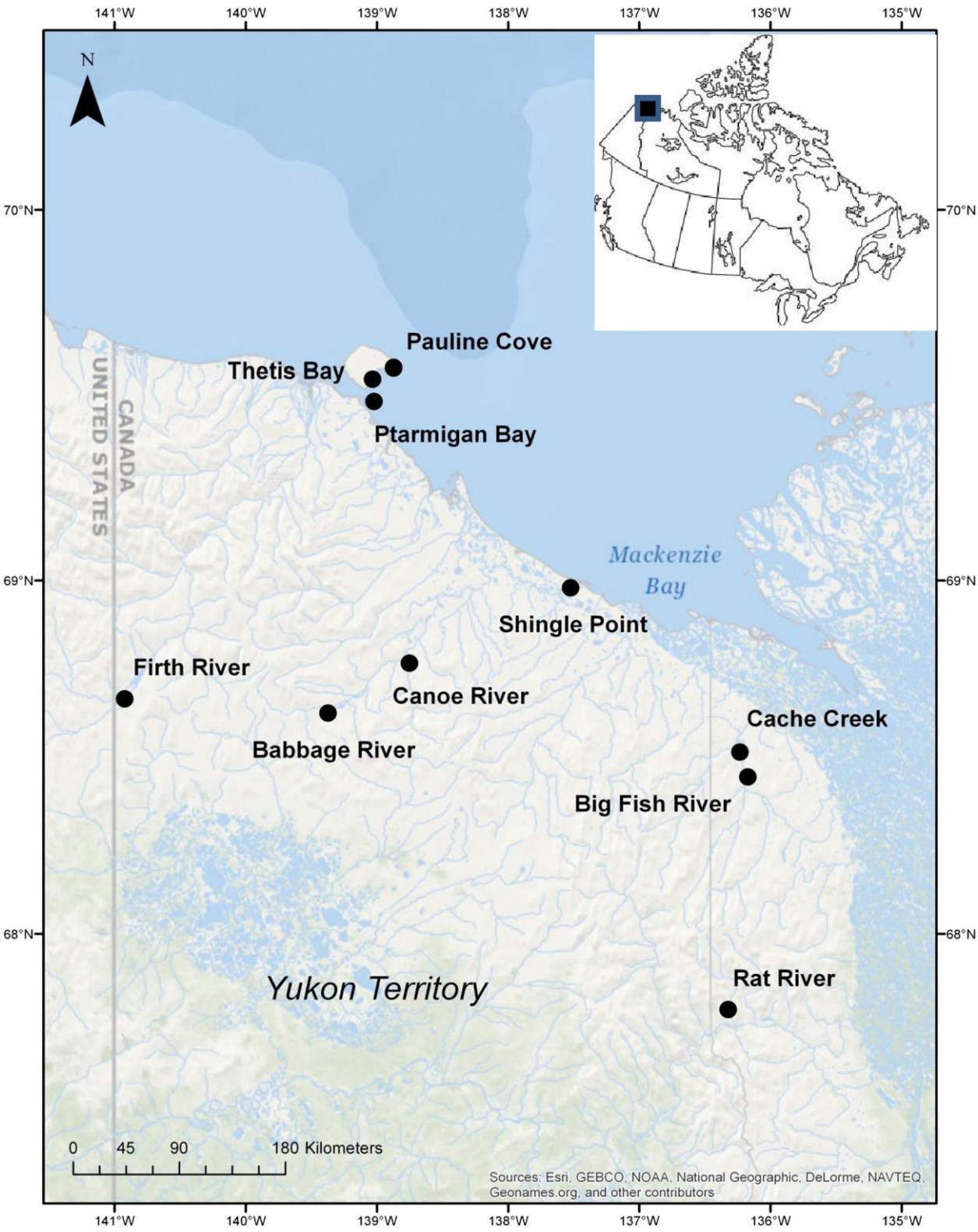


Figure 2.1: Location of sampling sites for populations of Northern Dolly Varden used in this study. Inset in the upper right highlights the sampling area with respect to Canada as a whole.

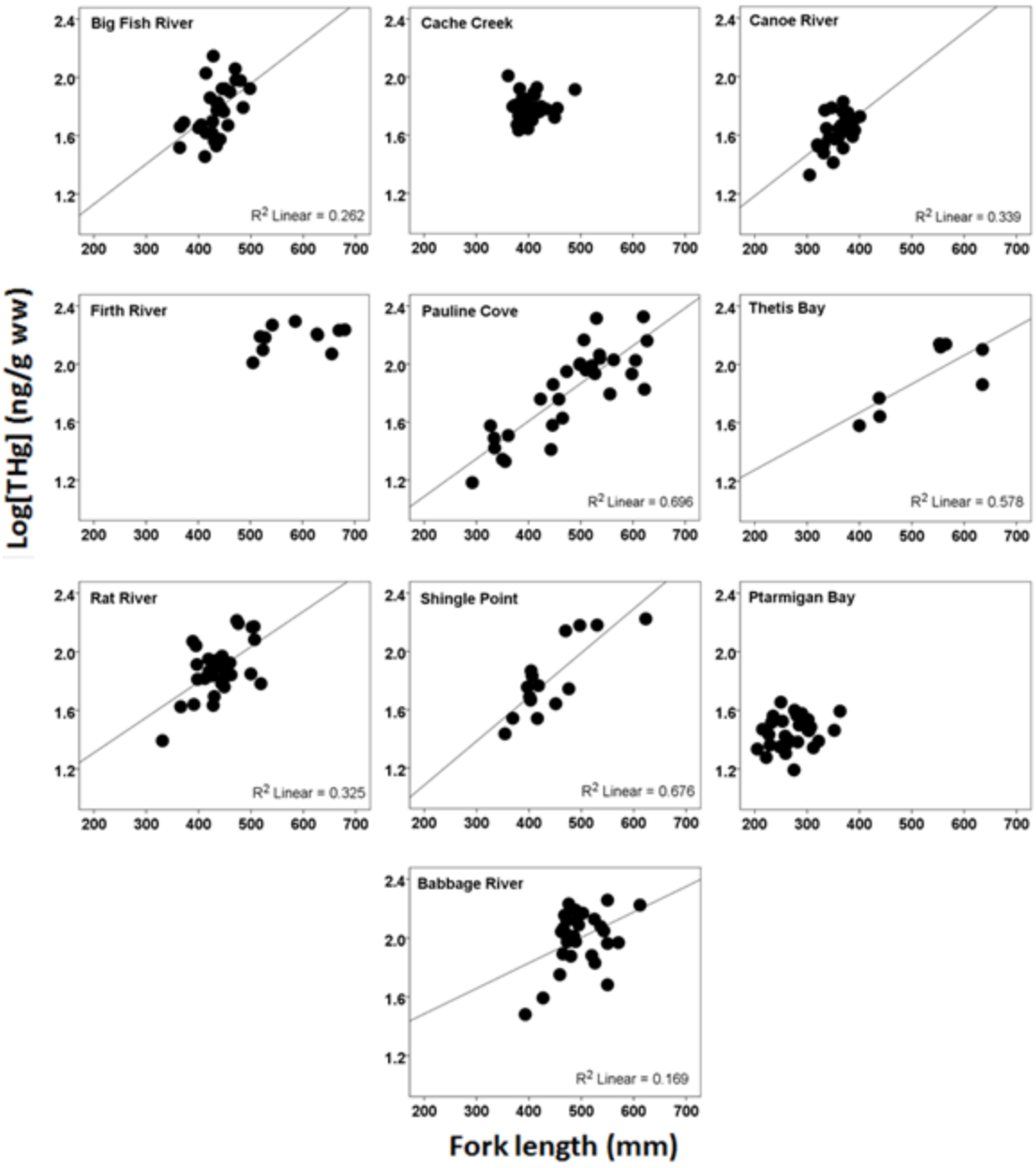


Figure 2.2: Log[THg] (ng/g ww) versus fork-length (mm) for each of the studied populations. Where a significant correlation exists, the estimated regression line is plotted as a solid line.

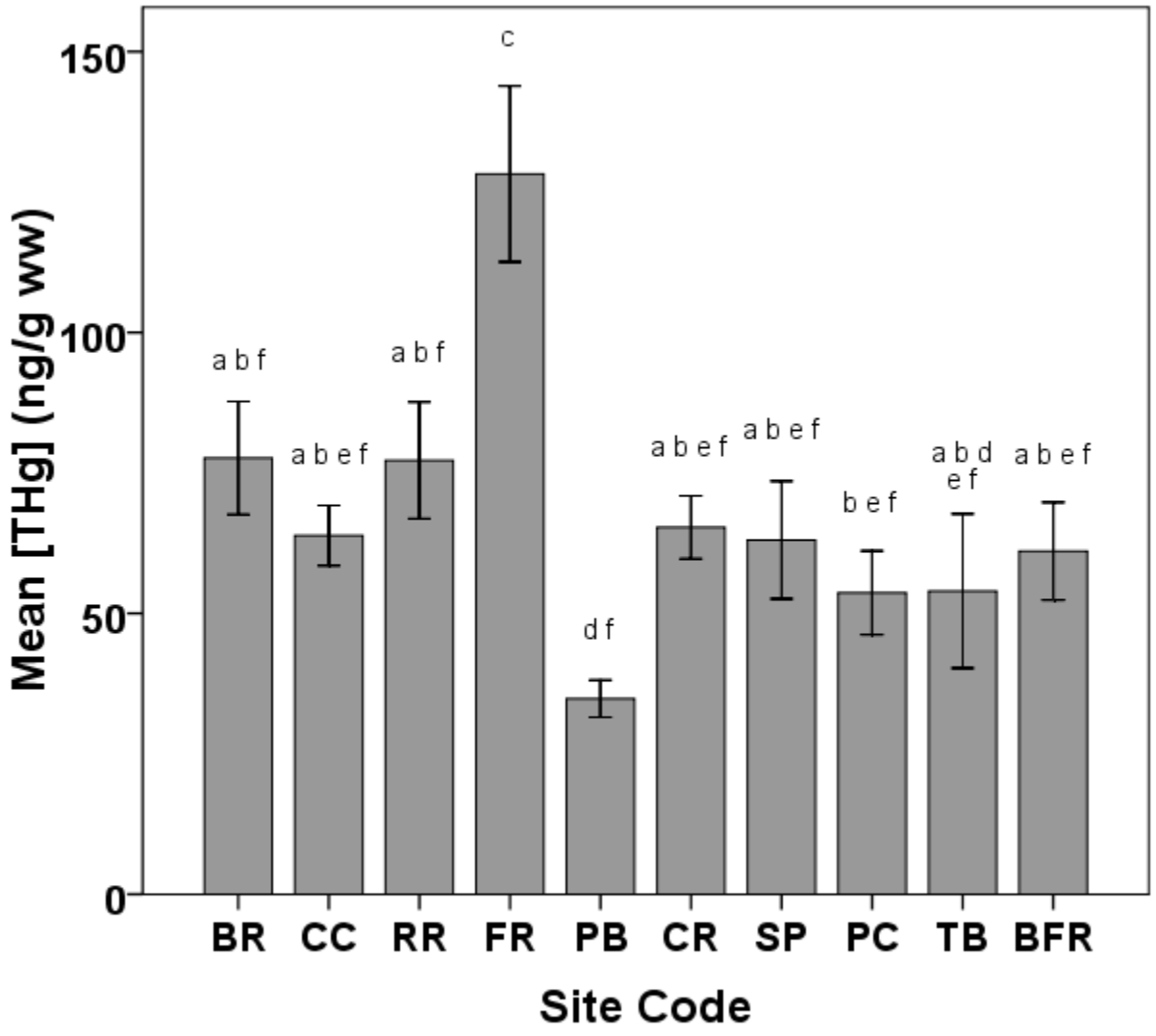


Figure 2.3: Mean length-adjusted THg (ng/g ww) for sampled sites. Site codes correspond to those given in Table 2.1. I-bars represent 95% confidence intervals. Sites with the same letter were not significantly different from one another (Tukey's HSD; $p > 0.05$).

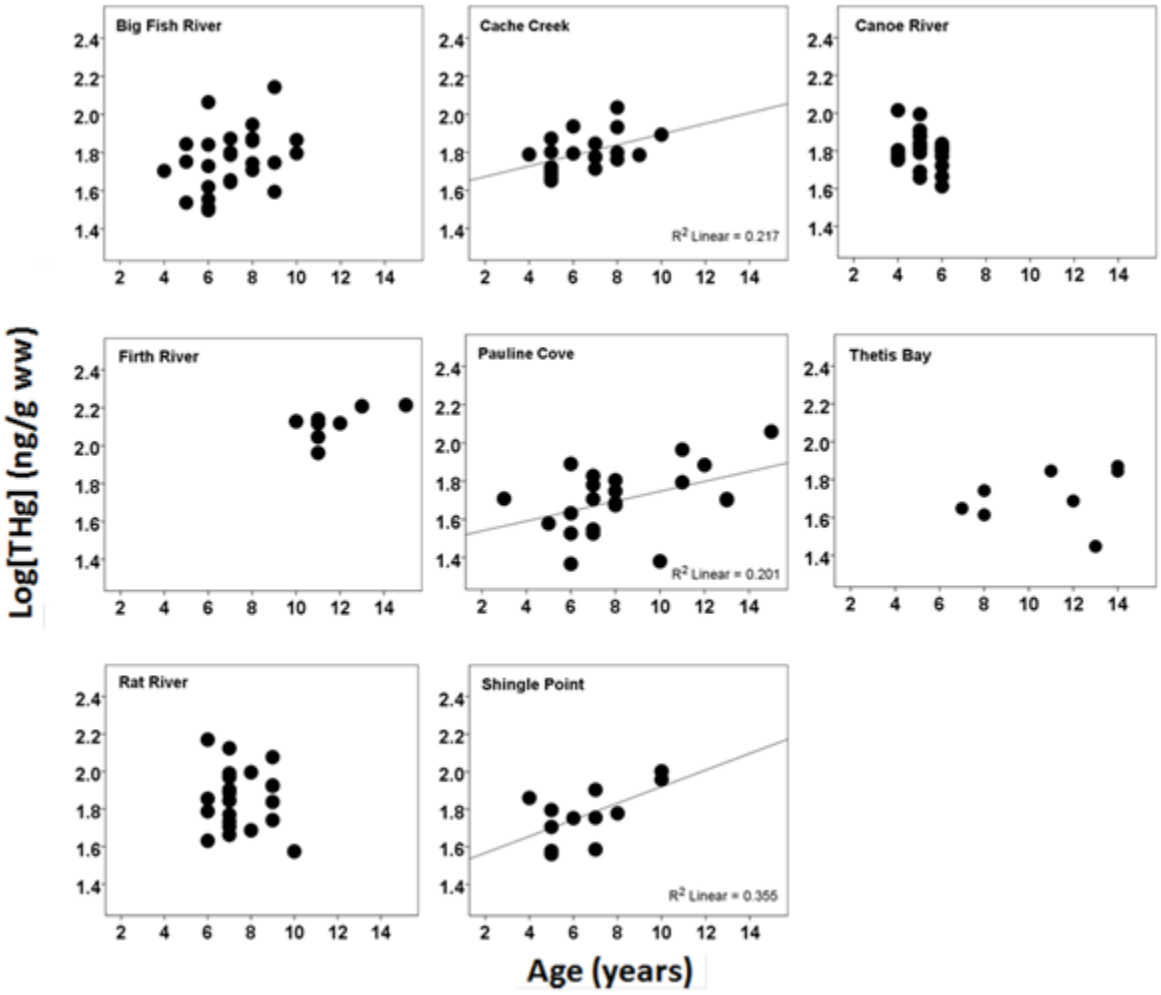


Figure 2.4: Length-adjusted log[THg] (ng/g ww) versus age for each of the studied populations. Where a significant correlation exists, the estimated regression line is plotted as a solid line.

Chapter 3: Life-history dependent variation of total mercury concentrations in Northern Dolly Varden from the Babbage River, Yukon Territory, Canada

Introduction

Mercury (Hg) is a heavy metal that, when methylated, can bioconcentrate and bioaccumulate as it moves through aquatic food webs, having harmful impacts on both the aquatic ecosystem and humans who depend on those systems (Morel et al., 1998; Braune et al., 1999; Booth and Zeller, 2005; Lockhart et al., 2005). Concerns over Hg contamination levels are most acute for northern peoples, e.g., Inuvialuit and Gwich'in, who rely on top predatory fishes such as Northern Dolly Varden (*Salvelinus malma malma*) for cultural and dietary purposes (Gallagher et al., 2011; Gallagher et al., 2012). As a result of their size and age, Northern Dolly Varden can accumulate high levels of Hg and may represent a consumption risk for northerners. While much is known about the factors that influence Hg levels in many northern fishes (e.g., Arctic charr, *S. alpinus*, lake charr, *S. namaycush*, northern pike, *Esox lucius*), relatively little is known about what controls the variation in Hg within and among Northern Dolly Varden populations.

Various biotic factors, some of which are life-history dependent, are known to influence fish Hg levels and are likely to be relevant for Northern Dolly Varden. Such factors include: fork-length (Grieb et al., 1990; Berninger and Pennanen, 1995), age (Bache et al., 1971; Kim, 1995), growth rate (Harris and Bodaly, 1998), trophic level (Cabana and Rasmussen, 1994) and carbon sources (Power et al., 2002). Larger and older fish are commonly associated with higher Hg concentrations due to longer periods of accumulation and slower, size-dependent excretion of Hg (Bache et al., 1971; Van Walleghem et al., 2013) and the greater ability to feed on larger, more contaminated prey as they age and grow (Stafford et al., 2004). Faster growth rates linked with size have been associated with lower Hg levels as a result of growth dilution (Greenfield et

al., 2001; Swanson and Kidd, 2010); although there is equivocal empirical support for growth dilution in field studies (Ward et al., 2010). The dilution occurs when a fish spends less energy on consumption and respiration to efficiently reach a given size (de Freitas et al., 1974; Norstrom et al., 1976; Greenfield et al., 2001). As diet is the main Hg exposure route in fish, both trophic level (Kidd et al., 1995; Atwell et al., 1998; Lockhart et al., 2005) and carbon source origin (Power et al., 2002) have been correlated with Hg concentrations in fish.

Northern Dolly Varden are a riverine charr species distributed north of the Alaska Peninsula and west of the Mackenzie River (Reist et al., 2002). Northern Dolly Varden show evidence of three separate life-history types: isolate, resident, and anadromous (Gallagher et al., 2012). Isolates are completely separated from the other forms by physical migration barriers or distance, and spend their entire lives in their natal stream with no access to the sea (Kowalchuk et al., 2010a; Reist and Sawatzky, 2010; Gallagher et al., 2012). Residents have access to the sea, but stay in their natal stream for their entire lives and are often found alongside the anadromous form (Kowalchuk et al., 2010a; Gallagher et al., 2012). Both the resident and isolate forms are considered non-anadromous and, therefore, are found and feed only in freshwater (Gallagher et al., 2012). By contrast, anadromous Northern Dolly Varden begin migrating in late June between 3-5 years of age to the Beaufort Sea each summer to feed (Gallagher et al., 2012). The anadromous fish return to freshwater for over-wintering and spawning, typically beginning in late August (Gallagher et al., 2012). Due to feeding in the marine environment, anadromous fish are larger in size and typically have faster growth rates than freshwater-feeding conspecifics (Gallagher et al., 2012).

Within a species, differences in life history often hold implications for measured contaminant concentrations because of differences in habitat occupancy and/or feeding tactics.

As a consequence, life history associated differences in total mercury (THg) concentrations have been studied in northern fishes such as Arctic (e.g., Rigét et al., 2000; Evans et al., 2005a,b; Swanson et al., 2011; van der Velden et al., 2012, 2013a,c) and lake charrs (e.g., Swanson et al., 2011), with anadromous fishes typically having lower THg concentrations as compared to similarly sized and aged landlocked fish (Evans et al., 2005a,b; Lockhart et al., 2005; Swanson et al., 2011; van der Velden et al., 2012, 2013a,c). For example, Rigét et al. (2000) reported anadromous Arctic charr to have 10-15 fold lower THg tissue concentrations than resident Arctic charr, van der Velden et al. (2013a,c) reported THg values 1.8 to 6.3 times higher in landlocked fish versus site-paired anadromous counterparts, and Swanson et al. (2011) noted that anadromous and lake-resident Arctic charr had significantly lower THg levels than landlocked conspecifics. Limited comparable studies with other species (e.g., Japanese eel, *Anguilla japonica*, and northern crayfish, *Orconectes virilis* (Hagen)) have similarly demonstrated the importance of life history as a determinant of tissue heavy metal contamination levels, either as a result of differences in migration or habitat occupancy tactics (Pennuto et al., 2005, Le et al., 2010). In some instances, however, comparisons between resident and anadromous charr life-history types have yielded differing results. For example, Swanson and Kidd (2010) and Swanson et al. (2011) noted that for both Arctic charr and lake charr, anadromous THg concentrations were not significantly different from those of lake-resident or landlocked fish, respectively.

Explanations for the differences among life-history types are limited, but suggestions include differences in feeding (Rigét et al., 2000) and physiology (e.g., maturity, Le et al., 2010) and/or Hg or energy density variations in the marine and lacustrine prey items (van der Velden et al., 2013c). On the other hand, Swanson and Kidd (2010), who found no differences in THg

concentrations between marine and freshwater prey items, suggested growth rates, C:N ratio, and ecosystem-specific differences in Hg biomagnification rates as explanations of the among life-history differences in measured THg tissue concentrations.

Given the limited information available on among life-history type differences in THg tissue concentrations, a key objective of this study was to compare THg concentrations among anadromous, resident, and isolate Northern Dolly Varden (NDVC). Specifically, the hypotheses tested were that: [i] NDVC life-history types would evidence significantly different patterns of THg tissue concentrations; [ii] THg differences within and among life-history types of NDVC would be related to differences in feeding ($\delta^{13}C$) and trophic position ($\delta^{15}N$); [iii] THg levels in NDVC would increase with size; and [iv] THg and growth rate would be negatively correlated.

Methods

Sample Collection

Archived muscle tissue samples (*n* = 62) of Northern Dolly Varden were obtained from the Department of Fisheries and Oceans Canada (DFO) in Winnipeg, Manitoba. All life-history types were sampled from the Babbage River located in the Yukon North Slope (Fig. 3.1), with anadromous (*n* = 30) and resident (*n* = 10) NDVC sampled at 68° 37’ 59”N and 138° 43’ 12”W and isolate (*n* = 22) NDVC sampled at 68° 37’ 44”N and 139° 21’ 14”W. Isolate and resident samples were caught in September 1988 and anadromous samples were caught from July to October of 1991 as they were returning from summer feeding in the Beaufort Sea. Isolates were distinguished by their size and capture location in the upper Babbage River, while anadromous and resident NDVC were distinguished by visual examination, with resident NDVC being

significantly smaller in size and exhibiting dark colouration/parr marks (Sandstrom et al., 1997; J. Johnson, personal communication).

Approximately 5 g of the muscle tissue was dissected from a standard area located just behind the dorsal fin and above the lateral line from whole frozen fish stored in individualized polyethylene bags. Biological data (fork-length (mm), weight (g), and age (year)) data were also obtained from the DFO capture records for each fish. Ages were determined using the whole otolith method (Sandstrom et al., 1997). The outer surface of the otolith was hand-ground until the core/nucleus was visible and the otolith was then examined in a petri dish under a stereoscopic microscope using water or a clearing agent (e.g., oil of wintergreen) to elucidate the annuli. Ages were not available for the anadromous NDVC and were inferred from a von Bertalanffy length-at-age model (e.g., Wootton, 1998) estimated using non-linear regressions (Bates and Watts, 1988) from the individual age-length data available for Northern Dolly Varden captured from the Babbage River from 1986-1993 (Sandstrom et al., 1997). Model estimation employed the Levenberg-Marquardt algorithm in Statistica, version 8 (Statsoft Inc., Tulsa, OK) using the default convergence criterion and varying starting values as a check for convergence stability (Bates and Watts, 1988). Maximum cut-off ages for males and females, respectively, of 11 and 14 years (DFO, 2003; and references therein) were assumed based on the ranges of ages reported in Sandstrom et al. (1997).

Stable isotope analyses

All muscle samples were measured for stable isotope ratios of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) following methods described in Guiguer et al. (2002). Briefly, the samples were dried at 50°C for 48 h and pulverized to an homogenate with a Retsch MM 301 ball mill grinder (F. Kurt

Retsch GmbH Co., Haan, Germany) and weighed on a Mettler Ultra micro balance (UMX2, Mettler-Toledo GmbH, Greifensee, Switzerland). Between 0.275 – 0.300 mg of prepared material was used for stable isotope analyses (SIA) completed with 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) at the Environmental Isotope Laboratory, University of Waterloo (Waterloo, Ontario). Machine analytical precision, respectively, for $\delta^{13}C$ and $\delta^{15}N$ was ±0.04‰ and ±0.3‰ and was determined by repeat analysis (duplicates run every $n = 11$) of laboratory working standards cross-calibrated to International Atomic Energy Agency standards CH6 for $\delta^{13}C$ and N1 and N2 for $\delta^{15}N$. All resulting measurements were expressed using standard delta notation as parts per thousand differences (‰) with respect to the international reference standards of Vienna Peedee Belemnite for $\delta^{13}C$ (Craig, 1957) and nitrogen gas in the atmosphere for $\delta^{15}N$ (Mariotti, 1983).

Total mercury analyses

Total mercury concentrations for a sub-sample (0.1-0.2g) of non-homogenized muscle tissue were measured on a Milestone Direct Mercury Analyzer, DMA-80 (Milestone S.r.l., Sorisole, Italy) using thermal decomposition followed by atomic absorption spectroscopy following U.S. EPA method 7473 (U.S. Environmental Protection Agency, 2007), with results expressed as ng/g wet weight (ww). Standard reference materials (SRMs) were run at the beginning and end of every batch of 20 samples, with no less than 5 blanks run in each sample batch. The method detection limit determined as 3x the standard deviation of the blanks was 0.03 ng Hg (approximately 0.2 ng/g ww). The certified reference materials and percent recoveries (mean percentage of certified value±1 standard deviation) used and determined in this study

included National Institute of Standards and Technology (NIST) 1566B, oyster tissue, (94.9±1.2%), NIST 2976, mussel tissue, (97.0±3.6%), NIST 2974a, freeze dried mussel tissue, (97.3±0.7%), and National Research Council of Canada (NRC) DORM-3, fish protein, (94.6±5.7%). In all analyses, a single sample was run in triplicate in each sample batch and the mean relative standard deviation of the triplicates was measured as 6.3%.

Methylmercury analyses

Methylmercury (MeHg) is the dominant, methylated, toxic form of mercury in organisms such as fish (Lindqvist and Rodhe, 1985). To determine whether the measurement of THg was an appropriate estimate of MeHg concentrations, random sub-samples (n = 20; same samples used for methylmercury analyses in Chapter 2) of fish were measured for MeHg. Muscle tissue samples were freeze dried for 48 hours and then ground using an acid washed glass mortar and pestle and placed into acid washed vials. Samples were weighed (±0.1 mg) before and after lyophilisation and the percent moisture was computed for each sample to facilitate conversion of wet weight (ww) and dry weight (dw) concentrations of THg and MeHg. Concentrations of MeHg and inorganic Hg (II) were determined at Quicksilver Scientific (Lafayette, CO, USA) using matrix spikes and certified reference materials as quality control measures. The limit of detection (LOD) and limit of quantification (LOQ) were respectively, 2.0 ng/g and 5.0 ng/g for both MeHg and Hg (II) in analyzed tissues samples.

Statistical analyses

All statistical analyses were performed using the statistical program SPSS (SPSS Inc., 2008) with Type I error set to α = 0.05. All data were tested for normality using residual plots

and Kolmogorov-Smirnov and Shapiro-Wilk statistics (Swanson et al., 2011). Data were log_{10}-transformed when residual plots did not illustrate variance homogeneity and when the significance values of the Kolmogorov-Smirnov and Shapiro-Wilk statistics were < 0.05 (Swanson et al., 2011).

Fish growth rate was determined using the following equation:

(1) Growth rate (mm/year) = fork-length (mm) / age (year)

Linear regressions were performed to determine the significance of the relationship between selected variables and partial correlation coefficient analyses were used to evaluate the significance of a single variable when other variables were held constant (e.g., Zar, 2010). Where necessary regression lines were compared following procedures described in Zar (2010). An analysis of variance (ANOVA) was performed to determine significant differences among life-history types and two sample t-tests corrected for variance differences were performed to determine significant differences between means (Zar, 2010).

Past studies have indicated either age (e.g., van der Velden et al., 2013c) or fork-length (e.g., Swanson et al., 2011 and Rigét et al., 2000) as the best variable to adjust when comparing THg concentrations among individuals. Relationships between THg and fork-length and age within and among life-history types were estimated using linear regression (e.g., van der Velden et al., 2013c). Log_{10}THg concentrations were age and length adjusted using age 4 individuals and a standard fork-length of 218 mm and compared among life-history types with a Tukey's honestly significant difference post hoc test. Age-4 was selected as it fell within the age ranges for all life-history types. Length standardizations were performed using methods as described by Fleming and Gross (1990):

(2) $$M_i = MO_i * (L/LO_i)^b$$

where M_i is the adjusted $\log_{10}$THg concentration for the i^{th} fish, MO_i is the observed $\log_{10}$THg concentration for the i^{th} fish, L is the mean length from all populations (218 mm), LO_i is the observed length of the i^{th} fish, and b is the slope of $\log_{10}$THg on $\log_{10}$fork-length.

A stepwise multiple regression using all life-history types was performed to determine the variables that best explained $\log_{10}$THg values among a candidate set of variables that included: life-history type, fork-length, age, growth rate, δ^{15}N, and δ^{13}C. Candidate variables considered for inclusion in the model were chosen from the set of relevant biological variables reported in the literature as being significantly correlated with THg, e.g., Rigét et al. (2000), Power et al. (2002), Gantner et al. (2009, 2010a,b), Swanson and Kidd (2010) and van der Velden et al. (2012).

As the magnitude of estimated regression coefficients depends on the variables included in the regression and their units of measure (Dunn and Clark, 1987), standardized regression coefficients were calculated following Cox (1987) to judge the relative influence of each independent variable on $\log_{10}$THg. Standardized regression coefficients (β_j) measure the amount by which $\log_{10}$THg changes in terms of units of its own standard deviation for a unit change in the standard deviation of the j^{th} independent variable, when all other independent variables are held constant. The βj coefficients, therefore, allow direct comparison to be made between variables in terms of their relative importance for explaining variation in $\log_{10}$THg and are useful for establishing which variables have the greatest influence on measured concentrations of $\log_{10}$THg in Northern Dolly Varden.

Sensitivity analysis on model variable selection was completed by varying the F-to-enter and exit criterion (Draper and Smith, 1981) to determine whether there was an effect on variable selection. In addition to sensitivity testing, AIC (Akaike Information Criterion) values were

calculated for the complete set of possible models that could be estimated from the considered set of candidate variables to ensure an appropriate balance between model parsimony and fit was achieved (Burnham and Anderson, 2002; Anderson, 2008). Subsequent to estimation, regression models were assessed for statistical adequacy by testing residuals for conformance to the underlying assumptions of linear estimation methods using standardized procedures (Dunn and Clark, 1987).

Results

The mean biological characteristics of the tested life-history groups (isolate, resident and anadromous) varied significantly: fork-length ($F_{(2,58)} = 596.23$, $p < 0.001$), weight ($F_{(2,57)} = 176.495$, $p < 0.001$), and age ($F_{(2,53)} = 18.56$, $p < 0.001$), with isolate and resident individuals differing significantly from the anadromous fish which were larger, heavier and older (Table 3.1). Growth rates similarly differed significantly among the life-history types ($F_{(2,52)} = 78.47$, $p < 0.001$).

Individual THg concentrations in the assessed Northern Dolly Varden ranged from 8.2-180.8 ng/g ww ($n = 62$), with an average percentage of MeHg of 95 ± 3.4% (mean ± standard deviation) in the $n = 20$ samples assessed. Mean THg differed among life-history types ($F_{(2,59)} = 51.68$, $p < 0.001$) by a factor of 4.9 with mean concentrations (± standard deviation) ranging from 22.1 ± 13.5 ng/g ww in isolates to 107.8 ± 27.4 ng/g ww in anadromous Northern Dolly Varden (Table 3.1). When THg concentrations were standardized for fork-length (218 mm) at a common age of 4 (THg_A), significant differences (Fig. 3.2) were found among life-history types ($F_{(2,13)} = 33.790$, $p < 0.001$, $r^2 = 0.839$). Residents (76.4 ± 10.8 ng/g ww) were significantly higher in THg_A than either anadromous (8.4 ± 0.2 ng/g ww; $p < 0.001$) or isolates (37.3 ± 5.3

ng/g ww; $p = 0.017$) and isolates were significantly higher in THg_A than anadromous ($p < 0.001$) fish.

Among life-history types, the stable isotope metrics also differed significantly ($\delta^{13}C$ ($F_{(2,59)} = 631.24$, $p < 0.001$); $\delta^{15}N$ ($F_{(2,59)} = 611.31$, $p < 0.001$)), with THg increasing as life-history type $\delta^{13}C$ and $\delta^{15}N$ increased (Fig. 3.3). Within life-history groupings, $\log_{10}THg$ did not vary significantly with any of the assessed stable isotope ($\delta^{13}C$, $\delta^{15}N$, C:N ratio) or biological (length, age, growth rate) measures for isolates and varied significantly only with $\delta^{15}N$ ($r = 0.761$, $p = 0.011$) for the residents. In contrast, within the anadromous life-history type, $\log_{10}THg$ varied significantly with all stable isotope and biological measures except the C:N ratio (Table 3.2), with correlations being strongest for $\delta^{15}N$ ($r = 0.675$, $p < 0.001$), growth rate ($r = -0.652$, $p < 0.001$) and age ($r = 0.553$, $p = 0.024$) (Fig. 3.4). Comparison of $\log_{10}THg$ versus $\delta^{15}N$ regressions for resident and anadromous life-history types indicated the common slope model applied ($t = 0.725$, $p = 0.473$), but that intercepts differed significantly ($t = 3.708$, $p = 0.001$).

Partial correlation coefficient analysis further indicated significant correlations between $\log_{10}THg$ and $\delta^{15}N$ or growth rate when controlling for the effect of relevant cross-correlations with $\delta^{15}N$, age or growth rate (Table 3.3). Correlations between $\log_{10}THg$ and age became insignificant once the effect of cross-correlations in $\delta^{15}N$ or growth rate were considered.

A stepwise multiple regression analysis that included life-history type and the relevant biological (age, length, growth rate) and stable isotope ($\delta^{13}C$, $\delta^{15}N$) variables known to be significantly correlated with $\log_{10}THg$ in at least one life-history type (Table 3.2), yielded a model that explained 85.1% of the variation in $\log_{10}THg$ (Table 3.4) and included life-history type, $\delta^{15}N$ and growth rate as the important explanatory variables. Standardized regression

coefficients indicated that $\delta^{15}N$ was the most important determinant of $\log_{10}THg$ followed by growth rate and life-history type (Table 3.4), with $\delta^{15}N$ being 1.62x as important for the determination of $\log_{10}THg$ as the other variables combined.

Similar stepwise regression analysis repeated using only anadromous fish yielded a model that explained 56.1 % of the variation in $\log_{10}THg$ (Table 3.4) and included $\delta^{15}N$ and growth rate as the important explanatory variables. As with the model including all life-history types, standardized regression coefficients indicated that $\delta^{15}N$ was the most important determinant of $\log_{10}THg$, being 1.14x as important for the determination of $\log_{10}THg$ as growth rate.

Discussion

Northern Dolly Varden dorsal muscle tissue samples varied significantly among life-history types, with the anadromous life-history type tending to have greater absolute levels of THg (ng/g ww) than isolate or resident life-history types. Differences among the life-history types varied significantly with the stable isotope measures of feeding ($\delta^{13}C$) and trophic position ($\delta^{15}N$), with $\delta^{15}N$ being the more important variable for explaining differences within and among life-history types. Tissue THg concentrations increased significantly with size only among the anadromous fish, where they were also significantly negatively related to growth. Overall, among-individual differences in THg were best explained by life-history type, trophic position and growth rate, with differences in trophic position dominating as an explanation of among-individual variation in measured THg.

Measured concentrations of THg in anadromous NDVC (108 ± 39 ng/g ww) were comparable with those previously reported for anadromous 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, Rigét et al., 2000; 40 ng/g, Swanson et al., 2011). Values for freshwater resident and for isolated life-history types of NDVC (56 ± 27 ng/g ww and 22 ± 14 ng/g ww, respectively) invariably fell at the lower end of the range of values reported in the literature for freshwater resident 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., 2010b; 97–185 ng/g, Marusczak et al., 2011; 147–1520 ng/g, Muir et al., 2005; 30–940 ng/g, Muir et al., 2009; 120–801 ng/g, Rigét et al., 2000; 55–179 ng/g, Rognerud et al., 2002; 190 ng/g, Swanson et al., 2011).

In contrast to data generally reported for related Arctic and lake charrs, (e.g., Arctic charr, Rigét et al., 2000; Evans et al., 2005b; Lockhart et al., 2005; Swanson et al., 2011; van der Velden et al., 2012, 2013c, and lake charr, Swanson et al., 2011), freshwater resident NVDC tended to be lower in THg than their anadromous counterparts, although standardization for both age and size reversed the resulting relative levels of THg in anadromous and non-anadromous life-history types. Differences in non-adjusted THg among life-history types were likely driven by the importance of the relationship between $\delta^{15}N$ and THg, which is expected to increase with trophic level (Cabana and Rasmussen, 1994; Kidd et al., 1995) and correlates with trophic level in other northern fishes (e.g., Atwell et al., 1998; Gantner et al., 2010b, van der Velden et al., 2012, 2013c). Thus the switch to anadromy corresponds with a switch to feeding primarily on marine crustaceans and fish (Armstrong and Morrow, 1980; Fechhelm et al., 1997) as opposed to the benthic invertebrates that dominate in the diets of freshwater resident NDVC (Armstrong and Morrow, 1980).

Trophic position and diet are important factors in influencing THg concentrations, due primarily to the biomagnification of THg along food chains (Mathers and Johansen, 1985; Cabana et al., 1994; Kidd et al., 2012). Thus $\delta^{15}N$ driven differences in THg among life-history types were expected, as organisms with a higher trophic level are more likely to be migratory and have a larger foraging range than those with a lower trophic level (Atwell et al., 1998). Although relationships between THg and $\delta^{15}N$ have been reported for many northern fishes (e.g., Evans et al. 2005a, Gantner et al., 2009; Kidd et al., 2012), and *Salvelinus* species in particular (e.g., Gantner et al., 2010b, Swanson et al., 2011, van der Velden et al., 2013c), they are not ubiquitous (Evans et al., 2005a; van der Velden et al., 2013c) as was noted for the among- life-history type comparison of Northern Dolly Varden. Nevertheless, for both resident and anadromous Northern Dolly Varden, ontogenetic shifts in diet associated with reduced reliance on benthivory and increased piscivory and/or marine feeding are accompanied by a significant increase, 2 and 4‰ respectively, in $\delta^{15}N$.

In fresh water, Northern Dolly Varden feed primarily on drift and benthic invertebrates (Nakano and Kaeriyama, 1995; Nakano et al., 1999; Hagan and Taylor, 2001) a foraging pattern that facilitates niche partitioning by Northern Dolly Varden in the low productivity environments they typically inhabit. Although the taxonomic form of Dolly Varden was not specified, dietary use of fish eggs, fry and small forage fishes (e.g., three-spine stickleback, *Gasterosteus aculeatus*) has been reported (Bond, 2013), with size-dependent usage of such dietary resources (Denton et al., 2009) likely explaining the correlated increase in resident THg with increasing $\delta^{15}N$ observed in the Babbage River fish. In contrast, movement to marine feeding in anadromous Northern Dolly Varden is initially associated with benthic foraging (decapod crustaceans, amphipods, isopods and annelids) in the nearshore (Morton, 1982; Fechhelm et al.,

1997), with a switch to piscivory as fish grow (Armstrong and Morrow, 1980). While larger, marine-captured Southern Dolly Varden continue to consume invertebrates, fish comprise the bulk of the diet (>70%, Morton, 1982) including: sandlance, *Ammodytes tobianus*, rock sole, *Lepidopsetta bilineata*, and starry flounder *Platichthys stellatus* (Townsend, 1943; Morton, 1982).

The upward trophic shift reflected in $\delta^{15}N$, in turn, is associated with an increase in dietary THg. For example, in freshwater environments mean THg ± standard deviation of baetid mayflies (gatherers) and hepta mayflies (scrapers) from selected streams in southeastern Alaska ranged from 0.034±0.015 (µg/g dw) to 0.050±0.021 (µg/g dw), respectively (Nagorski et al., 2014). Values approximate those given by Loseto et al. (2008) (mean ± standard error) for Beaufort Sea sampled marine zooplankton (0.035±0.005 µg/g dw) and *Calanus* spp. (0.025±0.003 µg/g dw) but are lower than preferred mysids (0.081±0.009 µg/g dw), decapod crustaceans (0.316±0.061 µg/g dw) and potential epi-benthic forage fishes such as starry and Arctic flounder (*Pleuronectes glacialis*) and fourhorn sculpin (*Myoxocephalus quadricornis*) that, respectively, had mean THg levels of 0.277±0.073 (µg/g dw), 0.255±0.044 (µg/g dw) and 0.587±0.076 (µg/g dw) (Loseto et al., 2008).

While shifts to marine feeding have the effect of raising THg in anadromous NDVC, the associated significant increases in body size resulting from marine feeding have the opposite effect of reducing THg concentrations. Growth has been found to significantly negatively influence THg concentrations in a number of northern fish species, including lake whitefish, *Coregonus clupeaformis*, (Doyon et al., 1998) and Atlantic salmon, *Salmo salar*, (Dutton, 1997; Ward et al., 2010), has been advanced as an explanation for among-population differences in lake charr, *Salvelinus namaycush*, THg concentrations (Swanson and Kidd, 2010), and

suggested as a possible explanatory variable among life-history type THg variation (e.g., Evans et al., 2005b, Swanson and Kidd, 2010, and Swanson et al., 2011). Nevertheless, field evidence for the growth dilution theory is rather inconsistent. For example, Stafford and Haines (2001) and van der Velden et al. (2012) found no evidence of a growth dilution in lake and Arctic charr, respectively.

For a given age and prey intake, fish that grow to a larger size will have lower THg concentrations because they accumulate more biomass relative to THg (e.g., growth dilution *sensu* Ward et al., 2010), particularly where the growth rate is associated with high growth efficiency (Trudel and Rasmussen, 2006). Dolly Varden are noted for their digestive flexibility and efficiency, which allows them to gorge and rapidly convert and store energy in fat reserves, somatic growth and gonad development over a limited period of time, e.g., weeks (Armstrong and Bond, 2013). The coupling of high growth rate and efficiency, therefore, acts to reduce overall THg levels in anadromous fish, albeit by amounts insufficient to overcome the effect of shifting to more contaminated prey that results from anadromous feeding. Mercury at the base of the food web has significant influence on the THg levels in higher trophic species (Simoneau et al., 2005), with differential basal THg concentrations between environments, e.g., marine and fresh water, thought to explain much of the difference in THg concentrations among life-history types that selectively use different environments for feeding (van der Velden et al., 2013a). Thus, THg in top predatory fishes is likely more dominated by the amount of MeHg available for uptake at the base of the foodweb than by differences in trophic position alone, with differences among systems often being determined by the percentage of wetlands within the drainage basin (Chasar et al., 2009). Smaller, steep gradient, nival-dominated drainage systems, such as the Babbage (4200 km^2, Forbes, 1983), typically have limited wetlands, with seasonal flows often

dominated by groundwater inputs. Furthermore, Arctic rivers typically transport more than half of their annual amounts of water, sediment and associated heavy metals within the short period of time defined by the spring flooding period (Rember and Trefry, 2004), with dissolved organic carbon (DOC) being highly correlated with discharge (Holmes et al., 2008) and often positively correlated with THg (Brigham et al., 2009). Thus within the freshwater environments occupied by Northern Dolly Varden, the physical factors that favour high environmental concentrations of MeHg/THg (e.g., DOC, wetlands) are generally limited. With exposure to high THg concentration periods also being limited to the spring run-off period, the overall environmental exposure to mercury for all freshwater aquatic biota is likely limited and the net accumulations observed should be low as observed in the freshwater types of Northern Dolly Varden tested here.

In contrast, in the marine environment the large amounts of relatively labile fluvial DOC discharged during the spring freshet fuel heterotrophic microbial metabolism on the shelf and stimulate marine primary production (Holmes et al., 2008). Once discharged, the fluvial DOC is incorporated into the nearshore marine sediment pool where Hg methylation occurs (Bloom et al., 1999). The resulting use of epi-benthic prey resources that feed in or near the sediments exposes them to significantly higher levels of Hg than those feeding in the water column (Morel et al., 1998) and, in turn, mediates the higher dietary exposures found in anadromous Northern Dolly Varden. Nevertheless, while basal foodweb THg concentrations in the marine environment are higher, similar $\log_{10}$THg-δ^{15}N slopes found for freshwater and anadromous fish suggest similar bioaccumulation rates as has been noted elsewhere for freshwater/marine comparisons (Campbell et al., 2005; Rigét et al., 2007; van der Velden et al., 2013a).

Table 3.1: Sample size (*n*), mean ± standard deviation (SD) and range of fork-length (mm), weight (g), age (year), THg (ng/g wet weight), adjusted THg (ng/g ww), $\delta^{13}C$ (‰), $\delta^{15}N$ (‰), and growth rate (mm/year) for each life-history type of NDVC studied. Life-history types with biological measures not significantly different from one another at the $\alpha = 0.05$ level of significance are indicated with uppercase superscripts (e.g., A, B).

Statistic	Fork-length (mm)	Weight (g)	Age (year)	THg (ng/g ww)	Adjusted THg (ng/g ww)	$\delta^{13}C$ (‰)	$\delta^{15}N$ (‰)	Growth rate (mm/year)
<u>Isolates (n=22)</u>								
Mean±SD	170 ± 25^{A}	62 ± 29^{A}	4.9 ± 1.5^{A}	22 ± 14^{A}	37 ± 5^{A}	-33.2 ± 1.5^{A}	7.3 ± 0.8^{A}	35 ± 6^{A}
Range	137 - 249	38 - 167	4 - 9	8 - 63	18 - 58	-37.3 to -30.6	5.9 to 8.7	21 - 42
<u>Residents (n=10)</u>								
Mean±SD	191 ± 27^{A}	80 ± 30^{A}	4.1 ± 0.9^{A}	56 ± 27^{B}	76 ± 11^{B}	-31.8 ± 1.2^{B}	9.2 ± 0.7^{B}	48 ± 9^{B}
Range	152 - 229	36 - 126	3 - 6	33 - 113	40 – 108	-33.9 to -30.3	8.2 to 10.6	38 - 69
<u>Anadromous (n=30)</u>								
Mean±SD	497 ± 45^{B}	1128 ± 305^{B}	6.8 ± 1.5^{B}	108 ± 39^{C}	8 ± 0^{C}	-22.7 ± 0.6^{C}	15.0 ± 0.9^{C}	75 ± 13^{C}
Range	393 - 612	643 - 1959	4 - 10	30 - 181	8 – 9	-24.6 to -21.8	13.0 to 16.5	55 - 115

Table 3.2: Pearson's correlation coefficients for log_{10}THg, stable isotope and biological variables for anadromous Northern Dolly Varden. All correlations were significant at the $\alpha = 0.01$ level of significance except those marked with an asterisk which were significant at the $\alpha = 0.05$ level and those that are underlined which were not significant.

Variable	$\delta^{13}C$	$\delta^{15}N$	Length	Age	Growth Rate	C:N Ratio
Log_{10}THg	0.470	0.675	0.411	0.553	-0.652	-0.357
$\delta^{13}C$		0.620	0.651	0.563	-0.487	-0.753
$\delta^{15}N$			0.492	0.546	-0.569	-0.404*
Length				0.899	-0.696	-0.457
Age					-0.905	-0.370
Growth Rate						0.309

Table 3.3: Sample partial correlation coefficients and associated *P*-values for anadromous Northern Dolly Varden for log_{10}THg (ng/g ww) concentrations versus $\delta^{15}N$, age and growth rate when controlling for the other variables.

	Variables controlled for					
Correlate	$\delta^{15}N$	Age	Growth Rate	$\delta^{15}N$ Age	$\delta^{15}N$ Growth Rate	Age Growth Rate
$\delta^{15}N$		0.534	0.487			0.502
P-value		0.003	0.007			0.006
Age	0.300		-0.113		-0.178	
P-value	0.114		0.560		0.364	
Growth Rate	-0.441	-0.425		-0.378		
P-value	0.017	0.021		0.047		

Table 3.4: Estimated multiple regression model coefficients, significance values (*p*), and standardized regression coefficients (β_j) for models explaining variation in $\log_{10}$THg (ng/g ww) as a function of biological and ecological factors for all life-history types combined and anadromous Northern Dolly Varden.

	Regression coefficient	*P*-value	Standardized regression coefficient (β_j)
	All life-history types		
	$r^2 = 0.851$	$F_{(3,51)} = 97.378$	
Intercept	16.968	< 0.001	
Life-history type	- 0.158	< 0.001	- 0.292
$\delta^{15}N$	0.107	< 0.001	1.063
Growth rate	- 0.006	< 0.001	- 0.363
	Anadromous Northern Dolly Varden		
	$r^2 = 0.561$	$F_{(2,27)} = 17.254$	
Intercept	0.996	0.117	
$\delta^{15}N$	0.096	0.007	0.450
Growth Rate	- 0.006	0.017	- 0.396

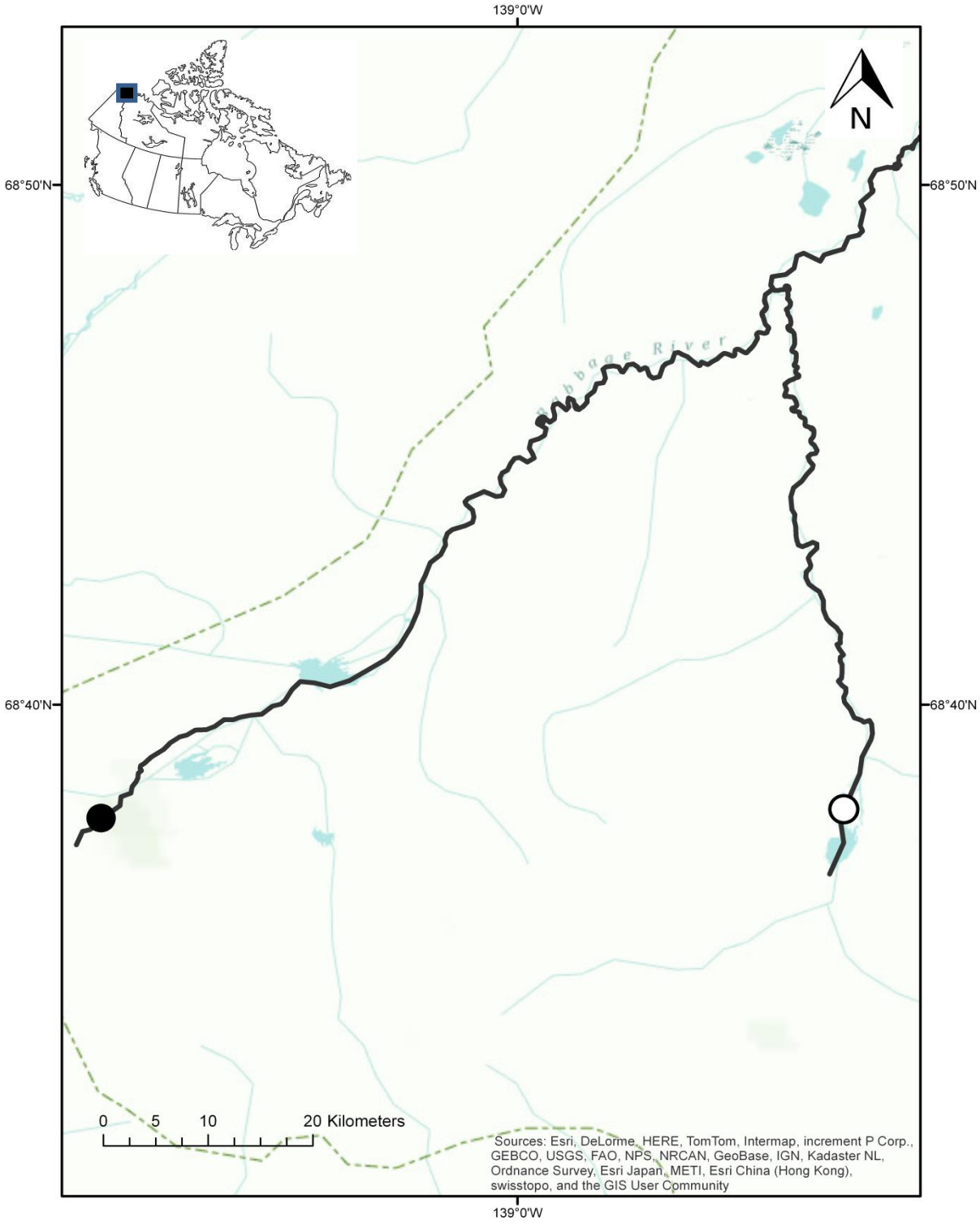


Figure 3.1: Map locations of isolate (black circle), resident (white circle) and anadromous (white circle) Northern Dolly Varden sampling sites along the Babbage River, Yukon Territory, Canada.

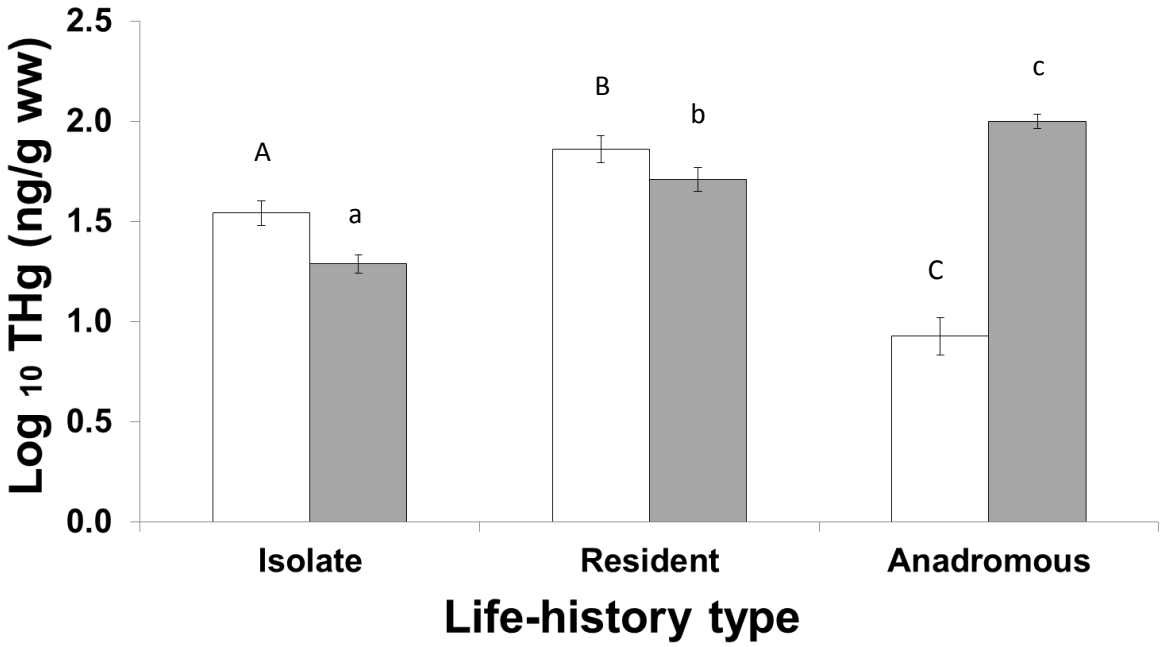


Figure 3.2: Age-4 LSM log_{10}THg (ng/g ww) ± standard error at a standardized fork-length of 218 mm (white bars) and unadjusted log_{10}THg (ng/g ww) ± standard error (gray bars) of Northern Dolly Varden life-history types. Capitalized letters indicate significant differences in adjusted THg concentrations and lower case letters indicate significant differences in unadjusted THg concentrations among life-history types.

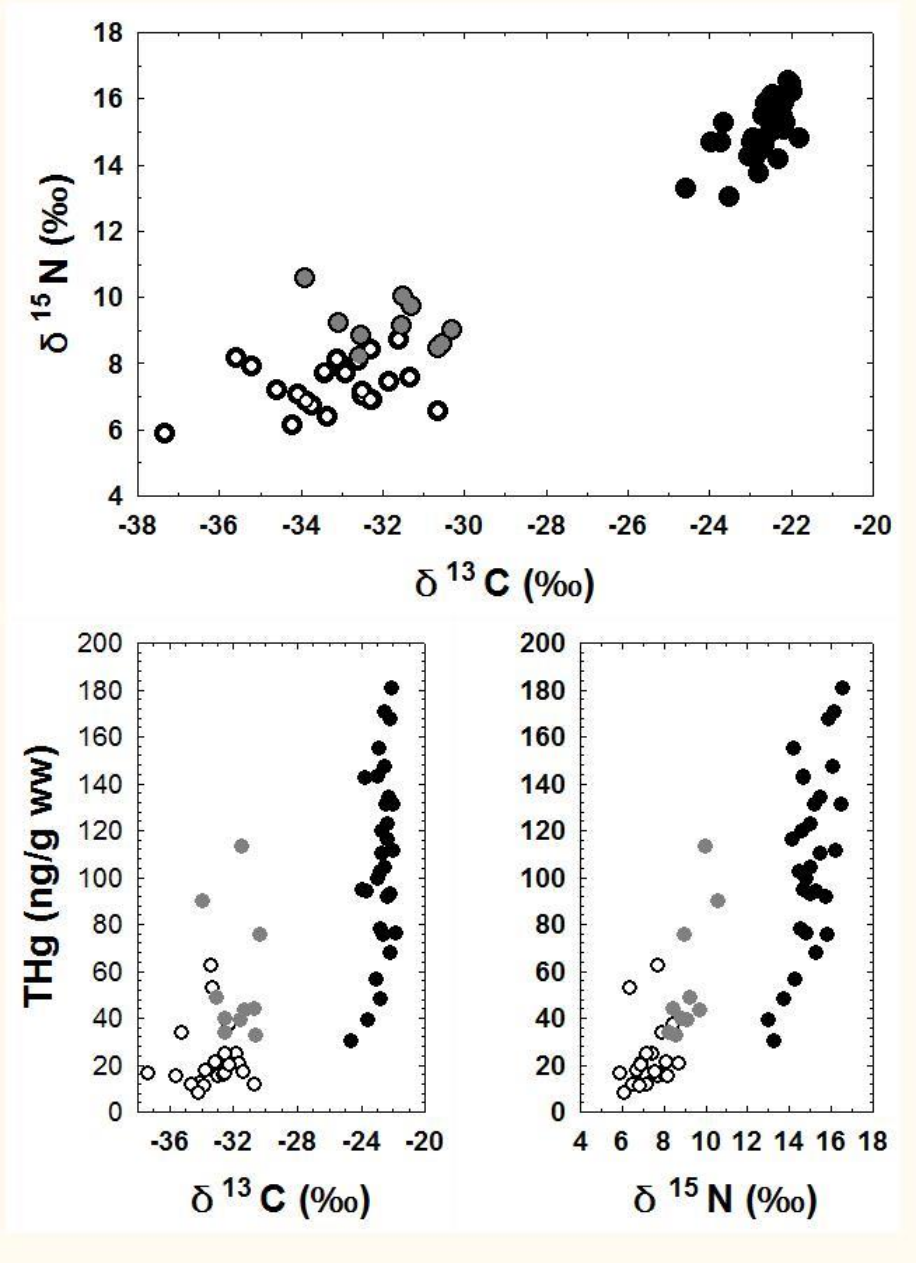


Figure 3.3: Individual isolate (white), resident (gray) and anadromous (black) Northern Dolly Varden stable isotope and THg bi-plots. Means of all life-history type stable isotope and THg values differ significantly ($p < 0.05$) and increase progressively from isolates through residents to anadromous fish.

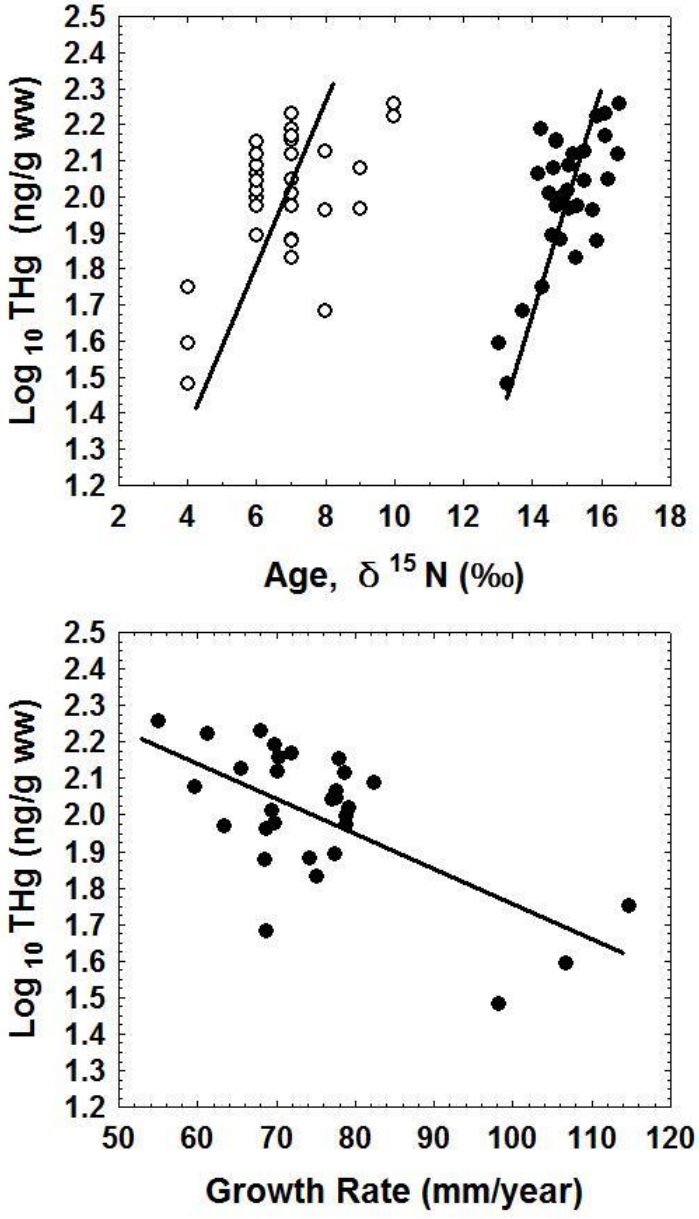


Figure 3.4: Plots of $\log_{10}$THg versus δ^{15}N, age and growth rate for anadromous Northern Dolly Varden. All correlations are significant ($p \leq 0.024$). Solid line plot estimated regression models.

Chapter 4: Comparison of contemporary and historical total mercury concentrations in Northern Dolly Varden from the northwestern Canadian Arctic

Introduction

As a result of long range atmospheric transport, high mercury (Hg) concentrations have been found in the remote, pristine aquatic ecosystems of the Arctic (Fitzgerald and Clarkson, 1991; Schroeder et al., 1998). The toxic form, methylmercury (MeHg), can bioaccumulate and biomagnify up food chains, resulting in harmful impacts to aquatic biota as well as the health of humans who harvest the biota (Morel et al., 1998; Braune et al., 1999). For example, aquatic biota MeHg levels are of particular concern to Inuit communities (e.g., Inuvialuit and Gwich'in) who consume large quantities of fish, such as Northern Dolly Varden (*Salvelinus malma malma*) for subsistence and cultural purposes (Papik et al., 2003; Gallagher et al., 2011).

Climate- and development-related impacts (e.g., global warming and oil and gas development) on the global Hg cycle are hypothesized to lead to higher Hg levels in Arctic biota (Reist et al., 2006a; Stern et al., 2012). Climate-induced changes to the Hg cycle may include increased Hg methylation rates due to increased water temperatures (Bodaly et al., 1993), longer ice-free periods as a result of changing thawing and freezing phenologies (Stern et al., 2012), increased direct deposition of Hg to the ocean surface and Hg re-emission as a result of reduced ice cover (Stern et al., 2012), and increased Hg deposition from increased precipitation (Macdonald et al., 2005 and references therein).

From 1961 to 2000, higher atmospheric temperatures were witnessed in the Arctic, with a greater warming evident in the western Canadian Arctic compared to the eastern Canadian Arctic (Macdonald et al., 2005). Since 2000, temperatures have continued to rise with the summers of 2005, 2007, 2010 and 2011 having been the warmest of all prior years back to 1400 and the summer of 2010 in all likelihood having been the warmest in the previous 600 years in the

Canadian Arctic (Tingley and Huybers, 2013). Temperatures are also predicted to continue increasing in the Arctic (Meier et al., 2014).

For Northern Dolly Varden, a charr species distributed north of the Alaska Peninsula in the Bering, Chukchi, and Beaufort sea drainages east to the Mackenzie River (Reist et al., 2002), temperature increases may decrease growth rates and increase metabolic stress (Reist et al., 2006b) leading to increased tissue Hg concentrations (Simoneau et al., 2005; Ward et al., 2010; Stern et al., 2012). Warmer waters may also alter Northern Dolly Varden food availability, geographic distribution (Reist et al., 2006c), and life-history (Reist et al., 2006b), all of which are known to control Hg tissue concentrations in aquatic biota (Loseto et al., 2008; Swanson and Kidd, 2010). Lastly, development activities (e.g., pipe-line installation and water withdrawals) could impede access to feeding and over-wintering areas (Mochnacz et al., 2010), leading to increased stress and starvation likely to be associated with increased Hg levels in fish.

Previous temporal studies of total mercury concentrations (THg) in northern Canadian fish, such as Arctic charr (*Salvelinus alpinus*) (Lockhart et al., 2005; Muir et al., 2005; Gantner et al., 2009; van der Velden et al., 2013b), lake charr (*Salvelinus namaycush*) (Evans et al., 2005a; Lockhart et al., 2005), northern pike (*Esox lucius*) (Evans et al., 2005a; Lockhart et al., 2005), broad whitefish (*Coregonus nasus*) (Evans et al., 2005a), walleye (*Sander vitreus*) (Braune et al., 1999; Evans et al., 2005a; Lockhart et al., 2005), and burbot (*Lota lota*) (Lockhart et al., 2005), have reported no significant broad-scale trends in THg over time, leading to the conclusion that regional changes in climate-driven factors have so far had limited impacts on mercury exposure in many northern fishes (van der Velden et al., 2013b). Nevertheless Carrie et al. (2010) reported an almost two fold increase in THg from 1985 to 2008 in burbot (*Lota lota*) from the Mackenzie River, Canada, despite consistent atmospheric deposition trends.

Notably absent from the list of fish species for which temporal THg change studies have been completed is Northern Dolly Varden. Given both the importance of Northern Dolly Varden as a consumption staple and the environmental changes occurring in the northwestern Canadian Arctic, documentation of existing trends in THg accumulation patterns in Northern Dolly Varden is critical. Accordingly, the purpose of this study was to investigate changes in THg from Northern Dolly Varden populations having an approximate 3 decade span of sampling (1986-2013) and to test the hypotheses that: [1] similar to other studied northern fish species (e.g., Lockhart et al., 2005; Muir et al., 2005; Gantner et al., 2009; van der Velden et al., 2013b), significant changes in THg have not occurred in Northern Dolly Varden since the mid-1980s; [2] the relationships of THg with size, $\delta^{13}C$ and $\delta^{15}N$ have remained consistent through time; and, [3] because of the predicted temporal trends in THg postulated in hypothesis [1], there is no association between THg and observed increases in mean annual temperature from the 1980's to the present.

Methods

Sample Collection

Archived muscle tissue samples of Northern Dolly Varden were obtained from the Department of Fisheries and Oceans Canada, Winnipeg, Manitoba for populations that had been repeat sampled over time. Only two populations provided sufficient samples for temporal testing, anadromous Northern Dolly Varden (*n* = 100) from the Rat River, Northwest Territories, (67° 46' 48"N, 136° 19' 12"W) sampled in 1986, 1988 and 2011, 2013 and anadromous Northern Dolly Varden (*n* = 54) from the Firth River, Yukon Territory, (68° 40' 12"N, 140° 55' 12"W) sampled in 1986, 1988 and 2011, 2012 (Fig. 4.1). Only anadromous fish identified on the basis

of size, morphology and colouration (Reist, personal communication) were sampled, with restricted sampling removing the effects of life-history (Tran et al, in review). Where available supplemental biological data, including: fork-length (mm), weight (g), sex, and age (year) for each sample were also obtained from Fisheries and Oceans Canada.

Stable isotope analyses

All muscle samples were measured for stable isotopes of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$). The samples were dried at 50°C for 48 h and pulverized to an homogenate with a Retsch MM 301 ball mill grinder (F. Kurt Retsch GmbH Co., Haan, Germany) and weighed on a Mettler Ultra micro balance (UMX2, Mettler-Toledo GmbH, Greifensee, Switzerland). Approximately 0.300 mg of prepared material was used for stable isotope analyses (SIA) completed with 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) at the Environmental Isotope Laboratory, University of Waterloo (Waterloo, Ontario). Machine analytical precision, respectively, for $\delta^{13}C$ and $\delta^{15}N$ was ±0.1% and ±0.2% and was determined by repeat analysis of laboratory working standards cross-calibrated to International Atomic Energy Agency standards CH6 for $\delta^{13}C$ and N1 and N2 for $\delta^{15}N$. All resulting measurements were expressed using standard delta notation (δ) as parts per thousand differences (‰) with respect to the international reference standards, Vienna Peedee Belemnite for $\delta^{13}C$ (Craig, 1957) and nitrogen gas in the atmosphere for $\delta^{15}N$ (Mariotti, 1983).

Total mercury analyses

Total mercury concentrations for a sub-sample (0.1-0.2g) of frozen, non-homogenized Northern Dolly Varden tissue were measured on a Milestone Direct Mercury Analyzer (DMA-80) using thermal decomposition followed by atomic absorption spectroscopy following U.S. EPA method 7473 (U.S. Environmental Protection Agency, 2007), with results expressed as ng/g wet weight (ww). Standard reference materials (SRMs) were run at the beginning and end of every batch of 20 samples, with no less than 5 blanks run in each sample batch. The method detection limit determined as 3x the standard deviation of the blanks was 0.07 ng Hg (approximately 0.5 ng/g ww). The certified reference materials and percent recoveries (mean percentage of certified value ± standard deviation) used included: National Institute of Standards and Technology (NIST) 1566B, oyster tissue, (95.6±1.3%), NIST 2976, mussel tissue, (103.9±5.4%), NIST 2974a, freeze dried mussel tissue, (101.8±0.7%), and National Research Council of Canada (NRC) DORM-3, fish protein, (111.1±8.2%). In all analyses, a single sample was run in triplicate in each sample batch and the mean relative standard deviation of the triplicates was measured as 11.4%.

Methylmercury analyses

To determine whether the measurement of THg was an appropriate estimate of MeHg, random sub-samples (n = 20; same samples used in Chapter 2 and 3) were measured for MeHg. Muscle tissue samples were freeze dried for 48 hours and then ground using an acid washed glass mortar and pestle and placed into acid washed vials. Samples were weighed (±0.1 mg) before and after lyophilisation and the percent moisture was computed for each sample to facilitate conversion of wet weight (ww) and dry weight (dw) concentrations of THg and MeHg. Concentrations of MeHg and inorganic Hg (II) were determined at Quicksilver Scientific

(Lafayette, CO, USA) using method QS-LC-CVAF-001, matrix spikes and certified reference materials as quality control measures. The limit of detection (LOD) and limit of quantification (LOQ) were respectively, 2.0 ng/g and 5.0 ng/g for both MeHg and Hg (II) in analyzed tissue samples.

Meteorological data

Meteorological data consisting of monthly mean temperatures for the period 1980-2013 were obtained from the Environment Canada National Climate Data and Information Archive (www.climate.weatheroffice.gc.ca), and supplemented with compiled airport weather station data (www.tutiempo.net/en), for weather stations proximate to each study river. Station data obtained included Old Crow, Yukon (67°34'14.00" N, 139°50'21.00" W), located 112km from the Firth River and Aklavik, NWT (68°13'24.00" N, 135°00'21.00" W), located 145km from the Rat River. Obtained data were used to compute mean summer (May to September) seasonal averages that were regressed against year to determine whether significant increases in temperature had occurred over the study period.

Statistical analyses

All statistical analyses were performed using the statistical program SPSS (SPSS Inc., 2008) with Type I error set to $\alpha = 0.05$. All data were tested for normality either with the use of residual plots or the Shapiro-Wilk statistic as appropriate (Swanson et al., 2011). THg data were $\log_{10}$-transformed when residual plots did not illustrate variance homogeneity or the Shapiro-Wilk statistic indicated non-normality (Swanson et al., 2011).

River-specific log[THg] versus length relationships were investigated using linear regression, with multiple models compared using analysis of covariance (ANCOVA). Data were compared between time periods 1986-1988 and 2011-2013, to increase the sample sizes in both rivers, using a two-sample t-test appropriately adjusted for variance homogeneity (Zar, 2010). Linear mixed-effects models were used to evaluate the effect of time period (1986–88 or 2011–13) on fish mercury concentration and fork-length among sample sites. Mixed-effects models were further used to explain variation in individual log[THg] using time period, fork-length, and available stable isotope data. Compliance with model assumptions was verified using diagnostic plots (e.g., fitted versus residuals, normal Q-Q plots of residuals and estimated random effects; Pinheiro and Bates, 2000; Zar, 2010).

Results

Rat River mean THg ranged from 79 ng/g ww in 1986-1988 to 109 ng/g ww in 2011-2013, while in the Firth River, THg ranged from 126 ng/g ww in 1986-1988 to 178 ng/g ww in 2011-2012. Mean THg, biological data, and $\delta^{15}N$ and $\delta^{13}C$ values in Northern Dolly Varden from each year in the Rat and Firth rivers are shown in Table 4.1. From $n = 20$ samples, the average percentage of MeHg was 95 ± 3.4% (mean ± standard deviation).

A linear mixed-effects model estimated from all available data with site as a random effect found that sampling period had a significant effect on log[THg] ($F_{(1,151)} = 22.197$, $p < 0.001$) as did sampling site ($F_{(1,151)} = 42.698$, $p < 0.001$). After fork-length, $\delta^{13}C$, and $\delta^{15}N$ were included as covariates, the model yielded an estimate of log[THg] as 1.86 ± 0.49 ng/g ww (mean ± standard error) for the historical period and 1.99 ± 0.53 ng/g ww for the contemporary period (Fig. 4.2), with time period ($F_{(1,144)} = 6.552$, $p = 0.012$), fork-length ($F_{(1,144)} = 30.955$, $p < 0.000$),

$\delta^{13}C$ ($F_{(1,144)}$ = 10.561, p = 0.001), and $\delta^{15}N$ ($F_{(1,144)}$ = 6.188, p = 0.014) having a significant effect on log[THg]. No significant effect of sampling site was found (p = 0.784). A mixed-effects model also determined a significant effect of sampling period on fork-length ($F_{(1,147)}$ = 43.376, p < 0.001), $\delta^{13}C$ ($F_{(1,151)}$ = 190.031, p < 0.001), and $\delta^{15}N$ ($F_{(1,151)}$ = 44.040, p < 0.001) of Northern Dolly Varden.

Firth River

Mean (± standard deviation) THg in 1986-1988 (126 ± 45 ng/g ww) was significantly different from THg in 2011-2012 (178 ± 47 ng/g ww) (Table 4.2). Northern Dolly Varden fork-length and trophic position in 1986-1988, which averaged (± standard deviation) 536.9 ± 73.2 mm and 15.1 ± 0.7 ‰, were not statistically different from the mean values in 2011-2012, which averaged 578.3 ± 75.7 mm and 15.1 ± 0.5 ‰ (Table 4.2). Carbon sources, on the other hand, significantly decreased from 1986-1988 (-22.7 ± 0.8 ‰) to 2011-2012 (-24.4 ± 0.6 ‰) (Table 4.2).

Between periods, a positive significant log[THg] relationship with fork-length was found for 1986-1988 ($F_{(1,39)}$ = 44.686, p < 0.001, r^2 = 0.534), but not for 2011-2012 (p = 0.755) (Fig. 4.3a). Both time periods exhibited different slopes (significant interaction for time period * fork-length, p = 0.003) and intercepts (p < 0.001) (Fig. 4.3a).

Trophic position and carbon sources significantly increased with log[THg] in 1986-1988 ($\delta^{15}N$, $F_{(1,42)}$ = 17.155, p < 0.001, r^2 = 0.290; $\delta^{13}C$, $F_{(1,42)}$ = 19.663, p < 0.000, r^2 = 0.319), while no significant relationships were found in 2011-2012 (p > 0.05 for both $\delta^{13}C$ and $\delta^{15}N$) (Fig. 4.4a,b). Log[THg] versus $\delta^{13}C$ relationships illustrated significantly different slopes between

time periods (interaction for time period * $\delta^{13}C$, $p = 0.013$) and significantly different intercepts ($p = 0.018$), while log[THg] versus $\delta^{15}N$ relationships illustrated the same slopes (no interaction for time period * $\delta^{15}N$, $p = 0.262$) and intercepts ($p = 0.771$) (Fig. 4.4a,b).

Rat River

Mean (± standard deviation) THg in 1986-1988 and 2011-2013 were 79 ± 42 ng/g ww and 109 ± 44 ng/g ww, respectively. Significantly higher THg values were found in 2011-2012 compared to 1986-1988 (Table 4.2). Fork-length and trophic position significantly increased from 452.2 ± 64.4 mm and 13.7 ± 0.7 ‰ in 1986-1988 to 538.8 ± 50.2 mm and 15.0 ± 0.9 ‰ in 2011-2013 (Table 4.2). Similar to the Firth River, carbon sources decreased over time from -23.6 ± 0.9 ‰ in 1986-1988 to -25.9 ± 1.0 ‰ in 2011-2013 (Table 4.2).

Significant log[THg]-length relationships were found for both the historical and contemporary periods (1986-1988, $F_{(1,58)} = 37.722$, $p < 0.000$, $r^2 = 0.394$; 2011-2013, $F_{(1,38)} = 15.052$, $p < 0.000$, $r^2 = 0.284$) (Fig. 4.3b). Both time periods exhibited the same slopes (no interaction for time period * fork-length, $p = 0.461$) and different intercepts ($p < 0.001$) (Fig. 4.3b).

A significant increasing relationship between log[THg] and $\delta^{13}C$ occurred only in 2011-2013 ($F_{(1,38)} = 16.610$, $p < 0.001$, $r^2 = 0.304$; 1986-1988, $p = 0.091$) while a significant positive relationship between log[THg] and $\delta^{15}N$ occurred in 1986-1988 ($F_{(1,58)} = 34.729$, $p < 0.001$, $r^2 = 0.375$) as well as 2011-2013 ($F_{(1,38)} = 8.003$, $p = 0.007$, $r^2 = 0.174$) (Fig. 4.4c,d). Log[THg] versus $\delta^{13}C$ relationships illustrated the same slopes between periods (no interaction for time period * $\delta^{13}C$, $p = 0.423$) and different intercepts ($p < 0.001$) while log[THg] versus $\delta^{15}N$

relationships illustrated significantly different slopes (interaction for time period * $\delta^{15}N$, p = 0.011) and similar intercepts (p = 0.796) (Fig. 4.4c,d).

Temperature

Over the 34 year period, large fluctuations in mean summer temperature were observed at both the Old Crow (Firth River) and Aklavik (Rat River) weather stations, with means ranging from 7.2 to 11.4°C at Old Crow and 5.8 to 11.3°C at Aklavik (Fig. 4.5). Significant warming trends were also observed at both weather stations between 1980 and 2013 (all linear regression $p < 0.001$), with slope estimates suggesting annual average increases in mean summer temperatures ranging from 0.065 °C/year (Old Crow) to 0.068 °C/year (Aklavik). Similar warming trends were found using data between periods (Rat River, $F_{(1,4)} = 7.820$, $p = 0.049$, $r^2 = 0.662$; Firth River, $F_{(1,3)} = 23.807$, $p = 0.016$, $r^2 = 0.888$).

Discussion

Unadjusted THg revealed a significant increase from 1986-1988 to 2011-2013 for fish from both the Rat and Firth rivers. Models including fork-length, $\delta^{13}C$, and $\delta^{15}N$, similarly indicated a significant increase over the study period. Relationships between log[THg] versus fork-length and log[THg] versus $\delta^{13}C$ have remained constant over time in the Rat River, but not in the Firth River, while relationships between log[THg] versus $\delta^{15}N$ have remained constant in the Firth River, but not in the Rat River. Lastly, the increase in mean temperatures from 1986-1988 to 2011-2013 paralleled the increasing trends in THg over time.

Mean historical THg concentrations from the Rat River (1986: 74 ng/g ww, 1988: 83 ng/g ww) were found to be lower than other studied fish from a similar location and time (lake trout: 110-530 ng/g, Lockhart et al., 2005; northern pike: 110-200 ng/g, Lockhart et al., 2005; walleye: 180-460 ng/g, Lockhart et al., 2005; burbot: 110-300 ng/g, Lockhart et al., 2005). However, Rat River NDVC THg concentrations were comparable to lake whitefish THg (40-110 ng/g, Lockhart et al., 2005). Similarly, mean contemporary THg concentrations from the Rat River (2011: 108 ng/g ww, 2013: 109 ng/g ww) were also found to be lower than lake trout (170-220 ng/g, Evans et al., 2013), burbot (120-150 ng/g, Evans et al., 2013), and northern pike (200-230 ng/g, Evans et al., 2013).

The modelled temporal increase suggests an approximate 7% increase in THg that contrasts with reported findings elsewhere, where temporal studies have tended to conclude that there has been no consistent change in THg concentrations in fish over time (e.g., Lockhart et al., 2005, Muir et al., 2005, Gantner et al., 2009, Rigét et al., 2011, Kirk et al., 2012, van der Velden et al., 2013b, and Sturluddotir et al., 2014). Fish mercury concentrations are generally thought to be positively related to atmospheric mercury deposition (Hammerschmidt and Fitzgerald, 2006). Although stable or decreasing global atmospheric mercury concentrations have been reported since the 1970's in the Canadian Arctic (Steffen et al., 2005), the THg response of any fish population to changing mercury inputs will be site-specific, depending on a variety of physical, chemical, and biological factors (Munthe et al., 2007). Thus in spatially proximate studies of temporal trends in a single species, contrasting trends in THg levels have been reported that depend on site-specific factors, e.g., population age and size structure, food web structure, fish community (van der Velden et al., 2013b). Furthermore within the region of this study, an increasing trend in mercury concentrations in Mackenzie River burbot has been reported over a

comparable (1985-2008) period of time associated with warming temperatures and reduced ice cover that may have led to increased THg exposure (Carrie et al., 2010).

Oceanic summer feeding by Northern Dolly Varden may also be influencing the temporal trends observed here. Dolly Varden are known to feed in the highly productive nearshore zone (Sandstrom et al., 2009), but have been reported well offshore (DeCicco, 1992, 1997) and may be migrating for feeding to waters characterized by higher prey abundance that enable greater bioenergetic efficiency (Fechhelm et al., 1997). With increasing loss of sea ice and the warming of oceanic environments associated with sea-ice loss (Macdonald et al., 2005), the expanse of open-water feeding areas will have increased as will the spatial extent of areas with large inventories of Hg^{II} in the upper water column that could result in increased methylation rates (Outridge et al., 2008). Further enhancing the opportunity for MeHg uptake has been the documented increase in freshwater associated discharge of Hg to the nearshore marine environments (Leitch et al., 2007), with the result that climate-driven increases in discharge of the Mackenzie River during the spring have been associated increased levels of mercury in higher trophic-level biota in the Mackenzie Delta and Beaufort Sea areas of the western Arctic Ocean post 2005 (Leitch et al., 2007). Given the importance of the Mackenzie River as a source of Hg for the Beaufort Sea, the proximity of the study populations to the river and the spatial movement of marine feeding Dolly Varden, it is probable that temporal trends in Dolly Varden THg from the Rat and the Firth rivers have been similarly influenced as reported in the data from this study.

While increasing temperatures were correlated with observed increases in Dolly Varden THg, temperature itself is unlikely to have much direct effect on tissue THg values given the importance of the marine feeding phase for growth. While increases in temperature may be

associated decreased growth and increased metabolic stress (Reist et al., 2006b) during the freshwater residency portion of the life-history that have the potential to increase tissue THg, the impact of changes in the marine environment probably more than outweighs those in the fresh water.

While an overall trend in increasing THg was observed and can be ascribed to large-scale drivers of ecosystem change (e.g., sea-ice loss, changes in river discharge) associated with climate change as noted above, the influence of local scale factors was clearly evident in the differential pattern of change in THg-fork-length, THg-δ^{13}C and THg-δ^{15}N relationships in the two rivers. Thus, while regional drivers (e.g., mean annual temperature, atmospheric mercury deposition) may dominate overall THg trends, site-specific factors (e.g., catchment area) will have some importance for determining THg (e.g., Rose et al., 1999, Macdonald et al., 2005). Accordingly, continuous monitoring of populations is important if we are to hope to fully understand the factors that affect variations in THg in key northern fishes, both in time and space.

Table 4.1: Sample size (*n*), mean ± standard deviation and range of fork-length (mm), age (year), THg (ng/g ww), $\delta^{13}C$ (‰), and $\delta^{15}N$ (‰) of Northern Dolly Varden for each period in the Rat and Firth rivers.

River	Time period	*n*	Statistic	Fork-length (mm)	Age (year)	THg (ng/g ww)	$\delta^{13}C$ (‰)	$\delta^{15}N$ (‰)
Rat River	1986-1988	60	Mean ± Stdev	452 ± 64	7.7 ± 1.3	79 ± 42	-23.6 ± 0.9	13.7 ± 0.7
			Range	295-619	5-11	23-174	-25.6 to -21.8	12.1-15.3
	2011-2013	40	Mean ± Stdev	539 ± 50	7.5 ± 1.8	109 ± 44	-25.9 ± 1.0	15.0 ± 0.9
			Range	435-641	5-13	38-214	-27.7 to -24.2	13.0-16.4
Firth River	1986-1988	44	Mean ± Stdev	537 ± 73	10.4 ± 2.0	126 ± 45	-22.7 ± 0.8	15.1 ± 0.7
			Range	391-681	5-15	33-254	-25.1 to -20.8	13.5-16.2
	2011-2012	10	Mean ± Stdev	578 ± 76	8.5 ± 1.8	178 ± 47	-24.4 ± 0.6	15.1 ± 0.5
			Range	500-710	6-11	112-265	-25.4 to -23.5	14.5-16.0

Table 4.2: Comparisons of mean THg (ng/g ww), fork-length (mm), $\delta^{13}C$ (‰), and $\delta^{15}N$ (‰) for the 1986-1988 and 2011-2013 periods determined from a two-sample t-test.

Site	Variable	*t*	*df*	*p*
Rat River	THg	-3.458	98	0.001
	Fork-length	-7.177	98	0.000
	$\delta^{13}C$	12.057	98	0.000
	$\delta^{15}N$	-7.774	98	0.000
Firth River	THg	-3.282	52	0.002
	Fork-length	-1.530	48	0.133
	$\delta^{13}C$	6.389	52	0.000
	$\delta^{15}N$	0.108	52	0.915

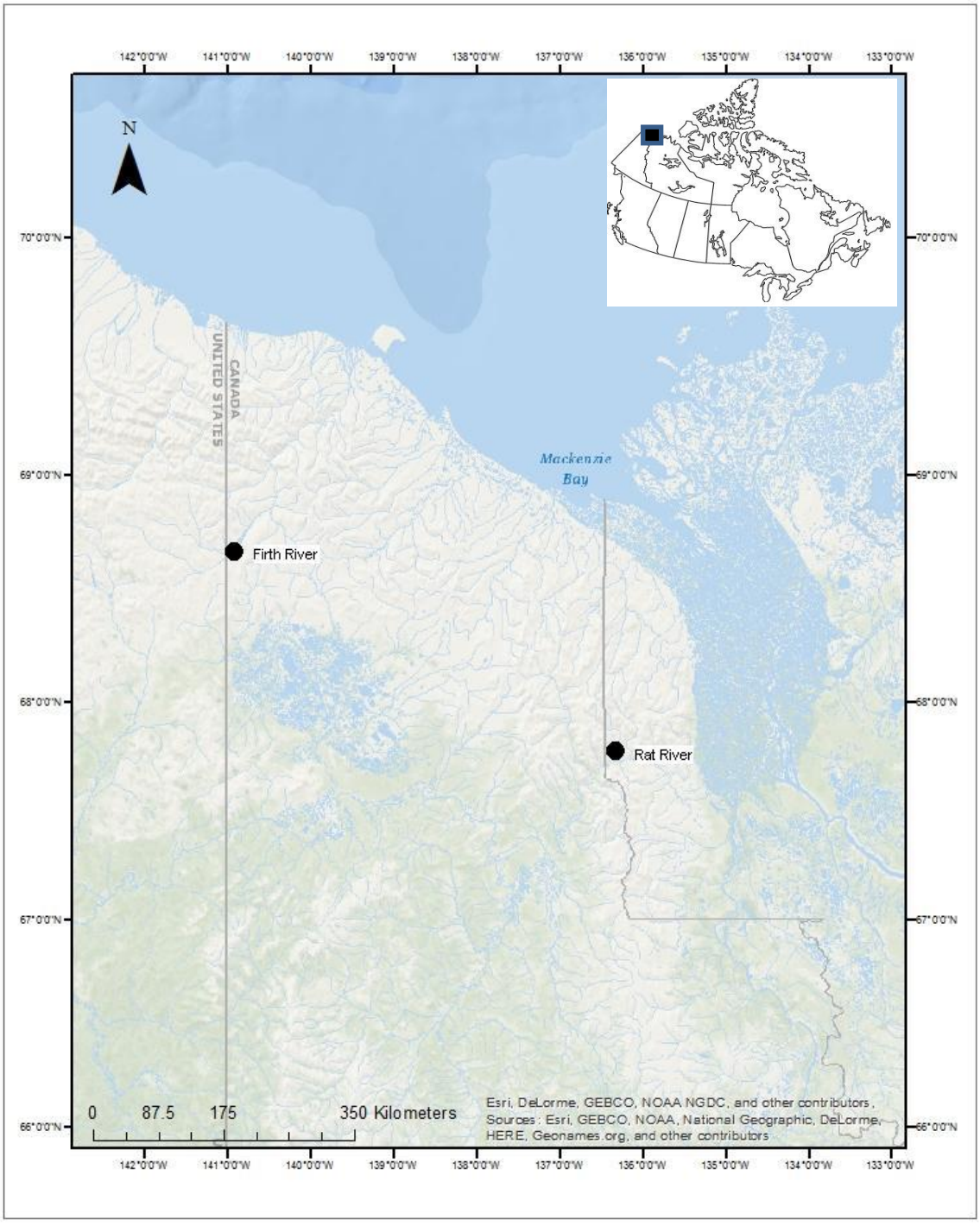


Figure 4.1: Map of sampling sites in the Rat River, Northwest Territory and the Firth River, Yukon Territory

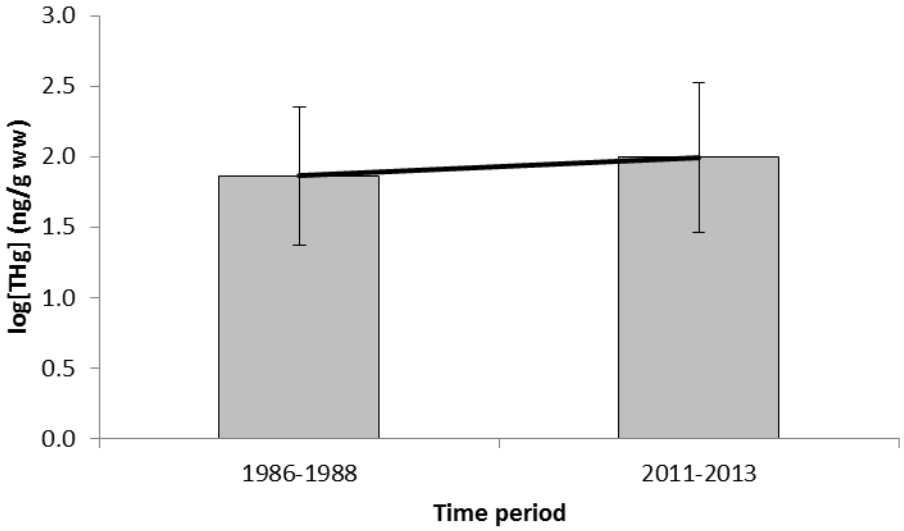


Figure 4.2: Log[THg] (ng/g ww) ± standard error versus time period estimated from the mixed model.

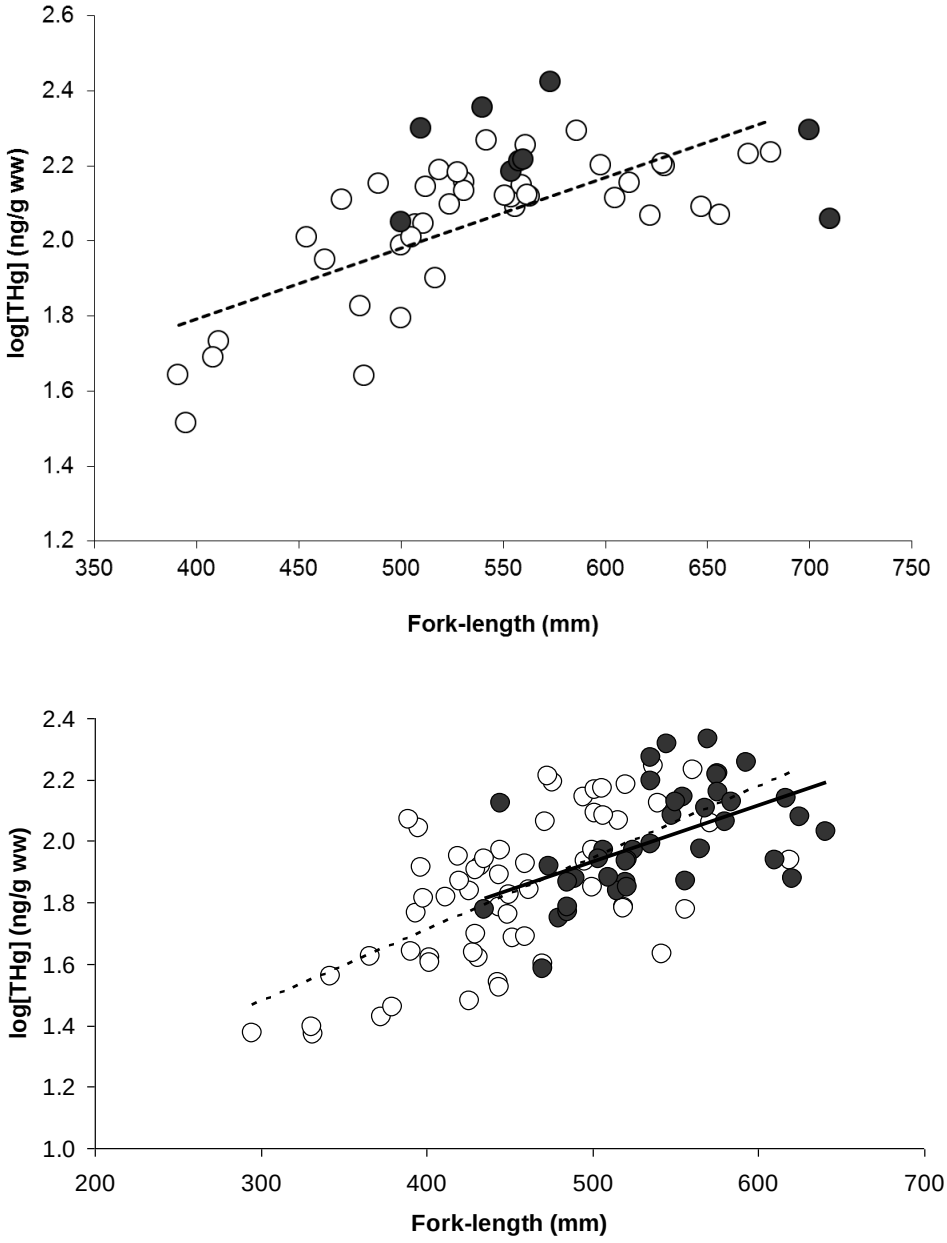


Figure 4.3: Log[THg] (ng/g ww) versus fork-length (mm) relationships for the 1986-1988 (white circles) and 2011-2012 (black circles) time periods for the (a) Firth River and the 1986-1988 (white circles) and 2011-2013 (black circles) time periods for the (b) Rat River. A dotted trend line indicates a significant relationship for the historical period and a solid black trend line indicates a significant relationship for the contemporary period.

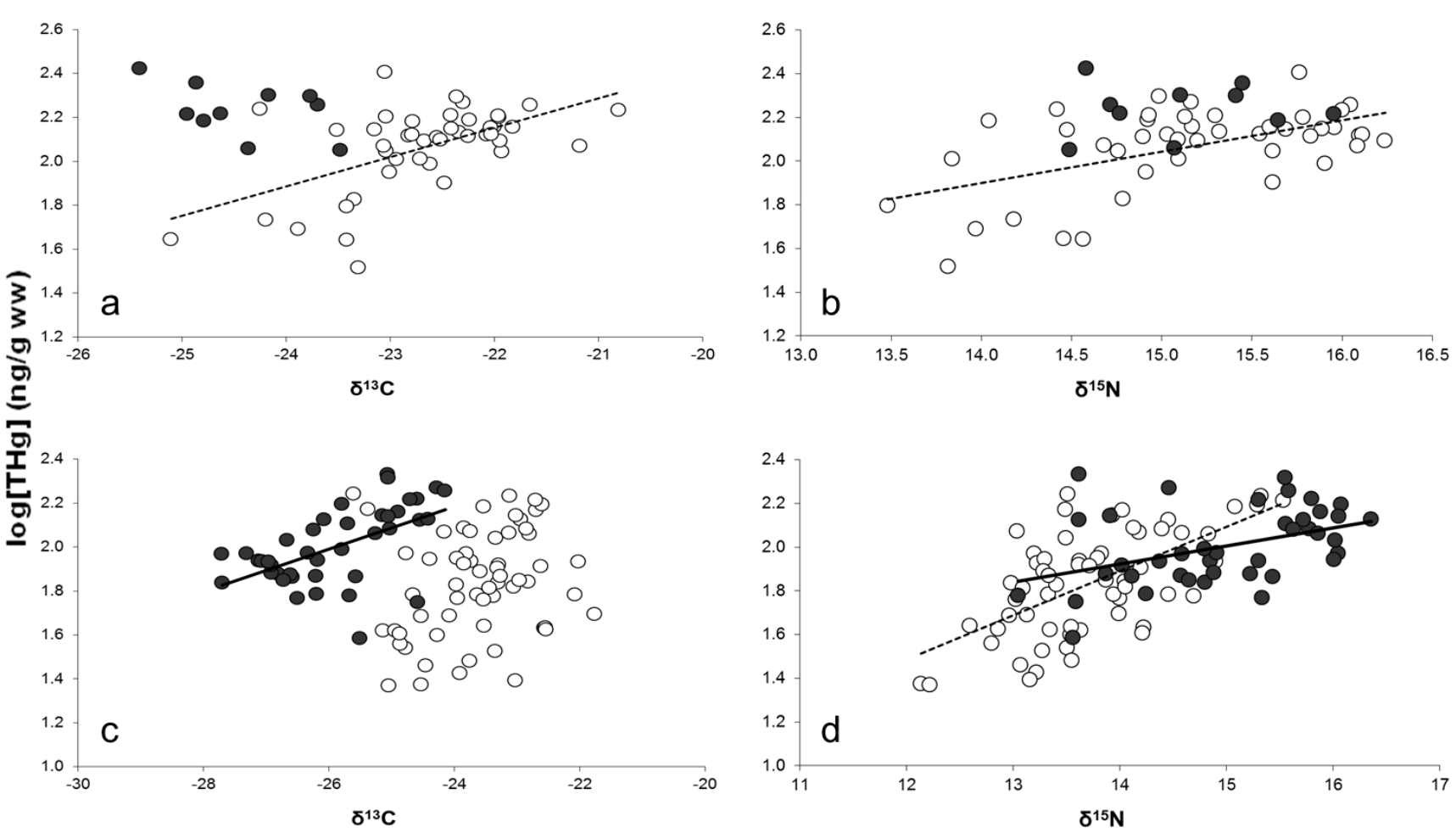


Figure 4.4: Log[THg] (ng/g ww) versus $\delta^{13}C$ (‰) for the historical and contemporary time periods for the (a) Firth River and (c) Rat River. Log[THg] (ng/g ww) versus $\delta^{15}N$ (‰) between periods for the (b) Firth River and (d) Rat River. White circles represent the historical period and black circles represent the contemporary period. A dotted trend line indicates a significant relationship for the historical period and a solid black trend line indicates a significant relationship for the contemporary period.

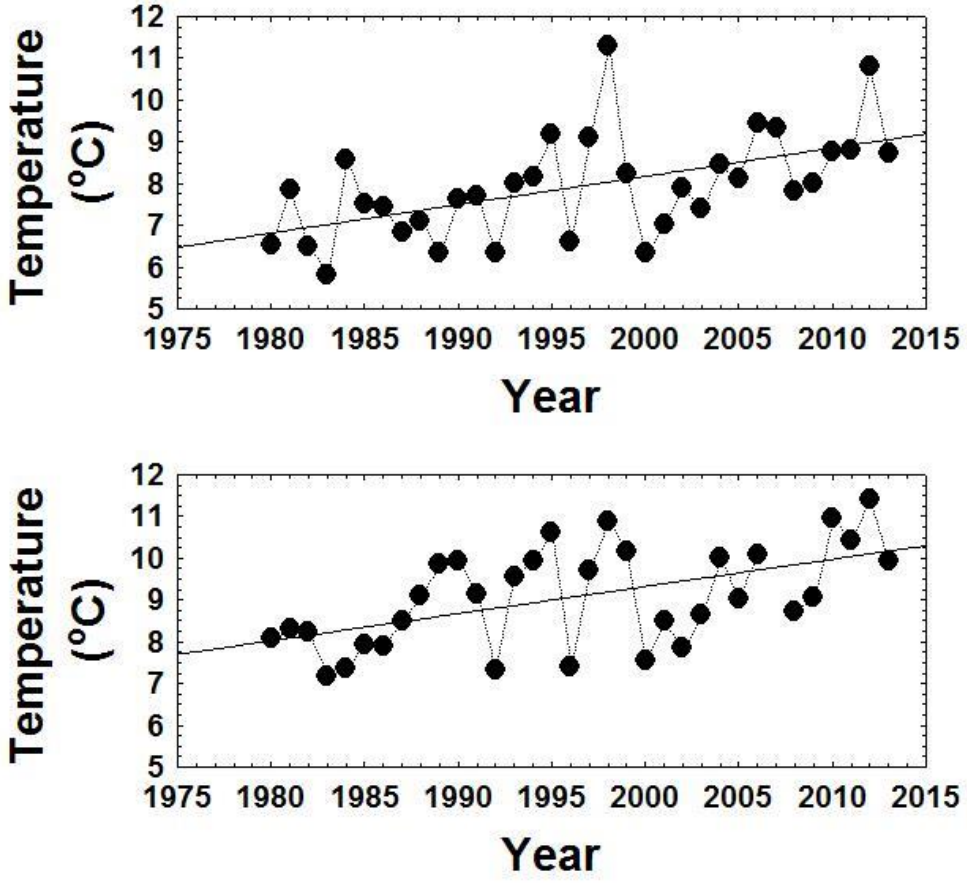


Figure 4.5: Plots of mean summer (May to September inclusive) temperatures (°C) for Aklavik, top panel, and Old Crow, bottom panel, weather stations located proximate to the Rat and Firth rivers, respectively. Lines plot the significant regression ($p < 0.001$) of mean summer temperatures versus time.

Chapter 5: Conclusions and Future Directions

Summary

The research presented in this thesis addressed several knowledge gaps concerning the mercury contamination levels present in Northern Dolly Varden. Chapter 2 reported on the spatial variability in Northern Dolly Varden THg tissue concentrations. In the time period for which samples were tested (1988–1991), observed mean THg levels in Northern Dolly Varden from all sites were well below the human consumption guidelines (0.5 ppm ww) (Health Canada, 2008), with no readily apparent patterns evident in the among-population differences. Variation among individuals within a population and between populations was significantly influenced by size, with larger fish tending to have higher concentrations of THg. However, a partial correlation analysis controlling for size determined that the effect of length did not dominate the effect of age. Although the index of trophic level ($\delta^{15}N$) included in the study suggests a role for trophic status in determining THg, variations among individuals in terms of their use of different foraging habitats or tactics appears less important for determining THg concentration levels than for other studied charr species (e.g., Arctic charr). While not complete in the sense that it provides only limited spatial coverage, the data reported do provide a useful historical baseline against which future changes in THg levels in Northern Dolly Varden may be statistically compared to determine whether and/or where significant changes or increases have occurred.

Chapter 3 compared THg concentrations among isolate, resident, and anadromous life-history types of Northern Dolly Varden. Life-history driven differences in THg in Northern Dolly Varden were evident, although trophic position and growth rates were both more important for explaining among individual differences in THg tissue concentrations. In contrast to studies of other northern fish species, such as Arctic charr, where anadromous life-history types have

lower THg relative to freshwater conspecifics, anadromous Northern Dolly Varden had higher absolute THg than their freshwater counterparts. Adjustment for age and length, however, indicated anadromous fish to have lower relative THg values. The reversal of ranks in terms of THg levels among life-history types ultimately depends on two counteracting mechanisms. The first mechanism raises overall THg as a result of the switch to marine feeding and the use of higher trophic level prey organisms that have previously bioconcentrated THg. The second mechanism lowers overall THg as a result of somatic growth dilution, whereby marine feeding allows fish to accumulate biomass faster relative to Hg and reduces standardized levels of THg relative to non-anadromous fish.

Chapter 4 investigated temporal trends in Northern Dolly Varden THg from periods 1986-1988 to 2011-2013 in the Rat and Firth rivers. THg in Northern Dolly Varden increased over a 27 year span, although the mean THg levels were lower in the Rat River and lower or comparable in the Firth River to other fish from a similar time and place. Time period, fork-length, $\delta^{13}C$, and $\delta^{15}N$ had the strongest influence on individual THg. Relationships between log[THg] versus fork-length and log[THg] versus $\delta^{13}C$ have remained constant over time in the Rat River, while relationships between log[THg] versus $\delta^{15}N$ have remained constant in the Firth River. In addition, while warming temperature trends correlated with the increasing trends in THg, the effect is not direct but acts through changes in freshwater discharge and sea ice cover that driven by a changing climate.

Study significance

This thesis is the first to our knowledge to examine the spatial, life-history, and temporal trends of THg concentrations in Northern Dolly Varden from the Canadian Arctic. Mercury

levels in Arctic ecosystems are of particular concern because of potential bioaccumulation impacts on human populations that rely on fish as a key component of their diet (Lockhart et al., 2005). Northern Dolly Varden are valued by the Inuvialuit and Gwich'in communities for subsistence and cultural purposes (Gallagher et al., 2011). Furthermore, concerns for Hg accumulation have risen in aquatic ecosystems due to the potential impacts of climate change on Hg dynamics. Increased glacier melt rates (Fisher et al., 2011), changes in precipitation and temperature patterns and increased shoreline erosion (Phillips et al., 2011) associated with freshwater discharge (Leitch et al., 2007) have all been implicated in likely increases in Hg accumulation rates in Arctic aquatic biota under climate change scenarios. Northern Dolly Varden are highly vulnerable to climate change as a result of habitat impacts (Mochnacz et al., 2010) and because they feed and grow in the marine areas of the Beaufort Sea being increasing affected by climate changes associated with likely increased Hg loadings (e.g., Leitch et al. 2007; Macdonald and Loseto, 2010). In an era and for an area being marked by significant ecological change (e.g., climate change) and the increased pressure of industrial development (e.g., oil and gas development), the utility of historical baseline information cannot be understated.

This study has also filled in some of the basic knowledge gaps of THg in Northern Dolly Varden and highlighted the importance of continued baseline monitoring for understanding future levels of contamination. In the late 1980's, Northern Dolly Varden THg concentrations were not found to illustrate any spatial trends (Chapter 2), however, from 1986-1988 to 2011-2013, an increase in THg concentrations were found in two Northern Dolly Varden populations (Rat River and Firth River) (Chapter 4). In addition, life-history type has been found to play a significant role in determining THg levels, with anadromous Northern Dolly Varden exhibiting higher THg concentrations than isolate and resident Northern Dolly Varden (Chapter 3). The

thesis further highlights the importance that size and feeding behaviour have on impacting the THg levels in Northern Dolly Varden, thereby providing information which may be useful in setting future consumption guidelines.

Future research

While the thesis has provided valuable THg data on Northern Dolly Varden, there is still a need for more Northern Dolly Varden THg trend data as such data will allow for better predictions of future contamination levels. Incorporating additional sites into future studies would improve the spatial coverage of the database (Chapter 2), and the addition of more years of data would improve understanding of temporal trends (Chapter 4).

With the prediction that warming temperatures may increase THg loadings to the important marine feeding grounds, with likely consequences for increased Northern Dolly Varden THg levels, further work is required to monitor and understand the trends in the marine environment (Chapter 4). Due to the strong relationship between fish THg and diet, further research on Northern Dolly Varden THg concentrations that incorporates marine and freshwater food web data is needed. Further understanding into Hg concentrations of organisms incorporated in the marine and riverine food webs of Northern Dolly Varden would aid in understanding their THg patterns and provide valuable data to complement the already detailed studies that have been completed in the eastern Arctic for species like Arctic charr (e.g., van der Velden et al., 2013a,c). Chapter 3, for example, highlights the need for improved understanding of the relative importance of differences in the THg concentrations in marine and freshwater habitats and prey, habitat-specific bioconcentration rates and/or differences in physiology related to anadromy that may contribute to the observed pattern of differences in THg among Northern

Dolly Varden life-history types. Gathered data should include Hg, MeHg and carbon and nitrogen stable isotope values of organisms from throughout the food web, such as benthic invertebrates and forage fishes (e.g., van der Velden et al., 2013a,c). Monitoring of marine and riverine Northern Dolly Varden food webs over time would without doubt further help to explain and predict future THg patterns.

Lastly, continued monitoring of THg concentrations in Northern Dolly Varden from the Canadian Arctic is suggested, not only for the safety of Dolly Varden populations, but for the health and safety of the Indigenous peoples of northern communities who consume them.

References

Anderson, D. R. 2008. Model based inference in the life sciences: a primer on evidence. Springer Science, New York, NY. 184p.

Armstrong, J. B. and Bond, M. H. 2013. Phenotype flexibility in wild fish: Dolly Varden regulate assimilative capacity to capitalize on annual pulsed subsidies. Journal of Animal Ecology. doi: 10.1111/1365-2656.12066

Armstrong, R.H. and Hermans, M. 2007. Dolly Varden (*Salvelinus malma*). Southeast Alaska Conservation Assessment 8.8. December 11.
<http://home.gci.net/~tnc/HTML/Resource_synthesis.html>

Armstrong, R.H. and Morrow, J.E. 1980. The Dolly Varden. Pages 99-140 in Balon, E.K., ed. Charr: Salmonid fishes of the genus *Salvelinus.* Dr. W. Junk Publishers. The Hague, The Netherlands.

Armstrong, R.H. 1984. Migration of anadromous Dolly Varden charr in southeastern Alaska – a manager's nightmare. *In* Biology of the Arctic Charr: Proceedings of the International Symposium on Arctic Charr. *Edited by* L. Johnson and B. Burns. University of Manitoba Press, Winnipeg, Manitoba. Pp. 559-570.

Atwell, L., Hobson, K.A. and Welch, H.E. 1998. Biomagnification and bioaccumulation of mercury in an arctic marine food web: insights from stable nitrogen isotope analysis. Canadian Journal of Fisheries and Aquatic Sciences 55: 1114-1121.

Bache, C.A., Gutenmann, W.H. and Lisk, D.J. 1971. Residues of total mercury and methylmercuric salts in lake trout as a function of age. Science 172: 951-952.

Baker, M.R., Schlinder, D.E., Holtgrieve, G.W. and St. Louis, V.L. 2009. Bioaccumulation and transport of contaminants: migrating sockeye salmon as vectors of mercury. Environmental Science and Technology 43: 8840-8846.

Barber, R.T., Vijayakumar, A. and Cross, F.A. 1972. Mercury concentrations in recent and ninety year old benthopelagic fish. Science 178: 636-639.

Bates, D.M. and Watts, D.G. 1988. Nonlinear regression analysis and its applications. New York: John Wiley & Sons. Xiv + 365 pp.

Berninger, K. and Pennanen, J. 1995. Heavy metals in perch (*Perca fluviatilis* L.) from two acidified lakes in the Salpausselka esker area in Finland. Water, Air, and Soil Pollution 81: 283-294.

Bloom, N.S., Gill, G.A., Cappellino, S., Dobbs, C., McShea, L., Driscoll, C., Mason, R.P. and Rudd, J.W.M. 1999. Speciation and cycling of mercury in Lavaca Bay, Texas, Sediments. Environmental Science & Technology 33, 7-13.

Bodaly, R.A., Hecky, R.E. and Fudge, R.J.P. 1984. Increases in fish mercury levels in lakes flooded by the Churchill River diversion, northern Manitoba. Canadian Journal of Fisheries and Aquatic Sciences 41: 682-691.

Bodaly, R.A., Rudd, J.W.M. and Fudge, R.J.P. 1993. Mercury concentrations in fish related to size of remote Canadian Shield lakes. Canadian Journal of Fisheries and Aquatic Sciences 50: 980-987.

Bond, M. H. 2013. Diversity in migration, habitat use, and growth of Dolly Varden char in Chignik Lakes, Alaska. PhD Thesis, University of Washington, Seattle, WA.

Bond, M.H. and Quinn, T.P. 2013. Patterns and influences on Dolly Varden migratory timing in the Chignik Lakes, Alaska, and comparison of populations throughout the northeastern Pacific and Arctic oceans. Canadian Journal of Fisheries and Aquatic Sciences 70: 655-665.

Booth, S. and Zeller, D. 2005. Mercury, food webs, and marine mammals: implications of diet and climate change for human health. Environmental Health Perspectives 113: 521-526.

Braune, B., Muir, D., DeMarch, B., Gamberg, M., Poole, K., Currie, R., Dodd, M., Duschenko, W., Eamer, J., Elkin, B., Evans, M., Grundy, S., Hebert, C., Johnstone, R., Kidd, K., Koenig, B., Lockhart, L., Marshall, H., Reimer, K., Sanderson, J., and Shutt, L. 1999. Spatial and temporal trends of contaminants in Canadian Arctic freshwater and terrestrial ecosystems: a review. The Science of the Total Environment 230: 145-207.

Brigham, M. E., Wentz, D. A., Aiken, G. R. and Krabbenhft, D. P. 2009. Mercury cycling in stream ecosystems. 1. Water column chemistry and transport. Environmental Science and Technology 43: 2720-2725.

Brosset, C. 1987. The behaviour of mercury in the physical environment. Water, Air, and Soil Pollution 34: 145-166.

Bruce, W.J. and Spencer, K.D. 1979. Mercury levels in Labrador fish, 1977–78. Can Ind Rep Fish Aquat Sci 111. [iv-12].

Burnham, K. P. and Anderson, D. R. 2002. Model selection and multimodel inference: a practical information-theoretic approach, 2nd edition. Springer Science, New York, NY. 488p.

Cabana, G. and Rasmussen, J.B. 1994. Modelling food chain structure and contaminant bioaccumulation using stable nitrogen isotopes. Nature 372: 255-257.

Cabana, G. and Rasmussen, J.B. 1996. Comparison of aquatic food chains using nitrogen isotopes. Proceedings of the National Academy of Sciences 93: 10844-10847.

Cabana, G., Tremblay, A., Kalff, J. and Rasmussen, J.B. 1994. Pelagic food chain structure in Ontario lakes: A determinant of mercury levels in lake trout (*Salvelinus namaycush*). Canadian Journal of Fisheries and Aquatic Sciences 51: 381-389.

Campbell, L.M. Norstrom, R.J., Hobson, K.A., Muir, D.C.G., Backus, S. and Fisk, A.T. 2005. Mercury and other trace elements in a pelagic Arctic marine food web (Northwater Polynya, Baffin Bay). Science of the Total Environment 351-352: 247-263.

Carrie, J., Wang, F., Sanei, H., Macdonald, R.W., Outridge, P.M., and Stern, G.A. 2010. Increasing contaminant burdens in an arctic fish, Burbot (*Lota lota*), in a warming climate. Environmental Science & Technology 44: 316-322.

Casey, M.M. and Post, D.M. 2011. The problem of isotopic baseline: Reconstructing the diet and trophic position of fossil animals. Earth-Science Reviews 106: 131-148.

Chasar, L. C., Scudder, B. C., Stewart, A. R., Bell, A. H., Aiken, G. R. 2009. Mercury cycling in stream ecosystems. 3. Trophic dynamics and methylmercury bioaccumulation. Environmental Science and Technology 43: 2733-2739.

Cobb, D., Fast, H., Papst, M.H., Rosenberg, D., Rutherford, R. and Sareault, J.E. 2008. Beaufort Sea Large Ocean Management Area: Ecosystem Overview and Assessment Report. Canadian Technical Report of Fisheries and Aquatic Sciences 2780: ii-ix + 188.

Cooke, S.J., Hinch, S.G., Farrell, A.P., Patterson, D.A., Miller-Saunders, K., Welch, D.W., Donaldson, M.R., Hanson, K.C., Crossin, G.T., Mathes, M.T., Lotto, A.G., Hruska, K.A., Olsson, I.C., Wagner, G.N., Thomson, R., Hourston, R., English, K.K., Larsson, S., Shrimpton, J.M. and Van der Kraak, G. 2008. Developing a mechanistic understanding of fish migrations by linking telemetry with physiology, behavior, genomics and experimental biology: an interdisciplinary case study on adult Fraser River sockeye salmon. Fisheries 33: 321-338.

Coplen, T.B. 1994. Reporting of stable hydrogen, carbon, and oxygen isotopic abundances. Pure & Applied Chemistry 66: 273-276.

Cox, C.P. 1987. A Handbook of Introductory Statistical Methods. John Wiley and Sons, New York.

Craig, H. 1957. Isotopic standards for carbon and oxygen and correction factors for mass-spectrometric analysis of carbon dioxide. Geochimica et Cosmochimica Acta 12: 133-149.

Daviglus, M., Sheeshka, J. and Murkin, E. 2002. Health benefits from eating fish. Comments on Toxicology 8: 345-375.

de Freitas, A.S.W., Qadri, S.U. and Case, B.E. 1974. Origins and fate of mercury compounds in fish. Proceedings of the International Conference on Transport of Persistent Chemicals in Aquatic Ecosystems. National Research Council of Canada, Ottawa, Canada. pp. III.31-III.36.

DeCicco, A.L. 1992. Long distance movements of anadromous Dolly Varden between Alaska and the USSR. Arctic 45: 120-123.

DeCicco, A.L. 1997. Movements of postsmolt anadromous Dolly Varden in northwestern Alaska. American Fisheries Society Symposium 19: 175-183.

Deniseger, J., Erickson, L.J., Austin, A., Roch, M. and Clark, M.J.R. 1990. The effects of decreasing heavy metal concentrations on the biota of Buttle Lake, Vancouver Island, British Columbia. Water Research 24: 403-416.

Denton , K. P., Rich Jr., H. B. and Quinn, T. P. 2009. Diet, Movement, and Growth of Dolly Varden in Response to Sockeye Salmon Subsidies. Transactions of the American Fisheries Society, 138: 1207-1219.

DFO. 2003. Babbage River Dolly Varden. DFO Sci. Stock Status Report D5-62 (2002).

Doyon, J., Schetagne, R. and Verdon, R. 1998. Different mercury bioaccumulation rates between sympatric populations of dwarf and normal lake whitefish (*Coregonus clupeaformis*) in the La Grande complex watershed, James Bay, Québec. Biogeochemistry 40: 203–16.

Draper, N.R. and Smith, H. 1981. Applied Regression Analysis. 2nd Ed., John Wiley and Sons, New York, N.Y.

Dunn, O.J. and Clark, V. 1987. Applied statistics: Analysis of variance and regression. 2nd Ed., Wiley, New York, N.Y.

Dutton, M.D. 1997. Methyl mercury bioaccumulation: A study of factors influencing uptake and elimination in fish. Thesis (PhD) University of Waterloo – Ontario.

Evans, M. and Lockhart, L. 2001. An investigation of factors affecting high mercury concentrations in predatory fish in the Mackenzie River Basin. Department of Indian Affairs and Northern Development, Ottawa, Canada, 174-184.

Evans, M.S., Lockhart, W.L., Doetzel, L., Low, G., Muir, D., Kidd, K., Stephens, G. and Delaronde, J. 2005a. Elevated mercury concentrations in fish in lakes in the Mackenzie River Basin: The role of physical, chemical, and biological factors. Science of the Total Environment 351-352: 479-500.

Evans, M.S. and Muir, D. 2010. Temporal trends and spatial variations in persistent organic pollutants and metals in sea-run char from the Canadian Arctic. Synopsis of research conducted under the 2009–2010 Northern Contaminants Program. Ottawa: Indian and Northern Affairs Canada, p. 142-150.

Evans, M., Muir, D., Brua, R.B., Keating, J. and Wang, X. 2013. Mercury trends in predatory fish in Great Slave Lake: The influence of temperature and other climate drivers. Environmental Science & Technology 47: 12793-12801.

Evans, M.S., Muir, D., Lockhart, W.L., Stern, G., Ryan, M. and Roach, P. 2005b. Persistent organic pollutants and metals in the freshwater biota of the Canadian Subarctic and Arctic: An overview. Science of the Total Environment 351-352: 94-147.

Fechhelm, R.G., Bryan, J.D., Griffiths, W.B. and Martin, L.R. 1997. Summer growth patterns of northern Dolly Varden (*Salvelinus malma*) smolts from the Prudhoe Bay region of Alaska. Canadian Journal of Fisheries and Aquatic Sciences 54: 1103-1110.

Fisher, D., Zheng, J., Burgess, D., Zdanowicz, C., Kinnard, C., Sharp, M. and Bourgeois, J. 2011. Recent melt rates of Canadian Arctic ice caps are the highest in four millennia. Global and Planetary Change, doi: 10.1016.j.gloplacha.2011.06.005.

Fitzgerald, W.F. and Clarkson, T.W. 1991. Mercury and mono-methylmercury: present and future concerns. Environmental Health Perspectives 96: 159-166.

Fleming, I.A. and Gross, M.R. 1990. Latitudinal clines: A trade-off between egg number and size in Pacific Salmon. Ecology 71: 1-11.

Forbes, D. L. 1983. Morphology and sedimentology of a sinuous gravel-bed channel system: lower Babbage River, Yukon coastal plain, Canada. Special Publications International Association of Sedimentology 6:195-206.

Fry, B. 2006. Stable Isotope Ecology. Springer Science Business Media LLC., New York, NY. 308 pp.

Gallagher, C.P. Roux, M.-J., Howland, K.L. and Tallman, R.F. 2011. Synthesis of biological and harvest information used to assess populations of northern form Dolly Varden (*Salvelinus malma malma*) in Canada. Part II: Big Fish River. DFO Can. Sci. Advis. Sec. Res. Doc. 2010/115. V + 45 p.

Gallagher, C.P., Roux, M.J., Howland, K.L. and Tallman, R.F. 2012. Synthesis of biological and harvest information used to assess populations of northern form Dolly Varden (*Salvelinus malma malma*) in Canada. Part III: Comparison among populations. DFO Can. Sci. Advis. Sec. Res. Doc. 2011/128. vi + 81 p.

Gantner, N., Power, M., Babaluk, J.A., Reist, J.D., Kock, G., Lockhart, L.W., Solomon, K.R. and Muir, D.C.G. 2009. Temporal trends of mercury, cesium, potassium, selenium, and thallium in Arctic charr (*Salvelinus alpinus*) from Lake Hazen, Nunavut, Canada: Effect of trophic position, size and age. Environmental Toxicology and Chemistry 28: 254-263.

Gantner, N., Muir, D.C., Power, M., Iqaluk, D., Reist, J.D., Babaluk, J.A., Meili, M., Borg, H., Hammar, J., Michaud, W., Dempson, B. and Solomon, K.R. 2010a. Mercury concentrations in landlocked Arctic charr (*Salvelinus alpinus*) from the Canadian Arctic. Part II: Influence of lake biotic and abiotic characteristics on geographic trends in 27 populations. Environmental Toxicology and Chemistry 29: 633-643.

Gantner, N., Power, M., Iqaluk, D., Meili, M., Borg, H., Sundbom, M., Solomon, K.R., Lawson, G. and Muir, D.C. 2010b. Mercury concentrations in landlocked Arctic charr (*Salvelinus alpinus*) from the Canadian Arctic. Part I: Insights from trophic relationships in 18 lakes. Environmental Toxicology and Chemistry 29: 621-632.

Gantner, N., Veillette, J., Michaud, W.K., Bajno, R., Muir, D., Vincent, W.F., Power, M., Dixon, B., Reist, J.D., Hausmann, S. and Pienitz, R. 2012. Physical and biological factors affecting mercury and perfluorinated contaminants in Arctic charr (*Salvelinus alpinus*) of Pingualuit Crater Lake (Nunavik, Canada). Arctic 65: 195-206.

Glass, G.E., Sorensen, J.A. and Rapp, G.R. 2000. Methylmercury bioaccumulation dependence on northern pike age and size in 20 Minnesota lakes. Allelopathy Glass 772: 150.

Government of Canada. 2014. Climate. Accessed 17 July 2014. <http://climate.weather.gc.ca/>

Greenfield, B.K., Hrabik, T.R., Harvey, C.J. and Carpenter, S.R. 2001. Predicting mercury levels in yellow perch: use of water chemistry, trophic ecology, and spatial traits. Canadian Journal of Fisheries and Aquatic Sciences 58: 1419-1429.

Grieb, T.M., Driscoll, C.T., Gloss, S.P., Schofield, C.L., Bowie, G.L. and Porcella, D.B. 1990. Factors affecting mercury accumulation in fish in the upper Michigan Peninsula. Evironmental Toxicology and Chemistry 9: 919-930.

Guiguer, K.R.R.A., Reist, J.D., Power, M. and Babaluk, J.A. 2002. Using stable isotopes to confirm the trophic ecology of Arctic charr morphotypes from Lake Hazen, Nunavut, Canada. Journal of Fish Biology 60: 348-362.

Gupta, S., Barlow, M. and Donaldson, S. 2005. Chapter 13: Mercury exposure and human health effects: A Canadian perspective. In M.B. Parsons & J.B. Percival, Mercury Sources, Measurements, Cycles, and Effects Series Volume 34 (p. 259-286). Halifax, Nova Scotia: Minerological Association of Canada.

Gwich'in Renewable Resources Board. 2010. The Plan. Integrated Fisheries Management Plan for Dolly Varden (*Salvelinus malma malma*) of the Gwich'in Settlement Area and Inuvialuit Settlement Region Northwest Territories and Yukon North Slope 2011-2015 1: 1-25.

Hagan, J. and Taylor, E. B. 2001. Resource partitioning as a factor limiting gene flow hybridizing populations of Dolly Varden char (*Salvelinus malma*) and bull trout (*Salvelinus confluentus)*. Canadian Journal of Fisheries and Aquatic Sciences 58:2037-2047.

Hammerschmidt, C.R. and Fitzgerald, W.F. 2006. Methylmercury in freshwater fish linked to atmospheric mercury deposition. Environmental Science and Technology 40: 7764-7770.

Harris, R.C. and Bodaly, R.A. 1998. Temperature, growth and dietary effects on fish mercury dynamics in two Ontario lakes. Biogeochemistry 40: 175-187.

Health Canada. 2007. Mercury you health and the environment. Environmental and Workplace Health. <http://www.hc-sc.gc.ca/ewh-semt/pubs/contaminants/mercur/q47-q56-eng.php> downloaded November 15, 2011.

Health Canada. 2008. Human Health Risk Assessment of Mercury in Fish and Health Benefits of Fish Consumption. <http://www.hc-sc.gc.ca/fn-an/pubs/mercur/merc_fish_poisson-eng.php> downloaded March 31, 2014.

Hesslein, R.H., Hallard, K.A. and Ramlal, P. 1993. Replacement of sulfur, carbon, and nitrogen in tissue of growing broad whitefish (*Coregonus nasus*) in response to a change in diet traced by $\delta^{34}S$, $\delta^{13}C$, and $\delta^{15}N$. Canadian Journal of Fisheries and Aquatic Science 50: 2071-2076.

Holm, M., Jacobsen, J.A., Sturlaugsson, J. and Holst, J.C. 2006. Behaviour of Atlantic salmon (*Salmo salar* L.) recorded by data storage tags in the NE Atlantic – implications for interception by pelagic trawls. ICES CM 2006/Q:12, 16 pp.

Holmes, R.M., McClelland, J. W., Raymond, P. A., Frazer, B. B., Peterson, B. J. and Stieglitz, M. 2008. Lability of DOC transported by Alaskan rivers to the Arctic Ocean. Geophysical Research Letters 35, L03402.

Jackson, T.A. 1991. Biological and environmental control of mercury accumulation by fish in lakes and reservoirs of Northern Manitoba, Canada. Canadian Journal of Fisheries and Aquatic Sciences 48: 2449-2470.

Jarvela, L.E. and Thorsteinson, L.K. 1999. The epipelagic fish community of Beaufort Sea Coastal Waters, Alaska. Arctic 52: 80-94.

Jeitner, C. 2009. Metal concentrations (arsenic, cadmium, chromium, lead, mercury and selenium) in Dolly Varden (*Salvinus malma*) from the Aleutian Islands, Alaska. Thesis (MSc) Rutgers The State University of New Jersey – New Brunswick

Jennings, S. and Warr, K.J. 2003. Environmental correlates of large-scale spatial variation in the $\delta^{15}N$ of marine animals. Marine Biology 142: 1131-1140.

Jennings, S., Maxwell, T.A.D., Schratzberger, M. and Milligan, S.P. 2008. Body-size dependent temporal variations in nitrogen stable isotope ratios in food webs. Marine Ecology Progress Series 370: 199-206.

Jensen, S. and Jernelov, A. 1969. Biological methylation of mercury in aquatic organisms. Nature 223: 753-754.

Jewett, S.C., Zhang, X., Naidu, A.S., Kelley, J.J., Dasher, D. and Duffy, L.K. 2003. Comparison of mercury and methylmercury in northern pike and Arctic grayling from western Alaska rivers. Chemosphere 50: 383-392.

Joensuu, O.I. 1971. Fossil fuels as a source of mercury pollution. Science 172: 1027-1028.

Johnson, S.L., Power, J.H., Wilson, D.R. and Ray, J. 2010. A comparison of the survival and migratory behavior of hatchery-reared and naturally reared steelhead smolts in the Alsea River and Estuary, Oregon, using acoustic telemetry. North American Journal of Fisheries Management 30: 55-71.

Jonsson, B. and Jonsson, N. 2011. Ecology of Atlantic salmon and brown trout: habitat as a template for life histories, Fish & Fisheries Series 33. Dordrecht, the Netherlands: Springer-Verlag.

Kidd, K.A., Hesslein, R.H., Fudge, R.J.P. and Hallard, K.A. 1995. The influence of trophic level as measured by $\delta^{15}N$ on mercury concentrations in freshwater organisms. Water, Air and Soil Pollution 80: 1011-1015.

Kidd, K.A., Bootsma, H.A., Hesslein, R.H., Lockhart, L.W. and Hecky, R.E. 2003. Mercury concentrations in the food web of Lake Malawi, East Africa. Journal of Great Lakes Research 29: 258-266.

Kidd, K.A., Muir, D. C. G., Evans, M. E., Wang, X., Whittle, M., Swanson, H. K., Johnson, T., and Guildford, S. 2012. Biomagnification of mercury through lake trout (*Salvelinus namaycush*) food webs of lakes with different physical, chemical and biological characteristics. Science of the Total Environment 438:135-148.

Kim, J.P. 1995. Methylmercury in rainbow trout (*Oncorhynchus mykiss*) from Lakes Okareka, Okaro, Rotomahana, Rotorua and Tarawera, North Island, New Zealand. The Science of the Total Environment 164: 209-219.

Kirk, J.L., Lehnherr, I., Andersson, M., Braune, B.M., Chan, L., Dastoor, A.P., Durnford, D., Gleason, A.L., Loseto, L.L., Steffen, A. and St. Louis, V.L. 2012. Mercury in Arctic marine ecosystems: Sources, pathways and exposure. Environmental Research 119: 64-87.

Kling, G.W., Fry, B. and O'Brien, W.J. 1992. Stable isotopes and planktonic trophic structure in arctic lakes. Ecology, 73, 561-566.

Kowalchuk, M.W., Reist, J.D., Bajno, R. and Sawatzky, C.D. 2010a. Population structuring and inter-river movements of northern form Dolly Varden, *Salvelinus malma malma* (Walbaum 1792), along the North Slope of Canada and Alaska. DFO Can. Sci. Advis. Sec. Res. Doc. 2010/038. vi + 17 p.

Kowalchuk, M.W., Sawatzky, C.D. and Reist, J.D. 2010b. A review of the taxonomic structure within Dolly Varden, *Salvelinus malma* (Walbaum 1792), of North America. DFO Canadian Science Advisory Secretariat Research Document.2010/013. vi + 16 p.

Lacroix, G. 2013. Population-specific ranges of oceanic migration for adult Atlantic salmon (*Salmo salar*) documented using pop-up satellite archival tags. Canadian Journal of Fisheries and Aquatic Sciences 70: 1011-1030.

Le, D.Q., Chino, N., Shirai, K. and Arai, T. 2010. Trace metals in Japanese eel, *Anguilla japonica*, in relation to ecological migratory types and growth stages. Estuarine Coastal and Shelf Science 87: 405-410.

Leitch, D.R., Carrie, J., Lean, D., Macdonald, R.W., Stern, G.A. and Wang, F. 2007. The delivery of mercury to the Beaufort Sea of the Arctic Ocean by the Mackenzie River. Science of the Total Environment 373: 178-195.

Lindqvist, O. and Rodhe, H. 1985. Atmospheric mercury – a review. Tellus 37B: 136-159.

Lockhart, W.L., Stern, G.A., Low, G., Hendzel, M., Boila, G., Roach, P., Evans, M.S., Billeck, B.N., DeLaronde, J., Friesen, S., Kidd, K., Atkins, S., Muir, D.C.G., Stoddart, M., Stephens, G., Stephenson, S., Harbicht, S., Snowshoe, N., Grey, B., Thompson, S. and DeGraff, N. 2005. A history of total mercury in edible muscle of fish from lakes in northern Canada. Science of the Total Environment 351-352: 427-463.

Loseto, L.L., Lean, D.R.S. and Siciliano, S.D. 2004. Snowmelt sources of methylmercury to high Arctic ecosystems. Environmental Science and Technology 38: 3004-3010.

Loseto, L.L., Stern, G.A., Deibel, D., Connelly, T.L., Prokopowicz, A., Lean, D.R.S., Fortier, L. and Ferguson, S.H. 2008. Linking mercury exposure to habitat and feeding behaviour in Beaufort Sea beluga whales. Journal of Marine Systems 74: 1012-1024.

Macdonald, R.W., Harner, T. and Fyfe, J. 2005. Recent climate change in the Arctic and its impact on contaminant pathways and interpretation of temporal trend data. Science of the Total Environment 342: 5-86.

Macdonald, R.W. and Loseto, L.L. 2010. Are Arctic Ocean ecosystems exceptionally vulnerable to global emissions of mercury? A call for emphasized research on methylation and the consequences of climate change. Environmental Chemistry 7: 133-138.

Mariotti, A. 1983. Atmospheric nitrogen is a reliable standard for natural $\delta^{15}N$ abundance measurements. Nature 303: 685-687.

Marusczak, N., Larose, C., Dommergue, A., Paquet, S., Beaulne, J.S., Maury-Brachet, R, Lucotte, M., Nedjai, R. and Ferrari, C.P. 2011. Mercury and methylmercury concentrations in high altitude lakes and fish (Arctic charr) from the French Alps related to watershed characteristics. Science of the Total Environment 409: 1909-1915.

Mathers, R.A. and Johansen, P.H. 1985. The effects of feeding ecology on mercury accumulation in walleye (*Stizostedion vitreum*) and pike (*Esox lucius*) in Lake Simcoe. Canadian Journal of Zoology 63: 2006-2012.

Matta, M.B., Linse, J., Cairncross, C., Francendese, L. and Kocan, R.M. 2001. Reproductive and transgenerational effects of merthylmercury or aroclor 1268 on *Fundulus heteroclitus.* Environmental Toxicology and Chemistry 20: 327-335.

Meier, W.N., Hovelsrud, G.K., van Oort, B.E.H., Key, J.R., Kovacs, K.M., Michel, C., Haas, C., Granskog, M.A., Gerland, S., Perovich, D.K., Makshtas, A. and Reist, J.D. 2014. Arctic sea ice in transformation: A review of recent observed changes and impacts on biology and human activity. Reviews of Geophysics, doi: 10.1002/2013RG000431

Milestone. 2010. DMA Operator Manual MA122. Sorisole (BG) Italy: Milestone Srl, p 1-128.

Mochnacz, N.J., Schroeder, B.S., Sawatzky, C.D. and Reist, J.D. 2010. Assessment of northern Dolly Varden, *Salvelinus malma malma* (Walbaum, 1792), habitat in Canada. Canadian Manuscript Report of Fisheries and Aquatic Sciences 2926: vi + 48 p.

Morel, F.M.M., Kraepiel, A.M.L. and Amyot, M. 1998. The chemical cycle and bioaccumulation of mercury. Annual Review of Ecological Systems 29: 543-566.

Morrow, J.E. 1980. Analysis of the Dolly Varden charr, *Salvelinus malma,* of northwestern North America and northwestern Siberia. *In* Charrs, salmonid fishes of the genus *Salvelinus*. Edited by E.K. Balon. Dr. W. Junk Publishers, The Hague, Netherlands. p. 323-338.

Morton, W. M. 1982. Comparative catches and food habits of Dolly Varden and Arctic charrs, *Salvelinus malma* and *S. alpinus*, at Karluk, Alaska, in 1939-1941. Environmental Biology of Fishes 7:7-28.

Muir, D., Braune, B., DeMarch, B., Norstrom, R., Wagemann, R., Lockhart, L., Hargrave, B., Bright, D., Addison, R., Payne, J. and Reimer, K. 1999. Spatial and temporal trends and effects of contaminants in the Canadian Arctic marine ecosystem: a review. Science of the Total Environment 230: 83-144.

Muir, D., Günter, K. and Evans, M.S. 2009. Temporal trends of persistent organic pollutants and mercury in landlocked char in the high Arctic. In: Smith SL, Stow J, editors. Synopsis of research conducted under the 2008–2009 Northern Contaminants Program. Ottawa, ON: Indian and Northern Affairs Canada, p. 145-151.

Muir, D., Wang, X., Bright, D., Lockhart, L. and Kock, G. 2005. Spatial and temporal trends of mercury and other metals in landlocked char from lakes in the Canadian Arctic archipelago. Science of the Total Environment 351-352: 464-478.

Munthe, J., Bodaly, R.A., Branfireun, B.A., Driscoll, C.T., Gilmour, C.C., Harris, R., Horvat, M., Lucotte, M. and Malm, O. 2007. Recovery of mercury-contaminated fisheries. Ambio 36: 33-44.

Nagorski, S.A., Engstrom, D.R., Hudson, J.P., Krabbenhof, D. P., Hood, E., DeWild, J. F. and Aiken, G. R. 2014. Spatial distribution of mercury in southeastern Alaskan streams influenced by glaciers, wetlands, and salmon. Environmental Pollution 184: 62-72.

Nakano, S. and Kaeriyama, M. 1995. Summer microhabitat use and diet of four sympatric stream-dwelling salmonids in a Kamchatkan stream. Fisheries Science 61: 926-930.

Nakano, S., Fausch, K. D. and Kitano, S. 1999. Flexible niche partitioning via a foraging mode shift: a proposed mechanism for co-existence in stream-dwelling charrs. Journal of Animal Ecology 68:1079-1092.

Norstrom, R.J., McKinnon, A.E. and de Freitas, A.S.W. 1976. A bioenergetics-based model for pollutant accumulation by fish simulation of PCB and methylmercury residue levels in Ottawa River yellow perch (*Perca flavascens*). J. Fish. Res. Board Can. 33: 248-267.

Outridge, P.M., Macdonald, R.W., Wang, F., Stern, G.A., Dastoor, A.P. 2008. A mass balance inventory of mercury in the Arctic Ocean. Environmental Chemistry 5, 89. doi: 10.1071/EN08002.

Papik, R., Merschke, M. and Ayles, G.B. 2003. Inuvialuit traditional ecological knowledge of fisheries in rivers west of the Mackenzie River in the Canadian Arctic. Canada/Inuvialuit Fisheries Joint Management Committee Technical Report 2003-4: v + 22p.

Pennuto, C.M., Lane, O.P., Evers, D.C., Taylor, R.J. and Loukmas, J. 2005. Mercury in the northern cray fish, *Orconectes virilise* (Hagen), in New England, USA. Ecotoxicology 14: 149-162.

Peterson, B.J. and Fry, B. 1987. Stable isotopes in ecosystem studies. Annual Review of Ecology and Systematics 18: 293-320.

Phillips, V.J.A., St. Louis, V.L., Cooke, C.A., Vinebrooke, R.D. and Hobbs, W.O. 2011. Increased mercury loadings to western Canadian alpine lakes over the past 150 years. Environmental Science and Technology 45: 2042-2047.

Pinheiro, J.C. and Bates, D.M. 2000. Mixed-effects models in S and S-PLUS. New York: Springer.

Post, D.M. 2002. Using stable isotopes to estimate trophic position: models, methods, and assumptions. Ecology 83: 703-718.

Power, M., Klein, G.M., Guiguer, K.R.R.A. and Kwan, M.K.H. 2002. Mercury accumulation in the fish community of a sub-Arctic lake in relation to trophic position and carbon sources. Journal of Applied Ecology 39: 819-830.

Quinn, T.P., Graynoth, E., Wood, C.C. and Foote, C.J. 1998. Genotypic and phenotypic divergence of sockeye salmon in New Zealand from their ancestral British Columbia populations. Transactions of the American Fisheries Society 127: 517-534.

Reddin, D.G., Downton, P., Fleming, I.A., Hansen, L.P. and Mahon, A. 2011. Behavioural ecology at sea of Atlantic salmon (*Salma salar* L.) kelts from a Newfoundland (Canada) river. Fisheries Oceanography 20: 174-191.

Reist, J.D. 1986. An empirical evaluation of coefficients used in residual and allometric adjustment of size covariation. Canadian Journal of Zoology 64: 1363-1368.

Reist, J.D., Low, G., Johnson, J.D. and McDowell, D. 2002. Range extension of bull trout, *Salvelinus confluentus*, to the central Northwest Territories, with notes on identification and distribution of Dolly Varden, *Salvelinus malma*, in the western Canadian Arctic. Arctic 55: 70-76.

Reist, J.D. and Sawatzky, C.D. 2010. Diversity and distribution of chars, genus *Salvelinus*, in Northwestern North America in the context of Northern Dolly Varden (*Salvelinus malma malma* (Walbaum 1792)). DFO Can. Sci. Advis. Sec. Res. Doc. 2010/014. vi + 18 p.

Reist, J.D., Wrona, F.J., Prowse, T.D., Dempson, J.B., Power, M., Kock, G., Carmichael, T.J., Sawatzky, C.D., Lehtonen, H. and Tallman, R.F. 2006a. Effects of climate change and UV radiation on fisheries for arctic freshwater and anadromous species. A Journal of the Human Environment 35: 402-410.

Reist, J.D., Wrona, F.J., Prowse, T.D., Power, M., Dempson, J.B., Beamish, R.J., King, J.R., Carmichael, T.J. and Sawatzky, C.D. 2006b. General effects of climate change on arctic fishes and fish populations. A Journal of the Human Environment 35: 370-380.

Reist, J.D., Wrona, F.J., Prowse, T.D., Power, M., Dempson, J.B., King, J.R. and Beamish, R.J. 2006c. An overview of effects of climate change on selected arctic freshwater and anadromous fishes. A Journal of the Human Environment 35: 381-387.

Rember, R. D. and Trefry, J. H. 2004. Increased concentration of dissolved trace metals and organic carbon during snowmelt in rivers of the Alaskan Arctic. Geochimica et Cosmochimica Acta. 68: 477-489.

Rigét, F., Asmund, G. and Aastrup, P. 2000. Mercury in Arctic char (*Salvelinus alpinus*) populations from Greenland. Science of the Total Environment 245: 161-172.

Rigét, F., Braune, B., Bignert, A., Wilson, S., Aars, J., Born, E., Dam, M., Dietz, R., Evans, M., Evans, T., Gamberg, M., Gantner, N., Green, N., Gunnlaugsdottir, H., Kannan, K., Letcher, R., Muir, D., Roach, P., Sonne, C., Stern, G. and Wigg, Ø. 2011. Temporal trends of Hg in Arctic biota, an update. Science of the Total Environment 409, 3520-3526.

Rigét, F., Moller, P., Dietz, R., Nielsen, T.G., Asmund, G., Strand, J., Larsen, M.M. and Hobson, K.A. 2007. Transfer of mercury in the marine food web of West Greenland. Journal of Environmental Monitoring 9: 877-883.

Rognerud, S., Grimalt, J.O., Rosseland, B.O., Fernandez, P., Hofer, R., Lackner, R., Lauritzen, B., Lien, L., Massabuau, J.C. and Ribes, A. 2002. Mercury and organochlorine contamination in brown trout (*Salmo trutta*) and Arctic charr (*Salvelinus alpinus*) from high mountain lakes in Europe and the Svalbard Archipelago. Water, Air, and Soil Pollution 2: 209-232.

Rolff, C., Broman, D., Naf, C. and Zebuhr, Y. 1993. Potential biomagnification of PCDD/Fs – new possibilities for quantitative assessment using stable isotope trophic position. Chemosphere 27: 461-468.

Rose, J., Hutcheson, M.S., West, C.R., Pancorbo, O., Hulme, K., Cooperman, A., DeCesare, G., Isaac, R. and Screpetis, A. 1999. Fish mercury distribution in Massachusetts, USA lakes. Environmental Toxicology and Chemistry 18: 1370-1379.

Sandstrom, S.J., Lemieux, P.J. and Reist, J.D. 1997. Enumeration and biological data from the upstream migration of Dolly Varden charr (*Salvelinus malma*) (W.), from the Babbage River, Yukon north slope, 1990 to 1992. Canadian Data Report of Fisheries and Aquatic Sciences 1018: iv + 132 p.

Sandstrom, S., Harwood, L. and Howland, K. 2009. Status of anadromous Dolly Varden char (Salvelinus malma) of the Rat River, Northwest Territories, as assessed through mark-recapture and live-sampling at the spawning and overwintering site (1995-2007). Canadian Technical Report of Fisheries and Aquatic Sciences 2842: vi + 68p.

Schell, D.M., Bartnett, B.A. and Vinette, K.A. 1998. Carbon and nitrogen isotope ratios in zooplankton of the Bering, Chukchi and Beaufort Seas. Marine Ecology Progress Series 162: 11-23.

Schroeder, W.H., Anlauf, K.G., Barrie, L.A., Lu, J.Y., Steffen, A., Schneeberger, D.R. and Berg, T. 1998. Arctic springtime depletion of mercury. Nature 394: 331-332.

Schroeder, W.H. and Munthe, J. 1998. Atmospheric mercury – an overview. Atmospheric Environment 32: 809-822.

Scott, K.J. 2001. Bioavailable mercury in arctic snow determined by a light-emitting mer-lux bioreporter. Arctic 54: 92-101.

Scott, D.P. and Armstrong, F.A.J. 1972. Mercury concentration in relation to size in several species of fresh water fishes from Manitoba and Northwestern Ontario. Journal of the Fisheries Research Board of Canada 29: 1685-1690.

Simoneau, M., Lucotte, M., Garceau, S. and Laliberte, D. 2005. Fish growth rates modulate mercury concentrations in walleye (*Sander vitreus*) from eastern Canadian lakes. Environmental Research 98: 73-82.

Snowshoe, N. and Stephenson, S.A. 2000. Results of the 1999 Peel River fish contaminant study. Fort McPherson Renewable Resource Council, Fort McPherson, NT. 14 p.

SPSS Inc. Released 2008. SPSS Statistics for Windows, Version 17.0. Chicago: SPSS Inc.

Stafford, C.P. and Haines, T.A. 2001. Mercury contamination and growth rate in two piscivore populations. Environmental Toxicology and Chemistry 20: 2099-2101.

Stafford, C.P., Hansen, B. and Stanford, J.A. 2004. Mercury in fishes and their diet items from Flathead Lake, Montana. Transactions of the American Fisheries Society 133: 349-357.

Steffen, A., Schroeder, W., Macdonald, R., Poissant, L. and Konoplev, A. 2005. Mercury in the Arctic atmosphere: an analysis of eight years of measurements of GEM at Alert (Canada) and a comparison with observations at Amderma (Russia) and Kuujjuarapik (Canada). Science of the Total Environment 342: 185-198.

Stern, G.A. and Macdonald, R.W. 2005. Biogeographic provinces of total and methyl mercury in zooplankton and fish from the Beaufort and Chukchi Seas: Results from the SHEBA drift. Environmental Science & Technology 39: 4707-4713.

Stern, G.A., Macdonald, R.W., Outridge, P.M., Wilson, S., Chetelat, J., Cole, A., Hintelmann, H., Loseto, L.L., Steffen, A., Wang, F. and Zdanowicz, C. 2012. How does climate change influence arctic mercury? Science of the Total Environment 414: 22-42.

Stewart, D.B., Taptuna, W.E.F., Lockhart, W.L. and Low, G. 2003. Biological data from experimental fisheries at special harvesting areas in the Sahtu Dene and Metis settlement area, NT: Volume 2. Lakes near the communities of Colville Lake, Fort Good Hope, Norman Wells, and Tulita. Canadian Data Report of Fisheries and Aquatic Sciences 1126: viii + 101 p.

Sturludottir, E., Gunnlaugsdottir, H., Jorundsdottir, H.O., Magnusdottir, E.V., Olafsdottir, K. and Stefansson, G. 2014. Temporal trends of contaminants in cod from Icelandic waters. Science of the Total Environment 475-477: 181-188.

Swanson, H., Gantner, N., Kidd, K.A., Muir, D.C.G. and Reist, J.D. 2011. Comparison of mercury concentrations in landlocked, resident, and sea-run fish (*Salvelinus* spp.) from Nunavut, Canada. Environmental Toxicology and Chemistry 30: 1459-1467.

Swanson, H.K. and Kidd, K.A. 2010. Mercury concentrations in Arctic food fishes reflect the presence of anadromous Arctic charr (*Salvelinus alpinus*), species, and life history. Environmental Science & Technology 44: 3286-3292.

Swanson, H.K., Kidd, K.A. and Reist, J.D. 2010. Effects of partially anadromous Arctic charr (*Salvelinus alpinus*) populations on ecology of coastal Arctic lakes. Ecosystems 13: 261-274.

Tingley, M.P. and Huybers, P. 2013. Recent temperature extremes at high northern latitudes unprecedented in the past 600 years. Nature 496: 201-208.

Townsend, L.D. 1942. The Occurrence of Flounder Post Larvae in Fish Stomachs. Copeia 1942:126-127.

Tran, L., Reist, J.D. and Power, M. 2014. Total mercury concentrations in anadromous Northern Dolly Varden from the northwestern Canadian Arctic: A historical baseline study. Science of the Total Environment. Advance online publication. doi: 10.1016/j.scitotenv.2014.04.099

Tran, L., Reist, J.D. and Power, M. in review. Life-history dependent variation of total mercury concentrations in Northern Dolly Varden from the Babbage River, Yukon Territory, Canada. Science of the Total Environment.

Trudel, M. and Rasmussen, J.B. 2006. Bioenergetics and mercury dynamics in fish: a modelling perspective. Canadian Journal of Fisheries and Aquatic Sciences 63: 1890-1902.

Tutiempo. 2014. Weather Canada. <http://www.tutiempo.net/en/> downloaded July 20, 2014.

U.S. Environmental Protection Agency. 2007. Method 7473: mercury in solids and solutions by thermal decomposition, amalgamation, and atomic absorption spectrophotometry. <http://www.epa.gov/osw/hazard/testmethods/sw846/pdfs/7473.pdf > downloaded August 18, 2011.

van der Velden, S., Reist, J.D., Babaluk, J.A. and Power, M. 2012. Biological and life-history factors affecting total mercury concentrations in Arctic charr from Heintzelman Lake, Ellesmere Island, Nunavut. Science of the Total Environment 433: 309-317.

van der Velden, S., Dempson, J.B., Evans, M.S., Muir, D.C.G. and Power, M. 2013a. Basal mercury concentrations and biomagnification rates in freshwater and marine food webs: Effects on Arctic charr (*Salvelinus alpinus*) from eastern Canada. Science of the Total Environment 444: 531-542.

van der Velden, S., Dempson, J.B. and Power, M. 2013b. Comparing mercury concentrations across a thirty year span in anadromous and non-anadromous Arctic charr from Labrador, Canada. Science of the Total Environment. Advance online publication. doi: 10.1016/j.scitotenv.2013.11.147.

van der Velden, S., Evans, M.S., Dempson, J.B., Muir, D.C.G. and Power, M. 2013c. Comparative analysis of total mercury concentrations in anadromous and non-anadromous Arctic charr (*Salvelinus alpinus*) from eastern Canada. Science of the Total Environment 447: 438-449.

Van Walleghem, J.L.A., Blanchfield, P.J., Hrenchuk, L.E. and Hintelmann, H. 2013. Mercury elimination by a top predator, *Esox lucius*. Environmental Science and Technology 47: 4147-4154.

Ward, D.M., Nislow, K.H., Chen, C.Y. and Folt, C.L. 2010. Rapid, efficient growth reduces mercury concentrations in stream-dwelling Atlantic salmon. Transactions of the American Fisheries Society 139: 1-10.

WHO. 2004. Safety evaluation of certain food additives and contaminants. WHO Food Additives Series 52: 1-648.

WHO. 2007. Evaluation of certain food additives and contaminants. WHO Technical Report Series 940: 1-104.

Wootton, R.J. 1998. Ecology of teleost fishes. Dordrecht, Boston: Kluwer Academic Publishers. Vii + 386 pp.

Zar, J.H. 2010. Biostatistical analysis. Upper Saddle River, N.J.: Prentice Hall.