EFFECTS OF CLIMATE CHANGE ON MERCURY CONCENTRATIONS IN ARCTIC CHAR (*SALVELINUS ALPINUS*) IN THE HIGH ARCTIC

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ABSTRACT

EFFECTS OF CLIMATE CHANGE ON MERCURY CONCENTRATIONS IN ARCTIC CHAR (*SALVELINUS ALPINUS*) IN THE HIGH ARCTIC

Nikolaus Gantner, MSc
University of Guelph, 2008

Advisors:
Professor Keith Solomon, PhD
Professor Derek Muir, PhD
Professor Michael Power, PhD
Professor Dr. Roland Psenner

This thesis is an investigation of linkages of climate change and mercury concentrations in landlocked Arctic char and underlying food webs in Canadian Arctic lakes. Although the neurotoxin mercury is globally present in all environments, temporal and spatial trends in aquatic biota are often inconsistent, or do not exist for remote environments. Knowledge gaps include the influence of abiotic factors, possibly climate, and food web parameters on mercury accumulation in top predators. Furthermore, anthropogenic and natural mercury present in the environment are not well differentiated. To address these research needs, we collected landlocked Arctic char from 27 systems and food web organism from 18 lakes, and recorded abiotic lake characteristics expected to influence mercury cycling. Total mercury (THg) and monomethylmercury (MeHg) concentrations, and stable isotopes of carbon ($\delta^{13}C$), nitrogen ($\delta^{15}N$), and mercury ($\delta^{x}Hg$) were determined. Relationships among these measures and with environmental characteristics were investigated.

A temporal trend of mercury in Arctic char (muscle tissue) from the largest lake in the Canadian High Arctic from an existing dataset and newly collected char was investigated. Two abundant char morphotypes from Lake Hazen differed in mercury concentrations, and the resulting variability was accounted for using a statistical adjustment. Mercury concentrations did not change over the 16-year period (1990-2006), which is consistent with nearby measurements of gaseous elemental mercury at Alert (Nunavut, Canada).

Food chain lengths (FCL) of 18 lakes were characterized, biomagnification of mercury was investigated using $\delta^{15}N$, and results compared among regions. Food web biomagnification was confirmed using trophic magnification factors (TMFs) in 18 Arctic char bearing lakes (TMFs 3.5 – 64.3). Benthic coupling was apparent from char $\delta^{13}C$ signatures that resembled $\delta^{13}C$ in chironomid midges. TMFs and FCL partly explained THg in adult char, but no latitudinal or other climatic trend was apparent. Mercury concentrations in adult Arctic char were positively related to catchment-to-lake area, and intra-regional, and regional differences in Hg in Arctic char were determined. No climatic factor we investigated was found to affect Hg in char directly; however, predicted climatic changes could affect THg in char indirectly through changes in the catchment (such as melting of permafrost).

Stable isotope ratios of mercury revealed region specific mass-independent fractionation (MIF) of up to ~ 4.5 ‰ in Arctic char, which did reflect sediment ratios with a constant offset (MIF-factor). High MIF in zooplankton possibly reflected mercury assimilated from the water column. Char from Lake Pingualuk, known pelagic feeders, had high MIF, possibly reflecting an atmospheric Hg signal.

PREFACE

This thesis has been organized as a series of manuscripts. Chapter 2 has been accepted for publication in Environmental Toxicology and Chemistry and is currently in press. Chapters 3, 4, and 5 are in intended for submission to Environmental Toxicology and Chemistry (Chapter 3, 4), and Environmental Science and Technology (Chapter 5) . A general introduction (Chapter 1) and overall conclusion (Chapter 6) bracket the research chapters. As a result, some repetition of introductory and methodological material could not be avoided. Specific contributions of individual co-authors are highlighted where necessary; otherwise, co-authorship does represent intellectual contribution to the publication. All manuscripts listed below were written by Nikolaus Gantner, as indicated by primary authorship. Editorial comments and points of discussion were contributed by Drs Keith R. Solomon, Derek Muir, Michael Power, and Roland Psenner.

Chapter 2: Gantner N, Power, M, Babaluk, J, Reist, JD, Köck, G, Lockhart, WL, Solomon, KR, and Muir, DCG 2009a. Temporal trends of mercury, cesium, potassium, selenium, and thallium in Arctic Char (*Salvelinus alpinus*) from Lake Hazen (Nunavut): Effects of trophic position, size and age. Environmental Toxicology and Chemistry (*in press, accepted Aug 14 08*).

- J Babaluk and JD Reist (both Freshwater institute, Winnipeg, MB, Canada) determined the ages of Arctic char, and provided comments on the manuscript
- G Köck (Austrian Academy of Science) assisted in the collection of Arctic char.
- Mercury data and fish biodata from 1990 and 1992 were provided by WL Lockhart (North-South Consulting, Winnipeg, MB, Canada)

Chapter 3: Gantner, N, Power, M, Lawson, G, Iqaluk, D, Meili, M, Köck, G, Borg, H, Sundbom, M, Solomon, KR, and Muir, DCG; 2009b; Mercury Concentrations in Landlocked Arctic char (*Salvelinus alpinus*) from the Canadian High Arctic: Part I – insights from trophic relationships in 18 lakes; *in prep for submission to* Environmental Toxicology and Chemistry.

- G Lawson (NWRI, Burlington, ON) conducted the MeHg analysis in biota
- D Iqaluk assisted in char and food web collection at 15 sites between 2005-2007
- M Meili (Stockholm University, Sweden) provided TNW99 Arctic char THg and stable isotope data, and zooplankton samples for THg and MeHg analysis
- G Köck aided in field collections 2005 and 2007
- H Borg, M Sundbom (both Stockholm University, Sweden) made TNW99 Arctic char data (HB) and zooplankton samples (MS) available

Chapter 4: Gantner, N, Muir, DCG, Power, M, Reist JD, Babaluk, J, Iqaluk, D, Michaud, W, Meili, M, Köck, G, Dempson JB, Borg H, Hammar J, and Solomon, KR; 2009c. Mercury Concentrations in Landlocked Arctic char (*Salvelinus alpinus*) in the Canadian High Arctic: Part II – Spatial comparison of 27 populations; *in prep for submission to* Environmental Toxicology and Chemistry

- JD Reist (Freshwater Institute, Winnipeg) aided char sampling of 2007, TNW99
- J Babaluk (Freshwater Institute, Winnipeg) coordinated age-determination of char, and provided logistical support
- D Iqaluk assisted in char and food web collection at 15 sites between 2005-2007

- W Michaud assisted in fish dissections (2007) and stable isotope analysis
- M Meili, H Borg, M Sundbom (see Chapter 3)
- G Köck provided gill nets and aided in char collections and dissections 2001, 2005-2007
- JB Dempson supplied 2007 char samples from Labrador (3 lakes)
- J Hammar (Institute of Freshwater Research, Drottningholm, Sweden) made TNW99 Arctic char data available

Chapter 5: Gantner, N, Hintelmann, H, Zheng, W, and Muir, DCG; 2009d; Fractionation and variations of mercury isotopes in Arctic lake food webs; *in prep for submission to* Environmental Science and Technology.

- All MC-ICP-MS work was conducted in the laboratory of H Hintelmann (Trent University, ON, Canada), who also provided comments on the manuscript
- W Zheng (Trent University, ON, Canada) aided with analytical method, and commented on the manuscript

ACKNOWLEDGMENTS

The privilege to set foot into the Canadian Arctic is tremendous; greater still is being able to follow one's passions in this remote and pristine region. Dr. Derek Muir, thank you for this fantastic opportunity. Your always patient mentorship and thoughtful advice guided me throughout this great experience. Debbie and Pilipoosie Iqaluk who assured my safe return from three Arctic trips should be thanked next. *Nakurmiik*, Debbie, I learned a lot from you!

I thank my advisors, Dr. Keith Solomon (UoG) and Dr. Michael Power (UW), for your academic guidance and scientific input over the past years. Gratefully, Dr. Roland Psenner agreed to serve as a distant advisor for a second time (after my M.Sc.). Dr. Günter Köck established the connection with Derek [aka *Great Northern Muir* (S)] in 2003, which lead to my arrival in Ontario.

I owe Dr. Jim Reist and John Babaluk (FWI, DFO Winnipeg) for invaluable support and contributions to my thesis, and also for taking interest in my extracurricular activities and interests. At CCIW, Xiaowa was Xiaowa, Greg put the 'Me' in MeHg, and Jessica was 'total-ly' helpful. All three greatly contributed to this thesis by turning random samples into accurate data. Thank you!

Timely advice on collaborative efforts came from Dr. Markus Meili (ITM, Stockholm), defying all time zones – thank you! I was fortunate and humbled to be invited by Prof. Dr. Shinsuke Tanabe to speak at the CEMS International Symposium in Matsuyama in 2006. Thank you, Prof. Tanabe, for an exquisite experience in Japan. Dr. Reinhard Pienitz (U. Laval) and Dr. Sonja Hausmann (U. Arkansas) involved me in the Pingualuk Lake study, which proved to be an excellent scientific endeavour.

I thank all folks at Polar Continental Shelf Project for making my expeditions to the field safe, work at the Resolute Bay research station comfortable, and *yammie* (Helen and Karen - *Ulaku*!). I further thank: Bill Lyall and family for hosting us in Cambridge Bay during our 2006 sampling; all helicopter and Twin pilots for safe landings; all Parks Canada wardens at Quttinirpaaq National Park for their dedicated support and work; two anonymous *Tim Hortons* employees in Ottawa for saving our 2005 samples by storing them in their freezer; all ArcticNet ASA members (05-06).

In Guelph, I am grateful to Virginia, Joy, Linda, Ruth, and Debbie for their administrative work at EVB. I wholeheartedly thank the unselfish bunch that helped me following the tragic events of October '04. Chris and Christine, Richard B., Amanda, Derek, Mike, and Keith enabled me then to stay on track – thank you. Domo aurigato, Hisa, you are a great friend and teacher of the Japanese culture. Rich and Greg, thanks for hosting two great B-day bashes and countless other events. Ted, Andrea W., Jared, Ashley, Francois, Kelly, Tim C., Martin, Ève, Rebecca, and Jordana have greatly enriched my life, as have others (they know who they are). Tricia and Jordan, who would have thought I would ever be in a pumpkin carving contest – *as* the pumpkin!? Great rants and frequent reality checks were provided by Sabrina, thank you! The Sushi-wolves, '*alpha*'-Shawn, Colin, and Jeff made Bur(b)lington truly more enjoyable. In Waterloo, Wendy made long days elbow deep in fish guts actually fun. Craig and Cora, despite being chemists *and* at U of T, repeatedly assured that morning sessions of SETAC conferences were sparsely attended.

'Dr T.' (Tobias) and Laura, you kept my feet on the ground many times, I owe you in so many ways. Enduring friendship and support despite increasing geographic distance came from Roman, Markus, Klaus, Andi, Kle x 2, Lilly, and Christian, who all humbled me with visits. So did my charming brother Peter! Weekly phone calls from my now 89yr old Oma, Helene Gantner, kept me up-to-date – wie kann ich Dir nur danken?! The first fishing rod of my life was a present from my Opa, Dr. Heinz Zillig, while first training in fish dissection was provided by his late wife, Paula. Eferdinger Oma, Du fehlst mir.

Finally, I am taking a deep bow in highest gratitude and full respect in front of my beloved mother Ursula. Uschi, you have raised me and Peter on your own, after our father Gerhard deceased in an avalanche accident in 1979, a mere 6 months after I was born. Even in the most difficult days of your life, you selflessly assured only the best for us, eventually allowing us an up-bringing without too much worry. Gerd ist stolz auf Dich.

Thank you, Nakurmiik, Merci, Aurigato, Tack, Danke

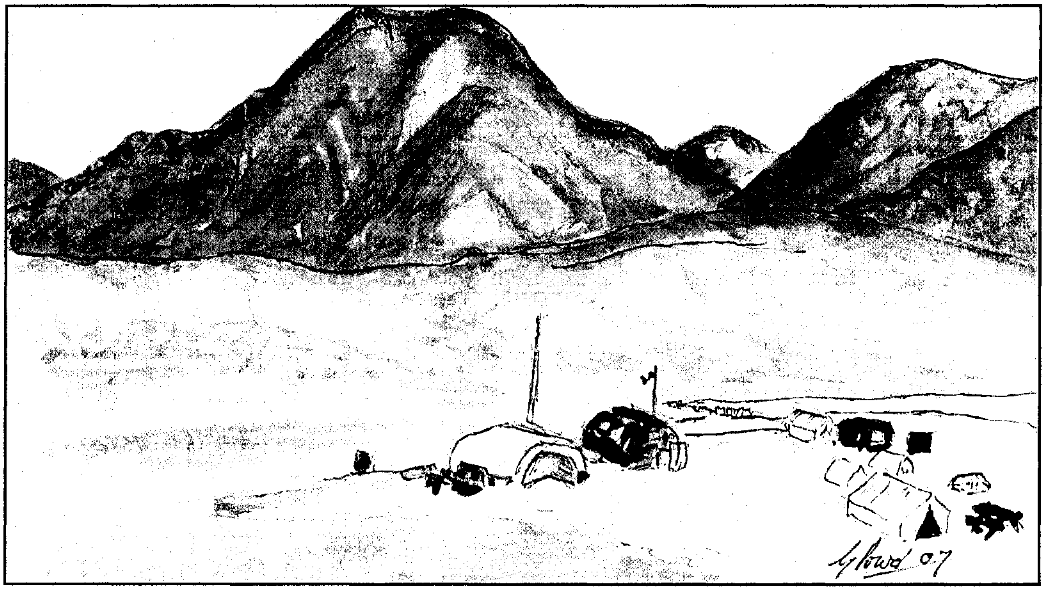

Figure 1: "*Hazen camp*" by Prof. Dr. Geoffrey Power, August 2007. The foreground shows the research facilities at Quttinirpaaq National Park's largest lake, Lake Hazen (82°N), Nunavut, Canada. Mt. Umingmak (Inuktitut for Muscox) is featured in the backdrop, as the viewer looks North. The painting was presented to me by the artist at the camp in 2007, for which I would like to thank Dr. G. Power by including it herein.

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1 INTRODUCTION AND LITERATURE REVIEW

Evidence for links between climate warming and mercury (Hg) accumulation in Arctic lake food webs is currently lacking. Mercury is a heavy metal that is bioaccumulative in its methylated form. Concentrations of Hg in the North American Arctic are increasing in some marine and freshwater biota, such as *Salvelinus alpinus* L. (Arctic char). It is expected that climate warming may increase mobilization and bioavailability of mercury in the Arctic. Global circulation models predict that major warming will occur over the next 70 years in the Canadian Arctic with increases in annual average temperatures of 3-5°C (ACIA 2004).

This thesis reports the results of studies on the transfer of mercury through food webs in High Arctic lakes, and broadens past investigations (e.g., Köck 2004; Muir et al. 2005) of temporal and spatial trends of mercury concentrations in Arctic char. Field studies in selected lakes were performed over a period of three summers (2005-2007). Archived samples were analyzed for mercury to determine temporal trends and establish possible links to climate change in the High arctic ecosystem.

The following introduction to the thesis reviews the current literature on mercury in aquatic ecosystems with a focus on Arctic freshwater environments, identifies research needs, and presents study objectives. The questions arising from identified knowledge gaps and the hypotheses derived from them are presented. Finally, the studied sites, materials, methods, and endpoints used to test the thesis hypotheses are outlined. The results are then presented in four research chapters (Chapters 2-5) with supporting discussion. A general discussion of the overall results, as well as conclusions from the work concludes the thesis.

1.1 Stressor characterisation

1.1.1 Chemistry of mercury

The heavy metal mercury, element number 80 in the periodic table, exists in various forms in the environment. The word “mercury” is derived from the Latin words “hydrargyrum” or “hydrargentum” which both mean liquid silver (Baird 1995). Mercury's atomic mass is 200.59 grams per mole and its specific gravity is 13.5 times that of water. Mercury has a melting point of -38.9°C, a boiling point of 357.3°C, and is the only metal to remain in liquid form at room temperature (Figure 2). Mercury is referred to as “elemental” or “metallic” mercury, expressed as Hg(0) in its pure form. At room temperature, it appears as a liquid, shiny, silver-white metal and its vapours are odour- and colour-less. In the environment, it is usually found as a component of other compounds or in inorganic salts as monovalent (Hg(I) or divalent (Hg(II) mercury. Mercury exists as a metallic vapour and liquid/elemental mercury, bound in mercury-containing minerals, as ions in solution, or bound to ionic compounds. It also appears as soluble ion complexes, as gaseous or dissolved non-ionic organic compounds or bound to inorganic or organic particles by ionic, electrophilic or lipophilic adsorption (U.S. EPA 1997).

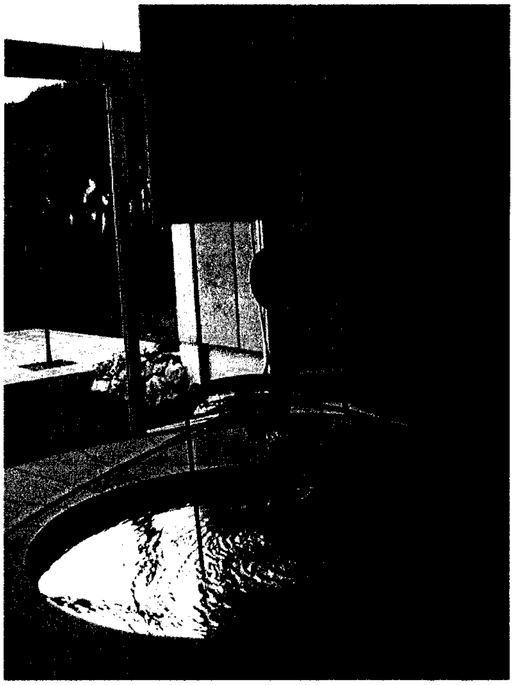

Figure 2. Alexander Calder’s mercury fountain at the Fundació Joan Miró in Barcelona, Spain. Liquid mercury is replacing water in this display. Picture by Finlay McWalter, Common Source licensed (Version 2.5).

Mercury is the most volatile of all metals, and its vapour is potentially toxic, with thresholds limit value (TLV) of 0.025 mg/m^3 (http://www.osha.gov/SLTC/healthguidelines/mercuryvapor/recognition.html). However, its liquid form is less toxic, as it is less readily uptaken. Its ability to form amalgams (compounds or solutions with other metals) is, or has been, used in various ways. In dental amalgam fillings, mercury was used until recent studies revealed that it was actually lost in small amounts from the filling over time, in part due to mechanical abrasion, and as a consequence can effect human health (Satoh 2000).

In nature, mercury has three possible conditions of electrical charge, or valence states. Elemental mercury (Hg^0) is a neutral species. Mercury is also found in two positively charged, or cationic, states, Hg^{2+} and Hg^{1+}. The mercuric cation is more stable and is generally associated with inorganic molecules, such as sulfur (in the mineral cinnabar), chlorine (mercuric chloride), oxygen and hydroxyl ions (Environment Canada 2004). It can also form organometallic species, such as monomethyl mercury or dimethyl mercury. These compounds are usually formed by sulfate-reducing bacteria under anaerobic conditions in sediments or soils.

Mercury has seven stable isotopes (^{196}Hg, ^{198}Hg, ^{199}Hg, ^{200}Hg, ^{201}Hg, ^{202}Hg, and ^{204}Hg), ratios among which have recently been investigated for their potential in source identification (Ridley and Stetson 2006; Bergquist and Blum 2007; Blum and Bergquist 2007; Jackson et al. 2008). Improvements in high precision multi-collector inductively coupled plasma mass spectrometry (MC ICP MS) allow measurements of heavier (~80 amu) isotopes. Detection of mass-dependent and mass-independent fractionation (MDF and MIF, respectively) of mercury isotopes is possible, owing to its chemical characteristics (e.g., active redox chemistry, ~ 4% difference in atomic mass) (Young et al. 2002; Ridley and Stetson 2006; Bergquist and Blum 2007). The mechanisms underlying this fractionation are thought to be a combination of the nuclear volume effect (NVE) (Bigeleisen 1996; Schauble 2007) and magnetic isotope effect (MIE) (Buchachenko et al. 2007; Jackson et al. 2008). MIF appears associated with Hg isotopes of odd mass numbers (^{199}Hg and ^{201}Hg). Experiments have demonstrated MIF and MDF of up to 5 ‰ (Kritee et al. 2007) through microbial activity and in redox experiments (Bergquist and Blum 2007). Both MDF and MIF have also been detected in nature where abiotic and biotic environmental processes can alter stable isotope ratios of mercury (Kritee et al. 2007; Ghosh et al. 2008). Understanding where fractionation occurs in the environment may be useful in linking mercury in the environment to its source (Jackson et al. 2004; Bergquist and Blum 2007), as isotopes may unravel a mercury 'fingerprint'.

1.1.2 Use of mercury

The unique properties of mercury have been exploited extensively by mankind throughout history. Mercury was first found in Egyptian tombs dating back 3500 years. It has important roles in the occult arts and human folklore and has been used in medicine, science and technology for millennia (Environment Canada 2004). More recently, mercury has been used by the mining industry to extract and refine gold and other metals (e.g., zinc) or has been mined as the main product from ores. Due to its unique properties (e.g., of being liquid at room temperature), and its ability to conduct electricity, mercury is widely utilized. Today, elemental mercury is used in small-scale mining of gold and silver, chlor-alkali production, in manometers, thermometers, electrical switches, fluorescent lamps, and amalgam fillings. Its compounds are, or have been, used in batteries, as biocides in the paper industry, or on seed grains, pharmaceuticals, and paints. It has also been used by the automobile industry in light switches (e.g., interior lights or trunk illumination), but the use was banned for environmental reasons. Across most of Asia, mercury is still used in a wide range of industrial processes. Asian and African emissions have even increased throughout the 1990s (Pacyna et al. 2006). Recent reports identify China and other Central and South-Eastern Asian countries as producers and emitters of large quantities of mercury into the atmosphere, while North American and European emissions have declined (Pacyna et al. 2006). Coal fired power plants are believed to be the largest source of atmospheric mercury in North America, followed by garbage incineration (UNEP 2002).

1.1.3 Health effects

1.1.3.1 Human toxicology

Mercury can adversely affect humans. The toxicity of mercury depends on the form of mercury present and the route of exposure to the body. Methylmercury is the most toxic form present. Once in the body, it can function as a neurotoxin hindering neuronal development, or cause adverse effects on the kidney. A review article by Satoh discusses the toxicity of mercury and its compounds (Satoh 2000). Tragically, the toxicity of mercury was brought to public attention when hundreds of lives were lost in Minamata (Japan) in the 1950s and 1960s. A local plant producing polyvinyl chloride, using Hg^{2+} as a catalyst, polluted Minamata Bay by releasing large amounts of mercury–containing residues. The diet of the local populations consisted largely of fish caught in the bay, with high fish consumption leading directly to mercury toxicity through dietary exposure. Since this tragedy, the symptoms of mercury poisoning are referred to as Minamata disease by the medical community. In the mid 1950s in Iraq, methylmercury was used as a fungicide on seeds subsequently used to manufacture bread flower instead of being planted. The resulting mercury poisoning led to the death of hundreds. In Canada, on the

Grassy Narrows First Nation in north-western Ontario in the 1970s, aboriginal fishing guides consuming large quantities of fish caught in the mercury polluted English River (Ontario) and were found to show signs of mercury poisoning (Wheatley 1979).

Exposure routes for humans can include inhalation of mercury vapour. When inhaled, mercury diffuses from the lungs into the bloodstream, crosses the blood/brain barrier, and enters the brain where it can cause damage to the central nervous system. Symptoms include impaired coordination and eyesight, and loss of tactile senses. Methylmercury usually enters the human body through diet. Once ingested, it enters the bloodstream via intestinal uptake. When concentrations in bloodstreams of pregnant woman are high, the development of the fetus can be affected (Baird 1995).

1.1.3.2 Exposure of northern aboriginal people and Inuit populations:

The continued consumption of indigenous food by the Inuit is necessary for three primary reasons: it is more nutritious than processed foods (from southern Canada), it is more economical than store brought food, and the sharing of indigenous food is critical to the reinforcement of social relationships (AMAP 1998; AMAP 2003; Fisk et al. 2003). Dietary of Inuit and other indigenous populations across the Arctic have been well studied (Wein et al. 1991; Wenzel 1991; Freeman et al. 1992; Kuhnlein 1995; Kuhnlein et al. 1996; Van Oostdam et al. 1999). Studies have concluded that while diet varies from season to season, region to region, community to community, and individual to individual, fish were a significant portion of the diet and hence a significant source of Hg exposure in all cases. The diet of local Inuit populations in the region of my study partly consists of fish of which Arctic char is the most heavily utilized species. Large anadromous Arctic char are the preferred form of char used, but landlocked char populations are also utilized. Where Lake trout (*Salvelinus namaycush* W.) and Lake

Whitefish (*Coregonus clupaeformis* M.) exist, both species are also harvested for consumption.

This thesis focuses mainly on the mechanisms of mercury accumulation and the temporal trends of mercury in Arctic char. However, some information is provided with respect to the potential exposure of humans to Hg (Section 6.3.2).

Although the prevailing theory concerning the uptake of methylmercury in humans has been challenged recently (Harris et al. 2003), the literature presents convincing evidence that elevated concentrations of mercury in humans are related to the consumption of contaminated organisms. The findings of Harris et al. (2003), however, suggest that a more detailed characterization of the mechanisms of mercury uptake is needed. Temporal trends of methylmercury concentrations in blood samples from young Inuit and First Nations persons in Canada have slowly decreased as shown by Dallarie et al. (2003). The current understanding of neuropathogenesis of mercury toxicity is summarized in Aschner and Walker (2002).

1.2 *Mercury in the environment*

The environmental mercury cycle has four strongly connected compartments – atmospheric, terrestrial, aquatic, and biotic (Figure 3). The atmospheric compartment is dominated by gaseous elemental mercury (Hg^0), while Hg^{2+} dominates the fluxes between the aquatic and terrestrial compartments. The terrestrial compartment is dominated by Hg^{2+} sorbed to organic matter in soils. The aquatic compartment is dominated by Hg^{2+}-ligand pairs in water and Hg^{2+} in sediments, and the biotic compartment is dominated by methyl mercury. Mercury is reactive in the environment and cycles readily among compartments (Hoffman et al. 2003; Wiener et al. 2003).

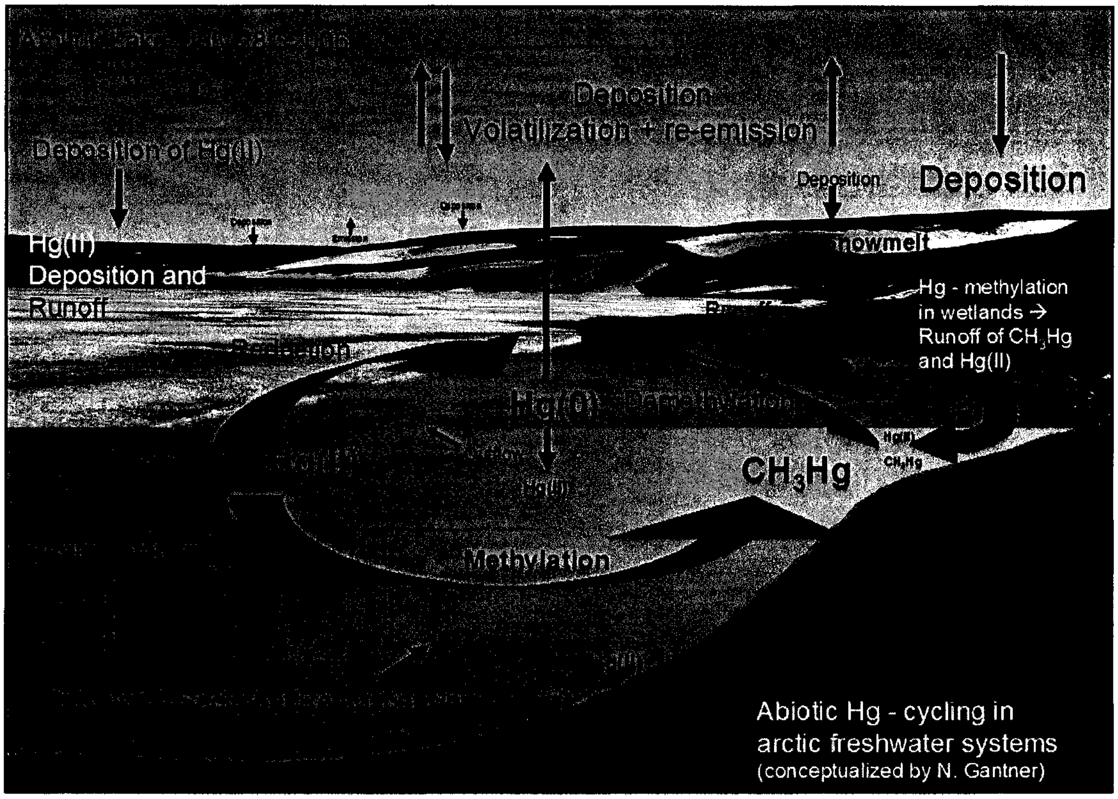

Figure 3. Mercury cycling pathways in aquatic environments are very complex. The various forms of mercury are converted from one to the next. Most important is the formation of methylmercury, the most toxic form. Ultimately, mercury ends up in the sediments, fish and wildlife, or recycles back to the atmosphere by volatilisation.

1.2.1 Mercury in the global context

Because it is an element, mercury is not biodegradable, and although its form and availability to living organisms may change over time, mercury persists in the environment. In their review of atmospheric mercury contamination, Fitzgerald et al. (1998) concluded that the knowledge of global mercury cycling has improved over the last decade, but that further high quality studies concerning the emissions, chemical speciation, and reactivity of mercury in the environment were needed. Most recently Lindberg et al. (2007) have identified progress since then, uncertainties, and research needs specifically related to the attribution of sources. The authors emphasized the importance of linking anthropogenic emission sources to studies of exposure in people, wildlife, and terrestrial and aquatic systems.

Mercury occurs in the environment in rocks and soils in its various inorganic forms. It is naturally released into the environment by weathering and enters (aquatic) ecosystems in low concentrations. This "background" signal (Fitzgerald et al. 1998) must not be underestimated or left out, particularly when attempting to interpret low concentrations of mercury. Knowledge of the underlying bedrock of studied sites will help the interpretation of results (Lindberg et al. 2007).

Global anthropogenic sources are described in the UNEP Global Mercury Report (UNEP 2002). An updated global mercury budget for 2000 is now available (Pacyna et al. 2006). The authors concluded that the global emissions have remained constant since 1990, although some continents (North America and Europe) reduced emissions, while others (Asia and Africa) have increased their emissions. Anthropogenic, rather than natural sources are at present more important in the mercury cycle, as current concentrations exceed (pre-industrial) background levels (Slemr and Langer 1992;

Pacyna and Pacyna 2002; Pacyna et al. 2003; Pacyna et al. 2006; Harris et al. 2007; Lindberg et al. 2007).

1.2.2 Mercury in the Arctic ecosystem

Atmospheric deposition of elemental mercury is believed to be the main entry pathway of mercury into Arctic lakes and other remote lakes throughout the Northern Hemisphere e.g., Landers et al. (1998), Fitzgerald et al. (1998; 2005), and Muir et al. (2008). Long range atmospheric transfer of airborne elemental mercury from anthropogenic sources such as industries in North America, Europe, and Asia is believed to be responsible for elevated concentrations in the Arctic. Direct linkage of contamination effect to this source is more difficult in the case of the distance sources that affect the Arctic, than in the case of near-source contamination. Nevertheless, global sources of anthropogenic mercury emissions have been identified (e.g., coal fired power plants, garbage incineration) and restrictions on emissions have been established and have resulted in reduced mercury emissions (Schuster et al. 2002; Pacyna et al. 2006). Schuster and co-workers used ice-cores from the Upper Fremont Glacier (Wyoming, U.S.A.) determine atmospheric deposition and the associated temporal trend over a period of 270 years. They described a maximum in the mid 1980s, but noted that in the last 10 years deposition has declined. Other work by Muir et al. (2003; 2008) in the Canadian Arctic, Kamman and Engstrom (2002), and Perry et al. (2005) (in north-eastern USA) have all shown declining fluxes of mercury in recent sediments from isolated lakes.

The fractionation of stable isotopes of Hg has been proposed as a novel approach to track mercury in the environment and, potentially, develop means to attribute it to a source (Jackson et al. 2004; Bergquist and Blum 2007; Lindberg et al. 2007). Evidence of fractionation of Hg in fish and other biota has recently been presented by several groups

(Jackson et al. 2006; Bergquist and Blum 2007; Jackson et al. 2008; Sonke et al. 2008). The method is not yet developed enough to identify the specific sources of Hg in a given receptor, but tracking of stable isotope ratios of Hg through environmental compartments is a promising tool towards this goal.

Mercury contamination in Arctic environments is not a new issue. Awareness of the problem was first raised in the late 1970s (Lockhart et al. 1992; Rasmussen 1994; Van Oostdam et al. 1999; Lockhart et al. 2005). In Canada, monitoring programs have since been established by the Federal Government (Northern Contaminants Program (NCP), http://www.ainc-inac.gc.ca/ncp/index_e.html; and Northern Ecosystems Initiative (NEI), http://www.mb.ec.gc.ca/nature/ecosystems/nei-ien/index.en.html). Various recent reports on studies designed to assess mercury contamination agreed on the importance of monitoring, modeling, and cautious interpretation of mercury concentrations in Arctic ecosystems (Dietz et al. 1998; UNEP 2002; AMAP 2005).

1.2.3 Mercury cycle in Arctic aquatic systems

Several transformations of mercury occur between its entry into aquatic systems through deposition in the water (as shown in Figure 3) and bioaccumulation along food chains. Input of mercury from anthropogenic sources to aquatic systems of the Arctic occurs mainly through deposition of atmospheric Hg^{2+}. Precipitation in the form of snowfall and rain are key means by which mercury reaches the systems either immediately or during snowmelt via run off in warmer periods of the year (Loseto et al. 2004; Semkin et al. 2005). Factors affecting inputs and the fate of elemental mercury (e.g., DOC, light attenuation) to Alaskan arctic lakes have been investigated recently (Tseng et al. 2004; Hammerschmidt and Fitzgerald 2006; Hammerschmidt et al. 2006). Local atmospheric Hg deposition has been shown to be correlated with the concentration in wild fish from southern lakes (Hammerschmidt and Fitzgerald 2006). Hammerschmidt et al. (2006) further suggest that the mercury depletion events (MDEs) described by Schroeder et al. (1998) and Lu et al. (2001) may increase deposition of atmospheric mercury to arctic lakes and catchments. During polar sunrise, sunlight interacts with bromine emitted from the arctic ocean to form bromine oxide, which then oxidizes gaseous elemental mercury Hg(0) to Hg^{2+} which is then deposited on the surface of snow, open lakes, or the ocean. The MDEs seem to be closely linked with stratospheric ozone depletion events (Ebinghaus et al. 2002). The net deposition of MDE, and thus its contribution to the cycling of Hg, is under debate, as there is also evidence that deposited Hg is reemitted to the atmosphere (Kirk et al. 2006).

Once Hg^{2+} has entered the aquatic system, it can follow various pathways and undergo transformations (see Figure 3). In the water column, it is adsorbed to phytoplankton and sinks to the bottom of the lake where it can be biotransformed (methylated) by sulfate-reducing bacteria. Methylation of mercury to methylmercury by bacterial communities in lake sediments is the point of entry into the food chain (Wren 1986). Once methylated, the resulting organic methylmercury compound is transferred, accumulated, and magnified up the food chain/web. The following sections discuss the fate and behavior of mercury in the Arctic aquatic environment.

A recent mini-review by Barkay and Poulain (2007) summarizes the biogeochemical cycle of mercury with specific focus on polar environments. Sediments of the lakes are considered hot-spots of methylation and therefore of great interest when characterizing the entry of mercury into the food chain. Methylation of Hg^{2+} occurs in the top layers of the sediments. A variety of microorganisms, particularly methanogenic (methane producing) and sulfate-dependent bacteria are mediators for this transformation

(Barkay and Poulain 2007). Conversion of Hg^{2+} to methyl mercury occurs under anaerobic conditions found in sediments, primarily in low pH environments with high concentrations of organic matter.

Low concentrations of methyl mercury in surficial sediments indicate that the fate of biotic Hg(II) and methyl mercury is different. Most biotically-produced methyl mercury is demethylated rather than buried in lake sediments (Watras et al. 1998). The same authors further conclude from observations in Wisconsin lakes that passive uptake of methyl mercury does not control bioaccumulation at the base of aquatic food webs in nature (i.e., phyto- and bacterio-plankton). Correlation of methyl mercury with pH and dissolved organic carbon (DOC) reflect the supply and bioavailability of methyl mercury to lower trophic levels (Ravichandran 2004); methyl mercury concentrations at higher trophic levels reflect uptake at low trophic levels and other factors, such as diet and growth. Findings in lakes in northern Ontario indicate a close linkage of DOC and methylmercury in biota (Harding et al. 2006). Although Arctic and particularly High Arctic lakes are considered to have ultra low DOC levels, the relationship between methylmercury and DOC must be considered (Wiener et al. 2003; Ravichandran 2004).

Sediments in Arctic lakes have been studied to reveal temporal trends in the atmospheric deposition of mercury (Fitzgerald et al. 1998; Lockhart et al. 1998; Muir et al. 2003). Recent results of mercury trend analyses in sediment cores from Alaska, Scandinavia, Russia, and northern Canada can also be found in Landers et al. (1998), Lucotte et al. (1995), and Muir et al. (2008) and indicate that concentrations and fluxes ([Hg]*sedimentation rate) of mercury in Canadian Subarctic and Arctic areas have declined in the 1990s (Hermanson 1998; Outridge et al. 2005) as in Sweden (Bindler et al. 2001) and mid-latitude lakes in North America (Engstrom and Swain 1997; Lockhart et al. 1998; Lorey and Driscoll 1999).

1.3 Characterizing the Arctic ecosystem

Knowledge of the prevailing abiotic and biotic conditions in the Arctic is necessary to understand what factors may influence a given ecosystem. The extreme environmental conditions of polar ecosystems include low temperatures, low light intensity, and limited liquid water inputs (Murray 1998). The presence of anthropogenic stressors, although often less intense than in Southern regions, put additional constraints on Arctic ecosystems. In relation to the biomagnification of contaminants in these systems, low productivity and cyclic productivity are also important.

1.3.1 Abiotic and biotic characteristics of Arctic Lakes

Figure 4: Candled ice near Hazen camp, Ellesmere Island in August 2007. *Picture © N. Gantner.*

The depressions in the landscape of the High Arctic, the basins of today's lakes, were carved out by retreating glaciers during the Pleistocene epoch. Following the retreat of the icecap, the land began to rise (isostatic rebound) and is still rising today, forming new lakes where gouged land rises out of the ocean (Peltier 2001; Peltier 2004). Consequently, lakes closer to the existing coastline are younger than those inland (Welch and Legault 1986). Many coastal High Arctic lakes and all Greenland lakes are ice-dammed (Van Hove et al. 2006). Many of these ice-dammed lakes have recently been lost due to climate warming (Prowse et al. 2006). About 20% of the central Canadian Arctic is covered with large numbers of small lakes. A number of hydro-electric dams have been built along major rivers in southern Arctic regions. The ice-free time in these lakes is very short and how long lakes remain open depends on temperature (mainly in June) and wind speed. The melting process involves gradual thinning and weakening, as is described in the AMAP report (Murray 1998), leading to candled ice formation (Figure 4). Solar radiation is responsible for ice and snow melting but, by the time lakes are ice-free, the intensity of solar radiation has already started to decline (Murray 1998). As a result, freshwater bodies do not warm up and the occurrence of thermal stratification is limited to shallow or wind protected lakes. The more southern Arctic lakes have greater warming and stratification than High Arctic lakes. Mixing of the water body in High Arctic lakes results mainly from wind. In larger High Arctic lakes (above 75°N) water temperatures rarely exceed 4-6°C (Van Hove et al. 2006). Water temperatures in small or shallow lakes, which also lose their ice quicker than large lakes, usually rise to 10°C, with a maximum of 15°C (AMAP 1998).

After air temperatures begin to drop in September, lakes lose heat quickly (Van Hove et al. 2006). As they are fully mixed, the whole water body often reaches temperatures near 0°C before ice begins to build up (Van Hove et al. 2006). Formation of ice continues through the winter months until May. Shallow lakes (under about 2 m) freeze to the bottom. On average, the ice cover (first year ice) of deeper lakes is around 2 m thick. Thus, Arctic lakes are frozen or partly frozen 9-12 months a year (Welch 1991).

High Arctic lakes are considered ultra-oligotrophic, their species diversity is low, their nutrient pathways are short, and the associated food webs are simple compared to southern lakes (Rigler 1978). Since lakes are often ice covered most of the year, light is very limited, resulting in low primary production. The nutrients needed for photosynthesis (i.e., P, N, and Si) are also low in concentration. As oxygen concentrations may also be low during the winter in shallow lakes, winterkill of fish has been reported for lakes in the High Arctic with mean depths of <1.8 m (Welch and Bergman 1985; Murray 1998). Such lakes are generally only occupied by fish on an ephemeral basis. Hydrology and its influence on contaminant transport to Arctic lakes and water bodies is summarized in the AMAP report (Brooks et al. 2005), and Arctic Climate Impact Assessment (ACIA) report (Wrona et al. 2005).

The limiting characteristics in High Arctic regions affect diversity and abundance of species. There are only a few species of small single-cell algae present, often large in numbers, which dominate low-nutrient lakes (Michelutti et al. 2003). These include chrysophytes and cryptophytes, but also a number of diatoms and dinoflagelate species. Much of the primary production during the short annual light conditions occurs on the bottom of the lake. For example benthic moss was described to be present in Char Lake, Cornwallis Island, Nunavut (Klaff and Wetzel 1971).

Zooplankton abundance is limited through nutrient limitation (bottom up control), low temperatures and poor feeding conditions (Hobson and Welch 1995). However, presence of zooplankton groups, such as rotifers, copepods, and cyclopoids are related to primary production, in that greater primary production through phytoplankton allows for greater zooplankton abundance. As prey, zooplankton represents an important source of food for all young Arctic char, and certain adult char populations depend on this food source (Knudsen et al. 2006). Lakes that do not support a fish population thus have a more abundant and more diverse zooplankton community (Smol et al. 2005). The importance of trophic position of zooplankton in relation to its mercury concentrations was recently described for marine food webs by Campbell et al. (2005). More specific descriptions of zooplankton communities can be found in the AMAP report (Murray 1998).

As one would expect, the benthic communities (zoobenthos) are also low in species diversity and biomass. They are characterized by a high variation of species among lakes, depending on abiotic factors of each lake. In fish-bearing High Arctic lakes, chironomid species are often the dominant group of benthic invertebrates present (Gajewski et al. 2005). They are important in the food web where they play a key role in nutrient and contaminant transfer to higher trophic levels (Sierszen et al. 2003). This strong benthic coupling in oligotrophic systems was also demonstrated for subalpine lakes (Vander Zanden et al. 2006). Water mites (Acari, *Hydracarina sp.*) are commonly found in char stomachs, but their role in the transfer may be limited, due to their presumably small contribution to the char diet. In High Arctic systems, Arctic char thus prey mostly on detritivorous invertebrates (emerging chironomid pupae) and zooplankton, receiving contaminants (mercury) through this pathway. Lakes in the southern areas of the Arctic often host more invertebrate groups, such as Cladocera, Phyllopoda, Trichoptera, Ephemeroptera, Notostraca, Oligochaeta, Ostracoda, Hydracarina, or Nematoda (Jørgensen and Eie 1993).

Fish species present in southern Arctic lakes may consist of Atlantic salmon, lake trout, several whitefish, sticklebacks (three-spine and nine-spine), and anadromous and non-anadromous Arctic char. In lakes north of McClure Strait, Barrow Strait and Lancaster Sound (~75°N) in the Canadian High Arctic, the area of focus in this thesis, Arctic char is often the only fish species present. Migrating or resident populations, differences in morphotypes, as well as feeding habits, have also been reported and are summarized in Power et al. (2008) (Section 1.5 herein).

Because species present in the Arctic aquatic systems are well adapted to the extreme conditions, but are 'stenök' (*german,* stenophile), they are vulnerable to environmental change. Due to the short food chain and few alternative nutrient sources, changes, such as temperature or other abiotic factors can directly affect life in these systems with resulting changes in movement and concentrations of contaminants.

1.4 Climate change

1.4.1 Trends, predictions, models

Global climate change is of concern for human health, economies, and countries around the world in multiple ways. Effects of climate change on aquatic systems in southern regions of Canada, specially the boreal forest, have been studied through long term monitoring, modeling or ecosystem manipulations (Schindler 1997). For the Arctic, the most comprehensive recent climate impact assessments are the ACIA report (ACIA 2005) and Intergovernmental Panel on Climate Change (IPCC) report (IPCC 2007). Both ACIA and IPCC propose warming trends with regard to Canadian Arctic environments and identify potential stressors and effects on Canadian Arctic freshwaters based on their observations. The ACIA predicts warming of up to 4°C by 2100 across the Arctic and Subarctic region of North America based on modeling of a doubling of CO_2 concentration in the atmosphere. The IPCC report predicts, among many other changes, anomalies in mean summer air temperature of >1 °C in the northern latitudes, as well as probable changes in precipitation (IPCC 2007). Paleoclimate records for the last 400 years presented by Overpeck et al. (1997) show changes in circumpolar trends and increasing variability in climate patterns. Moritz et al. (2002) described dynamics and effects of recent climate change in the Arctic and predicted rising mean annual temperatures as one of many environmental changes in the future. Comprehensive summary reports on the state of the Arctic, such as the three above-mentioned (AMAP 2003; ACIA 2005; IPCC 2007), clearly indicate the lack of climate change effects studies in the Arctic. The results presented by the IPCC have since triggered inclusion of studies in environmental and socioeconomic changes in the Arctic in the Speech from the Throne in the parliament of Canada in 2007. Research initiatives such as ArcticNet (http://www.arcticnet-ulaval.ca) and the co-occurring International Polar Year (IPY 2007/08) are examples for current initiatives investigating many facets of changes in the Arctic, including the effects on aquatic systems.

1.4.2 Regional effects of climate change

There is an emerging body of evidence on how climate warming will affect High Arctic lakes and rivers. These lakes and river are believed to be sensitive sentinels of climate change in their freeze, melt, and hydrological cycles (Wrona et al. 2006; Bates et al. 2008). Significant trends have been demonstrated by Magnusson et al. (2000) and Semilentov et al. (2000), revealing earlier ice break up (6.5 days per century) and delays in freeze-up (5.8 days per century) over a 150 year period. This can be translated into an increase in temperature of 1.2°C per century. As these characteristics of melt/freeze-up change, pathways through which contaminants enter the lakes could be altered (Macdonald and al. 2000; Fisk et al. 2003; Macdonald et al. 2005) and result in higher accumulation of contaminants in biota. Recent evidence for effects of climate warming comes from diatom records obtained from Arctic lakes (Smol et al. 2005). Changes in species assemblages after 1987 and greater diatom species diversity found in recent years could be caused by reduced ice cover and/or a longer growing season (Michelutti et al. 2003; Van Hove et al. 2006). An increase in mercury inputs to the lakes could consequently increase the amount available to lower trophic level biota, and ultimately increase the mercury concentrations in Arctic char. Greater water temperatures increase mercury bioaccumulation in fish, as demonstrated in a feeding experiment in the late 1970s (Cember et al. 1978). The authors also stated that higher temperatures would have a greater effect on mercury concentrations in fish than those in the water.

1.5 Biology of Arctic char (Salvelinus alpinus)

1.5.1 Geographic distribution

The distribution limits of Arctic char have been influenced by geological and climatic events during the Pleistocene period (Scott and Crossman 1973; Crossman and McAllister 1986). With few exemptions (Baikal Lake area, Banks Island, and Alaska), the current range of Arctic char was ice covered during the Pleistocene glaciation. Present northern limits of distribution in Canada are described for Arctic char: Landlocked populations are known to exist in lakes near Alert (82° 31' 4" N) at the north end of Ellesmere Island; Anadromous Arctic char have been caught slightly further North, in Rambow Lake at 83° 00' N, 75° 27' W (J. Babaluk, Department of Fisheries and Oceans, Winnipeg, personal communication); Southern population limits are dispersed, with Eurasian populations in the European Alps (45° N) and headwaters of Lake Baikal (Parker and Johnson 1991), and North American populations extending as far south as Maine, US (Kircheis 1989).

1.5.2 Arctic char in the High Arctic

Arctic char belong to the salmonid family and represent the top aquatic predator in most High Arctic lakes, while co-existence with lake trout (*S. namaycush*) is common in southern Arctic regions (Fraser and Power 1984). Char can reach up to 15 kg and individuals reach up to >30 years in age. In lakes of the High Arctic, it is the only abundant fish species (Hammar 1989). Two partly linked ontogenetic life-cycle characteristics have been reported to occur in char in High Arctic lakes, polymorphism and landlocked versus anadromous behavior. Dietary shifts from benthic to pelagic to piscivorous feeding (cannibalism) have been reported (Hammar 2000; Svenning and Borgström 2005). Previous studies of Arctic char populations focussed on morphological traits, life history traits, genetics, or migratory patterns (Nordeng 1961; Parker and Johnson 1991; Malmquist et al. 1992; Reist et al. 1995; Brunner et al. 2001; Jonsson and Jonsson 2001; Adams et al. 2003; Klemetsen et al. 2003). To determine whether a population is anadromous or landlocked, radio tracking (Babaluk et al. 2001) and elemental analysis (Halden et al. 1995; Halden et al. 2000) have also been used.

1.5.2.1 Polymorphism – the 'char problem'

The occurrence of different forms of Arctic char, also known as 'the char problem', in Arctic lakes is long known and has been described by several authors (Johnson 1980; Nordeng 1983; Parker and Johnson 1991; Reist et al. 1995; Riget et al. 2000; Gulseth and Nilssen 2001), and has also been reported in other, more southern environments (O'Connell and Dempson 2002). Nordeng (1983) offered a solution to 'the char problem' based on rearing experiments using a population in Norway, stating that phenotypic plasticity allows char to segregate into all three forms (anadromous, small and large resident). A summary of historical reports of sympatric stocks can be found in Johnson (1980). The distribution of sympatric Arctic char populations is circumpolar, mainly between 45° N and 82° N but also in alpine lakes. The number of different forms

Figure 5: The large morphotype (top) and the small morphotype (bottom) of landlocked *Salvelinus alpinus* from Lake Hazen, Ellesmere Island, Nunavut.
Picture © N. Gantner

present in a lake seems to be dependent on the size and depth of the water body, with more morphotypes (up to 4) in larger and deeper lakes with more complex habitat structure (Riget et al. 2000; Jeppensen et al. 2001). The presence of at least two morphotypes in Lake Hazen on Ellesmere Island has been confirmed (Reist et al. 1995; Guiguer et al. 2002) (Figure 5), and a potential third (benthic dwarf) morphotype was collected in 2007 (J. Reist, unpublished data).

1.5.2.2 Migratory and non-migratory Arctic char

Patterns of anadromy in Arctic char are described in McDowell et al. (1987), and are believed to be correlated with productivity of aquatic systems (Gross et al. 1988). It is believed to be selected for when the benefits of migration (more food available in the sea) outweighs the physiological cost (Babaluk et al. 1997). Anadromous behavior is usually detected by observation of upstream or downstream migration. Lipid-rich anadromous char, where abundant, are key features of Indigenous people's diet across the Arctic (Figure 6). Differences in distribution of strontium (Sr) in char otoliths have been used to confirm migratory behavior. Because Sr concentrations in the ocean water are greater, uptake by fish is greater and the difference can be detected in the layers of deposition in the otoliths. This approach, based on scanning proton microprobe analysis, was used by Babaluk et al. (1997) on Arctic

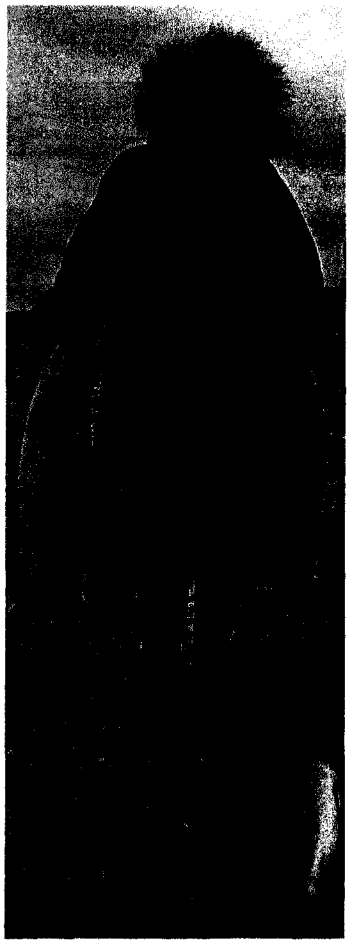

Figure 6: Anadromous Arctic char caught by local fishermen near Cambridge Bay, Nunavut
Picture © N. Gantner

char from Lake Hazen to demonstrate that the population is non-migratory. Tagging studies have also successfully shown whether char are resident or migratory (Babaluk et al. 2001). Non-migrating landlocked char are typically smaller in average size than their migrating counterparts. Stable isotopes of carbon and nitrogen have also been used to examine anadromy (Doucett et al. 1999).

1.5.3 Ontogenetic dietary shifts

The previously described morphotypes within a lake are also distinguishable through their stomach contents. Dietary uptake plays a crucial role in contaminant exposure. Fish feeding on macro-zoobenthos show different concentrations of mercury accumulation than those feeding on pelagic zooplankton (Power et al. 2002b). Ontogenetic shifts in diet from litoral to pelagic or from pelagic zooplankton feeding to piscivory and cannibalism are discussed by Griffiths (1994) and Riget et al. (2000). The coupling of pelagic and benthic food web components of Greenland lakes have been described by Jeppensen et al. (2001). Cannibalism and discrete bimodality of Arctic char populations in Sweden are discussed in Hammar (2000). The authors conclude that the dietary shift to cannibalism is an important cause of the bimodality. Finstad and co-authors (2003) implied that biased sampling using gill-nets is possibly causing artefacts that can be interpreted as bimodal size distributions. However, stable isotope analysis has been used to confirm bimodality in Scottish (Adams et al. 2003), Welch (McCarthy et al. 2004), Norwegian (Gulseth and Nilssen 2001), and Canadian freshwater systems (Guiguer et al. 2002).

Changes in diet during the life-cycle of the fish could be important information when using trophic position to analyze differences in uptake of mercury. Trophic position in the food web is important in terms of contaminant accumulation (Power et al. 2002b;

Gantner et al. 2009a). The higher an organism is in the food chain, the greater the concentrations of bioaccumulative contaminants (i.e., mercury) (Trudel and Rasmussen 2001). Stomach content analysis has been carried out to determine differences in feeding, but these reflect only the current spectrum of diet. Stable isotope ratios ($\delta^{13}C$, $\delta^{15}N$) analysis has been used successfully to distinguish life history of char (Guiguer et al. 2002), and can also be a powerful tool to investigate contaminant accumulation (Power et al. 2002b) (Section 1.7). Models to explain population dynamics and size-structured populations have recently been suggested and discussed by Parker (2001) and Claessen (2002). Both authors use landlocked populations of Arctic char in Arctic lakes as a model organism. Holarctic phylogenetic analysis of mitochondrial DNA sequences of Arctic char have been conducted by Brunner (2001) to determine genetic differences between populations.

1.6 Accumulation of mercury in Arctic char

The uptake of mercury (inorganic and organic mercury) by fish occurs mainly through the diet (Hall et al. 1997). As the top predator, Arctic char accumulate mercury via their prey (e.g., chironomids, other char). Subsequently, mercury concentrations in char depend on bioaccumulation (uptake over time) and biomagnification (related to trophic position) of methylmercury. Several recent studies report mercury concentrations in Arctic char for populations in North America and the Canadian Arctic (Borg et al. 2001; Köck 2004; Campbell et al. 2005; Evans et al. 2005; Muir et al. 2005), and for High Alpine and Norwegian lakes reported by Rognerud et al. (2002). In landlocked Arctic char, mercury concentrations are higher than in anadromous fish (Campbell et al. 2005). The changes in environmental conditions, feeding and subsequently growth when entering the ocean, leads to dilution, and therefore lowers mercury concentrations in anadromous population. Recent investigations indicate that mercury concentrations in fish in most Arctic lakes have not declined, and have increased in some lakes (Fisk et al. 2003; Muir and Köck 2003; Muir et al. 2008). Declines of mercury concentrations in fish (yellow perch) have been reported for temperate lakes (Hrabik and Watras 2002; Drevnick et al. 2008). All mercury found in landlocked Arctic char in these remote lakes ultimately originates from sources (geogenic or atmospheric deposition) within the watershed boundaries, making landlocked Arctic char a good indicator species for changes of processes in watershed or deposition of mercury.

1.7 Determination of trophic relationships in lake food webs

Stable isotope ratios of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) are widely used to determine habitat use and the trophic position of a species (Vander Zanden and Rasmussen 2001; Post 2002). Carbon isotopes ($\delta^{13}C$) are used to determine the source of C-uptake; with fractionation of ~0.1 ‰ (Fry and Sherr 1984), it can be used to distinguish carbon utilization, and thus habitat use by species (REF). Nitrogen ($\delta^{15}N$) can be used to determine trophic position, where fractionation of ~3.4 ‰ between trophic levels is commonly observed (Post 2002; Jardine et al. 2006). Investigating both isotope ratios simultaneously (plots of $\delta^{13}C$ versus $\delta^{15}N$) leads to maximized information on trophic position within a given system (Peterson and Fry 1987). Recently this method was applied to determine intra-species differences in $\delta^{13}C$ and $\delta^{15}N$ ratios and relate them to different morphotypes of Arctic char (Guiguer et al. 2002; Power et al. 2002a). Resolving the number of morphotypes and relating their mercury concentration could be a valuable addition to the knowledge on mercury transfer through the food web (Power et al. 2002b; Adams et al. 2003).

To investigate trophic relationships in food webs, several approaches are available: food chain length (FCL) or trophic level (TL) can be calculated from $\delta^{15}N$ from isotope ratios. Both FCL and TL are thought to give valuable information on predator-prey relationships, and thus on ecosystem function, within systems (Cabana and Rasmussen 1996). FCL is calculated using the formula: $FCL = (\delta^{15}N_{top\text{-}predator} - \delta^{15}N_{baseline}) / 3.4 + \lambda$ (λ = assumed trophic level of the baseline organism), and has been utilized to compare energy transfer in aquatic systems from a large geographical range (Cabana and Rasmussen 1996; Vander Zanden and Fetzer 2007). Prior to applying this method, lake food webs should be thoroughly characterized (e.g., plots of $\delta^{13}C$ versus $\delta^{15}N$), as the selection for the proper baseline for calculation of FCL is vital for ecotoxicological applications (Jardine et al. 2006; Vander Zanden and Fetzer 2007). TLs are calculated using the formula: $TL = 2 + (\delta^{15}N_{consumer} - \delta^{15}N_{zooplankton}) / 3.4$, where 2 is the assumed TL of zooplankton, and 3.4 is the trophic enrichment factor constant.

Merging the stable isotope and contaminants analysis has become a powerful tool to quantify contaminant transfer through food webs. Uses and limitations of this method in ecotoxicology are presented in a recent review by Jardine et al. (2006). Briefly, FCLs have been used to explain contaminant transfer through food webs, using $\delta^{15}N$ to infer predatory prey relationships (Cabana and Rasmussen 1994; Kidd et al. 1995; Vander Zanden and Rasmussen 1996; Atwell et al. 1998; Kidd 1998; Hoekstra et al. 2003). Trophic magnification factors (TMF) are calculated based on the antilog value of the regression slope between concentrations and TL (Jardine et al. 2006). TMFs have most recently aided Guildford et al. (2008) and Houde et al. (2008) to detect and explain biomagnification of organic pollutants.

1.8 Objectives and hypotheses

The general aim of the research described in this thesis was to examine factors affecting mercury transfer through landlocked Arctic char food webs, and to relate these factors to climate warming scenarios within the high Arctic Archipelago. Previous investigations of temporal trends in mercury concentrations in muscle of Arctic char (Köck 2004; Muir et al. 2005) were expanded upon by measuring archived and newly-collected landlocked Arctic char from a variety of High Arctic lakes, as well as the underlying food web from which tissues or whole organisms were obtained (Figure 7). In order to characterize the food web of the studied lakes, stable isotope analyses of char tissue samples, as well as all compartments of the food web, were used to determine trophic relationships within studied lakes.

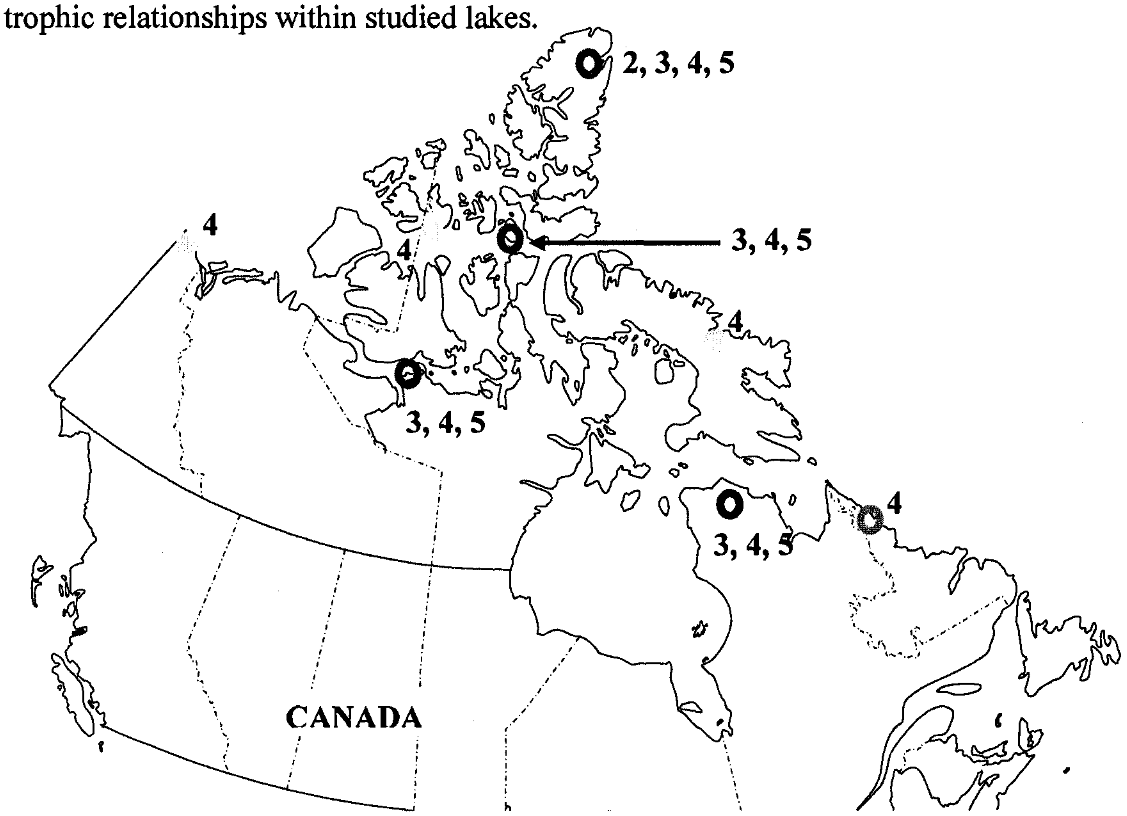

Figure 7: Approximate locations of sampling campaigns of landlocked Arctic char food webs are indicated by colored circles (multiple lakes) and dots (single lakes). Our three main sampling regions (2005-07) are indicated by red circles, blue circle indicate the Parc National des Pingualuit (2007) sites, green circle mark three Labrador locations (2007), and the three lakes added from the TNW99 expedition (1999) are marked in yellow dots. Digits on the map correspond to the chapters of this thesis, in which the site(s) appear(s). Lake names and locations are presented in each chapter. Detailed maps, photographs of lakes, and all 2005-2007 sampling dates are available in the Appendix of this thesis (Appx 1 - Appx 11, Appx 25).

Specific objectives and hypothesis of the individual research chapters are given below:

1. The objective of the research described in Chapter 2 was to describe the temporal trends of concentrations of mercury (and other elements of interest) in Arctic char muscle tissue. A mercury data set from 7 sampling collections in Lake Hazen, Nunavut, spread over a 16-year period was investigated to estimate the statistical significance of temporal trends. The trophic position, known to affect concentrations of mercury, of individual fish was taken into account.

> *The Null Hypothesis:* Mercury bioaccumulation in Arctic char from High Arctic lakes is not positively correlated with climate warming and the latter has not resulted in increases in measured mercury concentrations over time.

2. The objective of the research presented in Chapter 3 was to investigate lake food web structure and its effects on the bioaccumulation of mercury species, particularly methylmercury, in Arctic lake food webs. Concentrations of methylmercury in food web organisms (zooplankton, insects, juvenile and adult Arctic char) from 18 lakes were compared. Food web structure was analyzed and its influence on the bioaccumulation of methylmercury inferred. Trophic magnification factors were calculated and compared among lakes and regions.

> *The Null Hypothesis:* Methylmercury concentrations in food web organisms of landlocked Arctic char lakes are not positively correlated with the organism's trophic position ($\delta^{15}N$) and thus trophic level.

3. The objective of the research presented in Chapter 4 was to investigate spatial trends in the concentrations of mercury in Arctic char muscle, following suitable adjustment for co-variants (such as FCL, fish size, age, or trophic position). Mercury concentrations in Arctic char muscle tissue from 27 populations from across the Canadian Arctic were compared along latitudinal and longitudinal gradients. Results of Chapter 3 were incorporated in the analysis of spatial trends presented in this chapter.

> *Null Hypothesis:* There is no difference in mercury concentrations in Arctic char tissues from Arctic lakes along latitudinal and longitudinal gradients, with concentrations not being positively correlated along the gradient with the prevailing thermal regime.

4. The objective of the research presented in Chapter 5 was to investigate whether stable isotopes of mercury are fractionated in Arctic lake food webs. The fractionation of stable isotopes of mercury was investigated in sediments, zooplankton, insects, and Arctic char.

> *Null Hypothesis:* Stable isotopes of mercury do not undergo mass-independent fractionation during bioaccumulation in Arctic lake food webs. Therefore, Hg isotope signatures in Arctic char are not more enriched in isotopes of odd mass (^{199}Hg and ^{201}Hg) than sediments, chironomids, and zooplankton.

2 TEMPORAL TRENDS OF MERCURY, CESIUM, POTASSIUM, SELENIUM, AND THALLIUM IN ARCTIC CHAR (*SALVELINUS ALPINUS*) FROM LAKE HAZEN (NUNAVUT): EFFECTS OF TROPHIC POSITION, SIZE AND AGE

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2.1 Abstract

Arctic char (*Salvelinus alpinus* L.), the top predator in High Arctic lakes, is often used as a bioindicator of mercury (Hg) contamination in Arctic aquatic ecosystems. The present study investigated effects of trophic position, size, and age of Arctic char in Lake Hazen, the largest lake in the Canadian High Arctic (81°50'N; 70°25'W), on Hg bioaccumulation. In addition, several essential (Se, K) and non-essential elements (Tl, Cs), in char muscle tissue were examined to compare their behavior to Hg. Trophic position of Arctic char was identified by stable isotope ($\delta^{15}N$) signatures. Temporal trends of Hg from seven sampling campaigns over a 16-year period (1990 – 2006) were investigated, for the overall data and for one trophic class. Mercury concentrations were not correlated with age, but positively related to fork length and trophic position. "Large" char with greater $\delta^{15}N$ signature (> 12 ‰) had larger Hg concentrations (0.09 – 1.63 µg/g ww) than "small" char with smaller (< 12 ‰) $\delta^{15}N$ signature (0.03 – 0.32 µg/g ww), indicating that Hg concentrations increased with trophic position. Non-essential Cs and Tl showed similar relationships to age, length, and trophic position as Hg, indicating their potential to bioaccumulate and biomagnify. Essential elements Se and K did not show these relationships. Concentrations of Hg were adjusted using $\delta^{15}N$, leading to less within-year variability and a more consistent temporal trend. The $\delta^{15}N$-adjusted trend showed no decline in Hg in Arctic char from Lake Hazen (1990 – 2006) in the overall dataset and in the "small" morphotype. Trends for the same period prior to the adjustment were not significant for the overall data set, while a slight decrease was apparent in the "small" morphotype. The results confirm the need to consider trophic position and fish

size in the monitoring temporal trends of Hg particularly for species with different morphotypes.

2.2 Introduction

Deposition of mercury (Hg) on Arctic environments occurs through precipitation in the form of snow or rain, via dry-fall, and gas exchange directly from the atmosphere. Deposition of Hg may be altered, especially during Hg depletion events (St Louis et al. 2005) and/or in the presence of ultra-violet radiation. Runoff during spring snowmelt periods delivers mostly inorganic forms of mercury (Hg(0) or Hg(II)) into lakes and wetland areas within lake catchments (Loseto et al. 2004; Semkin et al. 2005) where it may be subsequently methylated by anaerobic bacteria (mainly by sulfate reducing communities in the sediment), to organic monomethylmercury (CH_3Hg+, or MeHg). MeHg can bioaccumulate in lower aquatic organisms and then biomagnify through the aquatic food web. Mercury monitoring work in Arctic predatory freshwater fish has recently been summarized (Fisk et al. 2003) and a limited number of long-term trend studies are available for sub-arctic locations (Muir et al. 2005; Evans and Muir 2006).

While reductions in industrial Hg emissions in the 1980s and 1990s in North America and Europe have resulted in decreased Hg concentrations in some fish species in temperate regions of the northern Hemisphere (Hrabik and Watras 2002), Hg concentrations in Arctic char in several High Arctic lakes have not declined and may be increasing (Muir et al. 2006). Concentrations may increase further via changes in aquatic ecosystems that are expected to alter accumulation pathways in food webs as a result of predicted climate warming (Schindler et al. 1995; Schindler 2001; Macdonald et al. 2003; Wrona et al. 2005). For example, Outridge et al. (2005; Outridge et al. 2007), found increasing Hg concentrations in lake sediments with increasing abundance of diatoms due to greater productivity, and suggested a climatic link to Hg deposition.

We hypothesize that climatic changes predicted for Arctic regions (ACIA 2005) may alter Hg loading to aquatic systems and the subsequent biomagnification and cycling of Hg within these systems. Food webs in Arctic lakes are generally short (Wrona et al. 2006), with low species diversity, and dominated by a single apex predator such as Arctic char (Johnson 1980). Landlocked Arctic char, which remain in one lake throughout their lifespan, are particularly vulnerable to changes in contaminant accumulation pathways and could thus serve as an indicator species for environmental monitoring purposes (Köck 2004). However, the interpretation of temporal trend data is often confounded by year-to-year variability in contaminant concentrations, accurate knowledge of fish trophic position, and variations in the size and age across sets of samples (MacCrimmon et al. 1983; Vander Zanden and Rasmussen 2001; Power et al. 2002b). For example, the effect of fish age and length on Hg bioaccumulation has been demonstrated for southern species, such as yellow perch (*Perca flavescens* L.), walleye (*Stizostedion vitreum* L.), lake trout (*Salvelinus namaycush* W.), and northern pike (*Esox lucius* L.) (MacCrimmon et al. 1983; Rodgers 1994; Evans et al. 2005; McIntyre and Beauchamp 2006). Concentrations of Hg are correlated to trophic position in lacustrine (Kidd et al. 1995) and Arctic marine food webs (Atwell et al. 1998). Furthermore, biomagnification leads to increases in Hg concentrations with increasing food chain length, or higher trophic position (Cabana and Rasmussen 1994; Vander Zanden and Rasmussen 2001) as has been demonstrated for lake trout in sub-arctic Quebec (Power et al. 2002b).

Although Hg is by far the most studied heavy metal contaminant of aquatic ecosystems, similar concerns exist for other potentially accumulative metals, in particular:

Selenium (Se), Cesium (Cs), Thallium (Tl), and Potassium (K). Cs and Tl are non-essential elements, while Se and K are essential to vertebrate metabolism. The pharmacology of essential metals is different from the non-essential metals as organisms have specific uptake and excretion processes to maintain a normal amount in the body (Windisch 2002). Campbell et al. (2005) have recently identified Rubidium (Rb) as a bioaccumulative element in Arctic char from Lake Hazen and demonstrated higher concentrations in piscivorous versus insectivorous char. Selenium is known to accumulate in food webs (Orr et al. 2006) and may interact with Hg (Mason et al. 2000; Belzile et al. 2006) to form stable mercuric selenide, SeHg, as discussed by Ikemoto et al. (2004), thereby influencing Hg accumulation. Radioactive Cs-137, as well as Cs/K ratios have been used as an indicator of trophic position in the past (Mearns et al. 1981). Potassium, which should be regulated by homeostasis, may provide a useful means of normalizing other elemental concentrations, if it is consistent over time and trophic position (Mearns et al. 1981). Finally, Tl is of interest because of its similarity to Hg in terms of its biomethylation potential and subsequent accumulation dynamics (Peter and Viraraghavan 2005).

Here, we used age and length of fish from seven collections of char conducted in Lake Hazen between 1990 and 2006, $\delta^{15}N$ data, and concentrations of total Hg, Se, Cs, Tl and K to investigate: First, which of the three factors (age, size, trophic position) best explains the variability in contaminant tissue concentrations; second, which of the factors can be used to statistically adjust the overall dataset of elemental concentrations and thereby reduce the year to year variability of Hg and accumulating elements; and third, whether trends of Hg concentrations particular to morphotype exist.

2.3 Materials and Methods

2.3.1 Study site

Lake Hazen (81°50'N; 70°25'W) is located in Quttinirpaaq National Park on Ellesmere Island, Nunavut, Canada. The lake, with an area of 537.5 km^2 (Inland Water Directorate 1973) and an estimated volume of $5.1x10^{10}m^3$ (G. Köck, Austrian Academy of Science, Vienna, Austria, unpublished data), is the largest lake in the Canadian High Arctic. It hosts a simple aquatic community dominated by Arctic char (Johnson 1983). Arctic char is the only fish species present in the lake (Babaluk et al. 1997; Babaluk et al. 2001) and, although there is an outlet to the sea, the char appear to be non-anadromous (Guiguer et al. 2002). Evidence from recent $\delta^{15}N$ isotope analysis (Guiguer et al. 2002) and size-at-maturity analysis (J. Babaluk, Department of Fisheries and Oceans, Winnipeg, MB, Canada, unpublished data; the present study) shows that up to three morphotypes of char are present, a "large" pelagic cannibal form, a "small" benthic insectivore, and possibly a benthic dwarf form (J. Reist, Department of Fisheries and Oceans, Winnipeg, MB, Canada, pers. comm.). Supporting water chemistry data were collected from 1990 – 1998 (Babaluk 1999) and more recently (1996 – 2003) by Environment Canada (Halliwell and Knight 2004).

2.3.2 Sample collection

Arctic char samples used in the present study were from fish captured between May and early August of 1990 (*n*=45), 1992 (*n*=20), 2001 (*n*=20), 2003 (*n*=20), 2004 (*n*=16), 2005 (*n*=7), and 2006 (*n*=22) by either angling through the ice, or with the aid of gill nets (36 - 42 mm mesh). A strict catch limit of 20 Arctic char per year is in effect at Lake Hazen for conservational purposes since 2001. Char were collected each year within 1 km of the Parks Canada Hazen camp (81° 49' N, 71° 19' W), except for samples from

2005 samples, when char were collected near the only outflow, the Ruggles River (81° 48' N, 70° 27' W). In 1990, 1992, and 2001 samples were frozen in the field and transported to Fisheries and Oceans Canada (Freshwater Institute, Winnipeg, MB, Canada) for processing. From 2003 – 2006 all char were dissected in the field shortly after capture. Fork length (nearest cm), weight (nearest g) and sex of char were recorded. Sagittal otoliths were collected for age determination. Sub-sampling of fish tissues included liver, kidney, and dorsal muscle tissue. Tissues were frozen and transported to the Canada Centre for Inland Waters (CCIW), Burlington, Ontario, Canada and stored at -20 °C until subsequent metals analyses. Collected otoliths were shipped to Fisheries and Oceans Canada (Winnipeg, MB) for ageing. Ages were determined from the otoliths using the break-and-burn method or thin section method as described by Chilton and Beamish (Chilton and Beamish 1982).

2.3.3 Multi-element analysis

All multi-element analyses were performed by the National Laboratory for Environmental Testing (NLET) at CCIW. Briefly, Arctic char muscle was sub-sampled and skinless samples were acid-digested in a high-pressure microwave oven. Total Hg was analyzed by cold vapor atomic absorption spectrophotometry (CV AAS). Char collected in 1990 were analyzed for total Hg by Department of Fisheries and Oceans (Winnipeg) also using CV AAS (Muir et al. 1993); all others by NLET. Both laboratories participated in annual interlaboratory studies of Hg, Se, and heavy metals over the time period of this study, starting in the early 1990s (results summarized in (Zhu 1997; Stokker 2003)). These interlaboratory studies demonstrated very good agreement among laboratories for the analysis of total Hg in fish tissues. The bioaccumulative elements, Se, Cs, Tl, and K, were determined by inductively coupled plasma mass spectrometry (ICP-

MS) (PQ-2, VG Elemental). Certified reference materials (DOLT-2 (dogfish liver), DORM-2 (dogfish muscle) and TORT-2 (lobster hepatopancreas; National Research Council of Canada, Ottawa ON) were analyzed for each batch of 20 samples. Results for Se and Hg averaged 95% and 129% of certified values, respectively.

2.3.4 Stable isotope analyses

Muscle from all fish collected from 1992 to 2004 was analyzed for stable isotopes of nitrogen (N) ($\delta^{15}N$) at the laboratory of L. Wassenaar (Environment Canada, National Hydrology Research Centre, Saskatoon) using a Micromass Optima (Waters, Milford, MA, USA) continuous-flow isotope-ratio mass spectrometer directly coupled to a Carlo Erba NA1500 elemental analyzer (Elemental Microanalysis, Okehampton UK). Samples from 2005 and 2006 were analyzed at the Environmental Isotope Lab (University of Waterloo, ON, Canada) using identical methods. Briefly, a small tissue sample was freeze-dried and a few milligrams ground to a fine powder prior to analysis on an isotope ratio mass spectrometer. At the Wassenaar lab, a laboratory working standard, Pharmamedium, was run every five to 10 samples for N analyses to ensure no variation existed among sample runs. Precision of the nitrogen isotope analyses was 0.4‰ (±2 standard deviations, (SD)). At the Environmental Isotope Laboratory, University of Waterloo, approximately 1 mg of ground material was used for stable isotope analyses performed on a Micromass VG Isochrom continuous-flow isotope ratio mass spectrometer connected to a Carlo Erba elemental analyzer. Machine analytical precision (0.3‰ for N) was determined by repeat analysis (*n*=25) of International Atomic Energy Agency N1 and N2 for $\delta^{15}N$.

2.3.5 Statistical analysis

The confidence level (type I error rate) for statistical significance was set to $\alpha=0.05$ for all analysis. The relationships of Hg, Se, Cs, Tl, and K with $\delta^{15}N$, fish length, and age were determined by linear regression analysis. Prior to regressing, metal concentrations were $\log_{10}$-transformed to assure skewness coefficient/SES<2, kurtosis/KES<2, and Shapiro-Wilk P value >0.01, all indicative of normality of distribution. For regression analysis, the software packages SigmaStat 3.0 and SigmaPlot 9.0 (SigmaPlot 2000) were used. Analysis of covariance (ANCOVA) was performed in SYSTAT 11 (SystatSoftware 2004) to test for trophic-, size- and age-effects over time using a general linear model (GLM). If no interaction was observed by ANCOVA (i.e., year * length, year * $\delta^{15}N$), relationships of Hg with length or $\delta^{15}N$ at each sampling time were consistent, and the resulting Least Square Means (LSM) were used to calculate adjusted Hg concentrations. This adjustment will be referred to as adjustment using ANCOVA in the paper. Tukey's Honestly Significant Differences (HSD) post-hoc test was used to determine significant differences of adjusted LSMs. For clearer graphical display, adjusted elemental LSM (±1 SD) concentrations (for Hg, Cs and Tl) for a given year were back transformed. Adjusted LSMs were regressed against year to investigate potential temporal trends and calculation of half life times from regression slopes. To estimate the power of the dataset and confirm the ANCOVA results, the PIA statistical analysis package of Bignert (2007) was also used.

2.4 Results and Discussion

2.4.1 Fish Age and Fork length

Ages were available for 148 of the 150 fish collected and these and other morphometric data are summarized in Table 1. Fork length was not closely correlated with age across the whole dataset ($r^2 = 0.122$, $p<0.001$), see Table 2, Figure 8B. This is, however, not surprising as multiple morphotypes of Arctic char with differing growth rates are present in Lake Hazen (Reist et al. 1995).

2.4.2 Total Hg concentrations

Concentrations of total Hg ranged from 0.03 to 1.63 μg/g wet weight (all 150 fish) over the entire 16-year period (Table 1). The lowest annual mean (all geomean) Hg concentration was observed in the most recent samples (2006 = 0.11 μg/g) and the highest in 1992 (0.29 μg/g). Intriguingly, large variability of Hg concentrations was observed within each year as well (Figure 8 and Table 1). This led us to further investigate of the factors that may explain the variability in the Hg concentrations.

The 75 "small" (insectivorous) char were first identified by morphology and stomach content in the field, and later classified by stable isotope signature using $\delta^{15}N$ cut off of 12 ‰ (Figure 8E). Hg concentrations in small char for each study year were less variable (0.029 – 0.324 μg/g) than the overall data. Mean concentrations of Hg in small char were less (0.108 μg/g) than the overall mean Hg (0.186 μg/g), whereas "large" char had greater mean concentrations of Hg (0.321 μg/g). The overall dataset and the subset of "small" char were then further investigated as described below.

Table 1: Age, length, and elemental data from Arctic char from Lake Hazen (Nunavut, Canada) over a 16-year period, classified into two morphotypes by $\delta^{15}N$.

Year	1990[a]		1992		2001		2003		2004		2005		2006	
n (*n* ageing)	45 (43)		20		20		20		16		7		22	
All fish														
	Avg/GM	Range	Avg/GM	Range	Avg/GM	Range	Avg/ GM	Range	Avg/ GM	Range	Avg/ GM	Range	Avg/ GM	Range
Age (years)	18.3±6.0	8.0 - 34.0	20.9±4.7	11.0 - 28.0	21.6±4.6	14.0 - 33.0	19.1±5.4	9 - 29	17.7±4.5	11 - 30	23.3±3.5	19 - 27	18.7±4.8	13 - 29
Length (mm)	400±80	277 - 580	439±92	342 - 618	454±130	290 - 683	447±11	344 - 670	460±6	370 - 630	493±12	370 - 680	359±45	310 - 462
$\delta^{15}N$ (‰)	10.9±1.7	9.7 - 14.2	11.6±2.1	7.4 - 14.1	11.5±2.1	8.7 - 14.1	11.3±1.8	8.5 - 13.5	12.4±1.7	9.1 - 13.9	11.9±2.3	8.8 - 13.9	10.0±1.8	8.2 - 13.9
Hg	0.158	0.043 - 0.463	0.288	0.111 - 0.898	0.282	0.069 - 1.630	0.163	0.06 - 0.555	0.223	0.04 - 0.561	0.278	0.072 - 0.723	0.105	0.029 - 0.400
Cs	-	-	0.005	0.002 - 0.012	0.005	0.002 - 0.01	0.006	0.002 - 0.012	0.007	0.003 - 0.011	0.005	0.002 - 0.009	0.004	0.002 - 0.014
K	-	-	4236	3500 - 5620	4064	3250 - 5110	4276	3610 - 4950	3963	3180 - 4360	3597	3330 - 3780	3741	3100 - 4010
Se	-	-	1.563	1.230 - 1.940	1.507	1.050 - 1.970	1.202	0.9 - 2.1	1.403	1.02 - 1.87	1.563	1.29 - 1.76	1.107	0.790 - 1.350
Tl	-	-	0.010	0.003 - 0.019	0.009	0.004 - 0.023	0.009	0.003 - 0.015	0.012	0.004 - 0.019	0.010	0.005 - 0.025	0.006	0.004 - 0.018
$\delta^{15}N<12$ ‰ small														
n	20 (19)		10		10		10		4		3		18	
Age (years)	17.0±5.7	8.0 - 26.0	21.7±3.6	13.0 - 26.0	20.7±4.3	14.0 - 28.0	20.2±4.3	12 - 27	19.3±3.4	16 - 24	22.3±4.2	19 - 27	19.6±4.7	13 - 29
Length (mm)	332±29	277 - 370	364±13	342 - 389	343±35	290 - 401	377±21	244 - 404	390±20	370 - 410	378±10	370 - 390	340±22	310 - 400
$\delta^{15}N$ (‰)	-	-	9.7±1.0	7.4 - 11.2	9.6±0.7	8.7 - 11.2	9.7±1.0	8.5 - 9.7	9.6±0.4	9.1 - 10.2	9.4±0.6	8.8 - 9.9	9.2±0.7	8.2 - 10.6
Hg	0.106	0.043 - 0.279	0.171	0.111 - 0.324	0.124	0.069 - 0.272	0.101	0.060 - 0.151	0.100	0.04 - 0.253	0.121	0.072 - 0.197	0.082	0.029 - 0.151
Cs	-	-	0.003	0.002 -	0.004	0.002 -	0.004	0.002 -	0.004	0.003 -	0.002	0.0024 -	0.003	0.002 -

				0.005		0.007		0.009		0.009		0.0025		0.005
K	-	-	4375	3680 - 5620	4102	3250 - 5110	4140	3610 - 4680	3819	3630 - 4280	3698	3670 - 3760	3724	3100 - 4010
Se	-	-	1.521	1.23 - 194	1.442	1.05 - 1.78	1.205	0.9 - 2.1	1.432	1.29 - 1.71	1.656	1.52 - 1.76	1.081	0.790 - 1.350
Tl	-	-	0.006	0.003 - 0.010	0.005	0.004 - 0.008	0.006	0.003 - 0.011	0.007	0.004 - 0.013	0.006	0.005 - 0.009	0.005	0.004 - 0.007
δ^{15}N>12‰ large														
n	25 (24)		10		10		10		12		4 (3)		4	
Age (years)	19.3±6.04	9.0 - 34.0	20.0±5.6	11.0 - 28.0	22.5±4.9	17.0 - 33.0	18.0±6.4	9 - 29	17.2±4.8	11 - 30	24.0±3.4	19 - 27	14.7±2.8	13 - 19
Length (mm)	462±6	371 - 580	514±73	410 - 618	564±86	388 - 683	517±121	356 - 670	484±60	400 - 630	580±90	460 - 680	443±21	420 - 462
δ^{15}N (‰)	-	-	13.6±0.4	13.1 - 14.1	13.4±0.4	12.9 - 14.1	12.8±0.5	12.1 - 13.5	13.3±0.4	12.5 - 13.9	13.8±0.2	13.4 - 13.98	13.3±0.4	13.0 - 13.91
Hg	0.216	0.087 - 0.463	0.488	0.224 - 0.898	0.638	0.166 - 1.63	0.264	0.111 - 0.555	0.291	0.180 - 0.561	0.518	0.247 - 0.723	0.334	0.229 - 0.400
Cs	-	-	0.009	0.005 - 0.012	0.008	0.006 - 0.010	0.010	0.007 - 0.012	0.009	0.007 - 0.011	0.008	0.007 - 0.009	0.010	0.006 - 0.014
K	-	-	4111	3500 - 4630	4018	3730 - 4450	4426	3770 - 4950	4009	3180 - 4360	3516	3330 - 3780	3811	3670 - 3990
Se	-	-	1.607	1.430 - 1.840	1.574	1.27 - 1.97	1.199	1.0 - 1.5	1.393	1.02 - 1.87	1.496	1.29 - 1.74	1.225	1.15 - 1.35
Tl	-	-	0.017	0.013 - 0.214	0.014	0.007 - 0.023	0.013	0.011 - 0.018	0.014	0.010 - 0.019	0.015	0.012 - 0.025	0.015	0.009 - 0.018

[a] Stable isotope data estimated based on 1992 relationship of length and δ^{15}N ($y = 19.661x - 40.16$)

Note: Averages ± 1standard deviation of age, length (fork length), δ^{15}N, and geometric means (GM) of Hg, Cs, K, Se, Tl in µg/g wet wt are given

Table 2: Linear regression parameters for relationships between Hg concentrations, $\delta^{15}N$ signature and fish length and age.

Regression	*n*	Slope	Intercept	r^2_{adj}	*p*
Length vs age	148	6.593	296.961	0.122	<0.001
$\log_{10}$Hg vs age	148	0.015	-1.031	0.042	0.007
$\log_{10}$Hg vs length	150	0.003	-1.926	0.627	<0.001
$\delta^{15}N$ vs age [a]	104	-0.024	11.801	0.000	0.566
$\delta^{15}N$ vs length $_{1992-2006}$	104	39.636	-14.512	0.611	<0.001
$\delta^{15}N$ vs $\log_{10}$Hg $_{1992-2006}$	104	0.153	-2.442	0.640	<0.001
$\delta^{15}N$ vs $\log_{10}$Hg $_{1992}$	20	0.112	-1.838	0.685	<0.001
$\delta^{15}N$ vs $\log_{10}$Hg $_{2001}$	20	0.194	-2.777	0.803	<0.001
$\delta^{15}N$ vs $\log_{10}$Hg $_{2003}$	20	0.140	-2.367	0.660	<0.001
$\delta^{15}N$ vs $\log_{10}$Hg $_{2004}$	16	0.124	-2.184	0.526	0.001
$\delta^{15}N$ vs $\log_{10}$Hg $_{2005}$	7	0.144	-2.265	0.691	0.013
$\delta^{15}N$ vs $\log_{10}$Hg $_{2006}$	22	0.147	-2.495	0.417	0.001

[a] $\delta^{15}N$ for 1990 not available

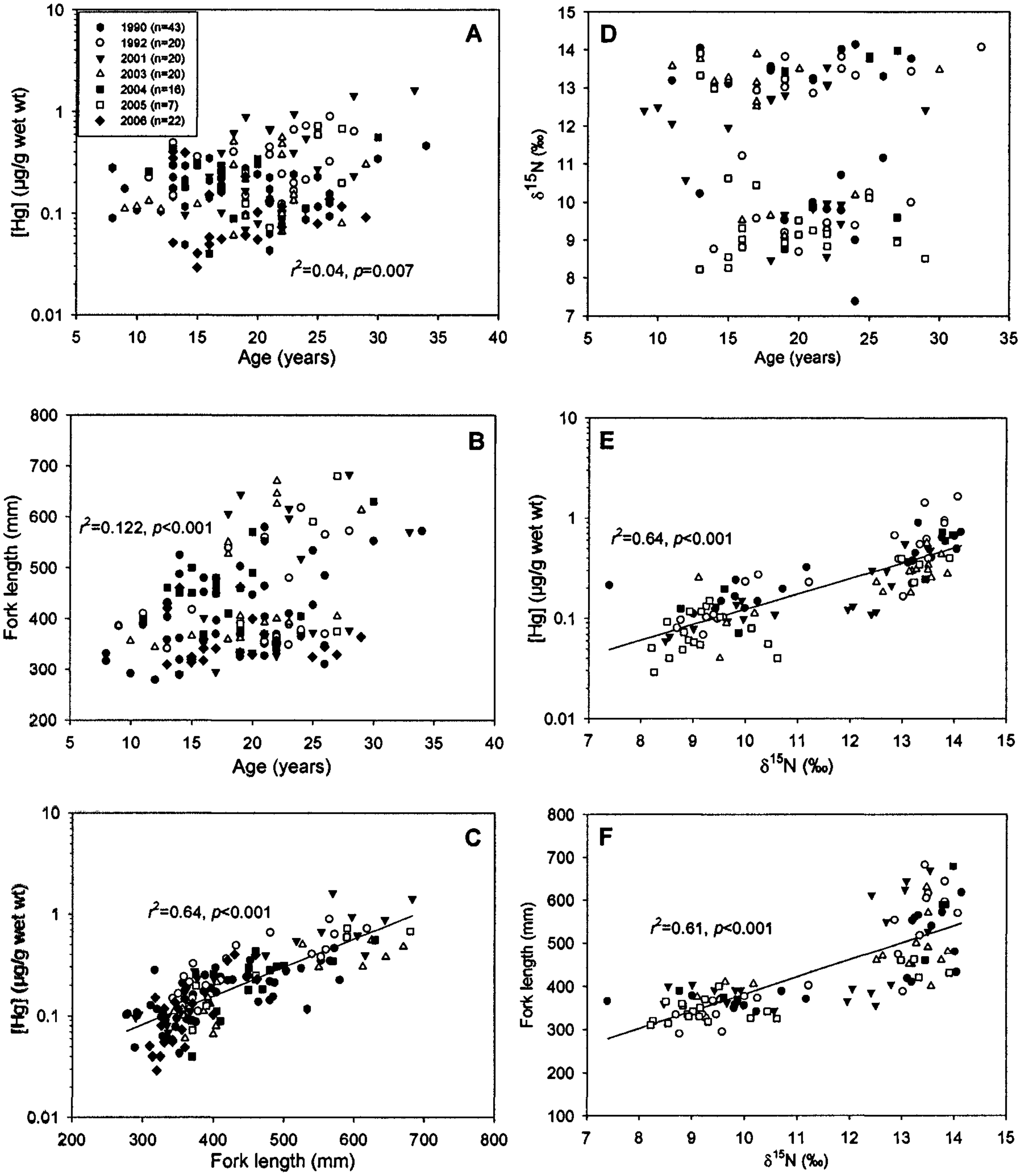

Figure 8: Linear regression plots for relationships of [Hg], age, length, and $\delta^{15}N$ of all fish collected during the study period. Symbols indicate individual fish from each sampling year, regression lines indicate significant ($p<0.05$) relationships.

2.4.3 $\delta^{15}N$ signatures

Stable isotope $\delta^{15}N$ signatures were available from 105 of 150 samples. Archived samples from 1990 (n=45) were not available; therefore $\delta^{15}N$ could not be determined. For this sampling year, a cutoff of 375 cm, derived from $\delta^{15}N$ – fork length comparison of other years was used for comparison of different morphotypes. $\delta^{15}N$ values for 1990 data were estimated from the length for char within the 1992 size range, using the strong $\delta^{15}N$ versus length relationship observed in 1992. The $\delta^{15}N$ isotopic signature ranged from 7.4 ‰ to 14.1 ‰ and the average for the overall data was 11.3 ± 2.0 ‰ (Table 1).

This wide range of stable isotope signature supported our hypothesis that feeding behavior differs among individual char, however, is this variability due to seasonal or multi-year changes in nitrogen inputs? Little is known about seasonal variability of mainly atmospheric N inputs to the Arctic. Ice-core records indicate slight increases in N deposition due to recent anthropogenic N enrichment of the atmosphere (Goto-Azuma and Koerner 2001). Benthic algae and epiphytes are the loci of the comparatively low N fixation of the atmospheric N in High Arctic Lakes (Gettel et al. 2005). Water chemistry data from recent multi-year monitoring of physiochemical parameters indicated little change over time and show Lake Hazen is a very stable system (Babaluk 1999). Ideally, the baseline N for a food web should be determined to be able to adjust for differences from year to year or site to site. In our study, baseline N and N delivery to the apex of the food chain in Lake Hazen appeared to be consistent among years during the observed period, the latter is apparent from the parallel slopes of the $\delta^{15}N$ – Hg correlations each year. Over the period from 1992 to 2006, a significant relationship between Hg and $\delta^{15}N$

was found ($p<0.001$ to $p=0.016$) overall, and regression slopes of each year were similar (Table 2).

2.4.4 Total Hg concentrations – effect of age

No significant relationship was found between age and Hg concentrations in Arctic char from Lake Hazen ($r^2=0.048$, p=0.007) (see Figure 8A). This is likely due to the presence of up to three different morphotypes of Arctic char in Lake Hazen (Reist et al. 1995). The weak predictability (Regression: $r^2=0.04$, p=0.007) of Hg concentration from char ages (Figure 8A) was also apparent from the classification in two groups by $\delta^{15}N$ (Figure 8E), where the average age of char feeding at a lower trophic level and of char feeding at a higher trophic level did not differ greatly (Figure 8D). Interestingly, in some cases, younger char feeding at a higher trophic level had higher Hg than older char feeding on lower trophic level (see Figure 8D, Table 1). Depending on the morphotype, Arctic char will grow at different rates and/or feeding behavior may vary. Age and feeding behavior are interlinked, as some species undergo dietary shifts during their first few years of life. This can result in age-related increases in contaminants, for example, modeling (Rodgers 1994) has shown that dietary shifts in young rainbow trout (*Oncorhynchus mykiss* W.) result in higher Hg concentration. In Arctic char, age could have more influence on Hg concentrations in earlier years of fish development (2 - 3 years) and/or during formation of morphotypes when dietary shifts (from planktivore to insectivore or insectivore to piscivore) lead to greater exposure to Hg. The weak correlations of age and Hg have also been reported for an Arctic char population in Resolute Lake, a small lake on Cornwallis Island (74° 41' N, 94° 57' W) (Muir et al. 2006). Because age and Hg are only weakly correlated, age could not be used as covariate in an ANCOVA to explain variability in our data.

2.4.5 Total Hg concentrations - effect of size

Fork length was positively correlated with Hg concentrations in Arctic char muscle tissue (r^2= 0.615, p<0.001) (see Figure 8C). This was expected, as a recent study by Evans et al. (2005) has confirmed this relationship in predatory fish (walleye, northern pike, and lake trout), and its usefulness to adjust Hg concentrations was demonstrated for sub-arctic systems. Consequently, we attempted the adjustment using ANCOVA with length as a covariate. However, there was a significant interaction (length*year) apparent. Thus, fork length could not be used for adjustment of Hg concentrations as the relationship between Hg concentration and length was not consistent across years.

Due to the polymorphism in this species, size of Arctic char alone is therefore not a good indicator of contaminant burden. The effects of polymorphism on the burden of PCBs has also been described for landlocked char (Hammar et al. 1993), and other fish species (Doyon et al. 1998; Berglund et al. 2001). Interestingly, Lake whitefish (*Coregonus clupeaformis* M.) show greater Hg accumulation rates in smaller, dwarf individuals. Doyon (1998) showed that this was due to higher food consumption of the faster-growing dwarf-type whitefish. For Atlantic salmon (*Salmo salar* L.), Berglund et al. (2001) showed that concentrations in $\delta^{15}N$ and PCB differed due to varying prey choice, representing three reproductive strategies.

2.4.6 Total Hg concentrations - effect of $\delta^{15}N$

Investigation of our regression plots (Figure 8E and Figure 8F) revealed at least two distinct groups of char in terms of $\delta^{15}N$ and size (fork length). Isotopic signatures of one group of fish were ≥12 ‰ and the other group consistently <12 ‰ (Figure 8F). Consequently, we chose a cut-off $\delta^{15}N$ value of 12.0 ‰ in terms of $\delta^{15}N$ (1992-2006 data) and 375 mm in terms of fork length (1990) to separate the two groups with fish with

$\delta^{15}N$ value ≤ 12.0 ‰ being "insectivorous" and those > 12.0 ‰ being "piscivorous." Hg and the concentrations of the selected accumulative elements (Cs, K, Se, and Tl) within these groups were then more closely investigated. To identify which of the two covariates ($\delta^{15}N$ or fork length) could be used to adjust Hg concentrations of the overall dataset, the variability of Hg concentrations of each morphotype (small-insectivorous and large-piscivorous) were examined as well.

Arctic char in Lake Hazen are mainly insectivorous, feeding mainly on chironomids, resulting in lower $\delta^{15}N$ signature, while some individuals may feed opportunistically on smaller char, which is reflected in the higher $\delta^{15}N$ signature. Morphological differences within the species have previously been described (Johnson 1980). These different types may vary not only in morphology, or growth rate, but also in foraging behavior. Despite these two apparent groups, the $\delta^{15}N$ to Hg relationship of all fish were consistent over time (no $\delta^{15}N$ * year interaction in the ANCOVA); thus, $\delta^{15}N$ adjustment using ANCOVA could be performed as described above.

2.4.7 Statistical adjustment and temporal trends

ANCOVA of Hg concentration ($\log_{10}$Hg) and fork length of over time (Figure 8C) revealed significant length * year interaction (r^2=0.72, p=0.029). Consequently, length could not be used to adjust Hg concentrations by ANCOVA for the purpose of temporal comparison of the overall dataset, despite its evident correlation (r^2=0.62, p<0.001) with Hg. ANCOVA using $\delta^{15}N$ as a covariate in the model in turn revealed no interaction (r^2=0.70, p=0.074), indicating parallel regression slopes, and allowing a subsequent ANCOVA without the interaction term $\delta^{15}N$ * year. The resulting LSMs of this analysis represent $\delta^{15}N$ - adjusted mean Hg concentrations, which we use here to compare Hg concentrations from year to year (Figure 9).

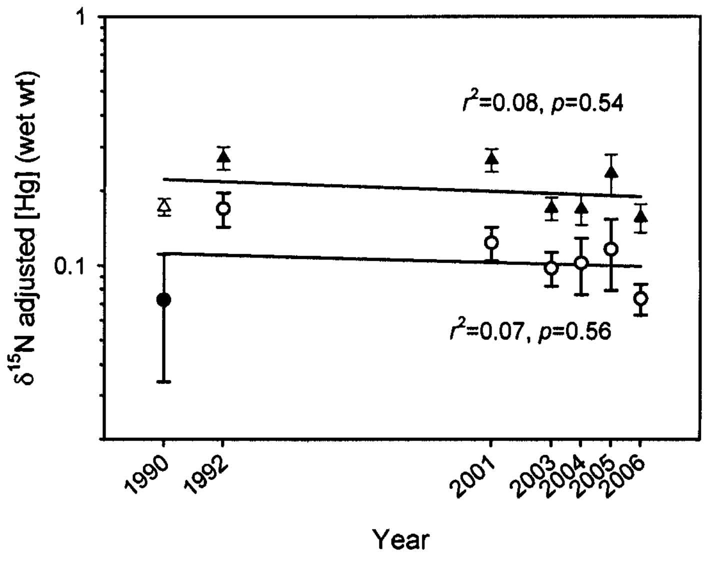

Figure 9: Temporal trends (1990 – 2006) of all char (▲) and "small" form char (○) are presented, 1990 Hg data (Δ, ●) adjusted with estimated $\delta^{15}N$.

Results of ANCOVA showed significant effects (r^2=0.68, p<0.001) of sampling year and Tukey's HSD post-hoc test found the following differences for $\delta^{15}N$ - adjusted Hg concentrations: 1992 and 2001 > 2005 > 1990, 2003, 2004, and 2006, but no significant difference between the period of 2003, 2004, and 2006. Concentrations of Hg in fish sampled in 2006 were similar to those in 1990 ($p_{\text{Tukey's 1990-2006}}$ = 0.49). The regression of LSMs against sampling years was also not significant in neither $\delta^{15}N$-adjusted (r^2=0.07; p=0.52; 1990 – 2006), nor unadjusted (r^2=0.25; p=0.177; 1992 – 2006) Hg concentrations, thus no half-life could be calculated. In summary, no overall temporal trend was detected for the period of 1990 and 2006 in the $\delta^{15}N$ – adjusted overall dataset.

Analysis of the data using PIA, a program widely used to examine temporal trend data (Bignert et al. 2004), showed no significant temporal trends (r^2= 0.07; p=0.67 and r^2=0.03; p=0.71) for both the unadjusted Hg, and the $\delta^{15}N$ – adjusted Hg results, in agreement with the ANCOVA. The lowest detectable change for a ten-year period with the current between year variation at a power of 80% was 17%; 21 years of data were required to detect an annual change of 5% (with a power of 80% and one-sided test at p=0.05) using the unadjusted data. While for the $\delta^{15}N$ – adjusted data, the lowest detectable change is 13% and 18 years were required to detect an annual change of 5% at a power of 80%. No non-linear trends were detected in either data set ($p_{\text{unadjusted}}<0.18$, $p_{\text{adjusted}}<0.08$).

2.4.8 Total Hg concentrations in "small" Arctic char

Temporal trends of log Hg concentrations of "small" insectivorous char (n=75) were also investigated, after adjustment for potential covariates (by ANCOVA). Over the period of 1990 to 2006, unadjusted concentrations varied slightly (r^2= 0.202, p=0.015), but were not different between the beginning and the end of the observed period ($p_{\text{Tukey's 1990-2006}}=0.832$). However, for the time period 1992-2006, a decrease was apparent ($p_{\text{Tukey's 1992-2006}}= 0.003$). Relationships of Hg and fork length as well as Hg and $\delta^{15}N$ in these "small" Arctic char morphs were consistent over time, as ANCOVA revealed no significant interaction for these two covariates (p=0.899, and p=0.125, respectively). The $\delta^{15}N$-adjusted time trend of the "small" char (Figure 9) revealed no decline. Here, regression of LSMs against sampling years (1990 – 2006) resulted in a negative relationship (r^2=0.07, p<0.001), indicating no decline in Hg concentration.

2.4.9 Effect of polymorphism of Arctic char on concentrations of Hg

Since dietary uptake is considered the predominant route of Hg into fish, the differences in feeding behavior contribute to the variability of the observed Hg concentrations. This is consistent with Hammar et al. (1993), who noted differences in PCB concentrations between landlocked char morphotypes. "Large" morphotypes were highly variable in their Hg concentration, and numbers of fish sampled per year were low, not allowing a conclusive trend to be established. The slight decrease in concentrations of Hg in "small" char from our dataset may therefore be more representative of the actual temporal trend of Hg in the Lake Hazen ecosystem.

2.4.10 Other factors affecting mercury in Arctic lakes

Over the past decades, concentrations of some persistent organic pollutants (POPs) have increased in arctic biota, while others have decreased (Evans and Muir 2006). For instance, total PBDEs in landlocked Arctic char muscle from Resolute Lake have increased over the period of 1996 to 2004, while concentrations of DDT have declined in this and other nearby lakes (Muir et al. 2005) and in Lake Hazen (Muir et al. 2005; Muir et al. 2006). This illustrates that lakes can respond in a detectable way to global trends of pollutants. In the case of Hg in landlocked Arctic char, a decrease was expected following reduced industrial Hg emissions, but was not observed in lakes on Cornwallis Island, Nunavut, Canada over the past 20 years (Muir et al. 2006). Recent evidence for declines of Hg concentrations in remote mid-latitude sites (Hrabik and Watras 2002) may reflect the reductions in emissions in the North America. However, similar reductions in Canadian High Arctic environments, which are influenced by Eurasian as well as North American sources (Pacyna et al. 2006), have not been observed. Steffen et al. (2005) found no decline in atmospheric Hg(0) since 1995 at Alert (Ellesmere Island, NU, Canada). This could, in part, be explained by the most recent global mercury budget established by Pacyna et al. (2006), which estimates that global emissions have not declined overall, likely due to increased Asian emissions.

Mercury input from lake catchments may influence concentrations of Hg in small lakes above the treeline (Fitzgerald et al. 2005; Semkin et al. 2005), and subsequently in biota. However, within a large lake such as Hazen, changes in Hg inputs may be more difficult to discern. This is supported by data from dated sediment cores from our group (D. Muir, Water Science and Technology Directorate, Burlington, ON, unpublished data), which indicated that Hg deposition to profundal sediments has been relatively constant in this lake for the past 100 years. However, the input from catchments may have a greater impact in other smaller lakes (Semkin et al. 2005), and may be enhanced under predicted warming of permafrost soil (Leitch et al. 2007), higher algal productivity (Outridge et al. 2007), and greater precipitation (Muir et al. 2006).

2.4.11 Essential and non-essential elements

Concentrations of Se, K, Cs, and Tl were measured in muscle tissues of all but the 1990 samples (Table 1). While the variability within year of Cs and Tl was similar to the Hg data, concentrations of the essential elements Se and K were more constant and showed less within-year variation. Consequently, non-essential Cs and Tl were further investigated and co-factors identified by using ANCOVA, as bioaccumulation was suspected to cause this variability. Relationships with age were not significant in all cases ($\log_{10}$[Cs] r^2=0.00, p=0.198; $\log_{10}$[Tl] r^2=0.02, p=0.185; $\log_{10}$[Se] r^2=0.02, p=0.063; $\log_{10}$[K] r^2=0.00, p=0.319). No significant temporal trends were apparent for Cs, Tl, K, or Se based on comparison of geometric mean concentrations.

Both essential elements were present in similar concentration in all sampled fish. Regression analysis did not reveal significant relationships of K or Se with either δ^{15}N ($\log_{10}$[Se] r^2= 0.08, p=0.001; $\log_{10}$[K] r^2= 0.00, p=0.787) or fork length ($\log_{10}$[Se] r^2=0.108, p<0.001; $\log_{10}$[K] r^2=0.00, p=0.97). Since K was not consistent with trophic position, we could not use K to normalize results for other elements. Se appeared not to bioaccumulate with trophic level of Arctic char, which is consistent with findings from a variety of invertebrate and fish species from western Appalachian streams (Ikemoto et al. 2004; Belzile et al. 2006), but contradicts studies showing Se accumulation (Orr et al. 2006). No further analysis was performed on Se and K due to low or no correlation with length or $\delta^{15}N$. Se remains interesting for its potential for complexation with Hg, and subsequent influence on Hg assimilation (Belzile et al. 2006). Further investigation of this relationship in Arctic food webs will require complete food web analysis for Se or other elements, and is beyond the scope of the present paper.

The two non-essential elements Cs and Tl showed similar relationships to $\delta^{15}N$ as those of Hg, which clearly showed their potential to biomagnify. Tl and Cs concentrations were also greater in "large" char. Regression analysis revealed that Cs and Tl are more strongly related to $\delta^{15}N$ ($\log_{10}$[Cs] r^2=0.68, p<0.001; $\log_{10}$[Tl] r^2= 0.77, p<0.001), than to fork length ($\log_{10}$[Cs] r^2= 0.45, p<0.001; $\log_{10}$[Tl] r^2=0.48, p<0.001).

Adjustment for $\delta^{15}N$ and length was thus possible for Cs and Tl, however, here we again present the $\delta^{15}N$-adjusted trends for Cs and Tl (Figure 10) to be consistent with reporting of Hg data (Figure 9). The subsequent ANCOVA analysis revealed no significant interaction among years for Cs (log[Cs] versus $\delta^{15}N$ * year: r^2=0.743, p=0.267 log[Cs] versus length * year; r^2= 0.542, p=0.184). ANCOVA analysis for Tl also revealed no interaction amongst years: log[Tl] versus N15 $\delta^{15}N$ * year: r^2= 0.782, p=0.715; log[Tl] versus length * year r^2=0.567, p=0.111). $\delta^{15}N$-adjusted Tl concentrations did not change over time (1992-2006) (r^2=0.775, p= 0.634). No temporal trend of Cs was apparent from our $\delta^{15}N$-adjusted data (r^2=0.715, p= 0.065); only the mean for 2005 was slightly lower than the otherwise constant concentrations. These multi-year findings for Cs confirm those of Campbell et al. (2005), who reported a significant relationship of log[Cs] with

δ^{15}N for a limited number of Arctic char from the same study site. The authors also provide evidence for bioaccumulation of another alkali metal, rubidium (Rb), which supports our findings regarding bioaccumulative behavior of non-essential elements. Further in-depth investigation of these non-essential, but accumulative and potentially toxic elements (Cs and Tl) in food webs should be considered.

2.5 Conclusions

Concentrations of Hg in Hazen char muscle did not change significantly over the 16-year period of the present study. Length and trophic position have been identified to be the factors mostly contributing to the year-to-year variability observed in Hg concentrations. Fish age was not significantly related to Hg concentration. Trophic position was found to be the most suitable covariate to statistically adjust the measured Hg concentrations. Following δ^{15}N-adjustment, the combined set of all fish collected (1990 – 2006) showed the same trend as the set of "small" morphotypes. A trend for the large type was not established, due to limited sample size. Our results indicate temporal trend studies need to take into account species-specific factors such as multiple morphotypes in the case of Arctic char or other polymorphic species (e.g., lake whitefish). As demonstrated, the correlation between δ^{15}N and Hg is greater and more consistent than with size and age in Arctic char from Lake Hazen. Based on these results, we recommend using δ^{15}N adjustment for mercury contamination for species with differing foraging behavior.

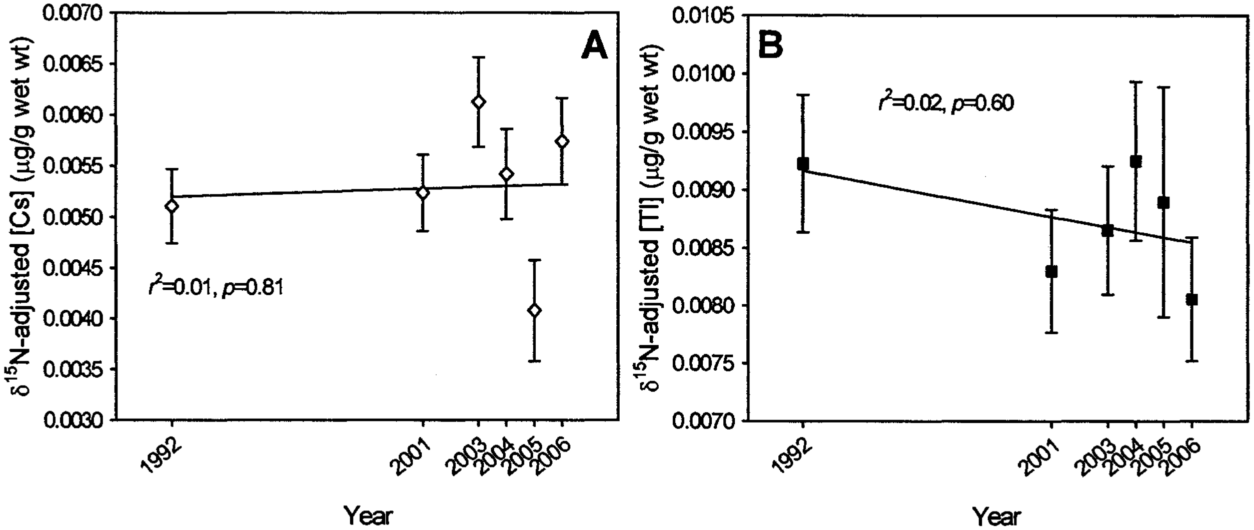

Figure 10: δ^{15}N-adjusted Cs (A) and Tl (B) concentrations (1992 – 2006). Symbols represent back-transformed mean (log transformed) concentrations +/- 1 SD

2.6 Acknowledgement

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3 MERCURY CONCENTRATIONS IN LANDLOCKED ARCTIC CHAR (*SALVELINUS ALPINUS*) FROM THE CANADIAN HIGH ARCTIC: PART I – INSIGHTS FROM TROPHIC RELATIONSHIPS IN 18 LAKES

In preparation for submission to Environmental Toxicology and Chemistry

3.1 *Abstract*

Concentrations of mercury [Hg] have increased slowly in landlocked char over a 10-15 year period in the Arctic. Hg fluxes to sediments also show increases in most Arctic lakes. We hypothesized that climate warming might increase the rate of Hg methylation and alter food chain biomagnification. Correlation of [Hg] with trophic level (TL) was used to investigate and compare Hg biomagnification in food webs from 18 lakes in the Canadian Arctic. Arctic char lake food webs were sampled from 2002-2007 and compared across longitudinal and latitudinal gradients. Food web samples analyzed for [Hg] (total Hg and MeHg), $\delta^{13}C$ and $\delta^{15}N$ included periphyton, zooplankton, benthic invertebrates, and Arctic char of varying size-classes. Trophic magnification factors (TMFs) were calculated for each lake food web and related to available lake characteristics. The relative content of MeHg increased 4.3 to 12.2 % in periphyton, 41 to 79 % in zooplankton, to 59 to 72 % in insects, and 74 to 100 % in juvenile and adult char. $\delta^{13}C$ signatures of adult char indicated coupling with benthic invertebrates. Cannibalism often had an elongating effect on food chains. Biomagnification was confirmed in all 18 lakes, with TMFs ranging from 3.5±1.1 to 64.3±0.8. Results indicate that TMFs and FCL are key factors in explaining inter-lake variability of [Hg] biomagnification up the lake food webs.

Key words: Arctic char, biomagnification, food chain length, diet, climate change

3.2 Introduction

Arctic char *(Salvelinus alpinus)* is the top predator in most High Arctic lake food webs and is thus regarded as a valuable indicator species for environmental monitoring studies of bioaccumulative pollutants, such as mercury (Hg) (Köck 2004; Muir et al. 2005; Gantner et al. 2009a). However, the interpretation of spatial and temporal trend data is often confounded by site-to-site or year-to-year variability of contaminant concentrations in the fish. The trophic position, size, and age of organisms have been identified as factors influencing the variability (MacCrimmon et al. 1983; Vander Zanden and Rasmussen 2001; Guiguer et al. 2002; Lockhart et al. 2005; Vander Zanden and Fetzer 2007; Gantner et al. 2009a) which makes regional and temporal comparisons difficult. Food web characteristics, such as food web length (Vander Zanden and Fetzer 2007), may also influence delivery of contaminants to top predators, and may vary in the Arctic as has been shown for more southern regions (Kidd et al. 1995; Bentzen et al. 1996; Kidd 1998; Guildford et al. 2008). However, information regarding the food web characteristics of Arctic char-bearing lakes is very limited. Typically, lacustrine arctic food webs are short and have low species diversity, although only a few lakes in polar regions have been thoroughly characterized (Rigler 1978; Stross et al. 1980; Hobbie 1984; Andrews and Rigler 1985; Hobson and Welch 1995; Jeppensen et al. 2001; Rautio and Vincent 2006). Zooplankton communities within the lakes are dominated by pelagic Copepoda (*Cyclopoda*) species (Rigler 1978). Benthic invertebrates are typically limited to a few species of Diptera (Chironomidae) (Gajewski et al. 2005), and Arctic char (*S. alpinus*) occupy the top trophic position of these systems, with some individuals displaying cannibalistic behavior, as known for European High Arctic systems (Svenning and Borgström 2005). In southern Arctic lakes, Gammaridae and Trichoptera may be present (Vanriel and Johnson 1995). Less documented is the influence of food web characteristics on contaminant transfer in arctic freshwaters (Macdonald et al. 2003; 2005).

Arctic environments receive Hg through wet deposition mainly in the form of snow, dry deposition of particulates, and gas exchange from the atmosphere (St Louis et al. 2005; Dastoor et al. 2008; Steffen et al. 2008). During snowmelt, both inorganic Hg (directly) and organic Hg (from wetlands) can be released into lakes (Loseto et al. 2004; Loseto et al. 2004; Semkin et al. 2005) where the former is methylated within the lakes to the more bioavailable monomethyl mercury (MeHg) species, mainly by sulfate reducing bacteria in the sediment. In all cases MeHg can bioaccumulate in aquatic organisms and biomagnify up the aquatic food web (Mason et al. 2000).

Future climate changes in northern/arctic regions may increase contaminant concentrations by altering their transfer along food webs (Schindler 2001; Macdonald et al. 2003). Recognizing the importance that the food web plays in delivering MeHg to top predators, we hypothesized that varying trophic characteristics and food web length may influence concentrations of MeHg in Arctic char among different lakes. To investigate this, we sampled the principal dietary items of landlocked Arctic char (benthos, zooplankton, and juvenile Arctic char) from 18 lakes across the Canadian Arctic, as well as water, sediment, and periphyton samples from each site. Samples were analyzed for stable isotope ratios ($^{13}C/^{12}C$ and $^{15}N/^{14}N$) to infer trophic level (from $\delta^{15}N$) and to distinguish the source of carbon ($\delta^{13}C$) for all biotic samples. All samples were analyzed for total Hg (THg) and the majority also for MeHg by standard methods. The relationship between these Hg measures and organism trophic signatures was then investigated using tropic magnification factors (TMFs) and food chain length (FCL). The aim in the analysis was to explain the observed variability in the concentrations of THg/MeHg among sites.

The present study investigates the influence of food web characteristics, utilizing stable isotope techniques to determine trophic relationships, on MeHg or THg transfer in Arctic char food webs under varying climatic conditions (i.e., over a large spatial range). A companion study (Gantner et al. 2009c) investigated geographical trends of THg concentrations in Arctic char populations from 27 lakes, utilizing some of the food web results of the present food web study.

3.3 Materials and Methods

3.3.1 Field collections

The majority of sampling was conducted during three expeditions to the Canadian Arctic (2005-2007) during the summer (July, August). Samples of zooplankton and Arctic char from three other lakes (Herbert, Desert, and Radar) from the Tundra North West expedition in 1999 (TNW99) and one from Nunavik, northern Québec (Pingualuk) were added for comparisons of food chain lengths. Additional Arctic char samples from Aquiatusuk samples were added from a 2002 collection. An overview map showing lake locations and methods of char collections is provided in Gantner et al. (2009c), while detailed maps of sampling areas and lakes are available in the Appendix (Appx 1 - Appx 12).

Surface water samples were obtained from ~ 0.5 m depth and filtrations for basic water chemistry were conducted on site. Periphyton was carefully scraped off rocks obtained in the littoral zone. Zooplankton samples (5-20 mL ww) were collected by multiple surface tows using a 40-200 μm mesh zooplankton net (with a 0.5 m diameter opening). Resulting bulk samples were separated into 250 - 500 μm and >500 μm size fractions (2005-2007), or into species/taxa (1999) in the field and frozen (-17°C) in whirl-pack® bags within one day of collection and kept at that temperature until analysed. Extreme low abundance of zooplankton at most High Arctic sites resulted in variable sample amounts available for analysis each year. When enough zooplankton material was available from a site and year, two size fractions (small = 250-500 μm, large >500 μm) were analysed. If not enough material was available from a site, zooplankton was analyzed as bulk zooplankton (bulk = >250 μm, by combining both fractions).

Emerging insects (Chironomids) were collected from the surface of columnar ice using aspirators that were modified to limit handling of samples and thus potential sources of contamination. Benthic invertebrates were sampled by the kick-sweep method at 0.5-1 m depth, parallel to ice free shore lines. Benthic invertebrate samples were classified to genera in the field and frozen in Nalgene™ Cryo-vials until analysed. Subsamples of bulk zooplankton and benthic invertebrates were preserved in 70% ethanol (2005-2007) or Lugol's solution (without HAc, TNW99) for taxonomic processing. The active layer of surficial sediment (top 1-2 cm) from the maximum observed lake depth was collected using a Ponar grab (2005-2007) or gravity (TNW99) samplers. Sediment samples were also kept frozen at -17C until analysis in pre-rinsed jars. Various sizes (1-10 cm) of juvenile char were collected by electro-fishing along the shoreline (5-30 per lake). Adult char were collected using gill nets or traditional ice-fishing methods. Inuit helped with all sampling on Ellesmere Island, Cornwallis Island, the Kent Peninsula, Victoria Island, and at Pingualuk. Laboratory dissections carried out at the facilities of the Polar Continental Shelf Project in Resolute Bay, Nunavut were also aided by local Inuit. All samples were freeze dried prior to analysis, except Arctic char muscle from 2005-2007 which was analyzed for THg on a wet weight basis. Dorsal muscle tissue (~10 g) and stomach contents were sub-sampled from adult char and frozen until analysed. Otoliths were removed from adult char for age determination.

3.3.2 Laboratory Analysis

MeHg was analyzed in all freeze-dried food web samples (periphyton, zooplankton, benthos, juvenile char) collected from 2005-2007 by acid leaching and solvent extraction followed by propylation with purge and trap, thermal desorption, gas chromatography - cold vapour atomic fluorescence spectroscopy (GC CV-AFS) detection. Analyses were conducted at the National Water Research Institute, Burlington, ON, Canada. The detection limit (3 SD) of this method was 0.36 pg, with a precision of 8.0 %, determined from the Relative Percent Difference between duplicates of all measured samples. Juvenile Arctic char and sticklebacks were analyzed as whole homogenates. For a subset of 24 juvenile char from 3 lakes, %-MeHg was determined (~74±6% of THg), and used to convert THg measurements in all lakes to MeHg ($[\text{MeHg}] = [\text{THg}]_{\text{measured}}*0.74$). All analyses of THg in muscle tissues of adult Arctic char are described in the materials and methods section of Chapter 4, (Gantner et al. 2009c). Moisture contents of biota for dry weight to wet weight conversions were determined from subsets of samples. Concentrations of THg measured in all adult char were converted to dry weight for calculation of TMF. As a consequence THg and MeHg concentrations in biota excluding adult Arctic char are presented as dry weight (moisture content 71-83%).

All fish and food web samples collected between 2005-2007 were dried at a constant temperature of approximately 50°C for 48 hours, pulverized to a fine powder using a Retsch MM 301 ball mill grinder and stored in glass desiccation vials until analysed. Approximately 0.3 mg of dried, ground tissue was used in the simultaneous analysis of stable C and N isotopes on a Delta continuous-flow isotope ratio mass spectrometer connected to a Carlo Erba elemental analyzer with an analytical precision of ±0.2‰ ($\delta^{13}C$) and ±0.3‰ ($\delta\ ^{15}N$) at the Environmental Isotope Lab (University of Waterloo, ON). Analytical precision was established through repeat analysis of internal laboratory standards calibrated against International Atomic Energy Agency standards CH6, N1 and N2. Analytical precision was additionally assessed by the repeat analysis of one in ten samples. All results are expressed here in conventional delta notation (δ) relative to Peedee Belemnite limestone for $\delta^{13}C$ (Craig 1957) and atmospheric nitrogen for $\delta^{15}N$ (Mariotti 1983). Isotopic compositions of TNW99 samples were determined using a Europa Scientific Hydra 20-20 continuous flow isotope ratio mass spectrometer with an analytical precision of ± 0.1 ‰ ($\delta^{13}C$ and $\delta\ ^{15}N$) at the Stable Isotope Facility of the University of California, Davis, USA (http://stableisotopefacility.ucdavis.edu/).

Trophic levels (TL) were calculated from $\delta^{15}N$ for all biota using periphyton (TL = 1, primary producer) as the $\delta^{15}N$-baseline (Vander Zanden and Rasmussen 2001), and 3.4 (‰) as the trophic enrichment factor (Minagawa and Wada 1984; Post 2002), using the formula: $TL = 1 + (\delta^{15}N_{consumer} - \delta^{15}N_{periphyton}) / 3.4$. For further details on the need for baseline adjustment see section 3.4.3 following.

Trophic magnification factors (TMFs) were then calculated from the relationship (slopes) of the regression of log transformed MeHg concentrations (dry weight) and TL (Fisk et al. 2001; Jardine et al. 2006) ($TMF = 10^{\wedge slope([MeHg]*TL)}$ or log[TMF]=slope([MeHg]*TL)). Food web length (FCL) was calculated from TL, representing the difference from lowest TL (primary consumer) to the top predator (Arctic char). Water chemistry parameters were determined according to standard methods of the Environment Canada laboratories (Environment Canada 1996) at the National Water Research Institute (NWRI) in Burlington, ON (Canada).

3.3.3 Statistical analysis

The confidence level (type I error rate) for statistical significance was set to $\alpha = 0.05$ for all analyses. The relationships of MeHg with $\delta^{15}N$, and fish length and age were determined by linear regression analysis. Prior to regressing, THg and MeHg concentrations were $\log_{10}$-transformed to satisfy normality criteria. Geometric means were calculated for THg and MeHg concentrations, whereas arithmetic averages of stable isotope results are presented. SigmaStat 3.1 (2004) and SigmaPlot 9.0 (2000) were used for regression analysis and analysis of variance (ANOVA). Tukey's Honestly Significant Difference (HSD) post hoc test was applied to identify pairwise significant differences in multiple group comparisons. Student's t-test was applied to determine differences when comparing two groups. Analysis of co-variance (ANCOVA) was performed using SYSTAT 11 (2004) to test for lake (spatial) or year (temporal) effects of trophic position, fish size, and age using a general linear model (GLM). Standard deviations of replicate samples were used to describe the data.

3.4 Results and Discussion

3.4.1 Arctic char prey items

In all 18 lakes, Arctic char was the top predator and the only fish species present. Exceptions included the four most southern lakes (Keyhole, located on Victoria Island, and, Notgordie, Little Nauyuk, and Gavia Faeces, located on the Kent Peninsula), where ninespine-sticklebacks (*Pungitius pungitius*) were present. Field observations of stomach contents indicated that Arctic char diet varied regionally (Appx 16, Appx 20, Appx 21). Opportunistic cannibalism of char was observed in stomach contents, and confirmed by greater δ^{15}N signatures in a portion of the char from 5 of 18 lakes (Hazen, Pingualuk, Char, North, and Resolute Lakes). No cannibalism of char was observed in lakes with abundant nine-spine sticklebacks. Sticklebacks, however, formed a portion of the char diet in Keyhole Lake as noted in earlier studies (Vanriel and Johnson 1995). Char were feeding on macroinvertebrates (Diptera (*Chironomidae*), Amphipoda (*Gammaridae*), Trichoptera (*Limnophilidae*), and Isopoda), pelagic zooplankton (Copepoda, Cladocera), and to some extent water mites (Acarina). Amphipoda, Trichoptera, and Isopoda were exclusively found in char (stomachs) collected in the four above-mentioned southern lakes. Water mites are known to live parasitic on chironomids, and could have been either co-ingested with chironomids, or targeted as a food source (Guiguer et al. 2002; O'Connell and Dempson 2002). In high Arctic lakes (Cornwallis Island and Ellesmere Island), all char were primarily feeding on emerging chironomid pupae and midges, as well as cannibalistic. The exceptions in this region were Meretta and Char Lakes. The former was inhabited by large Daphnidae (>1 mm) owing to earlier anthropogenic eutrophication (Douglas and Smol 2000) and the latter contained fairy shrimp species (Anostraca), which were also part of the respective char populations diet. The stomach contents of char were not analyzed quantitatively as they represent only a snapshot of the diet of an individual (Fry 2006). Gut content observations were used only to aid the interpretation of stable isotope and THg results, which are discussed below (Section 3.4.3).

3.4.2 THg and MeHg in food webs

Arctic char had the highest concentrations of mercury (THg) of studied biota in all study lakes. Concentrations of THg in adult char varied among lakes (from 0.07 to 1.31 μg/g THg ww) and were positively correlated to trophic position, length, and age. A detailed discussion of adult char THg can be found in Chapter 4, Gantner et al. (2009c). All juvenile char combined (*n*=138) had a mean (±1SD) THg concentration of 198±128 ng/g (dry weight). Juvenile Arctic char from Amituk Lake (*n*=12) had the greatest mean THg concentrations, 448±308 ng/g. The lowest THg concentrations were measured in Lake Hazen (*n*=6) juvenile char (54±25 ng/g). Individual results from each lake are presented in Appx 15. MeHg concentrations in juvenile char used for TMF calculations were determined for a subset of samples and calculated for all samples.

Mean THg concentrations in seven stickleback from two lakes (Notgordie and Keyhole) was 348±129 ng/g, which is comparable to the mean THg concentrations found in juvenile Arctic char (340±125 ng/g). Four Notgordie stickleback had a greater mean THg concentration than three Keyhole stickleback (t test, t=2.609, p=0.048, [df]=5), however samples size likely resulted in low power for the test (power=0.492). Notgordie Lake was the only site from which data on both juvenile Arctic char and sticklebacks were available, as electro-fishing for juvenile char was not successful at Keyhole Lake. In Notgordie Lake, juvenile char had lower $\delta^{15}N$ than sticklebacks (6.8±0.3 and 7.9±0.5 ‰, respectively; $p<0.05$), resulting in mean trophic levels of 2.6 and 2.9, respectively, and slightly lower THg concentrations (340±125 versus 450±109 ng/g) in Arctic char.

Samples of emergent chironomids were available from 12 lakes. Mean THg and MeHg concentrations of 212±100 ng/g and 151±94 ng/g were determined for all chironomids samples analyzed. The relative content of MeHg ranged from 9-100 % of THg in all samples combined (mean ± SD, 76±24 %). Greatest mean concentrations were measured in chironomids from Amituk lake (THg = 375±159 ng/g and MeHg = 268±40 ng/g, 3 year mean), whereas chironomids from Small Lake had the lowest concentrations (THg = 86 - 104 ng/g and MeHg = 59 - 80 ng/g) over 2 years (2005-2006).

Small and large size fractions were analysed to determine the effects of zooplankton size on THg and MeHg. The small zooplankton fraction had mean THg and MeHg concentrations of 52±19 ng/g (n=7) and 9±11 ng/g (n=10), respectively. The content of MeHg in the small zooplankton fraction was 25±19 % of THg. The large zooplankton fraction had mean THg concentrations of 36±15 ng/g (n=6) and mean MeHg concentrations of 12±13 ng/g (n=10), resulting in 32±26 % mean MeHg content. These difference between size classes were not significant for THg and MeHg content (t test, $p>0.05$). Bulk zooplankton (> 250 µm, n=17) had a mean THg concentration of 55±41 ng/g, and mean MeHg concentrations of 24±29 ng/g. A mean MeHg content of 42±26 % was determined for this subset. Mean concentrations of MeHg in zooplankton were highest in Amituk (45.6 µg/g) and lowest Lake G (1.4 µg/g).

Periphyton was collected for this study mainly to determine $\delta^{15}N$ baseline, however, a subset of samples from 10 lakes was available for THg and MeHg analysis. As expected, this plant material had the lowest mean THg (30±25 ng/g, n=10) and MeHg (2.0±3.1, n=8) concentrations of all biological tissues analyzed in the present study. The mean MeHg content in 8 samples ranged from 1.2 – 15 %, with a mean of 6.8 %.

3.4.3 Trophic relationships in food webs

The trophic hierarchy revealed from stable isotope analysis paralleled the Hg concentration hierarchy, with Arctic char having the highest $\delta^{15}N$ signatures, followed by sticklebacks (one lake), juvenile char, chironomids, zooplankton, and periphyton. The average (±1SD) $\delta^{15}N$ value in juvenile Arctic char was 8.7±1.4 ‰, resulting in a mean trophic level of 2.6±0.7. Variability (SD) of $\delta^{15}N$ within lakes could be due to the effect of omnivory (e.g., Notgordie) of adult char. The mean $\delta^{13}C$ value in juvenile Arctic char was -24.7±1.7 ‰. Sticklebacks (*n*=7) had mean $\delta^{13}C$ of -28.3 ± 1.8 ‰, which is similar to juvenile char from these two lakes (Appx 15). Chironomid $\delta^{15}N$ ranged from 4.4 ‰ (Lake Hazen) to 8.5 ‰ (12 Mile Lake) with a mean of 6.9±1.5 ‰ (Figure 11A), resulting in a mean TL of 2.1±0.4. The $\delta^{13}C$ signature varied among the 12 lakes (mean±SD, $\delta^{13}C$ = -24.1±2.6 ‰), which indicated variable, but mainly benthic sources of carbon. Variability of both $\delta^{13}C$ and $\delta^{15}N$ in chironomids among lakes could reflect natural variations of the biogeochemistry of the basin, or variable species assemblages (Grey et al. 2004), although the range of signatures is lower than the authors of this study report.

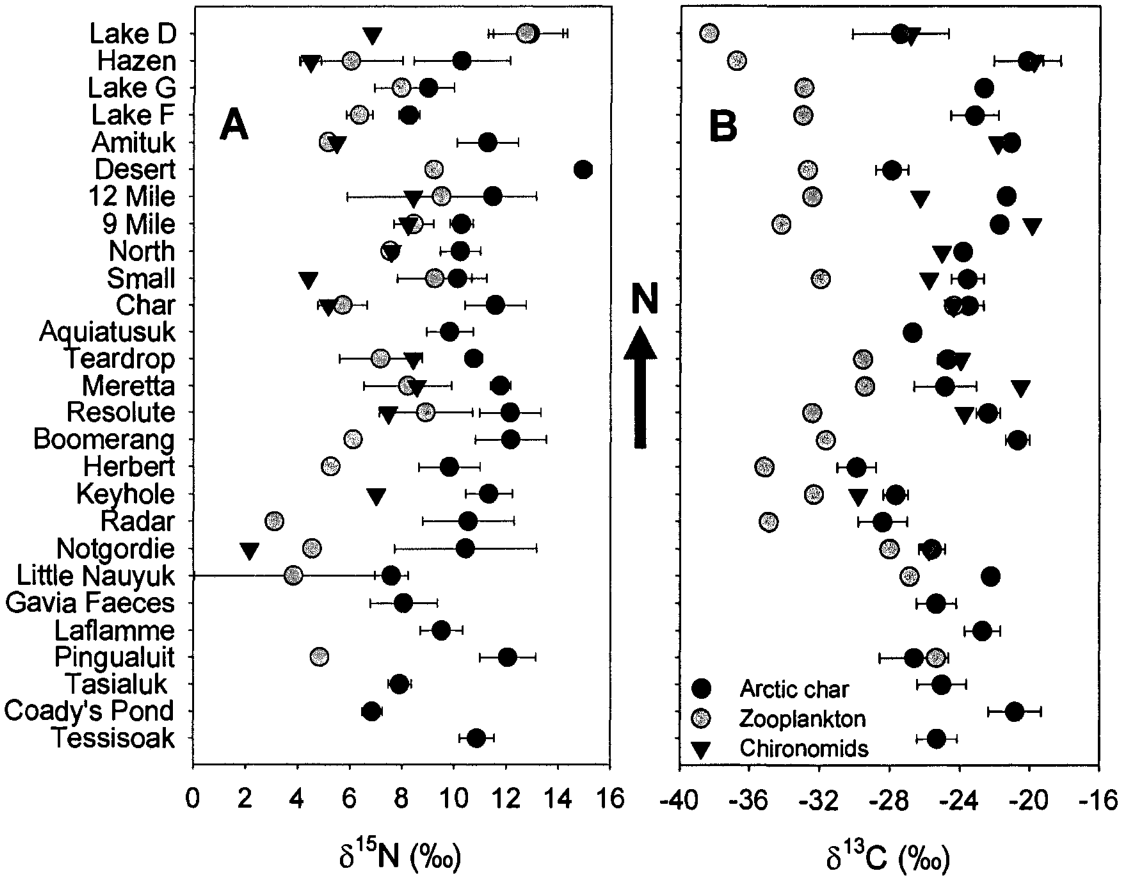

Figure 11: δ^{15}N and δ^{13}C signature in Arctic char, zooplankton, and adult chironomids of all lakes. For *n* of char refer to Table 3. Error barrs represent 1 standard deviation.

The available fractions of zooplankton (small, large, and bulk) were also analyzed for isotope ratios. The small fraction had mean δ^{15}N values of 6.9±2.7 ‰ (n=10), resulting in mean TL of 2.1±0.5, compared to the 9 available large fractions (>500 µm) (mean δ^{15}N = 7.7±2.6 ‰, mean TL=2.5±0.6, respectively). Whereas the variability in δ^{15}N was quite large, both size fractions did not vary greatly in δ^{13}C, i.e., from -31.3±3.3 ‰ (small) to -30.9±2.9 ‰ (large), respectively, indicating utilization of similar carbon sources independent of size and diet. As expected, combined bulk zooplankton (> 250 µm, n=17) had comparable mean δ^{15}N (7.6 ±2.1 ‰) and δ^{13}C (-30.7 ± 6.2 ‰) as each separate size fraction.

Periphyton, being a primary producer, generally had the lowest $\delta^{15}N$ (Appx 15), averaging 2.5±1.9 ‰ (17 Lakes). While the variability of $\delta^{15}N$ was quite considerable (from 0.3 to 6.5 ‰), neighbouring lakes had similar $\delta^{15}N$ signatures (12 Mile and 9 Mile Lake, Lake G and F, Gavia Faeces and Little Nauyuk; Appx 15). This suggests that the effects of biogeochemistry on $\delta^{15}N$ were region specific, and not catchment specific. The fact that $\delta^{15}N$ in periphyton ranged from 0.3 to 1.8 ‰ in Kent Peninsula lakes, 0.8 to 6.5 ‰ in Cornwallis Island lakes, and 0.8 to 2.2 ‰ in Ellesmere Island lakes underscores the importance to adjust for this baseline prior to comparing food webs among catchments or regions. Previous studies identify the need for N-baseline adjustment to account for variable anthropogenic inputs to each lake system, however, in remote Arctic lakes without considerable anthropogenic N inputs, we account for natural variation of N among catchments in order to compare regions. Food webs of Arctic lakes located within the same catchment may be compared from within each region using the absolute (unadjusted) $\delta^{15}N$ values, but this was not tested here.

Carbon signatures ($\delta^{13}C$) can be affected by variable inputs or sources of DIC related to lake size. However, sediment $\delta^{13}C$ values, available for 13 lakes, were not significantly related to latitude ($p>0.05$). Regional mean $\delta^{13}C$ in sediments did not differ between Cornwallis Island (8 lakes, mean $\delta^{13}C$ -26.3±2.2 ‰) and Kent Peninsula lakes (4 lakes, mean $\delta^{13}C$ -27.2±3.1 ‰) (t-test, $p>0.05$), while one sample from Ellesmere Island (Lake Hazen) had a $\delta^{13}C$ value of -27.4 ‰.

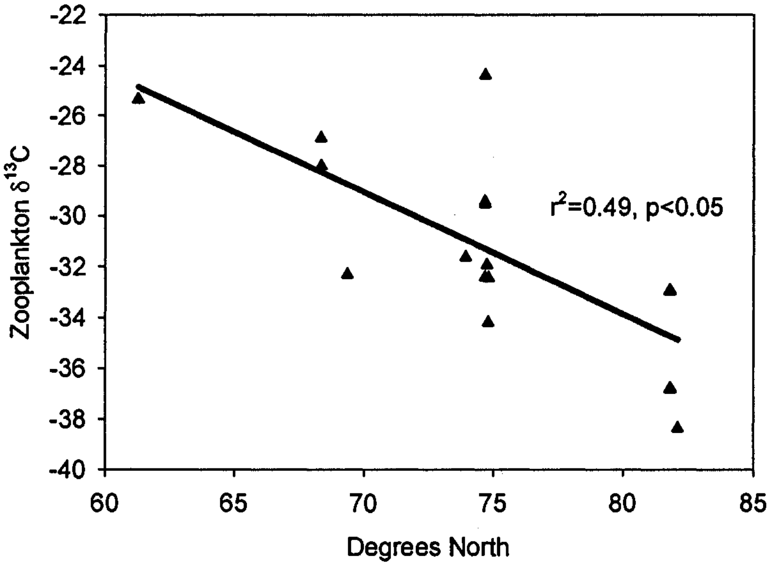

Figure 12: Relationship of zooplankton $\delta^{13}C$ and latitude ($\delta^{13}C$ = 4.6023 - 0.4808 x latitude). Lake codes are given in Table 3.

Strong coupling of adult Arctic char and benthic invertebrates (mainly chironomids) was evident from the $\delta^{13}C$ data. The $\delta^{13}C$ signature of adult char typically reflected that of benthic invertebrates present in a given lake, rather than that of zooplankton (Figure 11B). Arctic char appear to rely heavily on emerging/emergent chironomids during summer months, which is also supported by our observed stomach contents. Interestingly, $\delta^{13}C$ in zooplankton was correlated with latitude (linear regression r^2=0.49, p<0.05, n=18), in that $\delta^{13}C$ zooplankton was more depleted (negative) with increasing latitude (Figure 12). The importance of zooplankton as a prey item during the summer months for char from lakes in the present study may not be as high as is known for other systems (Malmquist et al. 1992; Riget et al. 2000).

3.4.4 Food chain length

Food chain length (FCL) varied significantly among the lakes (ANOVA, $p<0.05$), and ranged from 4.2±0.41 in Lake Hazen (Ellesmere Island, NU) to 2.1±0.12 in 9 Mile Lake (Cornwallis Island). FCLs presented here are consistent with literature values (Vander Zanden and Fetzer 2007). While the authors found a slight increase in FCL with higher latitude over a large geographic range (Northern Hemisphere), we could not detect such a relationship within our Arctic range. The effect that benthic or pelagic feeding has on FCL becomes apparent from Lake Hazen data. When determined for insectivorous small morphotypes, FCL was shorter (FCL = 3.1±0.6, n=38) than when using only large morphotypes (FCL = 4.2±0.4, n=11) (p<0. 05). The result for Lake Hazen FCL also provides an example how intra-specific differences in feeding behavior (i.e., cannibalism) can extend overall FCL (Paine 1965; Fox 1975; Hammar 2000). This food chain elongation as a result of cannibalism underscores the uniqueness of Lake Hazen char, and possibly Lake Pingualuk, where this effect is most pronounced. For the latter, more samples of smaller specimen would be needed to confirm the number of morphotypes present. The influence of FCL on THg in adult char is discussed in the accompanying study (Gantner et al. 2009c).

3.4.5 Trophic magnification factors

Biomagnification of MeHg was confirmed in all 18 lakes. Slopes of regressions between log[MeHg] and TL of biota were significant in each lake (Figure 13, Figure 14, Table 3), and averaged 1.01. Slopes varied between lakes, ranging from 0.54 to 1.81, subsequently TMFs ranged from 3.6 in Lake F to 64.3 in 9 Mile lakes. The TMFs presented for MeHg here are greater than those reported in marine ecosystems (slope of 0.22; TMF = 1.6) (Campbell et al. 2005) who determined Hg concentrations on a wet-weight basis, or for organic contaminants (mean 4±1.8) (Houde et al. 2008). Moisture content influences determination of Hg concentrations, thus all food web materials (periphyton, zooplankton, benthic invertebrates) were analyzed on dry weight basis (for MeHg). In char, moisture content was generally consistent at 77-80% in specimens >200 mm, a range that corresponds to literature suggestions for wet weight conversions in other salmonid species (Kelly et al. 2008). The high TMFs presented here are indicative of efficient uptake and transfer of MeHg through the short Arctic lake food web, which could be explained by high efficiency of energy transfer in Arctic lakes (Vanriel and Johnson 1995) compared to more southerly lakes. The transfer of MeHg from the loci of greatest production (sediments) occurs through an upwards cascade of only 2-3 TLs and as few as two species/groups (Chironomids and Arctic char), while zooplankton may be of lesser importance in Arctic lakes (Vanriel and Johnson 1995). TMFs are thus a measure of the whole lake ecosystem transfer function for Hg, and an important measure as they integrate information from the entire lake food web.

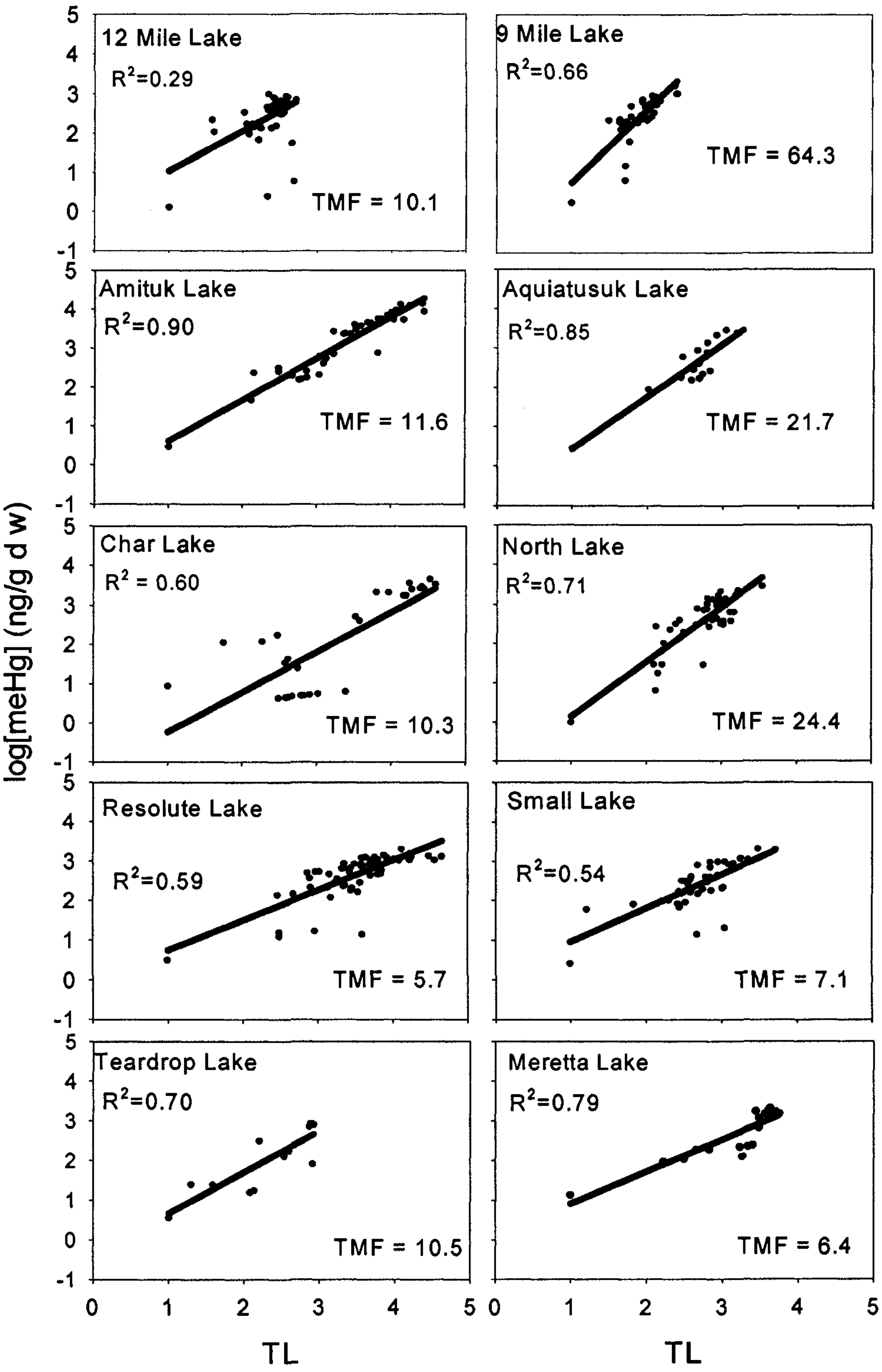

Figure 13: Regression plots (log[MeHg] vs. trophic level, TL) and trophic magnification factors (TMFs) of 10 lakes on Cornwallis Island

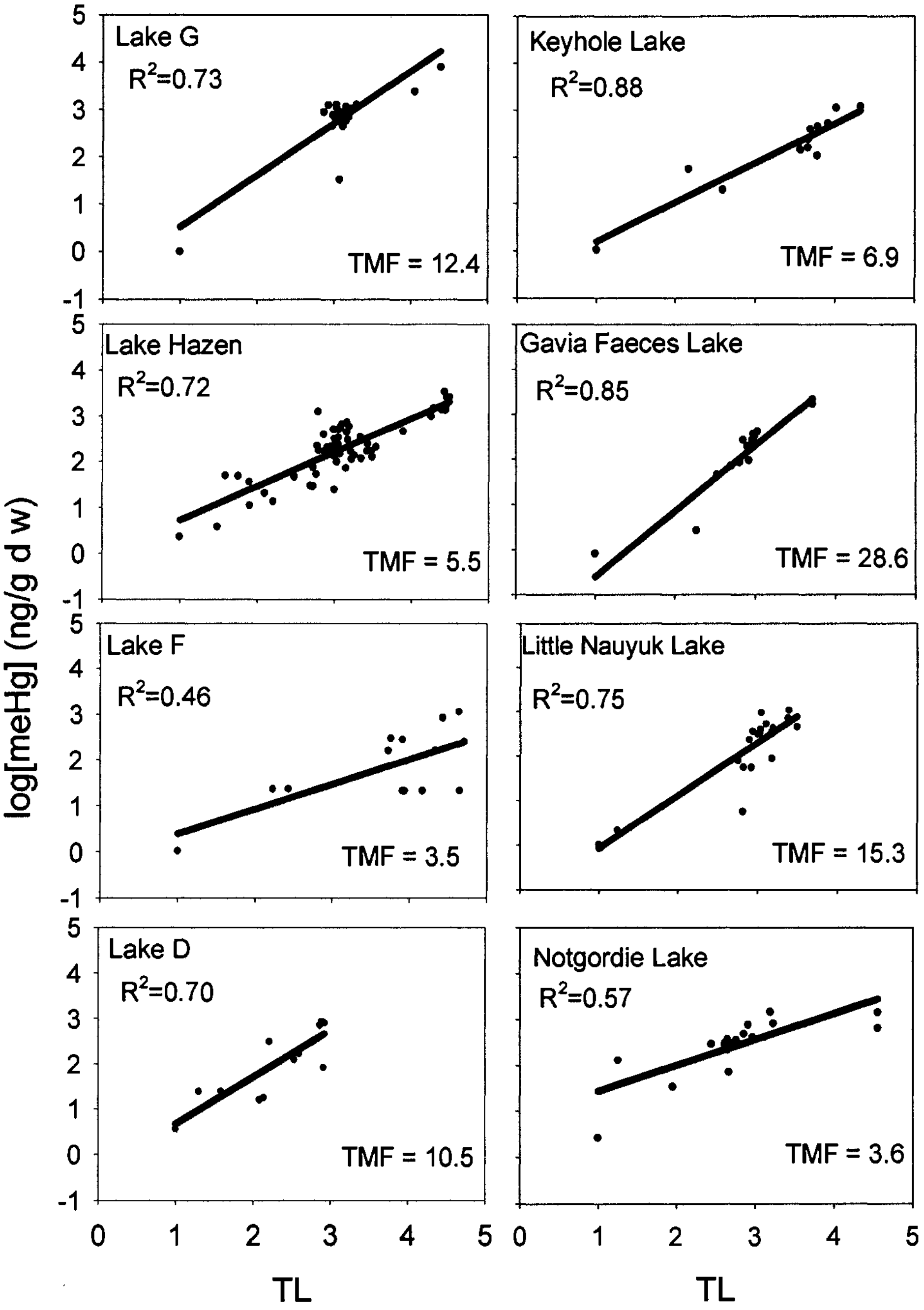

Figure 14: Regression plots (log[MeHg] vs. trophic level, TL) and trophic magnification factors (TMFs) of lakes from Ellesmere Island (left row) and the Kent Peninsula and Victoria Island lakes (right row).

Table 3: Trophic magnification factor (TMF) and linear regression parameters

Lake Name	Code		$n^{\#}$	TMF	SE_{slope}	R_{adj}^2	Slope	Intercept
Lake G	G	Ellesmere Island	35	12.4	0.8	0.73	1.09	-0.58
Lake D	D		14	10.5	0.9	0.70	1.02	-0.34
Lake Hazen	Hz		64	5.5	0.8	0.72	0.74	-0.01
Lake F	F		14	3.4	1.1	0.46	0.54	-0.14
9 Mile	9m	Cornwallis Island	51	64.3	0.8	0.66	1.80	-1.02
Aquiatusuk	Am		17	21.7	0.8	0.85	1.39	-1.24
Amituk	Aq		40	11.6	0.7	0.90	1.34	-0.91
Teardrop	Td		13	10.5	0.9	0.70	1.07	-0.47
12 Mile	12m		56	10.1	0.9	0.27	1.02	-0.34
North	N		45	24.4	0.9	0.71	1.02	-1.24
Small	S		43	7.1	0.9	0.54	1.00	0.05
Meretta	M		21	6.4	0.7	0.79	0.85	0.10
Char	C		30	10.3	1.2	0.60	0.81	0.11
Resolute	R		61	5.7	0.9	0.59	0.76	-0.01
Gavia Faeces	GF	Kent Peninsula	15	28.6	0.9	0.85	1.46	-2.06
Little Nauyuk	LN		20	15.3	0.9	0.74	1.18	-1.25
Notgordie	Ng		21	3.6	0.8	0.28	0.84	-0.65
Keyhole	K	Victoria I.	13	6.9	0.8	0.88	0.56	-0.87
Region			n^{**}					
Ellesmere Isl.			4	8.0	0.9			
Cornwallis Isl.			9***	12.0	0.8			
Kent Peninsula			3	15.8	0.9			
Victoria Island			1	6.9	0.8			

[#] number of organisms

** number of lakes

*** 9 Mile not included in this average

3.4.6 Benthic coupling

As demonstrated above, strong benthic coupling exists in the study lakes. This holds specifically true for Cornwallis Island and Ellesmere Island sites, where char depend on Chironomids as their principal prey during summer. Here, we have calculated TMF including information on the key food web species, it thus represents a measure of the entire species assemblage important for contaminant transfer. While TMFs have been successfully used to characterize biomagnification in more complex food webs with more predator prey interactions (Campbell et al. 2005; Houde et al. 2008), here TMFs are not fully predictive of Arctic char Hg concentrations. Measures describing the efficiency of contaminant uptake from one species (Chironomids) to another species (Char), such as biomagnification factors (BMF) may yield in better prediction of Arctic char Hg concentrations. In low productivity systems with high degree of benthic coupling, such as Arctic lakes, TMFs appear not to be predictive.

3.4.7 Relationships of TMFs and FCL with environmental factors

All lakes were ultraoligotrophic, with Chl a $< 5\mu g/g$. A summary of selected water chemistry data are presented in Appx 17. Linear regression results indicate no relationships between TMF and latitude, longitude, lake area (A), catchment area (C), C/A ratio, DOC, or chlorophyll a (all $p>0.05$, Appx 19). FCL and lake area showed a slightly positive correlation ($r_{adj}^2=0.15$, $p=0.07$), however, no other environmental factor or productivity measure (DOC, Chl a) was correlated with FCL (all $p>0.05$, Appx 19). The lack of predictability of TMFs from environmental factors which influence Hg is not surprising. TMF is a measure of efficiency of the biological compartments to transfer Hg, and lake abiotic characteristics may not directly influence this factor. Houde et al. (2008) suggest a connection between TMF and lake productivity measures in southern Canadian lake food webs, but we could not provide evidence for such a connection here. This may be due to the low productivity in all lakes of the present study, and the subsequently small differences in DOC and chlorophyll a among our lakes.

3.5 Conclusions

We have established trophic relationships and information on Hg transfer along the food web in 18 Arctic char-bearing lakes. Biomagnification of Hg (MeHg) occurs in all lakes to a varying degree, as demonstrated by TMF. The TMFs presented here were not related to a variety of environmental factors examined, such as latitude, indicating that this measure is not directly influenced by climatic drivers such as day length or mean air temperature. However, indirect effects could occur through changes in FCL. We applied this information in a parallel study of the geographic trends of concentrations of THg in adult Arctic char. Information on TMF and FCL presented herein can now be used to detect future alterations that may be caused by climate change. As TMFs are indicators of Hg transfer within each lake, this could be either continuously monitored in selected lakes, or revisited in the future for comparison. Food webs and other arctic lake characteristics may be altered by higher productivity, or invasion of more southern species into northern lakes, which are predicted consequence of climate change. Moreover, in order to detect changes in contaminant concentrations in Arctic char, the above factors need to be taken into account.

3.6 Acknowledgment

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05, 661-06, 651-07) and contributed to our team's safety and comfort during field work near Resolute Bay, Cambridge Bay, and Ellesmere Island. Parks Canada and staff aided greatly in sampling sites within Quittinirpaaq National Park on Ellesmere Island. We thank Jessica Epp (Environment Canada) for assisting in zooplankton identification. All work presented here was conducted under the terms of the Nunavut Research Institute (License: 0203206R-M), Department of Fisheries and Oceans (License: S-06/07-1015-NU), and the Ministre des Resources naturelles et de la faune du Québec (License: 9053.28). All work followed the of the University of Guelph (#06R083) animal care protocol. The Northern Ecosystem Initiative, Northern Contaminants Program, Network of Centres of Excellence Arctic Net, International Polar Year (IPY 07/08), Canadian Foundation for Climate and Atmospheric Sciences (Grant# GR-623), and Austrian Academy of Science (DOC-Stipend for lead author) provided funding for this research. We thank the reviewers for their comments on the manuscript.

4 MERCURY CONCENTRATIONS IN LANDLOCKED ARCTIC CHAR *(SALVELINUS ALPINUS)* IN THE CANADIAN HIGH ARCTIC: PART II – SPATIAL COMPARISON OF 27 POPULATIONS

In preparation for Environmental Toxicology and Chemistry.

4.1 Abstract

Among-lake variation in mercury (Hg) concentrations in landlocked Arctic char was examined in 27 char populations from across the Canadian Arctic to determine causes of observed variability. We collected a total of 520 landlocked Arctic char from 27 lakes, as well as sediments and surface water from a subset of lakes in 1999, 2002, and 2005-2007. Size, length, age, and trophic position ($\delta^{15}N$) of individual char were determined and relationships with total Hg (THg) concentrations investigated, to identify a common covariate for adjustment using ANCOVA. A subset of 216 char from 24 populations was used for spatial comparison, following adjustment to a common covariate (length). The influence of trophic position and food web length and abiotic characteristics such as location, geomorphology, lake area, catchment area, catchment-to-lake area ratio of the lakes on adjusted THg concentrations in char were then evaluated. Un-adjusted Hg concentrations varied significantly among regions. Arctic char from Amituk Lake (Cornwallis Island) had the highest Hg concentrations (1.31 μg/g wet wt), while Tessisoak Lake (Labrador, 0.07 μg/g wet wt) had the lowest. Concentrations of THg in char were positively correlated with size, $\delta^{15}N$, and age, respectively, in 88, 71, and 58% of study lakes. Length and $\delta^{15}N$ were correlated in populations from 67% of study lakes. Food chain length did not explain the length-adjusted THg concentrations in char. No relationships between adjusted THg concentrations in char and latitude or longitude were found, however, THg concentrations in char showed a positive correlation with catchment-to-lake area ratio. Our results indicate that a combination of biotic factors leading to inter-lake variability in food web structure may override abiotic influences on THg concentrations in char. Furthermore, we conclude that inputs from the surrounding environment (catchments/watershed) may influence THg concentrations, and will

ultimately affect THg concentrations in char as a result of predicted climate-driven changes that may occur in Arctic lake watersheds.

4.2 Introduction

Despite recent reduced emissions and use of mercury (Hg) and Hg-containing products in some parts of the industrial world, mercury remains of critical concern to human populations relying on subsistence fisheries as a key source of dietary protein (Lockhart et al. 2005; Mergler et al. 2007; Munthe et al. 2007). A recent global budget (Pacyna et al. 2006) indicates that the largest emitters of Hg are the rapidly growing economies in Asia (namely China and India), while North American and European emissions have declined since the 1990s. Overall, total global emissions have remained constant since 1980, totaling 2190 tons in the year 2000 (Pacyna et al. 2006). There is also a large body of literature that describes the sources of Hg in the atmosphere ranging from emission by coal fired power plants, garbage incineration (anthropogenic), to volcanic eruption (natural).

Long range atmospheric transport of Hg from these sources is believed to deliver atmospheric Hg to the Arctic, where it is deposited into the environment (Lindberg et al. 2007). Transfer of Hg species to the Arctic and into Arctic lakes, as well as mechanisms of bioaccumulation and food web transfer of monomethylHg (MeHg) are outlined in the accompanying study (Gantner et al. 2009b). The recent review by Munthe et al. (2007) summarizes contributing pathways and recent trends in Hg concentrations in fish. Mercury (Hg) in fish remains a dominant issue across the Arctic, where concentrations in predatory fish from arctic and sub-arctic regions of North America are frequently found to exceed Health Canada consumption guidelines (Evans et al. 2005; Muir et al. 2005; Jewett and Duffy 2007). In some small lakes, concentrations in fish have been increasing over time (Muir et al. 2005); however, in one of the largest Arctic lakes (Lake Hazen, Ellesmere Island Nunavut, Canada), concentrations remain unchanged over a 16-year period (1990-2006) (Gantner et al. 2009a). It has been suggested that documented increases of Hg concentrations in aquatic environments (Fisk et al. 2003) maybe linked to recent climate changes in the Arctic (Schindler 2001; Macdonald et al. 2003; Outridge et al. 2007). Increases of Hg in Arctic lake sediments have been attributed to anthropogenic sources (Fitzgerald et al. 2005). A combination of both climatic effects and the global distribution of Hg, due to its long residence time in the atmosphere (~ 1 year) (Lindberg et al. 2007), could be responsible for the lack of decline of concentrations in the Arctic, despite recent North American reductions. Studies near Alert on Ellesmere Island, Nunavut, have shown that there is no decline of gaseous elemental Hg (GEM) in the atmosphere (Temme et al. 2007; Steffen et al. 2008), while there is evidence from sediment cores that there are reductions in Hg deposition in lower latitude lakes in Canada (Muir et al. 2008).

A comparison of Hg species (organic + inorganic Hg fraction) over a wide geographical range (latitudinal and longitudinal) should improve our knowledge of biotic and abiotic factors contributing to Hg variability. Ultimately, these results can be applied to the study of pathways and delivery of this mostly atmospherically transported contaminant. However, spatial comparisons of Hg concentrations in fish are often constrained by the high variability among species, among populations within the same species, or among individuals within the same population (Lockhart et al. 2005; Muir et al. 2005; Jewett and Duffy 2007). Fish are exposed to Hg mainly through dietary uptake (Hall et al. 1997). Thus, food chain characteristics are important for any explanation of lake-to-lake variability, because biomagnification up the food chain appears to be lake specific and longer food chains may result in higher Hg concentrations (Gantner et al. 2009b). Other factors such as trophic position, fish size, and age (MacCrimmon et al. 1983; Rodgers 1996; Vander Zanden and Rasmussen 2001; Guiguer et al. 2002; Power et al. 2002b; Lockhart et al. 2005; Gantner et al. 2009a), contribute to the variability for Hg and other (organic) bioaccumulative contaminants (Kidd 1998; Guildford et al. 2008). Unless better understanding of the sources of variability in Hg concentrations is developed, meaningful comparisons among sites and regions will remain difficult.

Landlocked Arctic char (*Salvelinus alpinus*) is widely abundant in the Canadian Arctic (Scott and Crossman 1973), and is a keystone species throughout the Arctic in the lakes where it is found (Power et al. 2008). The distribution of Arctic char extends farther north than that of any other freshwater fish, and it is often the only species present in high latitude Arctic lakes (Johnson and Burns 1984; Hammar 1989; Parker and Johnson 1991; Power et al. 2008). Its circumpolar distribution within the Arctic allows widespread monitoring and large scale spatial comparisons of contaminant concentrations. Previous studies have shown that THg concentrations in Arctic char muscle tissue exceed Health Canada guidelines (subsistence consumption 0.2 μg/g wet wt), with landlocked populations typically having greater THg concentrations than migratory populations (Riget et al. 2000; Borg et al. 2001; Lockhart et al. 2005; Muir et al. 2005). Although ubiquitous and contaminated, to our knowledge there have been no large-scale spatial comparisons of Hg tissue concentrations in landlocked Arctic char, apart from reports on Hg data from anadromous char populations (Lockhart et al. 2005). The earlier study of Muir et al. (2005) investigated spatial variability only at local scales, using 6 lakes located near Resolute Bay, Nunavut, and did not include consideration of possible food web related effects on Hg concentrations. Here we set out to investigate large-scale spatial differences of Hg concentrations along longitudinal and latitudinal gradients in the Canadian Arctic using suitable adjustment factors to account for trophic position, fish length, and/or age effects. Abiotic characteristics such as those of watershed have been shown to affect Hg cycling in Alaskan arctic lakes (Hammerschmidt et al. 2006), and mid-latitude lakes (Munthe et al. 2007; Simonin et al. 2008) and will also be used to contrast spatial differences in Hg.

4.3 Materials and Methods

4.3.1 Site selection

We selected landlocked Arctic char populations from lakes in geographically distinct Arctic or sub-Arctic regions, attempting to cover the largest possible latitudinal range (Figure 15, Table 4). With exception of Ruggles River (Lake Hazen), all outflows of the study lakes do not allow for migration, and Lake Hazen char are confirmed to be lake resident (Babaluk et al. 2001). Selection of sites was constrained by accessibility and guided by past sampling campaigns, and/or current monitoring programs. The four most northern lakes are located in Quittinirpaaq National Park (~82°N) on Ellesmere Island (Nunavut, Canada). To our knowledge three char populations in this area (lakes F, G, and D) have never been sampled prior to this study. Information on limnological characteristics (Keatley et al. 2007) and diatom assemblages (Smith 2002) from nearby lakes and ponds has highlighted the characteristics of area lakes owing to local climate effects that make the area an "Arctic oasis". Twelve lakes are located near Resolute Bay (Nunavut, Canada) (~74°N), and were accessed either by All Terrain Vehicle or helicopter. Three populations in the Resolute Bay area (North, Small, and Resolute Lakes) are known to be utilized by the local community. The food web of Char Lake also in the Resolute area has also been previously studied in detail (Hobson and Welch 1995), and information on several contaminants (Hg and DDT, PCBs) in char is available (Lockhart et al. 2005; Muir et al. 2006) for some lakes of the Resolute area. Four lakes are located South-West of Cambridge Bay (~69°N) (Nunavut, Canada), and their resident char populations have been extensively studied by the Department of Fisheries and Oceans (DFO) in the 1970s and 1980s (Johnson 1994; Vanriel and Johnson 1995). Two lakes included in the study form part of the Parc National de Pingualuit (~61°N) on the

Figure 15: Approximate Sampling locations across the Canadian Arctic. Circles (O) indicate main sampling areas (multiple lakes per area were sampled); Dots (•) represent single lakes of the dataset (refer to Table 1 for names and location).

Ungava Peninsula (Quebec, Canada); Lake Pingualuk is a meteor-impact crater lake (Bouchard 1989). Three lakes from north Labrador (~56°N) represent the most eastern and southern lakes and were sampled in conjunction with a specialized International Polar Year survey carried out by Department of Fisheries and Oceans (St. John's, Newfoundland and Labrador) that are primarily focused on anadromous Arctic char. The most western lake (Herbert Lake, ~69°N, 139°W) is located in the Yukon Territory (Canada). A single lake (Radar Lake) is located on the north shore of eastern Baffin Island (~68°N, 66°W). With the exception of the three north Labrador lakes, all other lakes selected for the present study hosted exclusively landlocked Arctic char as the only fish species. An additional exception were the most southern study sites (Nauyuk Region Lakes), where ninespine stickleback (*Pungitius pungitius* L.) were also present. Samples of sticklebacks were only collected from the latter region.

4.3.2 Field collections

Between July and August of 1999, landlocked Arctic char and zooplankton samples were collected from lakes Radar, Herbert, and Desert Lakes during the Tundra North West (TNW99) expedition. Samples collected during TNW99 were shipped to ITM (Stockholm University, Sweden) on ice, and stored there at -20°C until analyzed. Sampling was supported by local Inuit guides, who also provided general information on the fish population of the sites. For all other study lakes Arctic char and food web samples were obtained in July and August of 2005, 2006, and 2007, following procedures outlined in Gantner et al. (2009b), with the exception of Lake Pingualuk and Lac Laflamme (collection in May). Briefly, adult char were collected using gill nets (32-46 mm mesh size) or traditional ice-fishing methods and/or angling through ice cover. An annual catch of 20 adult Arctic (maximum) char per lake was used for conservation purposes, set by the Canadian Federal Department of Fisheries and Oceans (DFO), with exception of Labrador sites. Typically, collections at each site occurred on a single day. If fishing results were not sufficient (n<7 adult char), a second sampling was added. All sampling conducted in the Resolute area was aided by the local Inuit. Sub sampling and fish dissections were performed immediately post-capture at the facilities of the Polar Continental Shelf Project in Resolute Bay, Nunavut. At all other sites, samples were either dissected in the field, put on ice, or frozen whole. Samples collected between 2005 and 2007 were shipped frozen to the National Water Research Institute (NWRI, Burlington, Ontario, Canada), and stored there at -20°C until analyzed. Between 2 and 49 Arctic char were available from 27 lakes, sampled between 1999 and 2007, making a total of 520 samples for the present study (Table 4).

Surface water samples were obtained near the centre of each lake using a pre-cleaned 2 L Teflon® container and clean techniques. Duplicate samples were transferred into acid-washed and rinsed amber (MeHg) and clear (THg) glass bottles (250 mL), and 3 mL HCl (0.5%) added to preserve the MeHg samples. Field blanks (2 per site) were brought to the field and were handled in accordance with the sampling procedure, except for filling of bottles. All water samples and blanks were stored at 4 °C in dark conditions until analyzed.

4.3.3 Laboratory Analysis

In fish, THg was analyzed using the direct combustion method performed with a Milestone Direct Mercury Analyser (DMA-80) according to U.S. EPA method 7473 (U.S. EPA). A method detection limit (=3 SD) of 2-5 ng/g dry wt and precision (=1 SD) in homogenous samples (at 100 * DL) of ±1-2% was determined for the processing of all TNW99 samples at (Department of Applied Environmental Science, ITM, Stockholm, Sweden). Equal precision, but lower detection limits (1-2 ng/g) were determined for samples analyzed at NWRI, Burlington laboratories (DMA 80). Arctic char muscle from the 2005-2007 collections was sub sampled (~0.1 - 0.2 g wet wt) following a brief thawing period, and measured without prior drying, following U.S. EPA method 7473 (U.S. EPA). Surficial sediment samples from 2005-2007 were analyzed for THg with identical methods, but freeze-dried prior to analysis. TNW99 char samples were freeze-dried and moisture content recorded, with a 0.05 g dry wt were used for analysis. Resulting Hg concentrations (dry wt) in TNW99 char samples were converted to wet wt concentrations using the mean water content of individual fish (77 ± 6% wt). Consequently, all reported concentrations herein are based on wet weight. Standard reference materials (DORM1, DORM3, DOLT, all National Research Council of Canada) were run daily to check for method accuracy, instrument performance, and matrix effects. Data were only used if the reference materials were within ±10 % of their certified value, or samples were re-analysed.

Stable isotope analysis of char muscle tissue and surficial sediments were completed following the methods described in the accompanying study (Gantner et al. 2009b). Briefly, sub samples of muscle were freeze dried and homogenized to a fine powder using a ball-mill grinder prior to analysis. Sediment sub-samples were acidified in petri-dishes using HCl (10%), then neutralized by adding MiliQ water (to pH ~6.5-7), and dried at 60°. Trophic magnification factors (TMFs) and food chain lengths (FCL) of a subset of 18 lakes (derived from food webs presented in the companioning study) were used here to infer their influence on THg concentrations in Arctic char muscle.

Total Hg in water samples was analyzed at NWRI, Burlington (Ontario, Canada), using atomic fluorescence spectroscopy (AFS). MeHg in surficial sediments were determined using gas chromatography (GC) for separation and AFS detection applying extraction methods outlined in Chapter 3, Gantner et al. (2009b).

4.3.4 Statistical analysis

The confidence level (type I error rate) for statistical significance was set to $\alpha=0.05$ for all analyses. All statistical analyses were conducted using software by Systat Inc. (SystatSoftware 2004). The relationships between the dependent variable THg and $\delta^{15}N$, fish length, or age were determined using linear regression analysis. Prior to analysis, THg concentrations were $\log_{10}$-transformed to improve the normality of the data distribution. For correlation and regression analyses SigmaPlot 9 and SigmaStat 3.1 were used. Analysis of covariance (ANCOVA) was performed using SYSTAT 11 to test for trophic, length, and age (fish) effects using a general linear model (GLM), following procedures described in Gantner et al. (2009a) and the resulting GLM model was used to adjust Hg concentrations for the effect of significant covariates. Adjusted Hg concentrations, back-calculated from least squares means (LSMs), were then used for graphical presentation. Tukey's Honestly Significant Difference (HSD) test was used to detect significant differences within regions with more than two sites. Student's *t* test was applied, when only two lakes were compared.

4.4 Results and Discussion

4.4.1 Unadjusted THg – 27 populations

Means of unadjusted THg concentrations in Arctic char ranged from 0.07 μg/g (Tessisoak Lake) to 1.31 μg/g (Amituk Lake) (Table 4). Char from Amituk Lake had the highest individual THg concentrations as well, with one char reaching 3.4 μg/g of THg. Mean Hg concentrations in Arctic char from 14 lakes were below the Health Canada guideline for subsistence consumption (of 0.2 μg/g), while mean THg concentrations in char from 10 lakes exceeded that guideline (Table 4). The THg concentrations presented here are comparable to previously reported literature values for six lakes from the Resolute Bay area (Muir et al. 2005). Thus, the present study is the most comprehensive summary of current concentrations of THg available for landlocked Arctic char across a large proportion of the geographic range of the Canadian Arctic.

Table 4: Lake, regions, locations, sampling years, and Arctic char (*n*, biodata, trophic signatures, and un-adjusted THg)

Lake	Region	Location		Years	*n*	Length (mm)		Weight (g)		Age (y)		δ^{15}N (‰)		δ^{13}C (‰)		[THg] (µg/g)	
		° N	° W			M	SD	M	SD	M	SD	M	SD	M	SD	GM	SD
Lake D	Ellesmere Island	82.11	67.48	2007	7	333	113	498	390	19	7	12.9	1.4	-27.5	2.7	0.08	0.09
Lake G		81.82	69.18	2007	34	287	73	260	460	19	3	9.0	1.0	-22.7	0.4	0.20	0.29
Lake F		81.81	69.35	2007	10	228	132	276	640	13	5	8.3	0.4	-23.2	1.4	0.11	0.19
Lake Hazen		81.83	70.42	'06-07	54	331	117	413	367	17	7	10.3	1.8	-20.2	1.9	0.08	0.16
Amituk	Cornwallis Island	75.05	93.75	'05-07	24	419	87	657	440	20	3	11.3	1.2	-21.1	0.4	1.31	0.89
Aquiatusuk		74.70	94.23	'05-07	10	441	43	741	259	31	18	9.8	0.9	-26.7	0.5	0.30	0.21
12 Mile		74.82	95.34	'05-07	49	334	28	484	323	16	1	11.5	0.3	-21.4	0.4	0.11	0.04
9 Mile		74.81	95.20	'05-07	34	321	26	225	54	16	3	10.3	0.4	-21.7	0.5	0.16	0.06
North		74.78	95.09	'05-07	23	352	58	415	120	17	3	10.2	0.8	-23.8	0.3	0.26	0.13
Meretta		74.68	94.92	'05-07	16	443	126	1261	713	7	2	11.8	0.4	-24.9	1.8	0.32	0.08
Char		74.70	94.88	'05+07	13	401	118	867	770	19	6	11.6	1.2	-23.5	0.9	0.46	0.26
Resolute		74.68	94.88	'05-07	40	374	66	475	218	19	5	12.2	1.2	-22.4	0.7	0.22	0.08
Small		74.76	95.06	'05-07	28	390	55	492	283	16	3	10.1	1.1	-23.6	0.9	0.12	0.11
Teardrop		74.68	94.99	2007	4	109	6	11	2	-	-	10.8	0.1	-24.7	0.6	0.19	0.01
Boomerang	Somerset	73.94	92.89	2007	8	461	33	648	173	16	2	12.2	1.4	-20.7	0.7	0.28	0.11
Keyhole	Victoria	69.38	106.24	2006	7	427	100	1167	840	9	6	11.3	0.9	-27.7	0.7	0.09	0.10
Gavia Faeces	Kent Pen.	68.34	107.73	2006	8	275	98	211	201	12	5	8.1	1.3	-25.3	1.1	0.11	0.18
Little Nauyuk		68.35	107.75	2006	15	349	50	349	157	13	4	7.6	0.6	-22.2	0.3	0.11	0.06
Notgordie		68.35	107.66	2006	5	442	34	1134	337	9	1	10.4	2.7	-25.6	0.7	0.23	0.09
Tasialuk	Labrador	56.74	62.69	2007	37	341	130	593	378	-	-	7.9	0.4	-25.0	1.4	0.11	0.07
Tessisoak		56.63	62.52	2007	8	113	28	17	15	-	-	10.9	0.7	-25.3	1.2	0.07	0.02
Coady's Pond		56.64	63.63	2007	33	308	111	430	356	-	-	6.9	0.4	-20.8	1.5	0.10	0.03
Laflamme	Ungava Pen.	61.32	73.71	2007	9	497	124	1563	875	14	5	9.5	0.8	-22.7	1.0	0.14	0.07
Pingualuk		61.28	73.66	2007	23[#]	400	129	532	495	19	5	12.1	1.1	-26.6	1.9	0.18	0.11
Herbert	Yukon T.	69.42	139.63	1999	9	295	79	186	109	-	-	9.8	1.2	-29.9	1.1	0.46	0.70
Desert	Melville I.	75.03	107.87	1999	2	540	62	1538	651	-	-	14.9	0.3	-27.9	0.9	1.24	0.89
Radar	Baffin I.	68.42	66.83	1999	10	380	154	695	811	-	-	10.6	1.8	-28.4	1.4	0.73	0.62

M = arithmetic mean; GM=Geomean; SD=standard deviation; [#]included are two A. char recovered from stomachs of two larger specimen

4.4.2 Trophic signatures ($\delta^{15}N$)

Detailed analysis of $\delta^{15}N$ signatures in Arctic char, are presented in Gantner et al. (2009b) and Table 4, and will only be discussed briefly here. The average $\delta^{15}N$ signature of Arctic char ranged from 6.7 ‰ in Coady's Pond (Labrador, N = 33) to 14.9 ‰ in Desert Lake (Melville Islands, Nunavut, N = 2). For the 18 lakes of known baseline (periphyton signature), char $\delta^{15}N$ signatures were baseline corrected ($\delta^{15}N_{A.\ char\ adjusted} = \delta^{15}N_{A.\ char\ unadjusted} - \delta^{15}N_{periphyton}$). The $\delta^{15}N$ signatures in char (adjusted and un-adjusted for baseline) were not related to latitude (linear regression $r^2 < 0.1$, Figure 16). Although both latitude and longitude themselves are correlated with climate, mean $\delta^{15}N$ does not appear correlated with broad-scale climatic patterns or drivers. FCL was also not related to latitude or longitude (p>0.05).

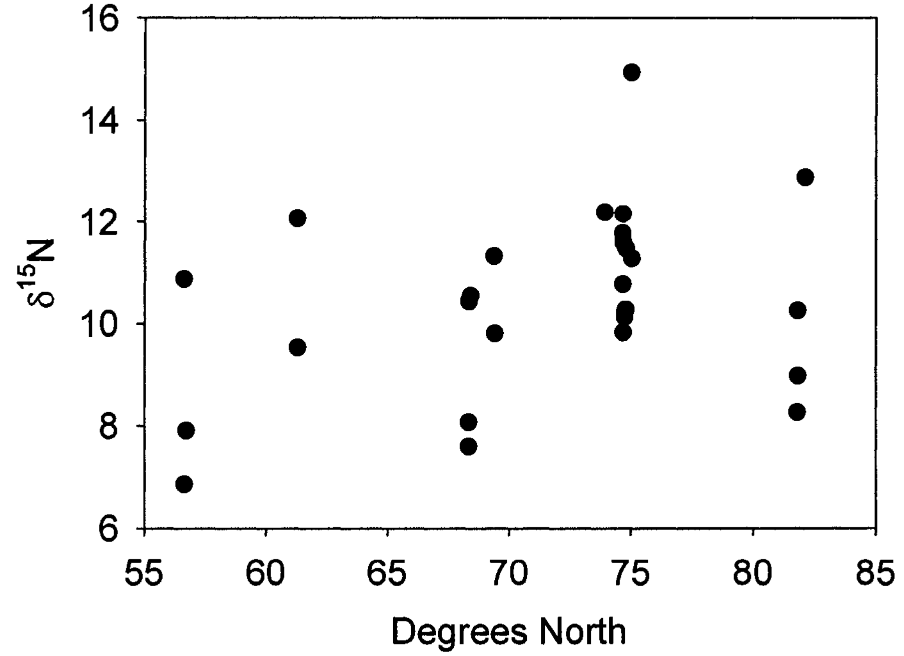

Figure 16: Mean trophic signature ($\delta^{15}N_{unadjusted}$) in Arctic char versus latitude (lake codes are given in Table 5).

4.4.3 Relationships of THg in char with length, age, and $\delta^{15}N$

Linear regression revealed positive relationships between the unadjusted THg and covariates known to influence Hg concentration in fish: fork length, age, and $\delta^{15}N$ (Table 5). In most lakes (17 of 27), THg in Arctic char was positively correlated with $\delta^{15}N$ in char muscle (linear regression r^2 range from 0.33 in Resolute Lake to 0.96 in Gavia Faeces Lake. In 21 of 27 lakes (linear regression range of $r^2 = 0.11$ in 12 Mile Lake to 0.88 in Lake F), THg was predicted by fork length. Fish ages were not available for seven lakes; however, in the remaining 14 lakes, THg was related to fish age. Noteworthy is the oldest Arctic char collected during our sampling campaign at Aquiatusuk Lake (Cornwallis Island) in 2005, as at 44+ years, it represents the oldest individual of this species aged by our age determination laboratory and as such may be the oldest recorded specimen for the species (J. Babaluk and R. Wastle, Fisheries and Oceans Canada, Arctic and Freshwater Institute, Winnipeg, pers. comm., Appx 6).

Size, age, and $\delta^{15}N$ were thus predictors of concentrations of THg for each lake, although no general rule could be established across all lakes of our study region, likely owing to the variability of underlying food webs (Gantner et al. 2009b) and the ecological plasticity of the individual char populations. This poses an issue for spatial comparisons. While small-scale comparisons (of lakes within one region) may work well with one of the presented covariates (Table 5), large scale comparisons, such as conducted here require statistical adjustment for a common covariate. Hence, a statistically adjusted subset of char was compared across the study region.

Table 5: Linear regression parameters of unadjusted THg and covariates for all Arctic char collected

Lake		log[THg] vs $\delta^{15}N$		log[THg] vs length		log[THg] vs age		length vs $\delta^{15}N$	
Name	**Abbrev.**	$p<0.05$	r^2	$p<0.05$	r^2	$p<0.05$	r^2	$p<0.05$	r^2
Lake D	D	-	-	√	0.52	-	-	-	-
Lake G	G	√	0.67	√	0.69	√	0.17	√	0.82
Lake F	F	√	0.61	√	0.88	-	-	√	0.75
Hazen	Hz	√	0.51	√	0.71	√	0.31	√	0.39
Amituk	Am	√	0.82	√	0.49	√	0.26	√	0.7
Aquiatusuk	Aq	√	0.71	√	0.59	√	0.40	√	0.6
12Mile	12m	-	-	√	0.11	-	-	-	-
9Mile	9m	√	0.45	-	-	√	0.14	√	0.11
North	No	√	0.40	√	0.26	√	0.14	√	0.22
Meretta	M	-	-	√	0.33	√	0.25	√	0.79
Char	C	√	0.74	√	0.74	√	0.31	√	0.61
Resolute	Rl	√	0.33	√	0.13	√	0.24	-	-
Small	S	√	0.59	√	0.28	√	0.57	√	0.24
Teardrop	Td	-	-	-	-	-	-	-	-
Boomerang	B	√	0.83	√	0.62	-	-	√	0.45
Keyhole	K	√	0.54	√	0.46	-	-	-	-
Gavia Faeces	GF	√	0.96	√	0.78	√	0.89	√	0.81
Little Nauyuk	LN	√	0.26	-	-	√	0.53	-	-
Notgordie	Ng	-	-	-	-	-	-	-	-
Tasialuk	Ta	√	0.40	√	0.63	-	N/A	√	0.24
Tessisoak	Te	-	-	√	0.73	-	N/A	-	-
Coady's Pond	CP	-	-	√	0.38	-	N/A	-	-
Laflamme	L	-	-	√	0.47	√	0.72	-	-
Pingualuk	P	√	0.38	√	0.57	√	0.71	√	0.19
Herbert	He	-	0.52	-	-	-	N/A	√	0.37
Desert	Dt	-	-	-	-	-	N/A	-	-
Radar	Rd	√	0.75	√	0.69	-	N/A	√	0.87

* '-' indicates not significant results

** *n* of Table 1 apply

Note: Abbreviation of lake names used as labels in Figures 3 and 4

4.4.4 Subset for spatial comparison:

In order to reduce variability of THg concentrations for spatial comparisons, we identified relationships (Table 5) of THg concentrations for the overall dataset (n=520) and selected covariates for adjustment using ANCOVA. As the slopes of the regression of THg concentration with δ^{15}N, length, age, and weight varied considerably from lake to lake, the adjustment using ANCOVA failed when attempted for all Arctic char (interaction was observed $p<0.05$). Varying sample size may have caused the interaction, as only 9 of 27 lakes had >20 char, while 10 lakes had <10 char. To standardize comparisons by lakes from all regions, we selected 5-10 similarly sized char from each lake, which were then adjusted using the ANCOVA model (see below). Data from three populations (Teardrop, Desert, and Tessisoak) were removed due to low sample size (n=2 and 4) or non-common fish sizes (Tessisoak). The remaining 216 char (ranging from 173 – 639 mm fork length) from 24 lakes were used for the spatial comparison in the present study.

Fork length and δ^{15}N were evaluated as covariates in the adjustment using ANCOVA, as ages were not available for all lakes. The GLM model log[THg] = lake + δ^{15}N + lake * δ^{15}N revealed interaction ($p<0.05$), indicating that there was no common slope of log[THg] and δ^{15}N to be found for the 24 lakes and thus δ^{15}N could not be used for ANCOVA adjustment. The same model was applied using baseline adjusted δ^{15}N values, again showing significant interaction ($p=0.00$). However, the model log[THg] = lake + fork length + lake * fork length revealed no interaction ($p=0.124$), and thus a common slope of log[THg] and length across lakes. The results also indicated a good fit for this model ($r^2 = 0.88, p<0.05$). Subsequently, the GLM model was re-run without an interaction term (log[THg] = lake + fork length), and the resulting the least squares means (LSMs) from the model were used to back calculate fork length-adjusted THg concentrations. The resulting Hg concentrations are thus adjusted to the length estimated by the grand mean (Houde et al. 2008), rather than to a given length. Hereafter, the results from this adjustment with fork length as the covariate will be referred to as length-adjusted THg concentrations.

4.4.5 Spatial comparison

Ellesmere Island

In the most northern region (Ellesmere Island), length-adjusted THg concentrations in char varied significantly among the four lakes (ANOVA $p<0.05$) (Figure 17). The highest ($p_{\text{Tukey's}}<0.05$) concentrations of THg were found in lake G (0.30±0.04 μg/g), followed by the neighbouring lake F (0.23±0.05 μg/g). Both sites had higher concentrations ($p_{\text{Tukey's}}<0.05$) than Lake Hazen (0.10 ±0.02 μg/g) and Lake D (0.10±0.02 μg/g). The latter two lakes did not differ significantly ($p_{\text{Tukey's}}>0.05$). Lake D is located upstream of Lake Hazen and is in the same watershed, but further from Lake Hazen, than either lakes (G and F). Lakes G and F drain into the outflow of Lake Hazen, and are thus not connected to the Hazen watershed. The similarity of lakes G and F is also apparent in their food chain lengths (Gantner et al. 2009b). Interestingly, we found the greatest concentrations of THg in char from the two smallest lakes (G + F) on Ellesmere Island, which is not the case for the other regions in the present study. These results suggest that lakes of the same watershed may have similar processes for delivery of Hg to the lake; this then leads to similar THg in char.

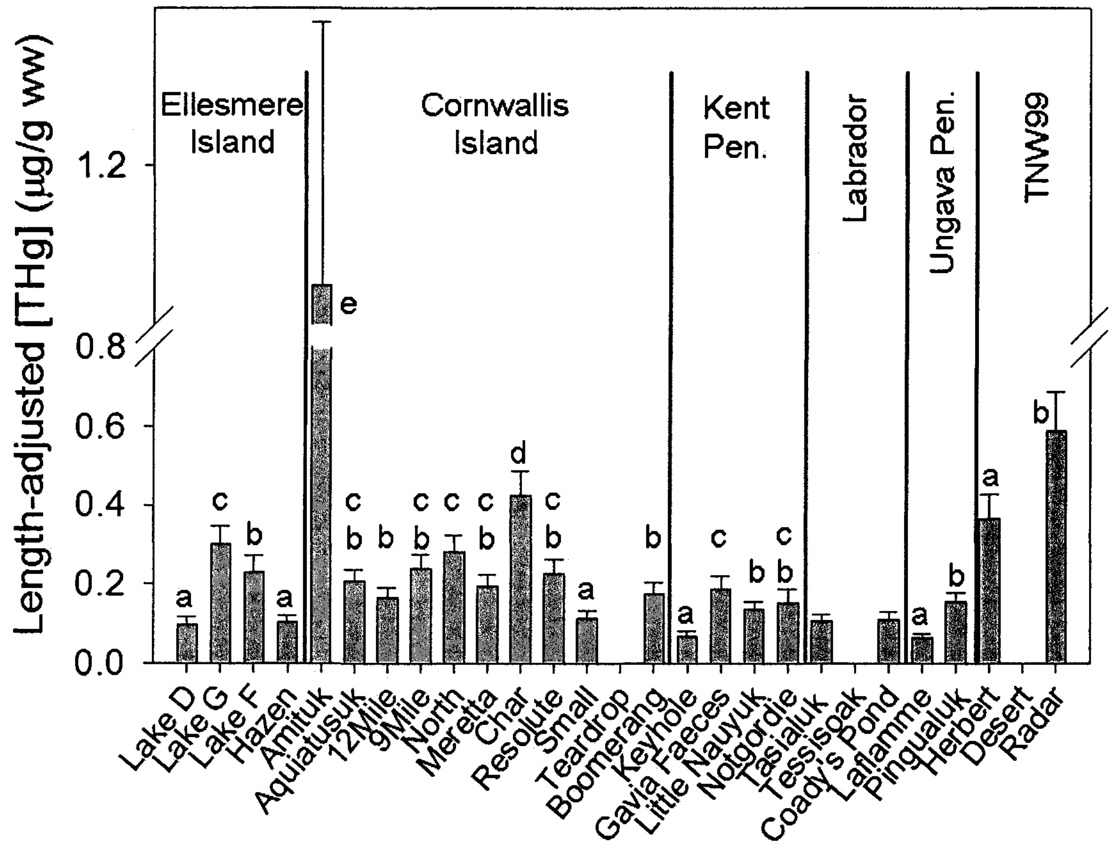

Figure 17: Length-adjusted THg concentrations in 24 Arctic char populations. Bars represent THg concentrations (µg/g), back calculated from ANCOVA LSMs. Significance (Tukey's test or Student's t test, $p<0.05$) of differences between populations within each region is indicated by different associated letter(s). Labrador populations did not differ ($p=0.49$).Teardrop, Tessisoak, and Desert lakes were removed due to small sample size prior to adjustment. Error bars represent 1 standard deviation.

Cornwallis Island

Resolute Bay area char (includes Cornwallis Island, Somerset Island, Melville Island) showed the greatest length-adjusted THg concentrations in the char from the present study area. Concentrations differed among all populations (ANCOVA $p<0.05$), and differences among lakes were found (Figure 17). Amituk Lake char, the char with the greatest individual un-adjusted THg concentration (3.4 µg/g), also have the greatest mean length-adjusted THg concentrations (1.12±0.17 µg/g) of lakes on Cornwallis Island, and of all populations in the present study. Next highest concentrations were found in Char and North Lakes (4.3±0.6, 0.28±0.4 μg/g), respectively. Char from Small Lake had the lowest length-adjusted THg concentrations (0.11±0.02 μg/g). Our results concur with the previous studies (Muir et al. 2005), which also indicate that Amituk and Char Lakes had the highest Hg concentrations in char. Although Hg release from upstream wetlands, input to, and export from Amituk Lake has been well studied (Semkin et al. 2005), it remains a unique example of extraordinarily high char THg concentrations in a remote lake.

Kent Peninsula and Victoria Island

Concentrations of Hg in char on the Kent Peninsula were generally lower compared to the more northerly sites (Figure 17). One explanation could be the more diverse benthic invertebrate communities (i.e., shorter FCL), or diverse feeding behavior (feeding on prey with lower Hg concentrations) among char within these lakes (Gantner et al. 2009b). Among lakes in this region, char from Keyhole Lake on Victoria Island had the lowest concentration. Concentrations in Little Nauyuk and Notgordie Lake did not differ ($p_{\mathrm{Tukey's}}>0.05$), neither did those of Notgordie and Gavia Faeces ($p_{\mathrm{Tukey's}}>0.05$). Gavia Faeces char had higher concentrations than Little Nauyuk ($p_{\mathrm{Tukey's}}<0.05$). TMF factors are high (Chapter 3) (Gantner et al. 2009b), food web lengths are longer than in other regions of the present study (Figure 18). The absence of cannibalism may be a further reason for lower mean THg concentrations. The fact that Keyhole lake char had lower THg concentrations compared to Kent Peninsula sites may be an indication that regional factors, such as underlying geology, may have played a role.

Easterly sites

Both sites on the northern Labrador coast, Coady's Pond and Tesialuk Lake had low length-adjusted Hg concentrations (Figure 17). The adjusted Hg concentrations did not differ between lakes (Student's $t = -0.695$, p=0.496). No food web information is available for these lakes. Since limited information on lake characteristics is available, these sites were excluded from most of the follow-on analysis. Lac Laflamme and Pingualuk char from the Ungava Peninsula had low concentrations of THg compared to the other sites in the present study. Pingualuk char had higher concentrations than Laflamme (Student's $t = -9.567$, $p<0.05$), which is explained by its FCL of ~4.2 (Figure 18). The char population from Radar Lake on Baffin Island showed the second highest length-adjusted THg concentrations at 0.59±0.1 μg/g wet wt in our study region. Although this lake is located near an abandoned DEW line radar station, previous studies have shown no signs of pollution with Hg or other metals in this lake (Borg et al. 2001). FCL in this lake was determined to be ~4.2 (using zooplankton as the baseline) (Gantner et al. 2009b), which is indicative of piscivory/cannibalism and could be the reason for higher Hg concentrations (Figure 18).

Yukon - westerly site

Finally, Herbert Lake, the most western lake of our study range, showed higher concentrations of THg compared to most lakes in the present study (0.37±0.06 μg/g). This is in contrast to the low MeHg concentrations in zooplankton detected in this lake (Gantner et al. 2009b). Other metals, especially cadmium (Cd), were previously measured in char muscle from this lake (Borg et al. 2001), and also exceeded concentrations found in other lakes of that study.

4.4.6 Food chain length and biomagnification

FCLs for an additional four sites were determined for TNW99 lakes and Lake Pingualuk using zooplankton $\delta^{15}N$ as the baseline, however no TMF were available from these 4 lakes. Linear regression analysis showed no relationship between TMF and FCL (Figure 18), following removing the outlier value (Studentized residual 5.554) of the 9 Mile Lake TMF (64.3).

Surprisingly, FCL alone was not predictive of length-adjusted Hg concentrations (linear regression, $r^2=0.20$, $p=0.251$) across our data set (22 lakes), see Figure 19E. TMF also did not predict length-adjusted Hg in Arctic char ($p>0.05$). Multiple regression of

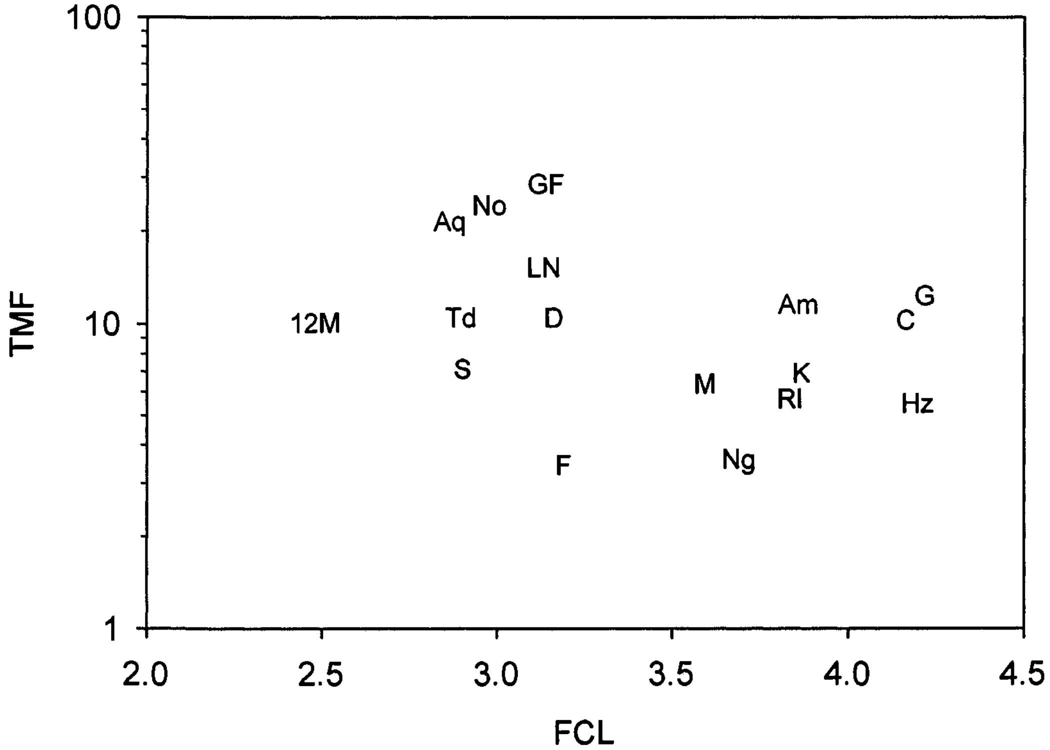

Figure 18: Relationship of trophic biomagnification factors (TMF) and food chain length (FCL) in 17 lakes (9 Mile Lake TMF was identified as outlier). The first or first two letters of the lake name are used and codes are given in Table 5.

log[length-adjTHg] and TMFs and FCL was not significant ($p > 0.05$). This may be less surprising than the previously described lack of relationship with FCL, as TMFs are measure that integrates the uptake of Hg through whole lake food webs. High TMFs can only lead to high THg in fish, if the food chain is long. The linkage is supported by our findings in Amituk and Char lakes where Hg in char is high (length-adjTHg = 1.12 and 0.42 μg/g wet wt, with FCL = 3.8 and 4.2, respectively) due to TMFs of ~10, whereas concentrations of THg in 9 Mile Lake char are low (0.24 μg/g wet wt, FCL = 2.1), despite high TMF (~64), due to a short food chain. We suggest that a combination of FCL and TMF measures may be useful to explain mechanisms within a given lake.

TMFs alone are not sufficient to predict Hg concentrations in char (no significant

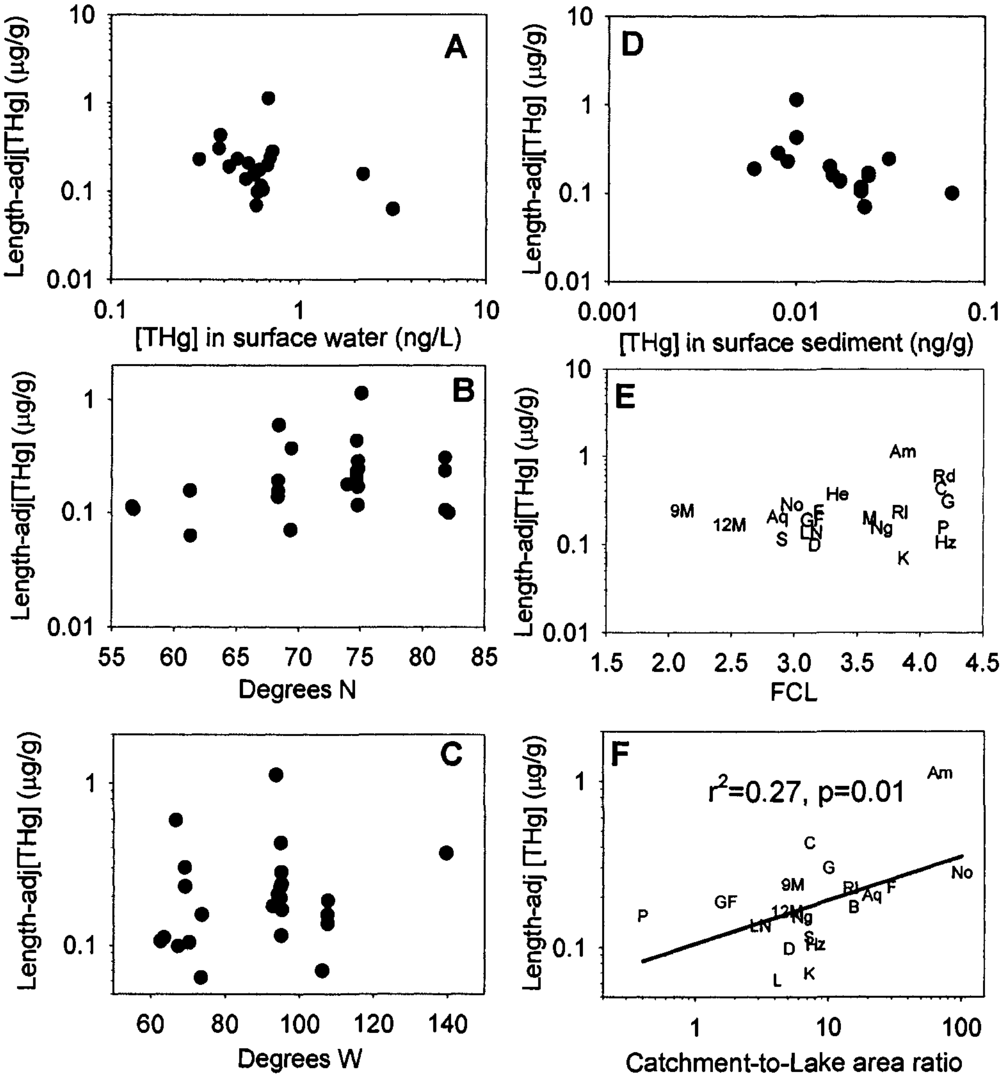

Figure 19: Relationships among concentrations of THg in water, sediment (d w), and length-adjusted [THg] in char muscle (w w) (A, D), char [THg] with latitude (B), longitude (C), FCL (E) and catchment-to lake area ratio (F). Regression lines are shown for significant relationships only. The first or first two letters of the lake name are used and codes are given in Table 5.

correlation between THg and TMF), as the results from this and our accompanying study revealed (Gantner et al. 2009b). While TMFs are indicative of the rate of biomagnification (slope of the regression), the extent of Hg uptake is ultimately determined by FCL. In addition, as TMFs are derived from the slopes of MeHg and TL regression of all organisms in the food web, they better indicate the efficiency of Hg

(MeHg) transfer through the food web, rather than the quantity of Hg delivered to a top predator. We conclude that TMFs are useful to describe efficiency in trophic uptake in lake food webs that are well characterized; however, TMF has to be applied in conjunction with a measure of food web structure, such as FCL. The relationship of TMF and FCL, as well as their influence on THg concentrations in adult char is discussed in detail in the accompanying paper (Gantner et al. 2009c).

The aforementioned observations led us to attempt to model the combined effect of TMFs and FCL on THg in char. Therefore, we multiplied the slope of the regression of biomagnification (i.e., log[TMF]) with FCL for each of the 18 lakes. The results of log[TMF] * FCL was then plotted against the length-adjusted THg concentrations (Figure 20). Again, the predictive capacity of log[TMF] * FCL calculation was low (linear regression r^2=0.12, p=0.09) for all 18 lakes. In addition a GLM model was run (log[adjHg] = logTMF + FCL = logTMF*FCL, resulting in similarly low predictability (r_{adj}^2=0.001) of Hg in char. This observation is not

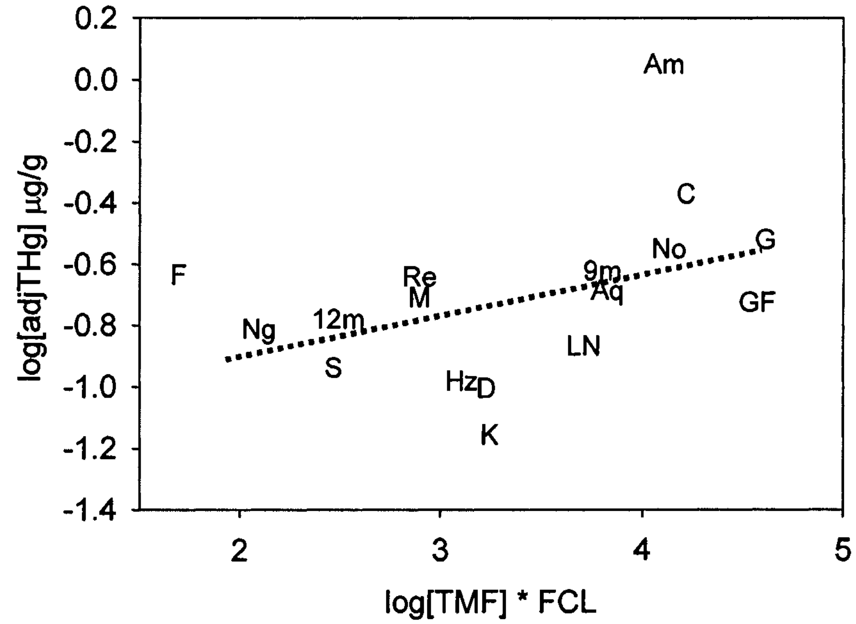

Figure 20: Modeled relationship of FCL-adjusted-[logTMF] and Length-adjusted[THg]. The dashed line indicates the observed trend (non-significant, p=0.09). The first or first two letters of the lake name are used and codes are given in Table 5.

only limited to MeHg accumulation (Guildford et al. 2008; Houde et al. 2008), and we suggest it to be considered in future bioaccumulation studies.

4.4.7 Hg species in water and sediments

Measurements of MeHg in water were available for 6 lakes, although they were typically near or below the detection limits of the analytical laboratories (~0.01 to 0.02 ng/L), making valid comparisons impossible. Concentrations of THg in water ranged from 0.29 to 0.72 ng/L in North Lake and Lake F, respectively. Length-adjusted THg concentrations in Arctic char, which should reflect >90 % MeHg according to literature values for salmonid species (Kelly et al. 2008), were not correlated with THg concentrations in surface water ($p>0.05$) or those of surface sediments (Figure 19A and Figure 19D). In surface water, Hg is mainly present in its inorganic forms (92 - 97 % of THg), which may explain the poor relationship. In addition, surface water samples were obtained as a single annual sampling event per year, and may not accurately represent water Hg concentrations for the whole lake or season.

Concentrations of THg in sediments ranged from 6 to 156 ng/g (all dry weight) in Notgordie and Pingualuk lakes, respectively, averaging 27 ng/g overall ($n=18$). MeHg available from 8 lakes ranged from 0.10 ng/g in Teardrop and Char Lake to 0.24 ng/g (North Lake), thus representing only ~0.5 – 1.0 % of THg. Sediment concentrations were expected to be correlated with fish Hg, as they represent the site of methylation and thus the entry point of bioavailable (organic) MeHg into food webs. However, sediment THg (log[THg]) and MeHg (log[MeHg]) were not predictive of THg in fish (linear regression $p>0.05$).

4.4.8 THg in char and environmental parameters

Relationships of length-adjusted THg concentrations in Arctic char with a number of known predictors of THg in fish revealed only a few of the putative predictors to be explanatory of our study data. Latitude appears to have no effect on Hg concentrations in landlocked char (linear regression $p>0.05$, Figure 19B). Nor was there any correlation between THg in char and longitude ($p>0.05$, Figure 19C). Catchment area and lake area did not predict THg concentrations in char (both linear regression $p>0.05$). However, catchment-to-lake area ratios (CA/LA) did show a positive relation (linear regression $r^2=0.27, p=0.01$) with THg in char, with greater CA/LA ratios leading to higher THg concentrations in char (Figure 19F, Table 6). The highest ratios were calculated for Amituk and North Lakes, which also had higher Hg concentrations in char. Lake Pingualuk, a meteor impact crater lake, has a minimal catchment area, and low THg concentrations. Multiple linear regression analysis using CA/LA (n=17) and FCL as independent variables revealed the relationship log[length-adjTHg] = -1.039 + (0.00585 * CA/LA) + (0.0662 * FCL), and weakly predictive of length-adjusted THg ($r^2=0.24$, $p=0.058$) . These results indicate that input of Hg from the surrounding land into lakes may have an effect on concentrations in char. The influence of CA/LA ratios on sediments have been previously described for mid-latitude lakes (Lorey and Driscoll 1999). Muir et al. (2008) suggest that increased aeolian inputs and/or greater erosion due to higher snowfall over the last 50 years could have affected sediment Hg concentrations in the central Canadian Arctic archipelago. Linkage of Hg deposition data and MeHg in a southern fish species (Largemouth bass, *Micropterus salmoides* L.) has been established by Hammerschmidt and Fitzgerald (2006), in multiple fish species in Eastern North America (Kamman et al. 2005), and in recent experimental studies in Northern Ontario, Canada (Harris et al. 2007). Moreover, the results of the present study highlights a possible linkage to climate vulnerability; melting permafrost or changes in precipitation under predicted climate scenarios (Prowse et al. 2006) could increase Hg inputs from catchments to lakes. Predicted dryer conditions within the watershed owing to warmer summers (Smol and Douglas 2007) could lead to drying out of wetlands and increased oxidative processes in normally wetted soil. Lakes of similar size within one watershed would respond similarly to change, as supported by our results from Ellesmere Island, Nunavut. Consequently, small lakes with large catchment areas may be most vulnerable to increased inputs owing to climate induced changes in precipitation and temperature. Catchment-or-lake area are indicative of contributions of Hg to sediments, however, in order to estimate the contribution of Hg from the atmosphere, catchment-specific precipitation needs to be taken in account.

Table 6: Relationships of log[length-adjTHg] with location and selected environmental factors

	linear regression parameters			
log[L-adj-THg] vs	**slope**	**intercept**	r_{adj}^2	p
Latitude			0.041	0.17
Longitude			-	0.36
Lake area (LA)			0.114	0.09
Catchment area (CA)			-	0.90
CA-LA ratio	0.2643	-0.9795	0.269	0.01
DOC			-	0.47
Chlorophyll a			-	0.69

Underlying geology can affect Hg concentrations in biota, as shown for Arctic fish by Lockhart et al (2005) and for ringed seals from Canada and Greenland (Riget et al. 2005). However, the geology on Cornwallis Island and Ellesmere Island is similar; it is composed of tertiary sediment over carbonate bedrock (Wheeler 1997) and all lakes on these islands can be considered to be on sedimentary bedrock (Lockhart et al. 2005). All three small lakes on Ellesmere Island are located on the Hazen plateau, which is known to be an 'Arctic oasis' due to its topo-climatic effects (Smith 2002). Kent Peninsula area lakes may be influenced by metamorphic bedrock due to the nearby continental shelf/mainland. This could hold true for Baffin Island and the North Slope of the Yukon (Herbert Lake) as well, as underlying geology may be an alternative explanation for the high THg concentrations in Radar Lake.

4.5 Conclusions

Mercury concentrations from 27 Arctic char populations from a large geographical area are summarized here, making this one of the most recent and comprehensive investigation of its kind for the Canadian Arctic. Our study thus expands our knowledge from previous studies on Hg concentrations in landlocked char (Köck 2004; Lockhart et al. 2005; Muir et al. 2005) not only by increasing number of lakes, but including information on food webs. We demonstrate intra-lake variability in THg concentrations due to difference in size, age, and trophic position of individual char within a lake, which concurs with the literature on other freshwater fish. We have established FCL for all lakes, providing insights on its use in explaining variability in Hg concentrations in top predators. No single factor predicting THg concentrations was derived for all lakes in the present study, indicating that a combination of above the factors in conjunction with biotic lake characteristics (such as TMFs and FCL combined) influence THg concentrations in Arctic char of a given lake. Spatial comparison of a statistically adjusted subset of samples indicated inter-lake variability of THg, but no relation to latitude or longitude. This could be because biological factors override indicators of climatic conditions (e.g., latitude). Catchment-to-lake-area ratios were related to THg concentrations in fish. This relationship highlights a possible linkage of Hg concentrations in fish and climate change because increased permafrost melt may result in greater inputs of Hg into lakes.

4.6 Acknowledgment

The authors wish to thank Deborah Iqaluk and Pilipoosie Iqaluk for invaluable assistance with field collections in 2005-2007. Polar Continental Shelf Project managers and employees provided logistical support (project numbers 2005-2007) and contributed to our team's safety and comfort during field work near Resolute Bay, Cambridge Bay, and Ellesmere Island. Milton Shears and co-workers collected all samples from Labrador. Parks Canada and staff aided greatly in sampling sites within Quittinirpaaq National Park on Ellesmere Island. Rick Wastle performed the ageing of char otoliths at the Freshwater Institute, Winnipeg, MB (Canada). John Chetelat and Marc Amyot provided MeHg data for surface water of 6 lakes. We thank Jessica Epp (Environment Canada) for assisting in THg analysis and determination of lake and catchment areas. This research was conducted under the terms of the Nunavut Research Institute (License: 0203206R-M), Department of Fisheries and Oceans (License: S-06/07-1015-NU), and the Ministre des Resources Naturelles et de la Faune du Québec (License: 9053.28). All work followed the of the University of Guelph animal care protocols (#06R083). The Northern Ecosystem Initiative, Northern Contaminants Program, Network of Centres of Excellence Arctic Net, International Polar Year (IPY 07/08), and Austrian Academy of Science (DOC-Stipend for lead author) provided funding for this research. Two anonymous reviewers have improved the quality of this work through their comments.

5 FRACTIONATION AND VARIATIONS OF STABLE ISOTOPES OF MERCURY IN ARCTIC LAKE FOOD WEBS

In preparation for submission Environmental Science and Technology

5.1 Abstract

Biotic and abiotic fractionation of mercury (Hg) isotopes has recently been shown to occur in aquatic environments. We determined isotope ratios (IRs) of Hg in sediments, biota (chironomids, zooplankton, Arctic char) of 10 Arctic lake food webs from 4 regions and investigated extent of Hg isotope fractionation. Samples were digested, and Hg isotope ratios were analyzed by multi-collector inductively coupled plasma mass spectrometry (MC-ICP/MS). Hg mass independent fractionation (MIF; Δ^{x}Hg) as well as mass dependant fractionation (MDF; δ^{x}Hg) were calculated and used to compare samples. Trophic signatures (δ^{15}N and δ^{13}C) were also examined to explain differences among species, lakes, and regions. We further investigated where fractionation may occur, and if the results were useful to distinguish sources from processes. IRs of Hg in sediment were consistent with data reported in the literature, and are characterized mainly by MDF and low MIF (Δ^{199}Hg -0.37 to 0.74 ‰). However, all biota were characterised by the evidence of MIF, most pronounced in zooplankton (Δ^{199}Hg up to 3.40 ‰) and fish (Δ^{199}Hg up to 4.87 ‰). Zooplankton showed greater degree of MIF than char in two lakes, possibly due to the uptake of Hg from the water column, as indicated by δ^{13}C data. Hg IRs in char varied among region, while char from lakes from each region showed similar degrees of MIF. A MIF-factor was derived representing the mean MIF difference between sediment and fish, and indicated that fish in two regions retain sediment signatures altered by a consistent factor. Due to its unique characteristics, e.g. minimal lake-to-catchment area, very high water retention time (~360 years), the meteor impact crater lake (Pingualuk) could reflect a "pure" atmospheric Hg signature, while others could reflect a mixture of terrestrial and atmospheric Hg inputs.

5.2 *Introduction*

The heavy metal mercury (Hg) is naturally present in seven stable isotopes (^{196}Hg, ^{198}Hg, ^{199}Hg, ^{200}Hg, ^{201}Hg, ^{202}Hg, and ^{204}Hg), which differ in relative mass by up to 4 %. Characterizing the fingerprints of Hg isotope (i.e., ratios) in the environment has been proposed as a tool of tracing sources of Hg pollution (atmospheric or point sources) (Blum and Bergquist 2007). In the past, stable isotope ratios of a number of elements (Li, Mg, Ca, Fe, Cu, Pb, Zn, Se, and Mo) have been used as isotopic tracers in the environment (Johnson et al. 2004). Hg isotopes are probably fractionated by industrial processes prior to deposition in the environment, resulting in specific isotopic ratios that may be specific to a region or even to an industry (Ridley and Stetson 2006; Jackson et al. 2008). While Hg isotope signatures could be highly useful in linking Hg found in the environment to a specific source (such as a certain industry, or industrial process, or a country), fractionation effects are likely to change Hg isotope ratios (IRs), which may obscure the initial fingerprint. While earlier studies have focused on detecting mass-dependent fractionation (MDF), more recently the discovery of mass-independent fractionation (MIF) of Hg in biota has received more attention. MIF is specifically associated with Hg isotopes of odd mass numbers (^{199}Hg and ^{201}Hg). This fractionation may be useful to track Hg transfer through the environment, since differences in MIF are more pronounced in biota than MDF (Jackson et al. 2008). Environmental fractionation of Hg, aided by biotic and abiotic (e.g. photochemical) processes (Ridley and Stetson 2006), has been experimentally demonstrated in water (Bergquist and Blum 2007), Hg resistant bacteria (Kritee et al. 2007), and naturally confirmed in sediments (Jackson et al. 2004; Xie et al. 2005; Foucher and Hintelmann 2006), ore deposits (Hintelmann and Lu 2003; Smith et al. 2005; Smith et al. 2008), as well as biota (Bergquist and Blum 2007; Ghosh et al. 2008; Jackson et al. 2008). Varying degrees of both MDF and MIF have been reported by these authors. Most recently, Jackson et al. (2006; 2008) reported biotic MIF in food webs (including fish) of three lakes in Canada (Lakes Cli, Shipiskan, and Ontario). MIF in several fish species was also documented by Bergquist and Blum (2007). The reported values of MDF range from ~6 ‰ by bacteria (δ^{202}Hg, experimental) (Kritee et al. 2007), ~5.5 ‰ in ores, to -1 ‰ to 4 ‰ in sediments (δ^{202}Hg, natural) (Foucher and Hintelmann 2006), and MIF values range from 2.5 ‰ (Δ^{199}Hg, photo reduction experiments) (Bergquist and Blum 2007), up to ~4 ‰ (Δ^{201}Hg) in various fish species (Bergquist and Blum 2007; Jackson et al. 2008). The precision of isotope ratio measurements by MC-ICP/MS allows detection of fractionation of about 0.2 ‰ or higher.

Two mechanisms are thought to underlie MIF of Hg: the nuclear field shift effect (Bigeleisen 1996; Schauble 2007), and the magnetic isotope effect (Buchachenko et al. 2007; Jackson et al. 2008). The latter may be mainly responsible for MIF observed in photochemical reaction experiments (Bergquist and Blum 2007). Ghosh et al. (2008) recently proposed calculating the relative contribution of these two effects to the MIF in peat and mosses using Δ^{201}Hg/Δ^{199}Hg ratios. Although the occurrence of MIF has been established, the exact molecular mechanisms for MIF in/by biota remain unclear (Bergquist and Blum 2007; Jackson et al. 2008).

It is of great interest to distinguish the sources and possibly geographic origin of atmospherically deposited Hg because it would allow better control of Hg emissions. The Arctic is an area of great concern because the diet of the indigenous peoples, particularly the Inuit (of Alaska, Canada and Greenland) who largely depend on a subsistence harvest of top predator species (fish, seals, and whales), which are known to contain high concentrations of Hg (Van Oostdam et al. 2005). The Arctic receives most of its anthropogenic Hg through the atmosphere (St Louis et al. 2005; Hedgecock et al. 2008), but inputs into lakes can also occur from natural sources (Air Pollution Control Directorate 1981). Semkin et al. (2005) showed that terrestrially derived Hg also plays a role in contributing Hg to arctic lakes through run off. From atmospheric modeling, we know that global and transpolar transfer of gaseous elemental Hg is possible (Dastoor et al. 2008; Jaffe and Strode 2008), owing to its long residence time (0.75 - 1 years) (Lindberg et al. 2007). The Canadian Arctic, and more specifically, High Arctic regions (north of 70° N), are known to be influenced by Eurasian sources (Brooks et al. 2005).

The relative contribution from these sources and their pathways are not fully understood, thus IRs, and in particular MIF have been proposed as a possible tool to trace sources of Hg in the environment. MIF signatures in fish may not need to be identical to atmospheric signatures to identify sources, as long as all fractionation steps between Hg(II) deposited in sediments leading to MeHg in fish are consistent among lakes. However, information on Hg isotopic ratios in Arctic samples is limited to one sediment core from Romulus Lake, Ellesmere Island, Nunavut, Canada (Jackson et al. 2004) and one study on Alaskan seabird eggs (Sonke et al. 2008).

Arctic lake food webs are low in biodiversity and the transfer of nutrients is coupled to benthos (Sierszen et al. 2003): from sediment (site of methylation) to benthic invertebrates (chironomids) to adult Arctic char. Zooplankton is part of the char diet, but may not contribute greatly to Hg uptake (Gantner et al. 2009b). Cannibalism is known to occur (Hammar 2000), and can effect Hg concentrations in adult char (Gantner et al. 2009a).

We set out to determine if: 1) Hg IRs in sediments and Arctic char vary by lake and region, 2) if there is fractionation along the food chain, and 3) whether the results can be used to distinguish sources from processes. Accordingly, we measured stable isotope ratios of Hg, N, and C in surface sediments (surface layer 0-2cm), zooplankton (bulk), emerging insects (chironomid midges), and top predator fish (Arctic char, *Salvelinus alpinus* L.) from 10 lakes across a wide geographic range in the Canadian Arctic. The $\delta^{15}N$ and $\delta^{13}C$ isotope data were used to infer trophic relations within the biota of each lake and were compared with the Hg isotope data to help clarify variations in the isotope composition of Hg along food webs.

5.3 Materials and Methods

5.3.1 Sampling sites

Three lakes in three primary sampling locations in the Canadian Arctic were selected over a range of latitudes and longitudes (Figure 21, Table 7). One site from a fourth region (Lake Pingualuk, 61°N) was included as a potential reference site for atmospheric Hg deposition. This deep lake fills a meteor-impact crater and has a minimal catchment (catchment-to-lake area ratio ~0.4), receiving most of its water through precipitation while ground water seepage is minimal (Appx 12). Lake D, Lake G, and Lake Hazen, located within Quttinirpaaq National Park on Ellesmere Island (Nunavut, Canada), represent sites the most northern region (81-82°N). Amituk Lake, 9 Mile Lake, and Resolute Lake are central Arctic sites on Cornwallis Island (74-75° N). Little Nauyuk Lake, Notgordie Lake, and Gavia Faeces Lake, all on Kent Peninsula, are western Canadian Arctic sites (~68°N). We compared our results among all lakes, among lakes by region, and among regions.

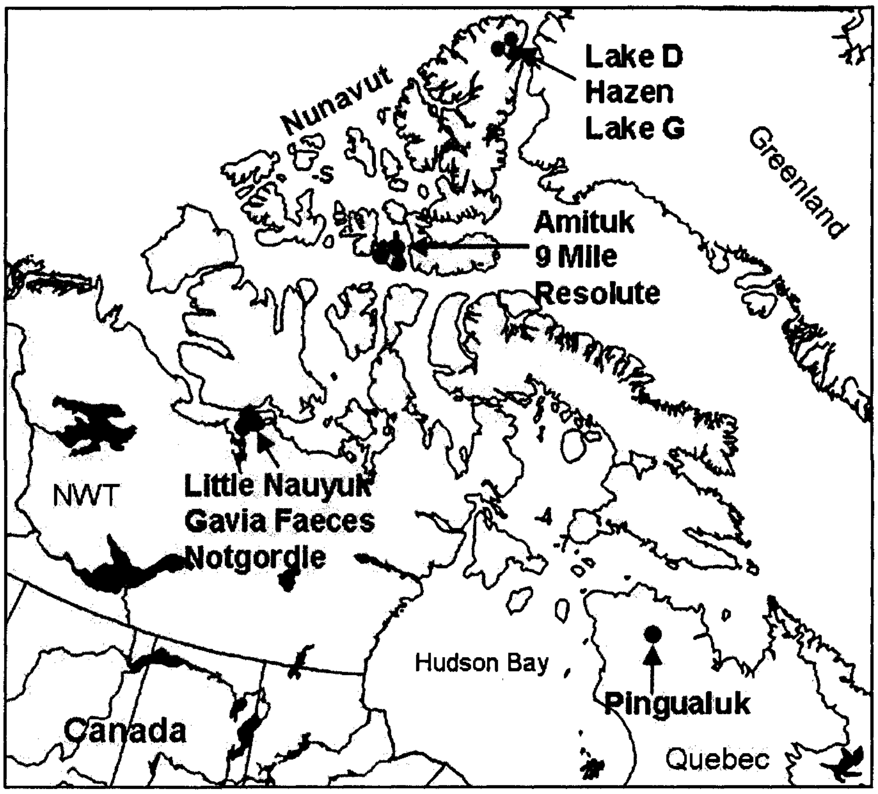

Figure 21: Map of northern Canada and part of Greenland showing the 10 sampled lakes (red dots indicate approximate locations of lakes) in four Arctic regions (3 in Nunavut, one in Nunavik, northern Quebec). Detailed location and lake characteristics are given in Table 7. NWT = North-West Territories.

5.3.2 Sample collection

From each of the ten lakes, six Arctic char were collected in July and August of 2006 and 2007 (with exception of Lake Pingualuk char, which were collected in May 2007). For eight lakes, the selection of Arctic char was based on similar $\delta^{15}N$ signatures and total Hg concentrations ([THg]) in muscle tissue (Appx 23) to reduce skewing of results through trophic variability or differences in [THg]. To test for influences of $\delta^{15}N$, two morphotypes of Arctic char (i.e., three cannibal and three piscivorous individuals) from Lake Hazen (Guiguer et al. 2002) were selected. For Amituk Lake, Arctic char were selected from a wide range of [THg] (0.9 - 3.0 μg/g ww), allowing us to investigate any effects [THg] may have on fractionation of Hg isotopes. All analysis for Arctic char (Hg IRs, [THg], $\delta^{15}N$, and $\delta^{13}C$) was performed on sub samples of dorsal muscle tissue.

Samples of the principal prey items of Arctic char (i.e., emerging chironomid midges and zooplankton) of known MeHg content were obtained from four lakes to investigate IRs. Zooplankton from Gavia Faeces and Notgordie Lakes, and Lake Hazen were collected in 2006 and 2007, respectively. Unfortunately, the amount of zooplankton obtained from Lake Pingualuk was not sufficient to allow the measurement of IRs. For Resolute Lake, zooplankton samples from 2005 to 2007 were analyzed to evaluate annual variability of IRs. Adult chironomid midges were collected from Resolute Lake and Lake Hazen. High Arctic chironomid species stop feeding several months prior to emergence (Danks 2004), thus Hg concentrations and IRs measured reflect body burden and not gut contents. Sediment samples were obtained using a Ponar sampler, and the upper 2 cm sub-sampled for analysis. Three sediment samples collected in 2007 were available from Lake Hazen, allowing investigation of spatial variability.

5.3.3 Analysis of THg and IRs

The THg content of fish and sediment samples was determined by direct combustion followed by analysis with a Milestone DMA 80 (Milestone Inc, Shelton, CT, USA; EPA method 7473). MeHg concentrations in zooplankton and chironomids were determined by GC CV-AFS, following methods described in (Gantner et al. 2009c). Stable isotope analysis of carbon and nitrogen was conducted using MC-ICP/MS at the Environmental Isotope Laboratory, University of Waterloo (Canada). Lake area (LA) and catchment area (CA) were available for all sites, allowing us to investigate the potential effect of inputs of Hg from the terrestrial surroundings (Outridge et al. 2005; Semkin et al. 2005; Hammerschmidt et al. 2006) of a lake by comparing CA/LA ratio and MIF.

For Hg isotope analysis, all samples were digested with a hot plate at ~120 °C using ~5 mL *aqua-regia* (HNO_3/HCl, 1:3 v/v) in pre-cleaned glass vials. Following digestion, the sample extracts were made up to 40 mL with Milli-Q water, aiming for final concentrations of 0.5, 1.0, or 2.0 ng/mL, to ensure sufficient detection for MC-ICP/MS analysis. Stable isotopes of Hg were measured with a Thermo-Finnigan *Neptune* MC-ICP-MS unit (ThermoScientific, Bremen, Germany), employing the method of Foucher and Hintelmann (2006). Here, we are using the delta notation (δ) as calculated from $\delta^{X}Hg = 1000 \times (^{198}Hg / ^{X}Hg_{sample} - ^{198}Hg / ^{X}Hg_{standard}) / ^{198}Hg / ^{X}Hg_{standard}$, and are reporting MDF in terms of $\delta^{202}Hg$ and MIF as $\Delta^{199}Hg = \delta^{202}Hg \times 0.252$ (unless otherwise noted). The theoretical mass dependant fractionation for all Hg stable isotopes was calculated following the kinetic fractionation law (2002), and was used to contrast MDF and MIF in samples from the theoretical MDF.

5.4 Results and Discussion

5.4.1 Surface sediments

The 13 sediments revealed mostly MDF of Hg, and little deviation from the theoretical MDF line for Hg isotopes (Figure 22). Ratios of $\delta^{202}Hg$ ranged from -2.03 to 0.25 ‰ (average -0.59 ± 0.66 ‰) overall. Regional comparison of sediment IR signatures from Ellesmere Island (n=6), Cornwallis (n=3) and Kent Peninsula (n=3) revealed small differences, with $\delta^{202}Hg$ averaging -0.94 ± 0.11 ‰ (average ± SE), 1.23 ± 0.41 ‰, and -1.02 ± 0.32 ‰, respectively. Our results are in agreement with literature values for two sub-arctic lakes of -2.3 and -1.6 ‰ $\delta^{202}Hg$ in Cli and Shipiskan (0 - 0.5 cm), respectively (converted to $^{202/198}Hg$ notation from Jackson et al. (2008)). However, sediments in subarctic lakes appear slightly more depleted in heavier isotopes. The Hg signal detected in lake sediments is probably a combination of Hg derived from the atmosphere directly and from the catchment (atmospheric + terrestrial/natural) of the lakes, and the isotope signatures of these sources may have been different. The upper ~2 cm of sediment layer

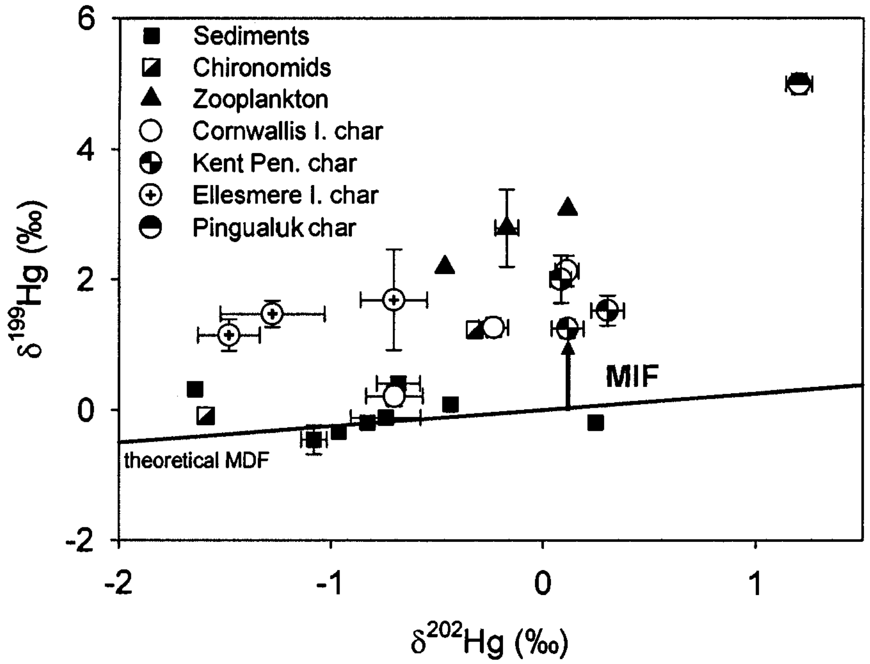

Figure 22: δ^{199}Hg versus δ^{202}Hg in sediments, Arctic char (by region), zooplankton, and chironomid samples (means ± 1SD). The continuous line represents the theoretical mass-dependant fractionation (MDF) calculated following Young et al. (2002). Deviation from this line (red arrow) represents mass-independent fractionation (MIF).

analysed in our study likely reflects a integrated Hg signal of 15 - 30 years of deposition and sedimentation (Muir et al. 2008), which could explain the variation among lake sediments. Variation of up to 1.5 ‰ in δ-values in the upper 15 cm was reported for one Arctic sediment core (Romulus Lake, Ellesmere Island) (Jackson et al. 2004), however, variation was small (<0.5 ‰) in the first 2 cm (~14 years). The fact that methylation of Hg mainly occurs in the top layer of the sediment, rather than throughout our sampled upper ~2 cm, may also have affected our results. Isotope ratios (Δ and δ) of sediments are available in the Appendix (Appx 24).

Interestingly, the sediment of Lake Pingualuk appeared to differ noticeably from the other samples, showing the only positive δ^{202}Hg signature (+0.250 ‰) of all sediment samples analyzed. This could be due to the relatively small catchment of this lake, and the very limited influence of terrestrially derived input of Hg (Table 7).

Table 7: Location and selected characteristics of lakes (Maps and photographs Appx 1-Appx 11)

Lake name	Region	Location		Geo-morphology	Lake area	Catchment area	CA/ LA
		° N	° W		km^2	km^2	km^2
Lake D	Ellesmere Island	82.11	67.48	Carbonate bedrock	2.2	10.9	5.1
Lake G		81.82	69.18		3.1	31.3	10.1
Hazen		81.83	70.42		538	4363	8.1
Amituk	Cornwallis Island	75.05	93.75	Carbonate bedrock	0.4	26.1	69.1
9 Mile		74.81	95.20		0.7	3.6	5.4
Resolute		74.68	94.88		1.3	18.7	14.7
Gavia Faeces	Kent Peninsula	68.34	107.73	Schist / Granite	0.2	0.3	1.7
Little Nauyuk		68.35	107.75		0.5	1.6	3.1
Notgordie		68.35	107.66		0.1	0.5	6.4
Pingualuk	Ungava Peninsula	61.28	73.66	Canadian shield	6.2	2.8	0.4

Overall, MIF in sediments was higher than expected, ranging from -0.371 to 0.736 ‰ (mean 0.088 ‰), respectively. The lowest MIF occurred in one of the northern lake sediments (Hazen; mean Δ^{199}Hg = -0.18 ± 0.20 ‰, n=4), while the greatest MIF in sediment was found in a southern lake (Notgordie; Δ^{199}Hg = 0.74 ‰, n=1). However, no relationship was found with MIF in sediments and latitude overall (linear regression $p>0.05$).

We did find a positive linear relationship between Δ^{199}Hg and Δ^{201}Hg (regression parameters: $\Delta^{199}Hg = 0.737 * \Delta^{201}Hg - 0.092$; $r^2=0.83, p<0.01$). Variability of Hg isotopic composition in sediment was low in terms of δ^{202}Hg (-1.12 ± 0.01 ‰) and Δ^{199}Hg (-0.2 ± 0.1 ‰), as indicated by three samples obtained from Lake Hazen in 2007 (mean of three samples ± SD).

The mean Δ^{201}Hg / Δ^{199}Hg ratio of sediments for the lakes as a whole was 1.06. The highest Δ^{201}Hg / Δ^{199}Hg ratio was calculated for the most northern lake sediments on Ellesmere Island (1.32), while Cornwallis Island lake sediments had a ratio of 0.99, and the most southern sediments on Kent Peninsula a ratio of 0.53. Pingualuk sediment had the highest ratio (2.03) of all individual lakes. These ratios are in very good agreement within the range (0.54 to 2.00) reported by Ghosh et al. (2008) for peat. According the authors, the Δ^{201}Hg / Δ^{199}Hg ratio can be used to infer the relative contribution of the two underlying effects leading to MIF fractionation, namely nuclear volume (NVE) and magnetic isotope (MIE) effect. Hg in Pingualuk sediments would thus be mainly fractionated by the NVE, while Kent Peninsula sediments are fractionated by the MIE.

5.4.2 Arctic char

The Hg signatures of 50 Arctic char muscle samples ranged considerably overall (δ^{199}Hg: 0.000 to 5.193 ‰, and δ^{202}Hg: -1.679 to 1.267 ‰) (Table 8). However, Arctic char from lakes within each region revealed strikingly similar degrees of fractionation (Figure 23). Char from Ellesmere Island lakes (Lake D, Lake G, and Hazen) showed the overall lowest fractionation (δ^{199}Hg: 0.860 to 2.026 ‰; δ^{202}Hg: -1.679 to -0.615 ‰), while Arctic char from Ungava Peninsula (Pingualuk) had the highest δ–values (δ^{199}Hg: 4.846 to 5.193; δ^{202}Hg: 1.129 to 1.267 ‰). Arctic char from lakes on Cornwallis Island and Kent Peninsula had similar fractionations (or deviations), ranging from δ^{199}Hg: 0.000 to 2.320 ‰ and δ^{202}Hg: -0.864 to 0.175 ‰, and δ^{199}Hg: 1.025 to 2.370 ‰ and δ^{202}Hg: 0.029 to 0.410 ‰, respectively (Appx 23).

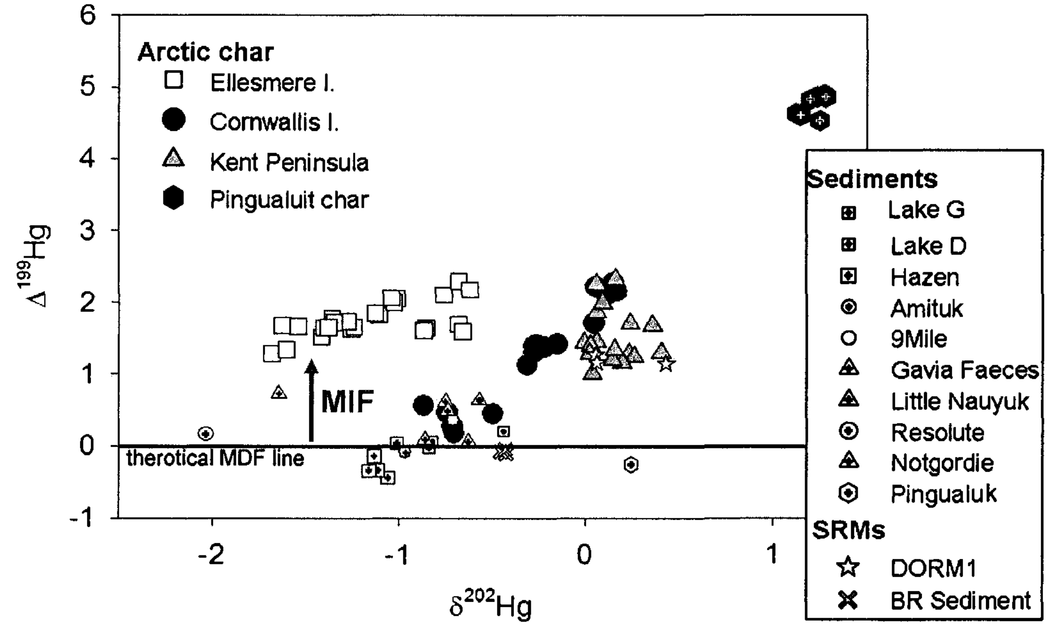

Figure 23: MIF Δ^{199}Hg versus δ^{202}Hg in Arctic char, sediments, and standard reference materials (SRMs).

Table 8: δ^{x}Hg and Δ^{x}Hg values for Arctic char from 10 lakes

Lake		δ^{199}Hg		δ^{200}Hg		δ^{201}Hg		δ^{202}Hg		Δ^{201}Hg		Δ^{199}Hg	
	n	‰	SD	‰	SD	‰	SD	‰	SD	‰	SD	‰	SD
Lake D	5	1.69	0.77	-0.29	0.10	0.97	0.59	-0.70	0.16	1.50	0.66	1.86	0.80
Lake G	5	1.15	0.25	-0.70	0.09	0.04	0.29	-1.48	0.15	1.15	0.18	1.52	0.21
Lake Hazen*	3S	1.63	0.13	-0.53	0.07	0.71	0.14	-1.08	0.05	1.53	0.10	1.91	0.12
	3L	1.30	0.05	-0.68	0.08	0.21	0.13	-1.47	0.19	1.31	0.02	1.67	0.01
	6	1.47	0.20	-0.60	0.11	0.46	0.30	-1.28	0.25	1.42	0.14	1.79	0.15
Amituk Lake	5	0.21	0.15	-0.37	0.12	-0.44	0.15	-0.70	0.13	0.08	0.14	0.39	0.16
9 Mile Lake	5	2.13	0.23	0.12	0.04	1.60	0.22	0.11	0.06	1.51	0.19	2.10	0.22
Resolute Lake	4	1.26	0.15	-0.07	0.06	0.78	0.16	-0.23	0.07	0.95	0.12	1.32	0.13
Gavia Faeces L.	5	1.52	0.23	0.25	0.04	1.29	0.20	0.30	0.08	1.06	0.19	1.45	0.23
Little Nauyuk L.	5	1.24	0.14	0.14	0.04	1.02	0.13	0.12	0.08	0.93	0.12	1.21	0.14
Notgordie Lake	5	2.00	0.36	0.13	0.02	1.52	0.31	0.08	0.05	1.46	0.29	1.98	0.35
Lake Pingualuk	5	4.99	0.15	0.70	0.03	4.60	0.14	1.20	0.06	3.70	0.12	4.69	0.15

* Three "small" (3S), three "large" (3L) ecotypes, and average of all six Hazen char are presented

Interestingly, δ^{202}Hg in fish decreased with increasing latitude. Linear regression analysis revealed a strong negative relationship (r^2=0.84, $p<0.05$) (Figure 24). This could be either due to different degree of fractionation of Hg during atmospheric transport to the Arctic, or reflect Hg derived from different air masses, or a combination of both.

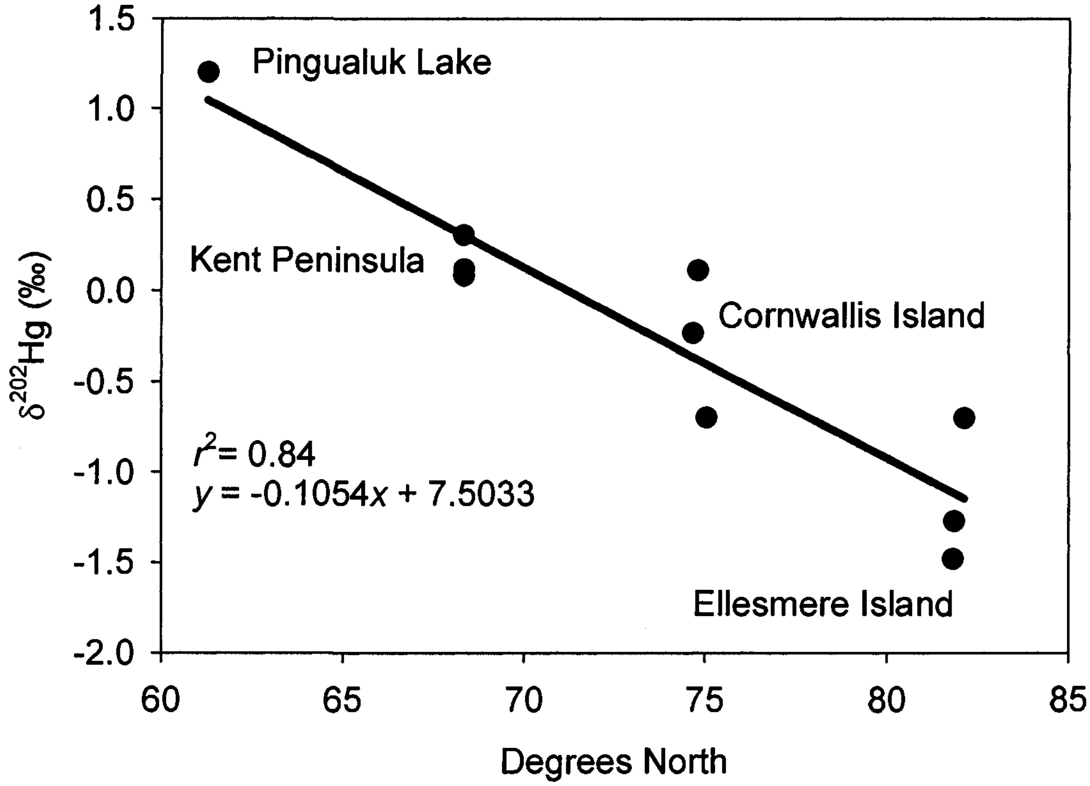

Figure 24: Negative relationship of δ^{202}Hg (in Arctic char) with latitude of lake.

MIF was evident in all Arctic char samples, Δ^{199}Hg ranging from 0.179 to 4.874 ‰ (Table 8). These results are in agreement with results for lake trout (*Salvelinus namaycush*) from subarctic lakes ($\Delta^{199/202}$Hg, 1.5 to 2 ‰) (Jackson et al. 2008) and burbot (*Lota lota*) from three Michigan lakes (Δ^{199}Hg, 3 - 5 ‰) (Blum and Bergquist 2007).

When comparing our regions, the lowest MIF occurred in the most northerly lakes on Ellesmere Island (range of Δ^{199}Hg: 1.283 to 2.203 ‰), while the greatest degree of

MIF was measured in Lake Pingualuk char (Δ^{199}Hg: 4.530 to 4.874 ‰). Again, Cornwallis Island and Kent Peninsula char had similar degrees of MIF (Δ^{199}Hg: 0.179 to 2.282 ‰; and 1.015 to 2.328 ‰, respectively). MIF in Pingualuk was higher than all three regions (ANOVA $p<0.05$, $p<0.05$), while there were no statistical difference amongst the other three regions ($p>0.05$). Amongst the Cornwallis Island char, Amituk Lake char ($n=5$) deviated from the other two sites within this region, as they showed aberrantly low MIF (Δ^{199}Hg: 0.179 to 0.563 ‰), which curiously resembles the range of MIF in sediments presented here. This may be due to the large catchment area of Amituk Lake, or different light conditions in this lake. The clear delineation of Δ^{199}Hg in char from the 4 regions is contrasted against δ^{202}Hg (Figure 23).

The relationship between Δ^{199}Hg and Δ^{201}Hg in all char combined was strong ($r^2=0.99$, $p<0.01$), and does not differ greatly among the four regions (Ellesmere Island $r^2=0.98$; Cornwallis Island $r^2=0.99$; Kent Peninsula $r^2=0.99$, Lake Pingualuk $r^2=0.97$, all $p<0.01$) (Figure 25). The mean Δ^{201}Hg / Δ^{199}Hg ratios for all char was 0.76±0.03 overall (excluding Amituk Lake char). Amituk Lake had a ratio of 0.08±0.43, and influenced the overall ratio greatly, prompting us to look at this site separately and exclude it. Mean Δ^{201}Hg / Δ^{199}Hg ratios of char were higher ($p_{\text{Tukey's}}<0.05$) on Ellesmere Island and at Pingualuk (both 0.79±0.02) than on Cornwallis Island (0.72±0.02, Amituk char removed) and Kent Peninsula (0.75±0.03). These ratios are lower than previously reported for fish-eating arctic birds (1.19) (Sonke et al. 2008), but lie with the range (0.54 – 2.00) suggested by Ghosh et al. (2008) to mark the contributions of the two mechanisms of MIF.

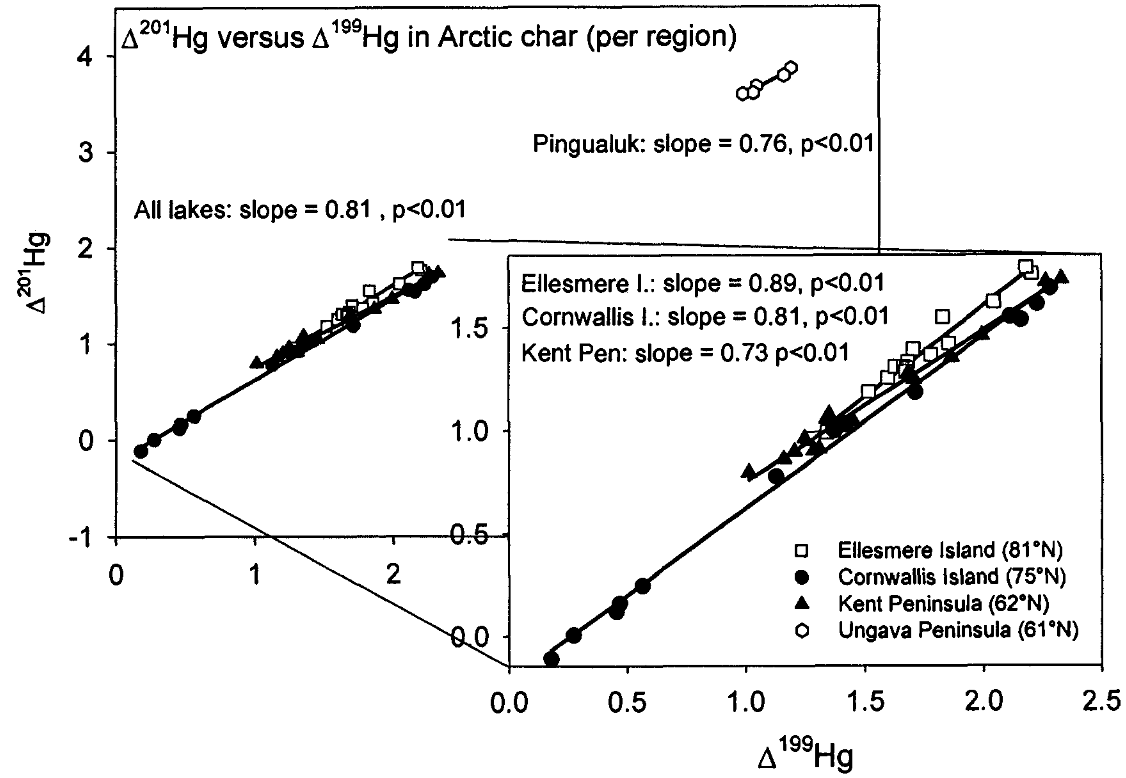

Figure 25: Regression equations of Δ^{201}Hg versus Δ^{199}Hg relationships for all lakes, and per region. The legend in the insert applies to both figures.

Previous studies showed that MIF varied among fish species (Bergquist and Blum 2007; Jackson et al. 2008), that influenced regional comparisons. Our study is the first to highlight regional differences in Hg isotope fractionation utilizing only one fish species over a large geographic range. We demonstrate here that the degree of Hg fractionation can vary considerably between geographic regions even within one species, which may be reflective of true regional differences of Hg signal. Among lakes of two of the three regions of which multiple lakes were sampled, however, fractionation was similar, indicating that regional Hg signals are consistent in neighbouring lakes. Region may be more important than trophic position of fish, as we demonstrate here (Figure 26, Appx 22). Watershed characteristics may need to be taken in account, as demonstrated by the differences between Amituk and the other two lakes on Cornwallis Islands. To test some of the hypotheses derived from this study, a comparison with a migratory char may reveal new insights, as they may reflect a marine Hg signal.

5.4.3 MIF in fish and sediments

Since >95% of all Hg in fish muscle is typically in the form of MeHg (Harris et al. 2003), fractionation of Hg isotopes during food web transfer should be associated with MeHg (Jackson et al. 2008). We compared the results of fractionation in char with sediment Hg IRs, where changes should reflect the integrated effects of processes occurring between Hg deposition (to sediments though sedimentation) and food web transfer to top predator.

We calculated the difference in MIF (Δ^{199}Hg and Δ^{201}Hg) between sediment signature and char signature, and refer to this measure as MIF-factor (MIF-factor = ($\text{MIF}_{\text{char}} - \text{MIF}_{\text{sediments}}$)), and compared the results amongst lakes and regions (Figure 26). Δ^{199}Hg in char and sediments was always significantly greater than Δ^{201}Hg in 7 lakes (t tests $p<0.05$), while 3 lakes showed equal MIF-factors of Δ^{199}Hg and Δ^{201}Hg (Lake D, Gavia Faeces, and Notgordie; t-test $p>0.05$). Significant differences in MIF-factors (Δ^{199}Hg and Δ^{201}Hg) were detected among all 10 lakes (ANOVA $p<0.05$), with Pingualuk showing the highest MIF-factor (~4.9 ‰ and ~4.2 ‰ for Δ^{199}Hg and Δ^{201}Hg, respectively) compared to all lakes ($p_{\text{Tukey's}}<0.05$), and lowest ($p_{\text{Tukey's}}<0.05$) in Amituk Lake (~0.5 ‰ and ~0.3 ‰ for Δ^{199}Hg and Δ^{201}Hg, respectively). Cornwallis Island lakes appeared most variable, showing significant differences ($p_{\text{Tukey's}}<0.05$), probably driven by the anomalous Amituk lake results. MIF-factors of lakes on Ellesmere Island did not differ among each other ($p_{\text{Tukey's}}>0.05$), and the same was determined for Kent Peninsula lakes ($p_{\text{Tukey's}}>0.05$) (Figure 26).

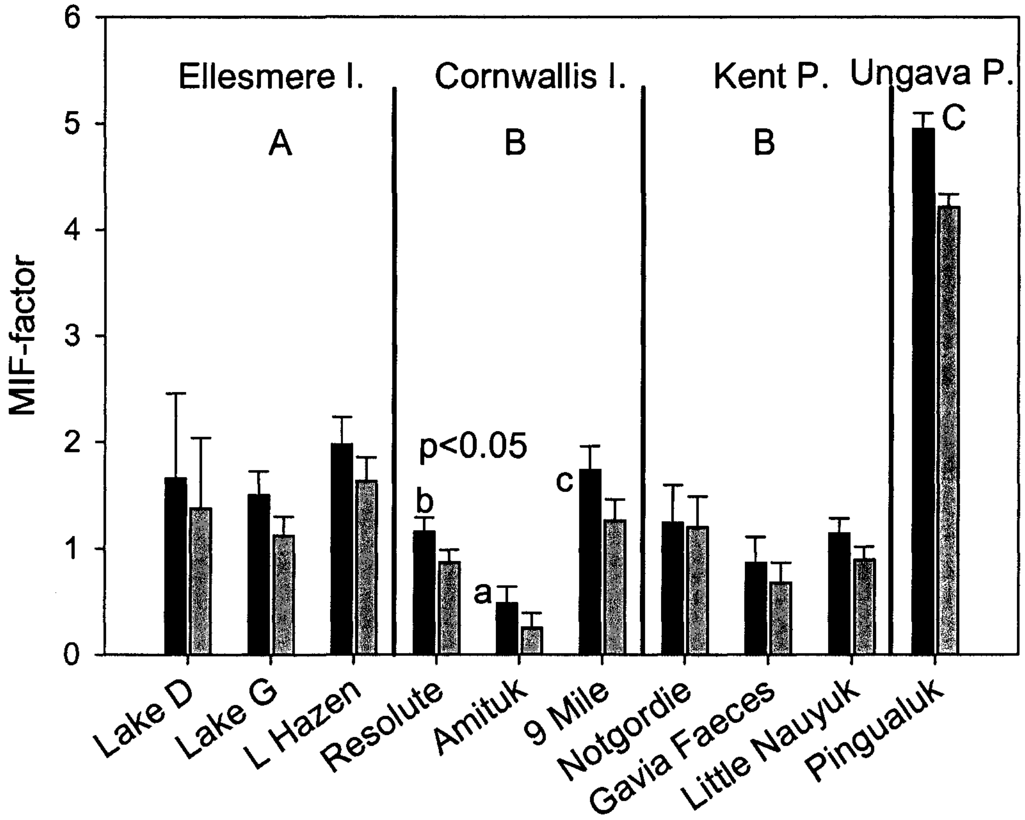

Figure 26: MIF-factor = ($MIF_{char} - MIF_{sediments}$) for $\Delta^{199}Hg$ (black bars) and $\Delta^{201}Hg$ (grey bars) in all 10 lakes. Capital letters indicate significant differences of regional mean MIF-factors (ANOVA $p<0.05$), small letters indicate significance of ANOVA Tukey's HSD within regions (statistic results shown refer to $\Delta^{199}Hg$).

When comparing mean MIF-factors of regions with each other, we did again find significant differences (ANOVA $p<0.05$). Ellesmere Island had higher MIF-factors than Cornwallis and Kent Peninsula sites ($p_{Tukey's}<0.05$), while the latter two did not differ ($p_{Tukey's}>0.05$). Consistency of this measure within regions could reflect consistent processes of fractionation along the food webs of neighbouring lakes. If these processes are consistent among lakes of one region, the MIF signal of sediments (sources) may thus be preserved, despite being altered by a MIF-factor resulting from fractionation effects during sedimentation or bioaccumulation and biomagnification up the food chain. The high degree of MIF in char indicates that MIF is either preserved in biotic samples despite bioaccumulation, or affected by bioaccumulation, which is in agreement with other studies (Jackson et al. 2004; Bergquist and Blum 2007; Jackson et al. 2008). The Hg isotopic signal carried in sediments may represent a mixture of natural geogenic and 'old' anthropogenic inputs of Hg (which ultimately come from atmospheric deposition), as well as newly deposited Hg. Sediment signatures may be altered over time, by methylation and de-methylation processes facilitated either by microbial activity (Jackson et al. 2004; Kritee et al. 2007), or photochemical reactions (Bergquist and Blum 2007). The degree to which the latter occur in the Arctic may be influenced by climatic drivers (ice-cover, light conditions, or water temperature).

The nature of Lake Pingualuk suggests that most of the Hg in the lake is derived from the atmosphere. Inputs are limited to the lake surface, as this crater lake receives its water mainly through precipitation (Bouchard 1989), and has a very small catchment-to-lake area ratio (0.4). In addition, the lakes great depth (>260 m) and the very steep shoreline indicates that Pingualuk Arctic char are not linked to the benthic environment or sediment. Stomach contents and $\delta^{15}N$ values indicate that adult Pingualuk char are cannibals, moreover their $\delta^{13}C$ signature implies a pelagic habitat, and their connection with the benthic environment is minimal (Gantner et al. 2009b). Thus, the high degree of Hg isotope fractionation in general, and MIF (~4.5 ‰) in particular, in Pingualuk char that were analysed suggest that the isotopic signature of this population is a 'pure' atmospheric Hg signal. Furthermore, Pingualuk was the only lake whose sediment yielded a positive $\delta^{202}Hg$ signature (0.250 ‰). This could be also due to its depth and its extraordinary clear water column (Secchi-depth of 33 m and a profundal depth of 87 m (= 1 % of surface light) (Bouchard 1989). Moreover, the sedimentation rate is extremely low (1.2 mm / 100 years) as determined most recently from sediment cores (R. Pienitz, University of Laval, Quebec, Canada, pers. comm.), indicating that Hg may remain in the water column (and thus exposed to light) much longer in this lake compared to other sites. This would allow photochemical reactions with Hg(0) or MeHg in the lake water, and subsequently MIF, to occur during sedimentation.

On the other end of the MIF spectrum (compared to all lakes in this study) is Amituk Lake. Arctic char here are known to have high [THg] (Muir et al. 2005). The THg profile in sediments cores suggest an anthropogenic signal (Muir et al. 2008), which may be derived via its large catchment (Semkin et al. 2005). Another study suggests that deposition of THg in sediments is a result of autochtonous carbon production and scavenging of Hg by algae (Outridge et al. 2007). However, the Amituk lake watershed is large (~26 km^2), compared to the lakes surface area (0.38 km^2), leading us to consider the effect of catchment inputs to the lake to be significant. The difference in fractionation (signal) of Hg in Amituk Lake char may thus be a result of the more terrestrially influenced Hg signal, as suggested by the low MIF in Amituk char.

5.4.4 Trophic relationships and MIF in food webs

Our δ^{13}C results confirm that zooplankton occupied a pelagic habitat and were subject to a pelagic dietary regime. Furthermore, zooplankton and chironomid midges had distinct δ^{13}C signatures, indicating utilization of different carbon sources (Figure 27). Zooplankton in the study lakes inhabit pelagic zones (δ^{13}C range: -35.7 ‰ to -29.7 ‰), while chironomid midges occupy benthic habitat (δ^{13}C range: -20.1 ‰ and -23.7 ‰). The

δ^{13}C values of Arctic char (-19.9 ‰ to -25.3 ‰) indicate that chironomids make up a large proportion of char diet (Figure 27A) (Gantner et al. 2009b; Gantner et al. 2009c).

High MIF in zooplankton was found in all four lakes. MIF in zooplankton of lakes Hazen and Resolute even exceeded the degree of MIF in Arctic char (Figure 27B, Appx 23, Appx 24). This is surprising, as previous studies in sub-arctic and mid-latitude lakes have shown zooplankton to have low MIF compared to fish (Jackson et al. 2008). Greater MIF in zooplankton may be due to a different source of Hg utilized by pelagic zooplankton. Zooplankton are likely receiving Hg directly from the water column (Kainz et al. 2002), in contrast to benthic invertebrates. Microcosm experiments using enriched Hg (odd isotopes ^{202}Hg and ^{200}Hg) undertaken during the METAALICUS experiment, also confirm rapid uptake of Hg by pelagic zooplankton (Orihel et al. 2008). MIF in zooplankton could thus reflect a (pelagic) water Hg signal, which maybe more representative of the atmospheric inputs than that of sediments. Possibly, MeHg present in the water column may also be photo-demethylated, resulting in a different MIF signature than sediments, as suggested by Lake Pingualuk sediment and fish results.

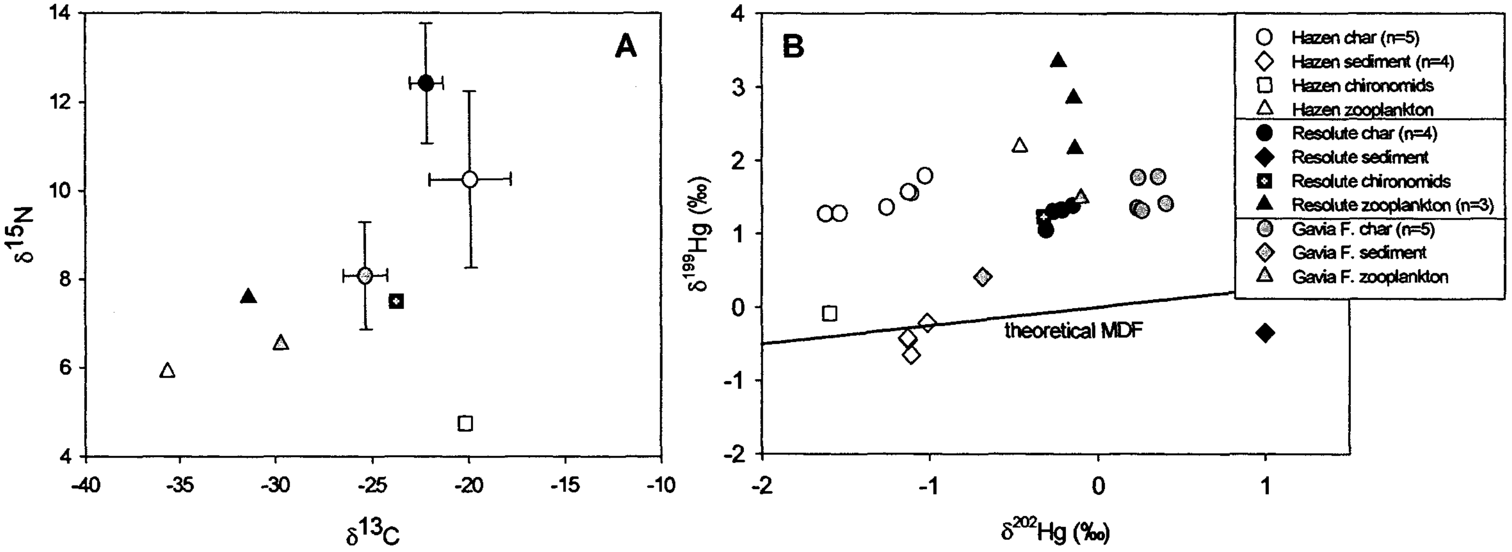

Figure 27: Three food webs (char, chironomids, zooplankton), one from each region are shown. A. Trophic signatures ($\delta^{15}N$, $\delta^{13}C$) indicate dietary relationships within the lakes; B. Isotope ratios ($\delta^{199}Hg$ versus $\delta^{202}Hg$) of the same biota samples as in A, and sediments (symbols of legend in B apply to A). Error bars represent 1 standard deviation.

Annual variation of δ^{X}Hg in three zooplankton samples from Resolute Lake (2005-2007) was considerable: (δ^{199}Hg: 3.343 ‰ in 2005 to 2.161 ‰ in 2006; δ^{201}Hg: 2.498 ‰ in 2005 to 1.445 ‰ in 2006), indicating some variability of the signal over time. It is possible that light conditions may have varied due to the extent of ice cover at time of sampling (July). Resolute Lake was mostly ice-covered 2005, partly ice-covered in 2006, and ice-free 2007. The degree of photo-demethylation (leading to stronger MIF) in the water column may have varied. During this period, mean (monthly) air temperatures in July of 3.8 °C (2005), to 4.3 °C (2006), and 7.4 °C (2007) were recorded at the nearby Environment Canada weather station (Appx 18). While these climatic indicators do not confirm our above hypothesis, they also do not eliminate the possibility that precipitation and thus optical properties of the ice-cover may have influenced light conditions in Resolute Lake prior to July. The degree of MIF in zooplankton varied between 2.2 – 3.4 ‰ (Δ^{199}Hg) and 1.5 – 2.7 ‰ (Δ^{201}Hg) over the same period (Figure 27B). This could indicate a response to variable proportions of terrestrial versus atmospheric inputs under the prevailing climatic conditions, as spring melt inputs are followed by dry summer conditions (i.e., overall low precipitation) and inputs via wind-blown dust (Semkin et al. 2005).

Chironomid midges showed MIF as well (Δ^{199}Hg: 0.0317 ‰ and 1.313 ‰) from Lake Hazen and Resolute Lake, although to a much lesser degree than zooplankton. At both sites, adult chironomid IRs were intermediate between those of sediment and Arctic char (Figure 27B). No chironomids were obtained at Lake Pingualuk and Kent Peninsula lakes. In contrast to pelagic zooplankton, chironomids are benthic organisms, most- or partly buried in the sediment throughout their larval stage. It is thus intuitive that these organisms would carry an Hg IR signal similar to the sediments, which is confirmed by the low MIF in chironomids. Jackson et al. (2008) report similar results for invertebrates from Lake Ontario (Ontario, Canada).

Modeling MIF

We applied a simple two source mixing model, attempting to predict MIF measured in chironomids. We hypothesised that benthos (i.e., chironomids) should have a mix of sediment MIF (low, as majority is THg) and fish MIF (high as a result of high MeHg). The model $[\text{MeHg}]_{\text{chironomid}} \times \text{MIF}_{\text{fish}} + [\text{Hg(II)}]_{\text{chironomid}} \times \text{MIF}_{\text{sediment}} = \text{MIF}_{\text{chironomid}}$ was applied, and compared to observed $\text{MIF}_{\text{chironomid}}$ (all Δ^{199}Hg). Predictions for Resolute Lake were near the measured value of 1.31, when using the average of all available chironomid MeHg concentrations (predicted $\text{MIF}_{\text{chironomid}} = 1.17$). However this model failed to predict the concentration in Hazen insects accurately (predicted $\text{MIF}_{\text{chironomid}} = 0.91$ versus measured MIF in Hazen chironomids was 0.32).

The two ecotypes of Arctic char from Lake Hazen revealed slightly different IRs (Table 8, Appx 22A). Cannibal char (large ecotype, δ^{15}N>12 ‰) had slightly higher δ^{199}Hg signatures (by ~0.2 - 0.4 ‰) than the insectivorous char (small ecotype, δ^{15}N<12 ‰) (t test [df] = 4, t = 4.111, p=0.015, n=3). Insectivorous char had also slightly lower MIF than large char (t test [df] = 4, t = 3.437, p = 0.026, n=3), which supports our argument that benthic feeding results in lower MIF in fish. A comparison of δ^{15}N and δ^{199}Hg of all Arctic char is available in the Appendix (Appx 22A).

In order to evaluate the influence of the amount of Hg (measured as THg) accumulated on IRs, char with a range of Hg concentrations (0.9 – 3.0 μg/g) were selected from the site with the highest concentration range (Amituk Lake, Cornwallis Island). Interestingly, IRs did not vary greatly among the samples of Amituk lake, indicating that the fractionation of Hg is consistent despite the great range of THg concentrations. The relationship of δ^{199}Hg and THg in all char samples is presented in the Appendix (Appx 22B).

5.4.5 Catchment–to-lake area and MIF

Although the "MIF-factor" was not related to ratio of catchment area (CA) / lake area (LA) overall (linear regression $p>0.05$) (Figure 28), results of Lake Pingualuk (which had the smallest CA/LA ratio and greatest MIF) and Amituk lake (which had the greatest CA/LA ratio and smallest MIF) indicate that this factor may nonetheless affect Hg isotope fractionation to some extent. The size of the catchment and precipitation determines the amount of terrestrial and atmospheric input a lake receives, but the design of this study did not allow detailed conclusions to be drawn from our observation. However, conventional Hg models assign catchment area a big influence (Hammerschmidt et al. 2006; Harris et al. 2007). On the other hand the influence may be

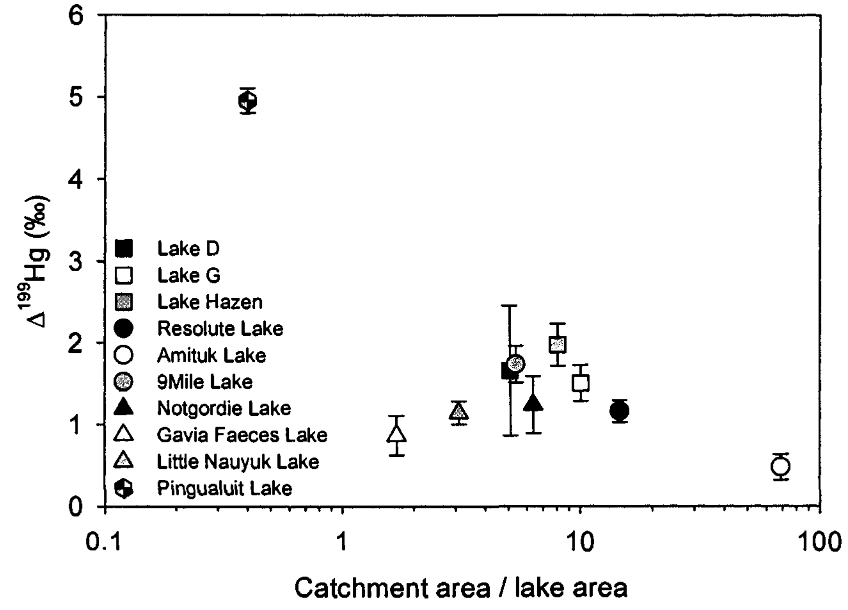

Figure 28: MIF-factor versus catchment-to-lake-area ratio (log scale). Although the relationship is not significant for all lakes, Amituk and Pingualuit represent the two exceptional lakes. Error barrs represent standard deviation (n=4-5).

greater in temperate lakes since Muir et al. (2008) did not see a relationship of Hg fluxes in dated sediment cores with CA/LA or catchment area in high arctic lakes but did see a relationship in mid-latitude lakes.

5.5 Acknowledgment

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6 GENERAL DISCUSSION

6.1 Mercury contamination in fish - a continuing issue

Mercury remains one of the most, if not the most, studied elements in the environment. The fascination for it stems from its unique physical and chemical properties (U.S. EPA 1997), which mankind has utilized, is utilizing widely today, and likely will be utilizing in the future. This heavy metal has aided in generating great wealth if one thinks about its use in the gold extraction process. It has eased the pain of millions in its various medicinal uses. Mercury has made daily life convenient for many of us through manifold use in industry and in numerous household applications. While mercury was historically, and until recently, best known for its beneficial applications, in the last 30 years it has come to have more negative public attention globally. Most notably, the human tragedy of Minamata, Japan in the 1970s raised great public health concerns. The understanding of the toxic potential/toxicity of mercury to humans has greatly advanced in the post-Minamata era. The toxicology of mercury has been thoroughly investigated over the past 40 years. Today, mechanisms of action (neuro-toxicity), routes of exposure (mainly dietary fish consumption), and many toxic effects (neuronal dysfunction, cardio-vascular diseases) on humans are thought to be well characterized (Satoh 2000). Fish consumption guidelines are now in place, thus the mercury exposure of people in many parts of the world is measured and regulated, e.g., in Canada (www.hc-sc.gc.ca and www.ec.gc.ca/MERCURY/EN/fc.cfm), in the United States of America (www.epa.gov/waterscience/fish/advice/mercupd.pdf), in the European Union (http://ec.europa.eu/environment/chemicals/mercury/index.htm), or by United Nations (http://www.chem.unep.ch/MERCURY/).

However, the effectiveness of such regulation varies, as both Hg emissions and concentrations in fish (and exposure of people) in some regions remain poorly monitored. Anthropogenic contributions of mercury in the environment have been quantified for point-sources and to some extent for dispersed inputs (atmospheric). The major emission sources to the atmosphere are well known (Pacyna et al. 2006) and emissions are either regulated under current policies (U.S. Clean Air Act www.epa.gov/air/caa/; or Clean Air Regulatory Agenda for Canada, www.ec.gc.ca/cleanair-airpur/clean_air_act-WS1CA709C8-1_en.htm), or efforts are being made to identify and regulate mercury emitters (see international mercury initiatives mentioned above).

However, there are new questions posed by stakeholders (private and industry) and policy makers, who ask: *How much* mercury is in our environment? *Where* is this mercury coming from? Are these mercury concentrations increasing or decreasing? What is natural, what is anthropogenic mercury? The interest of the former and the latter is to best manage this issue through adequate policies based on sound science.

To address these questions, scientific investigations of mercury have shifted from studies of toxicity and human exposure, to more mechanistic investigations of the environmental mercury cycle. Research now aims to explain spatial and temporal trends (both current and historic), sources, biological transfer of mercury, and/or the key issue of mercury methylation rates. Pathways of mercury transfer through the aquatic environment have been investigated (Harris et al. 2007), but not all aspects are fully understood. Key factors in this proposed cycle as they relate to cold climates are: deposition and re-emission of mercury from the atmosphere, or release from permafrost and bedrock, mercury methylation in lakes, bioaccumulation, and biomagnification along food chains, from low initial environmental (inorganic) mercury concentrations to elevated concentrations of MeHg in edible fish.

The attempt to characterize natural or 'background' concentrations of Hg in the environment, and to study 'simple' aquatic systems with minimal anthropogenic influence, has brought the Polar regions into the forefront for such research. Much to the surprise of many, concentrations of mercury in Arctic apex predators were often found to be higher than in Southern counterparts. Consequently, new concern about mercury in the Arctic environment stemming from these initial findings arose, and the impact on the local Inuit communities over the past two decades has been investigated (AMAP 2003; Van Oostdam et al. 2005). Abiotic and biotic conditions in the Arctic have been suggested to contribute to these findings, but few studies have validated or addressed these for Arctic freshwater environments. Finally, climate change in the Arctic is predicted to have more severe impacts on higher latitudes (ACIA 2005; IPCC 2007), and thus poses a possibly dominating additional stressor in both the physical and biological environments, as well as on Inuit populations.

The research presented in this thesis tested several hypotheses in order to address the above mentioned knowledge gaps about mercury transfer in Arctic freshwater systems. We designed field collections of Arctic char and food webs, and selected analytical endpoints to test hypotheses. Special common focus for all four studies was the investigation of possible linkages to climate change, however, each study addressed additional individual objectives.

6.1.1 Temporal trends of mercury in Lake Hazen char

As climate change may have an effect on the inputs of mercury, an initial objective of the research (Chapter 2) was to characterize the temporal trends of concentrations of mercury (and other elements of interest) in Arctic char muscle tissue. The focus was on a long term data set available from Lake Hazen, Nunavut. This work represents the most comprehensive temporal trend study conducted on mercury in Canadian landlocked Arctic char populations and, with results over a 17-year period is among the longest for Arctic fish in general (Bignert et al. 2004; Braune et al. 2005; Evans et al. 2005). The results are important as the detected trends (or lack thereof) did not confirm reported declines in concentrations of mercury in fish in remote mid-latitude lakes (Hrabik and Watras 2002; Drevnick et al. 2007). The data established for Lake Hazen can now be related to other studies conducted during a similar or overlapping time period (Muir et al. 2005). The comparison of trends may not be limited to Arctic char systems or even to comparisons with other freshwater systems alone, it can also be compared with sediment records or nearby atmospheric measurements (Steffen et al. 2005; Temme et al. 2007; Muir et al. 2008). The scientific information gained from this study at Lake Hazen that trophic position (which affects concentrations of mercury) can be accounted for by ANCOVA, thus improving trends analysis, is of interest for studies of other systems in two ways. First, similar statistical analysis could be applied to data sets where species are known to exist in multiple morphotypes, thus the use of available samples and contaminant data is maximized. Second, and alternatively, the finding that lower variability of mercury concentrations and a slight decline of mercury over time was detectable in insectivorous "small" Arctic char, leads us to the conclusion that these insectivorous forms may be better suited for monitoring, than the "large" piscivorous forms. Much of the variability in mercury associated with the inclusion of two morphotypes in temporal trend analysis could be avoided by sample selection, or even earlier in the process, by sampling only insectivorous char. This suggestion is limited to temporal trend work, as important ecological questions may be addressed through sampling of all abundant ecotypes. Overall, the importance of annual sampling in future temporal programs was underscored by our study. We failed to reject the null hypothesis set out a priori here, and did not detect a significant increase in mercury concentrations over the 16-year period (1990-2006).

6.1.2 Trophic magnification in lake food webs

Prior to this large scale food web study (Chapter 3), no reports have been available addressing the biomagnification of mercury along Arctic lake food webs with a single top predator. Most other Arctic food web studies were conducted in the marine environment (Campbell et al. 2005), or restricted to more southern regions and included multiple fish species (Power et al. 2002b; Guildford et al. 2008), which underscores the importance of our study. The only previous study of food web biomagnification in Arctic aquatic systems investigated the pelagic marine food web (Campbell et al. 2005). Past studies of mercury in landlocked Arctic char (Köck 2004; Muir et al. 2005) were conducted without taking into account the influence that differing food webs may have. The large number and wide range of sites presented in this study allows for inferences on a large geographical area. Several food webs included in this study had not been characterized previously (Lake D, F, G, Small, 12 Mile, Aquiatusuk, Teardrop), nor had contaminants been measured within them. Since biomagnification was apparent in all lakes, we reject the null-hypothesis set for this study. The fact that biomagnification was confirmed in all lakes may be less surprising given the results of other studies, and more generally, the properties of MeHg. However, these results lend themselves to future investigations as a baseline comparison of mercury biomagnification in lakes from the four studied regions of the Canadian Arctic. Moreover, the data represent comprehensive baseline information, which in the light of climate warming will aid the determination and attribution of future changes.

6.1.3 Spatial trends in Arctic char

The results presented on the geographical trends (Chapter 4) have expanded current knowledge about spatial variability of, and the mean concentrations of mercury in landlocked Arctic char. With 27 lakes, 24 of which were sampled within the period of this study (2005-2007), it also represents the most current inventory of landlocked char concentrations. In addition, we have established the key relationships that determine THg concentrations in char (and variability thereof) for each lake. Mean mercury concentrations in char presented here did not correlate positively with latitude or longitude (thermal regimes), hence we fail to reject the null-hypothesis for this study. Monitoring programs now have the opportunity to use these data to either revisit these sites, or refine monitoring programs in specific regions. Parallel studies of sea-run Arctic char (Evans and Muir 2006), or investigations of lake food webs with more complex fish species composition that include Arctic char (Swanson 2007), can utilize knowledge gained from our experience. The influence of diet and trophic position is valuable information for future studies. The results will directly aid the continuous monitoring efforts of the Northern Contaminants Program (NCP), in that results from North, Amituk, Char, and Resolute lakes presented in this study will also be included in the continuing temporal trend assessments studies at these sites under the NCP. This comprehensive investigation of regions across the Arctic will allow stakeholders as well policy makers to infer conclusions about future mitigation efforts towards global controls on atmospheric emissions of Hg.

6.1.4 Stable isotopes of mercury - a method of 'fingerprinting'?

The work on stable isotopes of mercury (HgSI) (Chapter 5) is the first evidence of species-specific degrees of mass independent fractionation (MIF) in Arctic char, zooplankton, and chironomids. In addition, the lake and region specific fractionation of HgSI in char was observed. This expands the small body of literature available on MIF and mass-independent fractionation (MDF) in biota (Bergquist and Blum 2007; Blum and Bergquist 2007; Jackson et al. 2008; Sonke et al. 2008), as well as abiotic environmental data (Jackson et al. 2004; Ghosh et al. 2008). We demonstrated low intra-lake variability of MIF in Arctic char, indicating that processes during uptake were similar in individual fish from each lake. We also showed that fish from neighbouring lakes within one region had similar MIF signals in fish, indicating similar processes within regions. Further, we distinguished regions based on MIF in Arctic char. MIF Arctic char from three lakes on Ellesmere Island, delineated more than Kent Peninsula lakes and Cornwallis Island lakes, while Pingualuk char showed the greatest MIF. Regional effects processes appear to be driving this differentiation, rather than differences in bioaccumulation. However, as we demonstrate increasing MIF in sediments, insects, and fish, we reject the null hypothesis of this study.

A reoccurring theme of this thesis resurfaces here, in that the only predators in these lakes are Arctic char, making conclusions on the fractionation of HgSIs along food webs from different regions more conclusive. While this methodology has yet to identify sources based on fractionation of stable isotopes of mercury, we have made considerable progress in source tracking through our efforts. This is of special interest to regulators of mercury emissions, and arguably emitting industries, as attribution of mercury contamination has been a major research gap, identified by many recent research programs. Future research should be directed towards linking the fractionation (and associated processes) in air, precipitation, catchments, water, and sediments in order to understand processes during transfer to lake sediments. Further, investigations of sediment methylation and de-methylation by photoreactions should be investigated.

6.2 Uncertainties and Limitations

While we have maximized our efforts to reduce the uncertainty of results, it is inevitable that any research identifies limitations and data needs. Field studies, such as those conducted and presented herein, inherently suffer from several constraints. Most constraints increase with latitude and logistical challenges of conducting research in such remote locations. These are further confounded by natural sample variability, and contamination, or loss of samples during transport. In this work, data were collected across 4 distinct Arctic regions aiming for a good representation of lakes within each region. Food web sampling was conducted over a very large geographical range (61 – 82 °N and 62 – 107 °W) and maximizing the number of lakes sampled at each region limited sampling time at each site to 4-6 hours. Because of this, it was not possible to only select lakes with similar catchments or size. Selection of sites was primarily constrained by logistics and accessibility. Prior to sampling a tentative site, we relied on knowledge of the char populations from earlier studies conducted in each area. Given the demonstrated influence of catchment, it is possible that fewer sites, with similar catchment size may have allowed stronger conclusions to be drawn.

Seasonality may have influenced our results for lakes sampled in 2006 on the Kent Peninsula in comparison to the two other major sites. While we know that the sampling on Ellesmere and Cornwallis was conducted under similar ice-free conditions (2005-2007), we sampled these southern lakes after ice-off. This is also the reason why no emerging insects were collected from this site. The year 2007 was amongst the warmest recorded in the Canadian High Arctic. This also constrained our sampling, e.g., of chironomids at Ellesmere Island in this year. As a matter of fact, our Ellesmere Island sampling team experienced the day (July 25 2007) on which the warmest outside air temperature was recorded at Eureka (20.7 °C) during a stop over at this weather station (79° 59"N, 95° 48"W) on Ellesmere Island.

6.3 Conclusions and recommendations

6.3.1 Biotic and abiotic linkage to climate change

This thesis represents the most comprehensive assessment of the current status of concentrations of THg and MeHg in landlocked Arctic char populations and underlying food webs to date. In addition, it includes discussion of the possible consequences of, or influence of, climate change effects on freshwater systems (Prowse et al. 2006; Wrona et al. 2006). Direct and indirect effects of climate change on mercury in char are possible. For example, our results indicate that greater zooplankton size results in higher MeHg concentrations in this char prey item (Chapter 3), which could lead to higher MeHg in Arctic char. Independent of size, zooplankton appear to take up Hg in the water column (Chapter 5), likely reflecting newly deposited and readily available mercury (Harris et al. 2007). Thus, increased or reduced deposition of Hg to lake surfaces may directly result in greater or smaller concentrations of Hg in zooplankton, as observed by Harris et al. (2007) for Hg isotopes applied to an experimental lake. The observed shift of trophic signatures ($\delta^{13}C$ and $\delta^{15}N$) of zooplankton with latitude may indicate that this group of organisms may be most sensitive to climatic conditions, thus most vulnerable to changes.

Specific knowledge gaps, such as lake-to-lake variability, were addressed in order to advance and improve monitoring of spatial and temporal trends (Chapter 2 and 4). Initiatives such as AMAP (2003) and others repeatedly called for studies that investigate food web accumulation and utilize modern techniques (e.g., stable isotope analysis) to explain this variability of mercury concentrations in fish. Factors causing variability in mercury concentrations in adult Arctic char have been identified (Chapters 2, 3, and 4).

While we addressed research questions specific to the Arctic, some comparisons to other environments (e.g., high Alpine ecosystems) may be valid. Results from chapters 2 and 5 are of greater general interest for the scientific community interested in global mercury cycling. Temporal trends of mercury in fish (Chapter 2) in high latitudes are now available for comparisons with studies of trends in other regions. The results presented in Chapter 5 are of interest for studies focused on relating atmospheric (i.e., anthropogenic) emissions to sources. Our mercury isotope ratio data from remote Arctic lakes may be the most useful data as a baseline on which to draw inferences about Hg at sites influences by local mercury sources.

We have further advanced the knowledge on cycling of mercury through biological compartments in the Arctic. Information on food webs of 17 small arctic lakes have been established, many of which were sampled and described for the first time. A specific strength of this study is the use of a single predatory fish species across the compared regions. The influence of fish species assemblage on mercury concentrations in top predators has been demonstrated (Power et al. 2002b; Swanson 2007).

In conjunction with this thesis, Environment Canada (NWRI, Burlington) has developed an analytical method to detect trace MeHg in small samples of biota, and thus expanded its capacity to analyse this element. This may allow refined investigations of concentrations of MeHg in lower trophic level organisms and their role in accumulation of MeHg in the future in the Arctic and other ecosystem.

Permafrost is deteriorating in some regions of the Arctic. As a consequence, nutrients and also contaminants stored in the soil will be mobilized or re-mobilized and enter aquatic systems (Prowse et al. 2006). Inputs of mercury from the surrounding landscape to aquatic systems have been documented to be a significant contributor to the mercury budget for larger freshwater or marine systems (Leitch et al. 2007; Outridge et al. 2008), however no such budgets are available for small Canadian Arctic lakes. We established that catchment-to-lake area ratio does affect Arctic char in our lakes. Since catchment and lake area are constant, changes of factors within the catchment and lakes will determine effects on char. Thus, increased melting of permafrost within the catchment of the lake, and subsequent inputs of stored (geogenic and anthropogenic) mercury could be indirectly linked to char mercury concentrations. Alternatively these could be the result of a second indirect effect, as the consequence of greater methylation of mercury in sediments and/or greater uptake by chironomids. In addition, changes in precipitation patterns, as predicted, by the IPCC, may result in change in amounts of mercury deposited within a catchment. This in turn, will have consequences on mercury in the aquatic food web.

We present a novel approach to investigate the tracking of mercury through the biological compartments of a lake. Stable isotope ratios are a promising tool that may one day close an important scientific knowledge gap. The fractionation of mercury isotopes in aquatic food webs has been confirmed in the Arctic for the first time. Our results indicate that processes that may occur prior to methylation of mercury (i.e., photochemical reactions), or methylation itself and lead to MIF are preserved during bioaccumulation. If light conditions do influence the degree of MIF, lake ice cover, and snow cover of lake ice may alter this effect. Lower precipitation or earlier ice-off through warmer air temperatures could thus result in changes in the MIF. According to our results, effects would be reflected in Arctic char muscle tissue.

In conclusion, the results of all chapters indicate that abiotic changes in the catchments are the most likely to result in changes in mercury transfer in lake biota. It appears less likely that changes in ambient air and water temperature will have direct effect on food web accumulation of mercury.

6.3.2 Consumption of Arctic char by local people

Although the research for this thesis was focused on mechanisms of mercury bioaccumulation in wildlife and not directly on consumption of Arctic char by humans, some general statements can be made relevant to the consumption of fish from the lakes that were studied. Results of THg concentrations in adult char (Chapter 4), which are possibly the best available measure for human exposure, allow the derivation of simple, but hopefully effective recommendations to fishermen, following Heath Canada guidelines (limited to the 27 landlocked Arctic char populations presented herein).

1. Do not eat Arctic char of any size from Amituk Lake.
2. Eat Arctic char from other lakes only two times each month.
3. If an Arctic char is caught in a lake, open the stomach. If the stomach is filled with small char or other fish, it is best to not eat that fish. Feed the fish to the dog, to make use of the kill. If black insects are found in the stomach, it can be eaten.

6.3.3 Future Work

Mercury in Arctic char populations

Current monitoring programs must be continued and sampling should be done on an annual basis. This is necessary to detect any change related to responses to climate change, as well as to improve the statistical power of temporal trend analysis (Bignert et al. 2004; Gantner et al. 2009a). We have established that there was no significant temporal trend of mercury in Arctic char from one large ecosystem, however, there are indications that smaller lakes may respond differently and more rapidly. Slight increases in mercury concentrations in char were detected in two lakes (Char and Amituk) near Resolute Bay (Cornwallis Island) over the past years (Muir et al. 2006; Muir et al. 2007).

Toxicological effects of mercury on Arctic char should be explored, for example in Amituk Lake, the lake with the highest individual mercury concentrations. Recent studies showed that liver function in pike (*E. lucius*) can be affected in individuals with high mercury concentrations (Drevnick et al. 2008). Feeding experiments in Arctic char have reported severe necrosis of the liver and alterations of cytoplasmic organization following exposure to MeHg (de Oliveira Ribeiro et al. 2002).

Pharmacokinetics of mercury may be explored to explain high concentrations of mercury in populations of landlocked Arctic char. We have confirmed here that landlocked Arctic char in most lakes have greater mercury concentrations than sea-run populations (Evans et al. 2005). Investigations of mercury in food webs and prey items in the marine environment, similar to our work, should reveal whether mercury is less available. Experiments need to be designed aiming at explaining the metabolism (uptake / elimination) of mercury by fish in freshwater and marine environment.

In conjunction with either above discussed matter, effects on char, or pharmacokinetics, investigation of measurements of Selenium (Se) could be considered. Some analyses of Se have been conducted on selected char from our study lakes, but only few measurements have been published for Arctic freshwater fish (Gantner et al. 2009a) (Chapter 2). Se has been proposed to have a protective effect by complex-forming with mercury (Ikemoto et al. 2004; Belzile et al. 2006), and is also known to bioaccumulate (Orr et al. 2006).

Exploring terrestrial-aquatic linkages

This work established that terrestrial parameters (watershed, catchments) can influence concentrations of Hg in char. Our study sites were heterogeneous and not all catchments were well characterized. Thus, future efforts should be focused on investigating systems with more abiotic information available. The watersheds and nutrient input to two neighbouring lakes on Melville Island (Nunavut) are currently being characterized by S. Lamoureux (PI) and co-workers (Cockburn and Lamoureux 2008), and are candidate lakes for such integrated monitoring efforts.

Exploring aquatic-terrestrial linkages and mercury exposure

Biotic linkages:

An estimate of exposure of terrestrial predator species via existing aquatic-terrestrial linkages could be investigated, hand in hand with export of mercury from lakes and the Arctic. The transfer of contaminants such as mercury from aquatic to terrestrial environments has gained recent interest (Brasso and Cristol 2008; Cristol et al. 2008), but are not commonly explored in the Arctic environment. A growing body of literature on toxicological effects of MeHg on birds and other wild life species is becoming available (Scheuhammer et al. 2007; Scheuhammer and Sandheinrich 2008). Several terrestrial predators are commonly present in the vicinity and consume biota from the lakes, thus aquatic-terrestrial linkages do exist in this region. Arctic birds rely seasonally on aquatic biota from Arctic lakes. Especially during the summer months, while most birds are breeding and feeding their young, this could be a significant route of exposure to their likely even more vulnerable offspring, as demonstrated for common loons (Evers et al. 2008). High concentrations of persistent organic pollutants in breeding and non-breeding birds have been reported (Fisk et al. 2003; Mallory et al. 2006; Braune et al. 2007). Several observations during our field work indicate aquatic-terrestrial linkages with potential of mercury exposure: Insectivorous birds, such as Snow buntings (*Plectrophenax nivalis* L.) were observed feeding on emerging chironomid midges on candled lake ice in the Quittinirpaaq National Park. As buntings breed during the Arctic summer, adults likely feed their offspring chironomid midges, the exposure to mercury is a possible. Fish-eating birds, such as Arctic loons (*Gavia arctica* L.) or Red Throated loon (*Gavia stellata* P.), who inhabit almost all lakes in the Resolute Bay area consume juvenile as well as adult Arctic char, for example, we observed loons diving for Arctic char that had been caught in our gill nets. Thus, loons maybe exposed to mercury from the aquatic systems. Arctic terns (*Sterna paradisaea* P.) were observed to follow the shoreline of the lake and to dive and feed for juvenile Arctic char. The catch is then brought and fed to the young immediately. Quite likely, there are other species of birds that consume either juvenile or adult char, or chironomid midges. Data on feeding behavior of these birds would be required to estimate uptake of mercury from their aquatic prey. Migratory birds are also known to be vectors of contaminants from southern regions into the Arctic (Blais et al. 2005; Evenset et al. 2007). Our observations may lead to the considerations of migratory birds as 'exporters' of mercury, as adult individuals and young of the year are leaving the Arctic in the fall months.

Exposure of sled dogs to mercury has been demonstrated in the Yukon (Dunlap et al. 2007), and study of the effects of contaminants in sled dogs in Greenland is ongoing (Sonne et al. 2008). We have observed that landlocked Arctic char are frequently fed to sled dogs in Resolute Bay, Nunavut. This is an example of human-induced linkage of our studied lake systems and the terrestrial environment. Our data on concentrations of mercury in adult and juvenile Arctic char, as well as chironomids midges could aid models of exposure of arctic bird populations and sled dogs to mercury.

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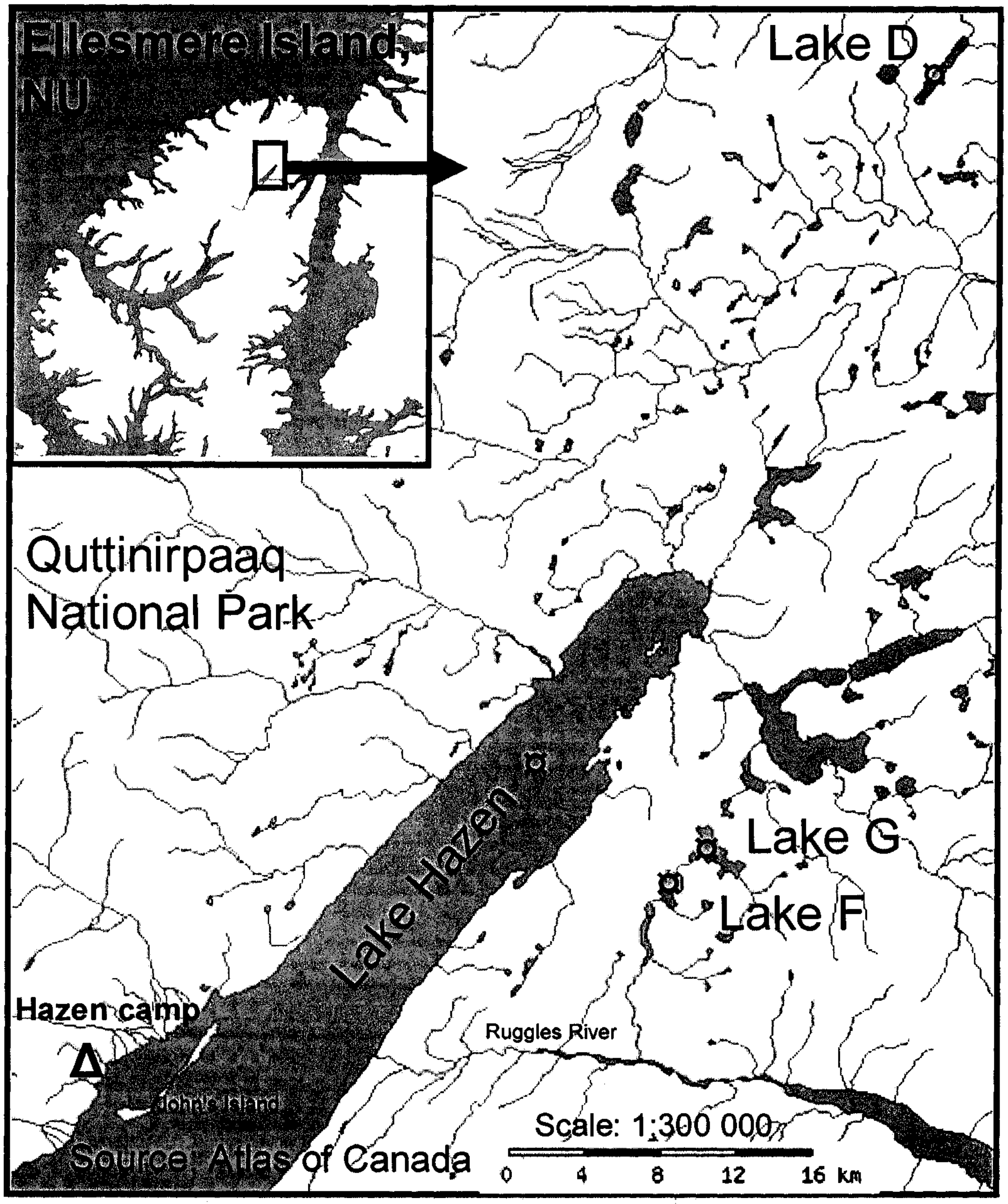
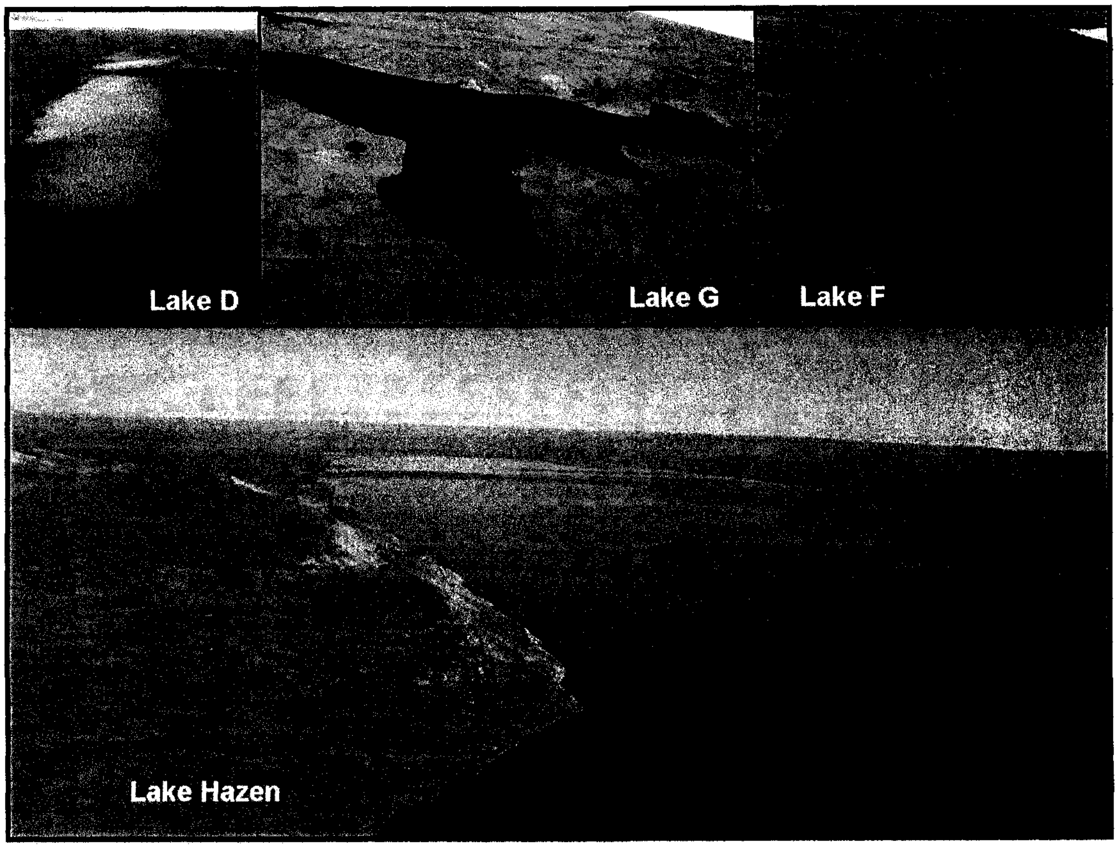
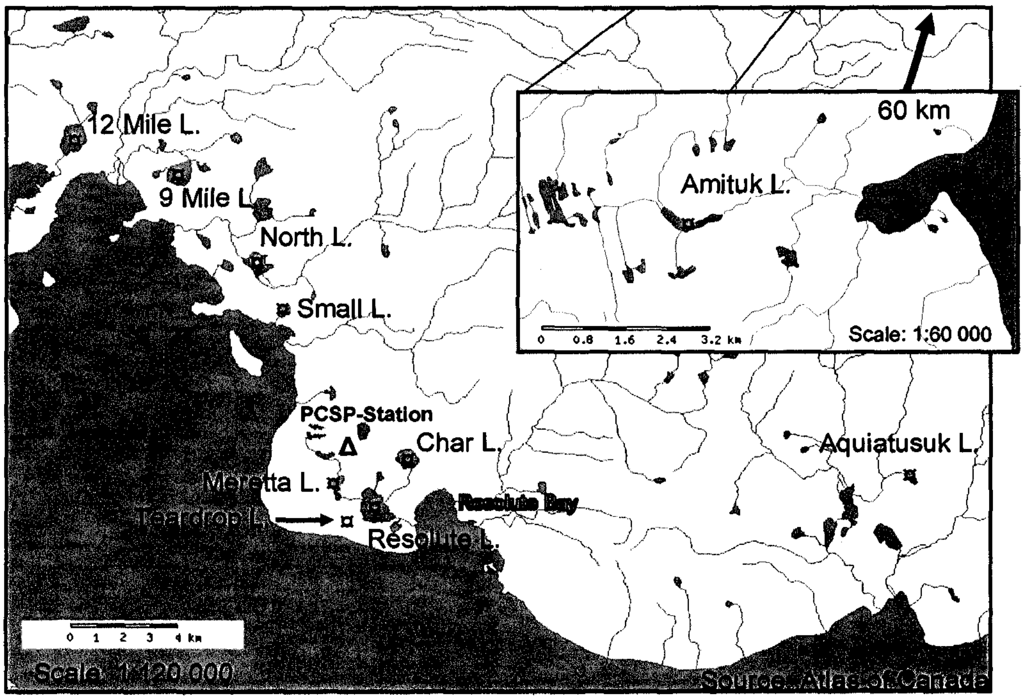
8 APPENDIX



Appx 1: Map of Ellesmere Island sampling locations located in the Quttinirpaaq National Park, Nunavut. The '¤' symbol indicates the lake.

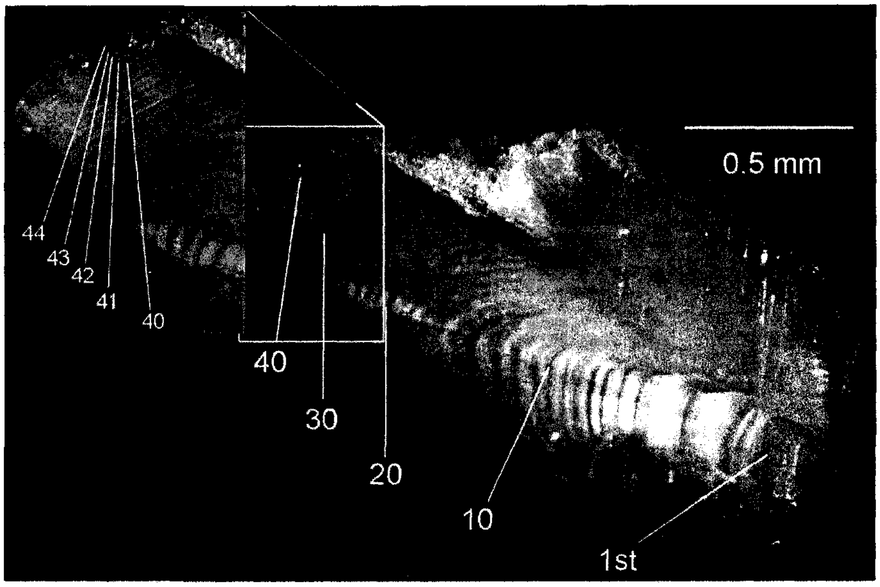


Appx 2: Aerial photographs of Ellesmere Island sampling locations and photographs of lakes (four lakes). *Pictures © N. Gantner*

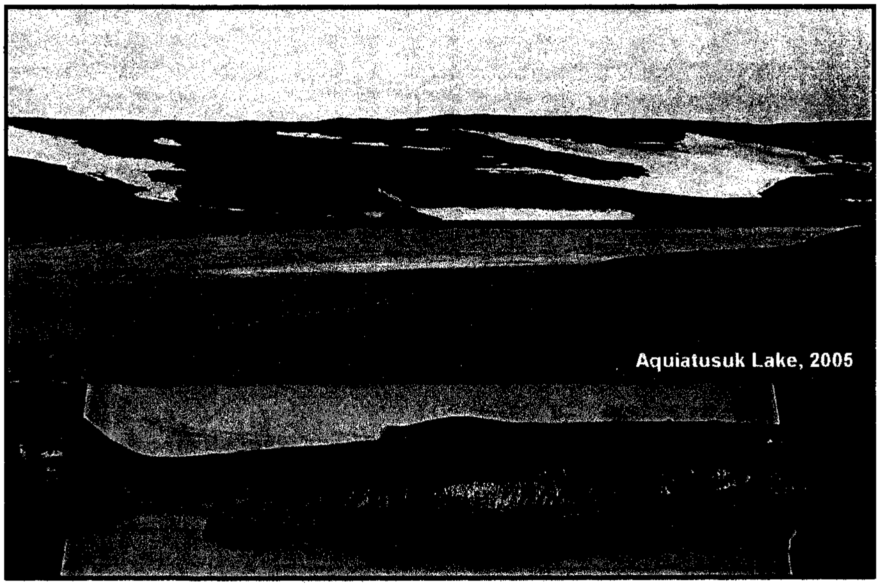


Appx 4: Map of southern Cornwallis Island indicating 10 sampling locations near Resolute Bay, Nunavut. Lakes are indicated by '¤'. The insert shows a map of Amituk Lake at the N/E coast of the island. NOTES: Teardrop Lake's position is approximated; scales of insert and main map differ.

Appx 4: Aerial photograph of Amituk Lake (2006) on Cornwallis Island, looking North. *Picture © N. Gantner*



Appx 5: Photograph (under microscope) showing a cross section through an Arctic char otolith (earbone used for age determination). At age 44+, this may be the oldest recorded specimen of the species (J. Babaluk and R. Wastle, Fisheries and Oceans Canada, Arctic and Freshwater Institute, Winnipeg, pers. comm.) this represents oldest Arctic char and was collected at Aquiatusuk Lake (Cornwallis Island) by D. and P. Iqaluk on August 8th 2005. *Picture provided by R. Wastle.*



Appx 6: Photograph of Aquiatusuk Lake (Cornwallis Island) (top) and a 44+ year old Arctic char (bottom) collected from this lake by D. and P. Iqaluk on August 8th 2005. The fork length of this male specimen was 461 mm, and a weight of 776 g was recorded. *Pictures © N. Gantner.*

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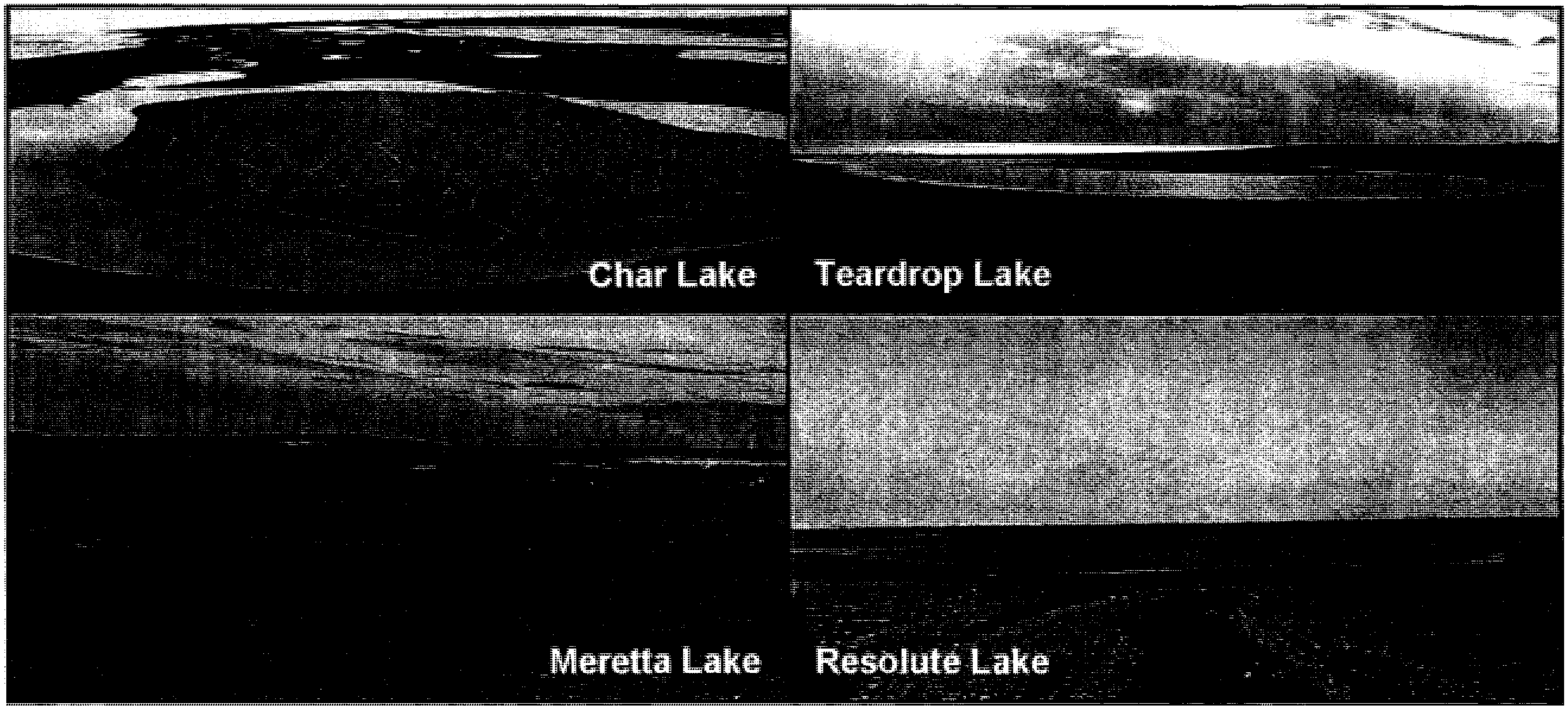
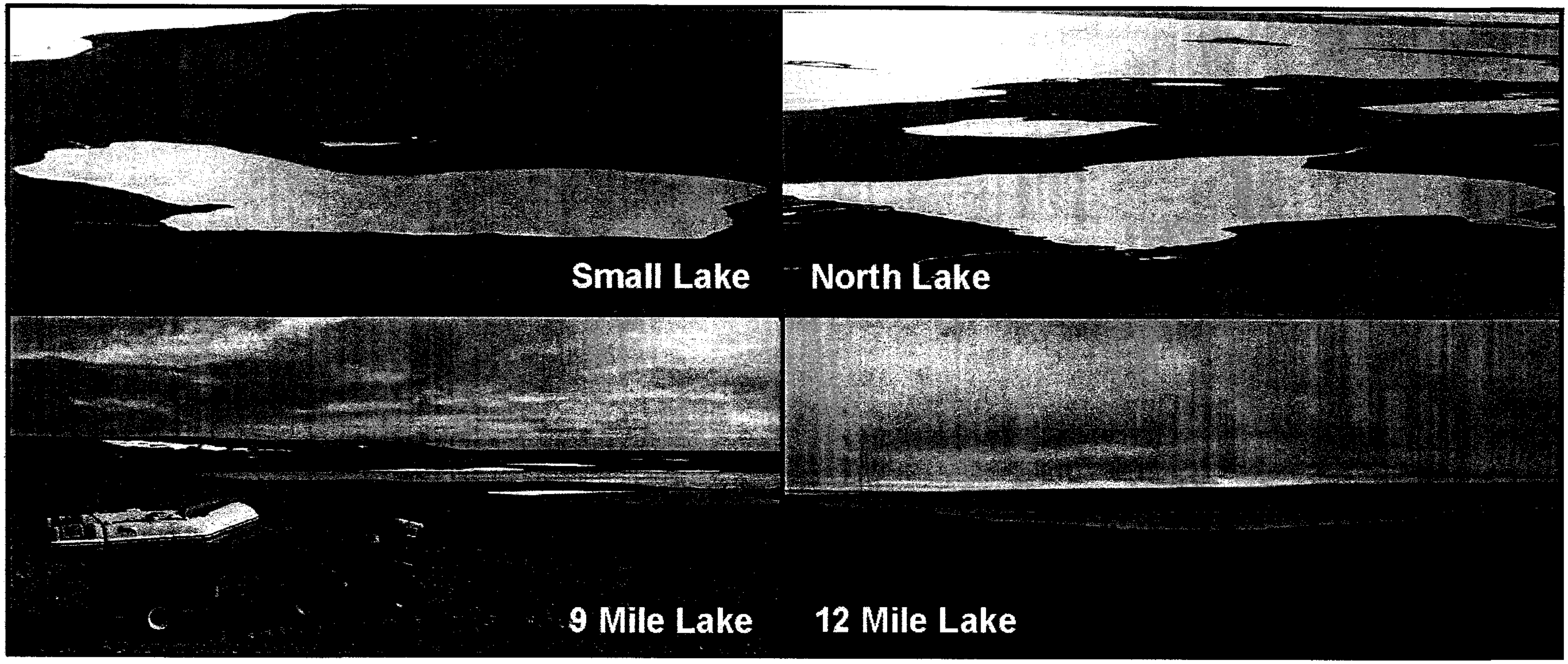
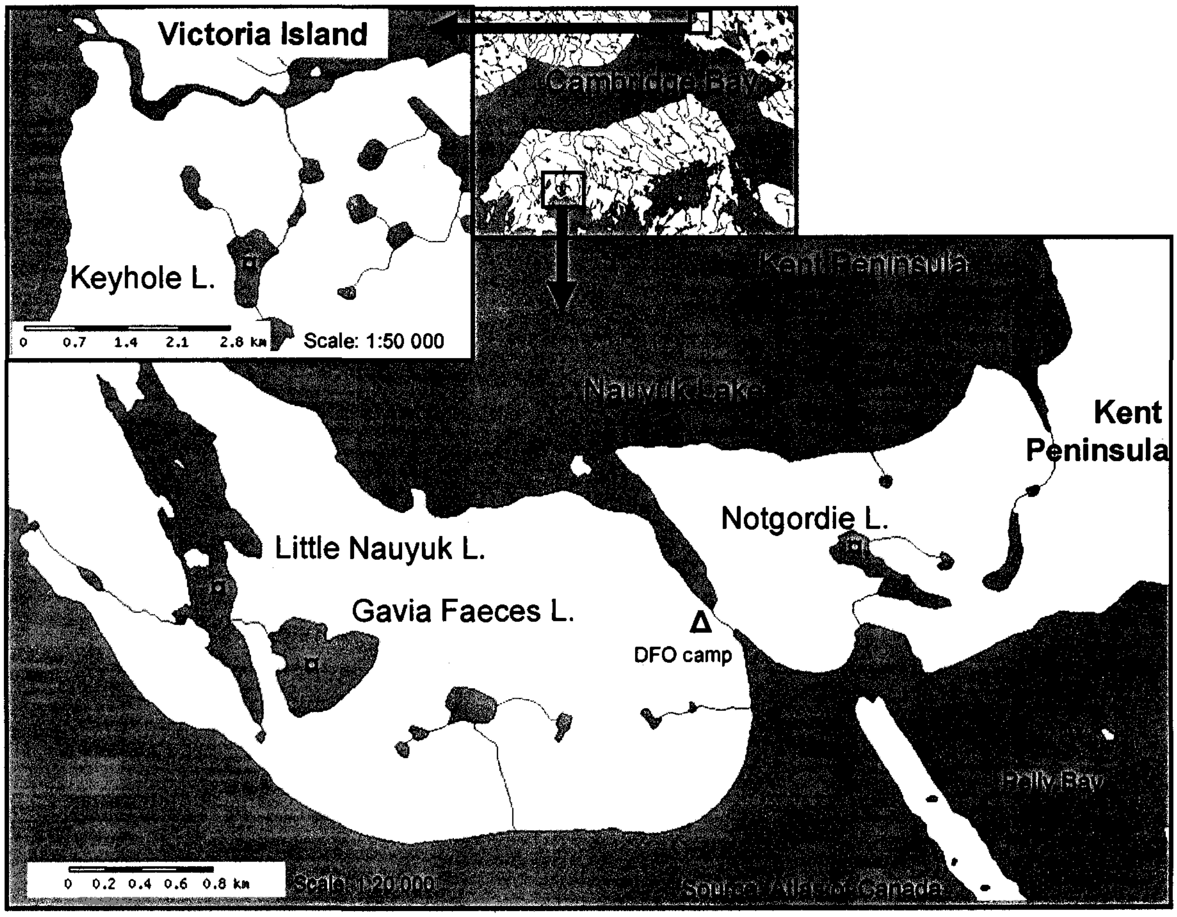


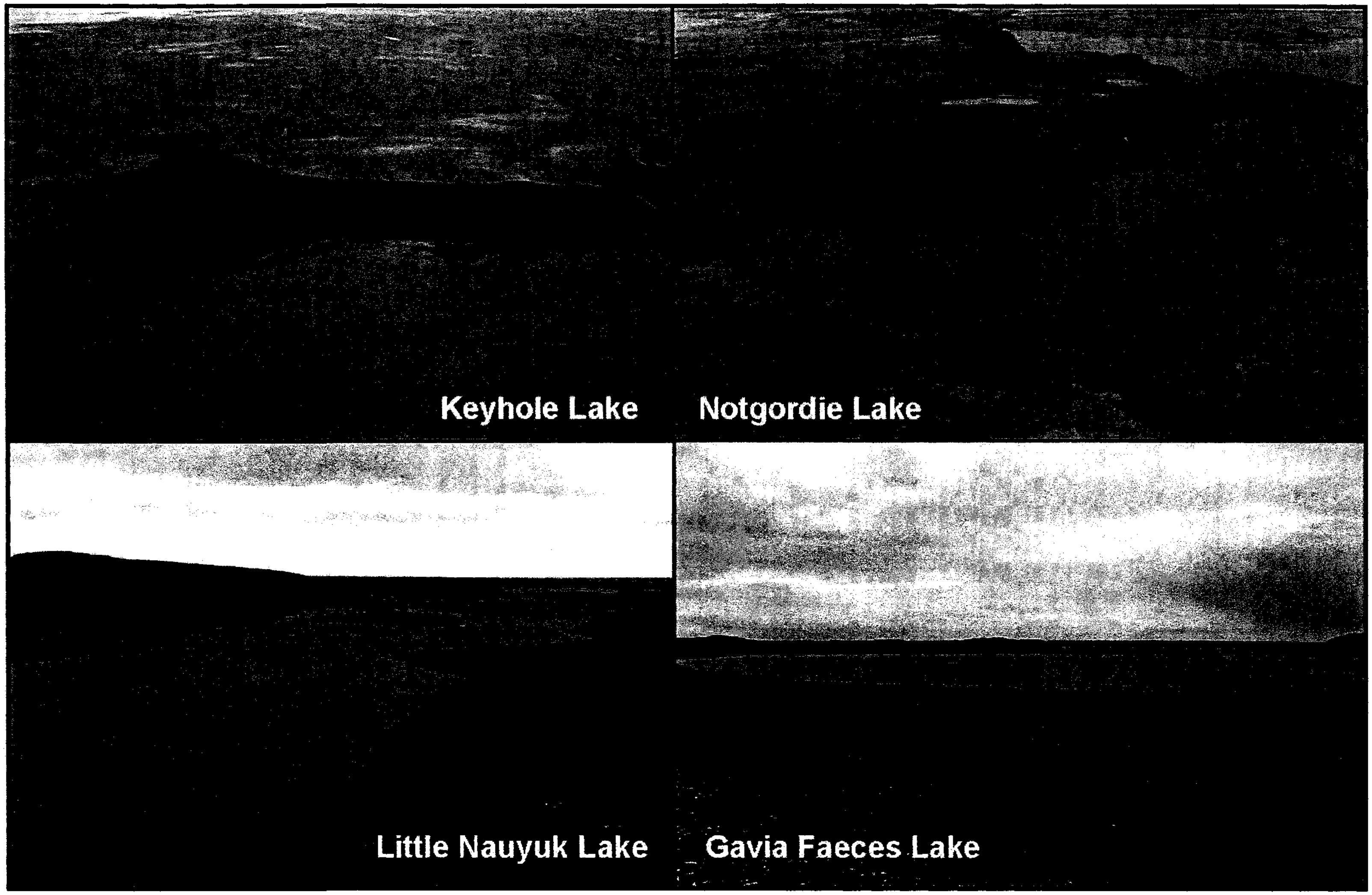
Appx 7: Photographs of Char, Meretta (both 2005), Teardrop, and Resolute (both 2006) Lakes. All four lakes are located 2 - 3 km west of the Hamlet of Resolute Bay (Cornwallis Island). Resolute Lake is also visible in the back-drop of Char Lake, and to the right of Meretta Lake. Resolute Bay (town) is visible in the far right backdrop of Meretta Lake. *Pictures © N. Gantner*



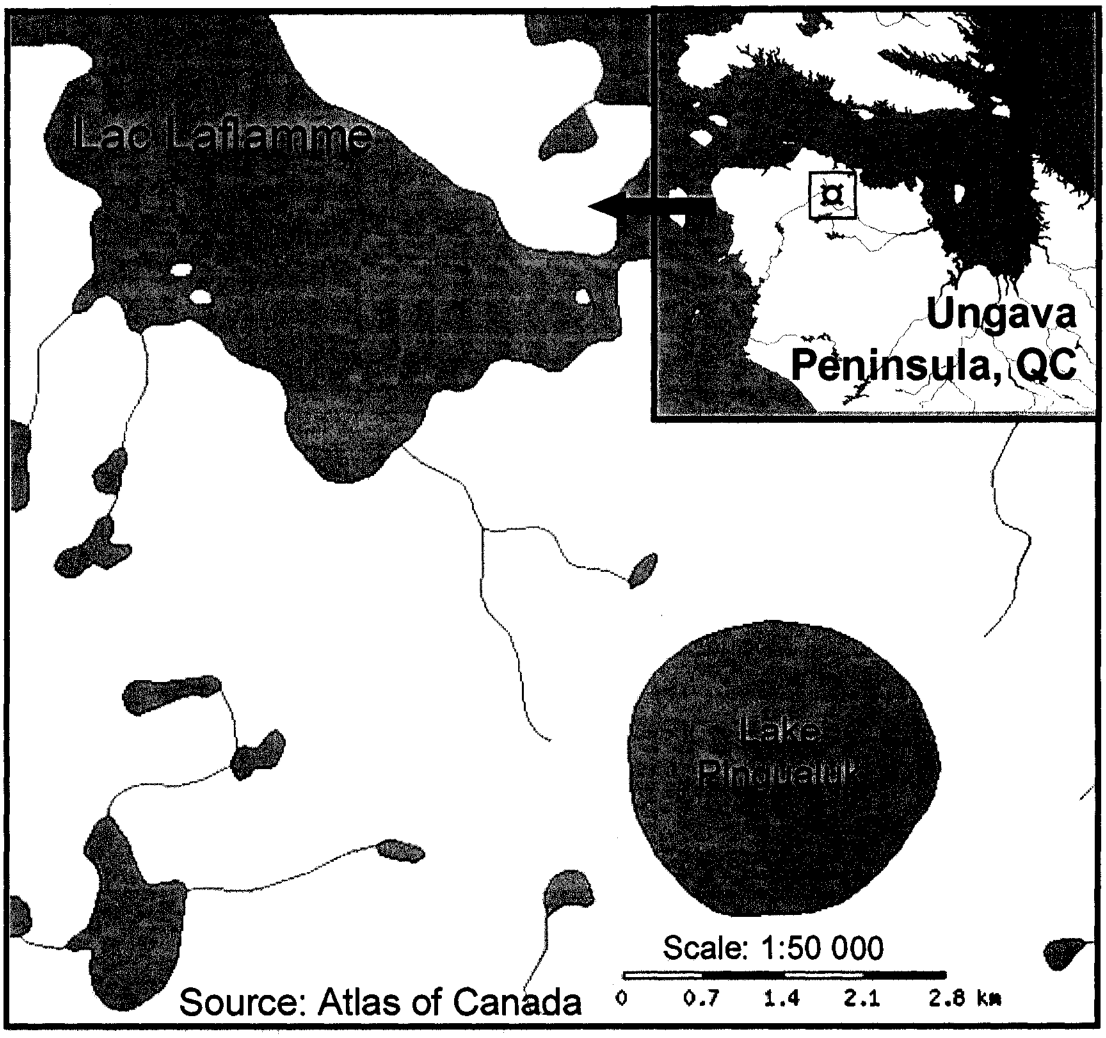
Appx 8: Photographs of Small, North (both 2007; aerial), 9 Mile, and 12 Mile (both 2006) Lakes. The four lakes are located ~9-18 km north-west of the Hamlet of Resolute Bay (Cornwallis Island). The lakes were accessed by All Terrain Vehicle, sampling equipment transported with trailer (see bottom left picture). *Pictures © N. Gantner*



Appx 9: Map of the southern Kent Peninsula (three lakes) and Victoria Island (one lake, insert). NOTE: Scales of insert and main map differ.

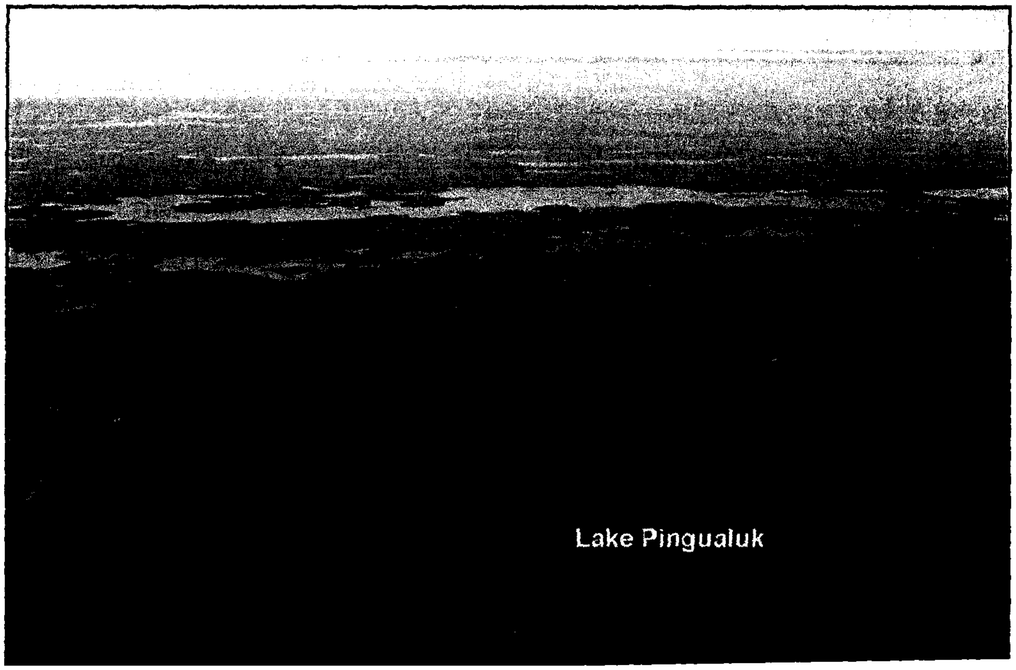


Appx 10: Photographs of Keyhole (Victoria Island), Notgordie, Gavia Faeces, and Little Nauyuk Lakes (all Kent Peninsula). All pictures were taken in 2006. *Pictures © N. Gantner*

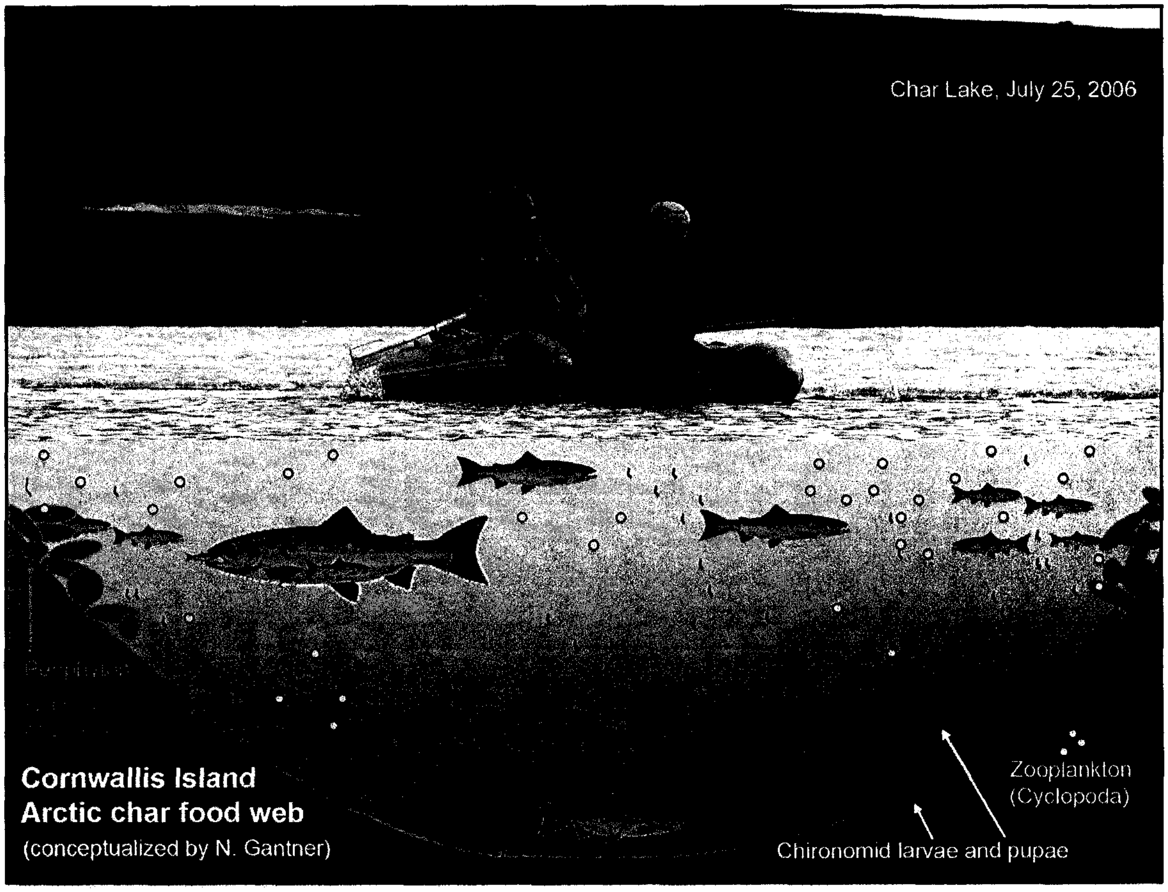


Source: Atlas of Canada

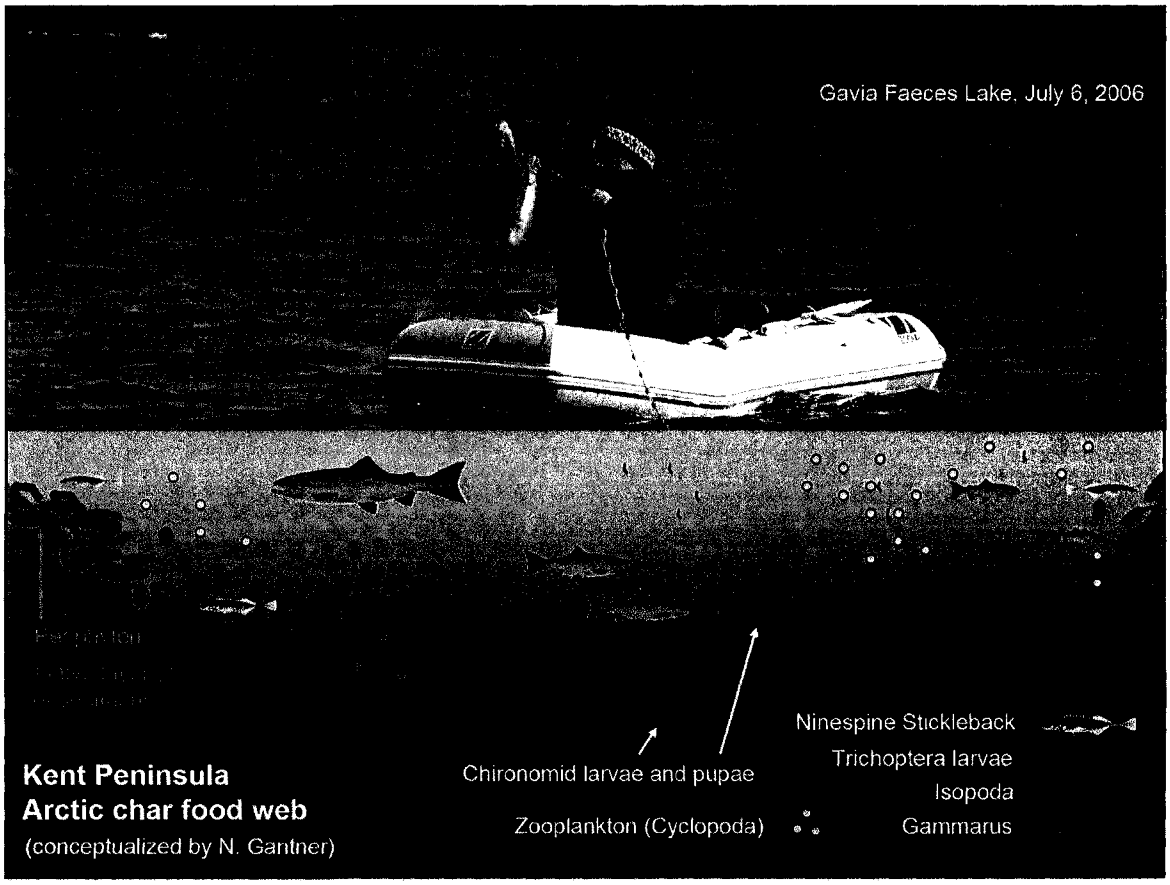
Appx 11: Map of the northern Ungava Peninsula showing Lac Laflamme and Lake Pingualuk (a meteor-impact crater).



Appx 12: Aerial photograph showing Lake Pingualuk (a meteor-impact crater lake) located in the Parc National des Pingualuit, Quebec. A meteor-impact 1.4±0.1 million years ago resulted in the ~3.4 km diameter "Crystal Eye of Nunavik". Arctic char were collected by Yakaa Yakaa and Peter Kaitainaq through the ice during sediment coring work led by Reinhard Pienitz and Sonja Hausmann in May, 2007.
Picture curtsey of Robert Fréchette, Kativik Regional Government



Appx 13: Schematic of a simple Arctic char food web for lakes on Cornwallis Island and Ellesmere Island. The picture shows D. Iqaluk and D. Muir during a zooplankton tow. NOTE: Sizes of biota are not proportional. *Picture and graphic © N. Gantner*



Appx 14: Schematic of a simple Arctic char food web for lakes on Kent Peninsula. The picture shows D. Iqaluk and N. Gantner removing an adult Arctic char from a gill net. NOTE: Sizes of biota are not proportional. *Picture and graphic © N. Gantner*

Appx 15: Samples, stable isotopes, Hg species, and TL used in TMF calculation

Region / Lake	Sample	N	Year	$\delta^{13}C$	SD	$\delta^{15}N$	SD	THg*	SD	MeHg	TL	SD
Cornwallis				‰		‰		µg/g		µg/g		
12 Mile	Arctic char	21	2005	-21.6	0.3	11.4	0.3	0.095	0.02	-	2.5	0.1
		13	2006	-21.3	0.5	11.6	0.2	0.129	0.04	-	2.5	0.1
		12	2007	-21.2	0.6	11.4	0.2	0.105	0.04	-	2.5	0.1
	Juvenile Arctic char	15	2005-2006	-23.6	1.0	10.5	0.6	185	87	-	2.2	0.2
	Chironomidae	1	2006	-26.3		8.4		218		219.4	1.6	
		1	2005	-23.6		8.5		109		109.6	1.6	
	Zooplankton >500µm	1	2007	-34.6		12.2		27		6.0	2.7	
	Zooplankton 250-500µm	1	2007	-34.1		11.0		-		2.5	2.3	
	Periphyton	1	2007	-17.5		6.4		-		1.3	1.0	
9 Mile	Arctic char	12	2005	-22.0	0.5	10.2	0.5	0.141	0.05	-	2.1	0.2
		11	2006	-21.6	0.4	10.3	0.4	0.174	0.07	-	2.1	0.1
		7	2007	-21.6	0.4	10.4	0.4	0.164	0.05	-	2.1	0.1
	Juvenile Arctic char	18	2005-2006	-22.1	1.5	9.3	0.5	263	75	-	1.8	0.1
	Chironomidae	1	2006	-19.9		8.2		225		226.2	1.5	
	Zooplankton 250-500µm	1	2007	-34.7		9.0		-		6.9	1.7	
	Zooplankton >250µm	1	2007	-34.7		9.0		27		16.0	1.7	
	Periphyton	1	2007	-19.9		6.5		-		1.9	1.0	
Amituk	Arctic char	6	2005	-21.1	0.5	10.5	1.1	0.853	0.27	-	3.6	0.3
		6	2006	-21.1	0.3	12.0	0.8	1.787	0.74	-	4.1	0.2
		11	2007	-21.1	0.4	11.3	1.2	1.287	0.91	-	3.9	0.3
	Juvenile Arctic char	12	2005-2006	-23.7	1.2	8.5	1.0	446	308	-	3.0	0.3
	Chironomidae	1	2006	-21.8		5.5		278		239.9	2.2	
		1	2005	-24.6		6.6		503		315.6	2.5	
		1	2005	-24.6		6.6		-		253.0	2.5	
	Zooplankton >250µm	1	2006	-18.3		5.4		-		45.6	2.1	
	Periphyton	1	2006	-19.9		1.6		3		0.5	1.0	
Aquiatusuk	Arctic char	2	2005	-26.2		10.7		0.593		-	3.1	
		7	2002	-26.8	0.4	9.6	0.8	0.235	0.20	-	2.8	0.2
	Juvenile Arctic char	6	2005	-27.8	0.5	9.1	0.5	278	69	-	2.6	0.1
	Chironomidae	1	2005	-26.6		6.9		116		86.7	2.0	

Appx 15: Samples, stable isotopes, Hg species, and TL used in TMF calculation

Region / Lake	Sample	N	Year	$\delta^{13}C$	SD	$\delta^{15}N$	SD	THg*	SD	MeHg	TL	SD
	Periphyton	1	2005	-		*3.5		-		2.7	1.0	
Char Lake	Arctic char	7	2005	-23.9	0.4	11.9	1.0	0.595	0.22	-	4.3	0.3
		6	2007	-23.0	1.0	11.3	1.4	0.336	0.30	-	4.1	0.4
	Juvenile Arctic char	12	2005-2006	-25.7	1.4	7.0	1.4	133	0.87	-	0.7	0.0
	Chironomidae	1	2006	-24.4		5.2		133		120.4	2.3	
		1	2005	-26.2		5.9		200		174.3	2.5	
	Zooplankton >500µm	1	2007	-31.2		6.3		58		43.1	2.6	
	Zooplankton 250-500µm	1	2007	-31.1		6.2		71		34.0	2.6	
	Zooplankton >250µm	1	2005	-31.9		6.8				25.4	2.7	
	Char Fary Shrimp	1	2005	-28.2		3.4				113.3	1.7	
	Periphyton	1	2006	-28.7		0.8				9.1	1.0	
North	Arctic char	11	2005	-23.8	0.4	10.3	0.8	0.261	0.16	-	3.0	0.2
		6	2006	-23.9	0.4	10.1	0.3	0.233	0.04	-	3.0	0.1
		5	2007	-23.9	0.1	9.8	0.3	0.305	0.11	-	2.9	0.1
	Juvenile Arctic char	16	2005-2006	-25.6	1.7	9.5	1.0	394	180	-	2.8	0.3
	Chironomidae	1	2006	-25.0		7.6		166		101.7	2.2	
		1	2005	-26.9		8.2		313		315.6	2.4	
	Zooplankton >500µm	1	2007	-		7.3		-		17.7	2.2	
	Zooplankton 250-500µm	1	2007	-		7.2		-		6.5	2.1	
	Zooplankton >250µm	1	2006	-		7.5		42		29.2	2.2	
		1	2005	-35.7		7.1		-		29.2	2.1	
	Periphyton	1	2007	-17.1		3.4		-		-	1.0	
Resolute	Arctic char	16	2005	-22.3	0.7	12.7	1.0	0.221	0.10	-	4.0	0.3
		8	2006	-22.2	0.9	12.4	1.3	0.226	0.08	-	3.9	0.4
		14	2007	-22.6	0.6	11.4	0.9	0.200	0.07	-	3.6	0.3
	Juvenile Arctic char	16	2005-2006	-25.2	0.9	10.0	0.8	388	218	-	3.2	0.2
	Chironomidae	1	2006	-23.7		7.5		156		136.5	2.5	
		1	2005	-24.1		8.2		198		147.2	2.7	
	Zooplankton >500µm	1	2007	-32.7		11.3		29		14.1	3.6	
	Zooplankton 250-500µm	1	2007	-31.7		9.2		31		17.2	3.0	
	Zooplankton 250-500µm	1	2006	-31.4		7.6		61		12.3	2.5	

Appx 15: Samples, stable isotopes, Hg species, and TL used in TMF calculation

Region / Lake	Sample	N	Year	$\delta^{13}C$	SD	$\delta^{15}N$	SD	THg*	SD	MeHg	TL	SD
	Zooplankton >500µm	1	2006	-33.8		7.6		44		15.6	2.5	
	Periphyton	1	2006	-		3		52		3.2	1.0	
Small	Arctic char	13	2005	-23.7	1.1	10.6	0.9	0.135	0.12	-	3.0	0.3
		5	2006	-23.1	1.3	10.3	1.2	0.202	0.14	-	2.9	0.4
		8	2007	-23.7	0.8	9.3	1.0	0.077	0.07	-	2.7	0.3
	Juvenile Arctic char	12	2005-2006	-24.1	0.8	8.9	0.6	178	67.09	-	2.5	0.2
	Chironomidae	1	2006	-25.8		4.4		86		59.4	1.2	
		1	2005	-27.1		6.5		104		80.3	1.8	
	Zooplankton >250µm - 1	1	2007	-31.3		9.4		-		14.3	2.7	
	Zooplankton >250µm - 2	1	2007	-33.7		10.6		30		20.5	3.0	
	Periphyton	1	2007	-15.9		3.7		-		2.6	1.0	
Teardrop	Arctic char	4	2007	-24.7	0.6	10.8	0.1	0.185	0.01	-	2.9	0.0
	Juvenile Arctic char	3	2005-2006	-25.7	1.1	10.05	0.7	162	56	-	2.7	0.2
	Chironomid midges	1	2006	-24.0		8.4		362		319.1	2.2	
	Zooplankton >500µm	1	2006	-27.6		5.3		-		24.9	1.3	
	Zooplankton 250-500µm	1	2006	-30.0		6.3		82		24.9	1.6	
	Zooplankton >250µm	2	2007	-30.5		8.1		-		17.4	2.1	
	Periphyton	1	2007	-20.7		4.3		71		3.7	1.0	
Meretta	Arctic char	12	2006	-24.9	1.8	11.8	0.3	0.328	0.08	-	3.6	0.1
	Juvenile Arctic char	3	2005-2006	-22.3	0.1	10.9	0.3	313	16	-	3.3	0.1
	Chironomidae	1	2006	-20.5		8.6		188		186.0	2.7	
		1	2005	-21.3		9.2		244		190.6	2.8	
	Daphnidae >1000µm	1	2007	-28.9		10.7		153		127.4	3.3	
	Zooplankton >250µm - 1	1	2007	-29.3		8.0		-		109.0	2.5	
	Zooplankton >250µm - 2	1	2007	-29.1		7.1		-		93.2	2.2	
	Periphyton	1	2007	-23.8		3.0		-		11.2	1.0	

Region / Lake	Sample	N	Year	$\delta^{13}C$	SD	$\delta^{15}N$	SD	THg*	SD	MeHg	TL	SD
Ellesmere								µg/g		µg/g		
Lake G	Arctic char	33	2007	-22.7	0.0	9.0	0.4	0.197	0.30	-	3.2	0.1
	Zooplankton	1	2007	-32.4		8.7		33		1.4	3.1	

Appx 15: Samples, stable isotopes, Hg species, and TL used in TMF calculation

Region / Lake	Sample	N	Year	δ^{13}C	SD	δ^{15}N	SD	THg*	SD	MeHg	TL	SD
	Periphyton	1	2007	-12.7		1.6					1.0	
Hazen	Arctic char	18	2006	-19.8	2.2	9.9	1.8	0.102	0.11	-	3.3	0.5
		31	2007	-20.4	1.8	10.5	1.9	0.073	0.19	-	3.4	0.6
	Juvenile Arctic char	6	2006	-21.8	1.5	8.1	0.6	54	25	-	2.7	0.2
	Chironomidae	1	2006	-20.2		4.7		94		49.4	1.8	
		1	2007	-19.4		4.2		232		50.5	1.6	
	Zooplankton 250-500µm	1	2006	-35.7		5.9		32		21.2	2.1	
	Zooplankton >250µm 1	1	2007	-33.0		6.3		113		14.0	2.2	
	Zooplankton >250µm 2	2	2007	-37.2		5.2		-		23.8	1.9	
	Periphyton	1	2006	-30.0		2.2		65		2.4	1.0	
Lake F	Arctic char	9	2007	-23.2	1.4	8.3	1.5	0.116	0.19	-	3.2	0.4
	Zooplankton >250µm	4	2007	-33.0	1.0	6.36	0.51	57	0.00	34.5±1.85	2.8	0.2
	Periphyton	1	2007	-9.5		0.8		11		3.0	1.0	
Lake D	Arctic char	7	2007	-27.5	2.7	12.9	1.4	0.079	0.09	-	4.2	0.4
	Water Mite		2007	-37.4		12.7		-		22.0	4.2	
	Chironomidae	1	2007	-28.2		6.1		263		24.0	2.2	
		1	2007	-26.9		6.8		263		24.0	2.4	
	Zooplankton >250µm	3	2007	-38.4	0.15	12.7	1.4	-		22.0	4.2	0.4
	Periphyton	1	2007	-19.5		1.9		-		1.1	1.0	
Kent Peninsula												
Gavia Faeces	Arctic char	8	2006	-25.3	1.1	8.1	1.2	0.109	0.18	-	3.1	0.4
	Juvenile Arctic char	4	2006	-24.9	1.3	6.7	1.3	91	0.20	-	2.7	0.2
	Zooplankton 250-500µm	1	2006	-28.1		5.1		48		2.7	2.3	
	Zooplankton >500µm	1	2006	-28.1		5.1		22		2.7	2.3	
	Periphyton	1	2006	-27.6		0.8		69		0.8	1.0	
Little Nauyuk	Arctic char	13	2006	-22.2	0.3	7.6	0.6	0.104	0.06	-	3.1	0.2
	Juvenile Arctic char	4	2006	-23.6	1.2	6.9	0.6	93	22	-	2.9	0.2
	Zooplankton 250-500µm	1	2006	-25.3		1.2		64		2.2	1.2	
	Zooplankton >500µm	1	2006	-28.4		6.5		53		5.8	2.8	
	Periphyton	1	2006	-23.3		0.3		48		1.0	1.0	
Notgordie	Arctic char	5	2006	-25.6	0.7	10.4	2.7	0.980	0.39	-	3.7	0.8

Appx 15: Samples, stable isotopes, Hg species, and TL used in TMF calculation

Region / Lake	**Sample**	**N**	**Year**	**$\delta^{13}C$**	**SD**	**$\delta^{15}N$**	**SD**	**THg***	**SD**	**MeHg**	**TL**	**SD**
	Juvenile Arctic char	11	2006	-26.4	0.6	6.8	0.3	340	125	-	2.6	0.1
	Stickleback	4	2006	-27.0	0.8	7.9	0.5	450	109	-	2.9	0.1
	Trichoptera	1	2006	-25.8		2.2		271		132.6	1.3	
	Zooplankton >250µm	1	2006	-28.0		4.6		89		35.0	2.0	
	Periphyton	1	2006	-23.0		1.3		49		-	1.0	
Keyhole	Arctic char	7	2006	-27.7	0.7	11.3	0.8	0.094	0.10	-	3.9	0.2
	Stickleback	3	2006	-29.6	0.7	10.5	0.2	268	55	-	3.6	0.1
	Trichoptera	1	2006	-29.8		7.0		38		19.9	2.6	
	Zooplankton >250µm	1	2006	-32.3		5.6		133		55.9	2.2	
	Periphyton	1	2006	-15.6		1.6		18		1.1	1.0	

* **Note:** THg concentrations for adult Arctic char is given in µg/g ww, all others ng/g dw

Appx 16: Arctic char summer prey items as determined from stomach contents (by region)

	Char population	Prey item	Taxonomic group	Prey-Species	Prey-Habitat
Ellesmere Island (4 Lakes)					
	Hazen	Fish	Salmonidae	*S. alpinus* (<15 cm)	litoral
	all lakes	Benthos	Diptera (pupae)	*Chironomus sp.*	benthic
	all lakes	Zooplankton	Copepoda	*Cyclops (scutifer)*	pelagic
Resolute area (10 Lakes)					
	Amituk, Char, North, Resolute	Fish	Salmonidae	*S. alpinus* (<12 cm)	litoral
	all lakes	Benthos	Diptera (pupae)	*Chironomus sp.*	benthic
	all lakes	Zooplankton	Copepoda	*Cyclops (scutifer)*	pelagic
	Meretta		Cladocera	*Daphnia sp.*	bentho-pelagic
	Char		Branchiopoda	*B. paludosa*	bentho-pelagic
Kent Peninsula and Victoria Island (4 Lakes)					
	Keyhole, Gavia Faeces	Fish	Gasterosteidae	*P. pungitus*	litoral
	Little Nauyuk	Benthos	Isopoda	*M. entomon*	benthic
	Keyhole, Notgordie		Trichoptera	*G. praeterita* (W)	benthic
	all		Diptera (pupae)	*Chironomus sp.*	benthic
	Notgordie	Zooplankton	Cladocera	*Daphnia sp.*	bentho-pelagic
	Notgordie, Little Nauyuk		Amphipoda	*Gammarus sp.*	bentho-pelagic
	all		Copepoda	*Cyclopoda sp.*	pelagic
Ungava Peninsula (1 Lake)					
	Pingualuk	Fish	Salmonidae	*S. alpinus* (<25 cm)	litoral
		Zooplankton	Copepoda	-	pelagic

Appx 17: Selected water chemistry parameters for 18 lakes

Lake	Region		DOC		Chl a		SO_4^{-2}		Cl^-		SIO_2		POC		PON	
		Sampling Years	µg/L	SD	µg/L	SD	µg/L	SD	µg/L	SD	µg/L	SD	µg/L	SD	µg/L	SD
Lake D	Ellesmere	2007	2.8		1.2		1.2		0.5		0.65		0.25		0.03	
Lake G			1.6		1.0		20.5		2.1		2.66		0.31		0.02	
Hazen		2005-2007	0.6	0.2	0.9	0.1	10.1	1.0	0.3	0.1	0.88	0.12	0.08	0.04	0.01	0.00
Amituk	Cornwallis	2005-2007	1.7	1.9	0.3	0.1	2.5	2.5	2.1	0.3	0.25	0.06	0.07	0.02	0.01	0.01
Aquiatusuk		2005+2007	0.7		0.7		9.8		3.2		0.28		0.16		0.02	
12 Mile		2005-2007	1.2	0.3	0.7	0.5	2.3	0.5	5.1	1.1	0.20	0.04	0.12	0.04	0.02	0.01
9 Mile		2006+2007	1.6		0.6		0.8		4.1		0.56		0.23		0.03	
North		2005-2007	1.3	0.5	2.1	1.1	1.5	1.8	2.7	1.6	0.21	0.04	0.12	0.06	0.01	0.01
Meretta			1.4	0.2	1.3	1.1	3.9	2.4	9.0	2.7	0.16	0.05	0.15	0.04	0.02	0.01
Char			1.0	0.2	0.7	0.5	11.8	1.6	10.4	1.0	0.64	0.10	0.11	0.03	0.01	0.01
Resolute			1.1	0.1	0.9	0.8	13.1	2.8	19.3	2.4	0.47	0.08	0.16	0.06	0.02	0.01
Small			1.8	0.3	1.8	1.8	2.0	0.4	13.0	2.8	0.48	0.15	0.22	0.09	0.03	0.02
Teardrop		2006+2007	2.1		1.1		8.6		17.3		0.36		0.16		0.02	
Boomerang	Somerset	2007	0.7		1.3		2.7		1.8		0.58		0.09		0.02	
Keyhole	Victoria	2006	5.6		1.1		4.4		33.1		0.40		0.34		0.03	
Gavia Faeces	Kent	2006	4.3		0.3		5.8		53.2		2.46		0.34		0.03	
Little Nauyuk			2.7		< 0.1		4.9		71.2		2.50		0.41		0.03	
Notgordie			6.8		0.6		12.7		118.0		3.02		0.41		0.04	
Laflamme	Ungava	2007	1.7		0.3		0.6		0.9		0.31		0.37		0.05	
Pingualuk			0.7		< 0.1		0.8		1.1		0.28		0.13		0.02	

Appx 18: Ambient air temperature (°C) in July at four main sampling regions

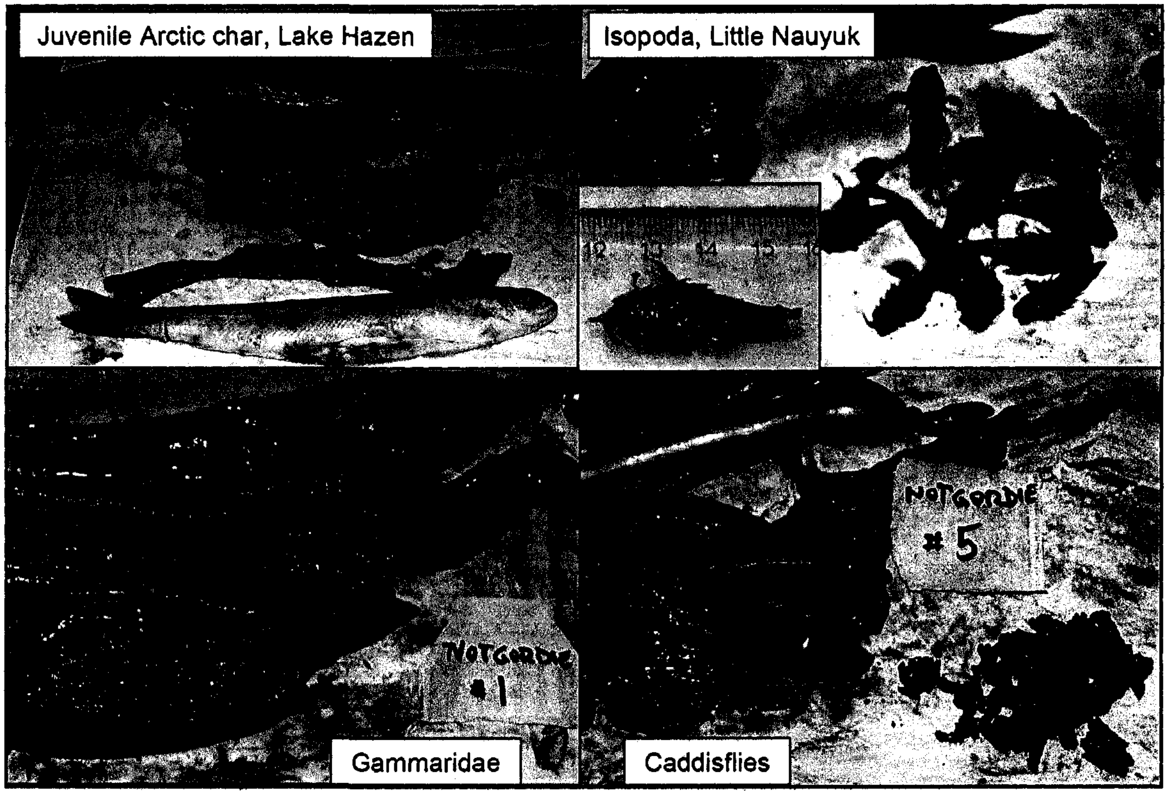
Mean air-temperature	2005	2006	2007
Ellesmere Island - Alert weather station	3	3.7	2.4
Cornwallis Island - Resolute Bay	3.8	4.3	7.4
Kent Peninsula/Victoria Island - Cambridge Bay	7	9.3	11.9
Nain, Labrador	12.1	12.0	10.7

*Source: Environment Canada

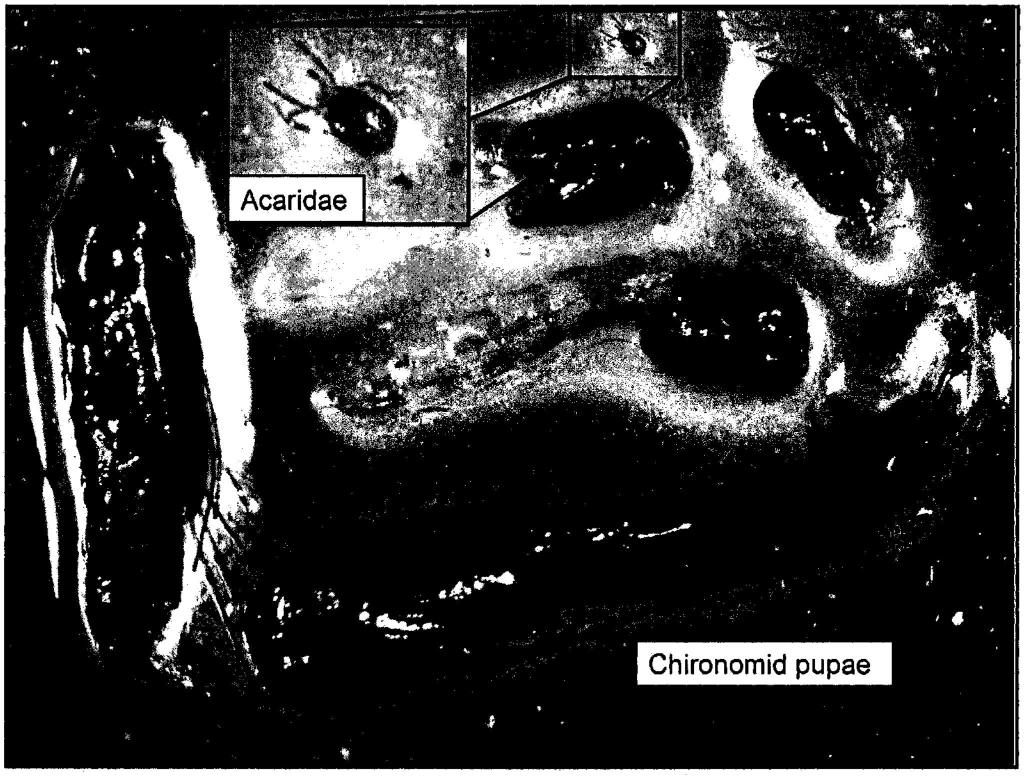
Appx 19: Relationships of TMF and FCL 18 lakes with location and selected environmental factors

	Linear regression parameters			
TMF versus	r_{adj}^2	p<0.05	slope	intercept
Latitude	-	0.58	-	-
Longitude	-	0.47	-	-
Lake area (LA)	-	0.70	-	-
Catchment area (CA)	-	0.53	-	-
CA-LA ratio	-	0.87	-	-
DOC	-	0.56	-	-
Chl a	-	0.52	-	-

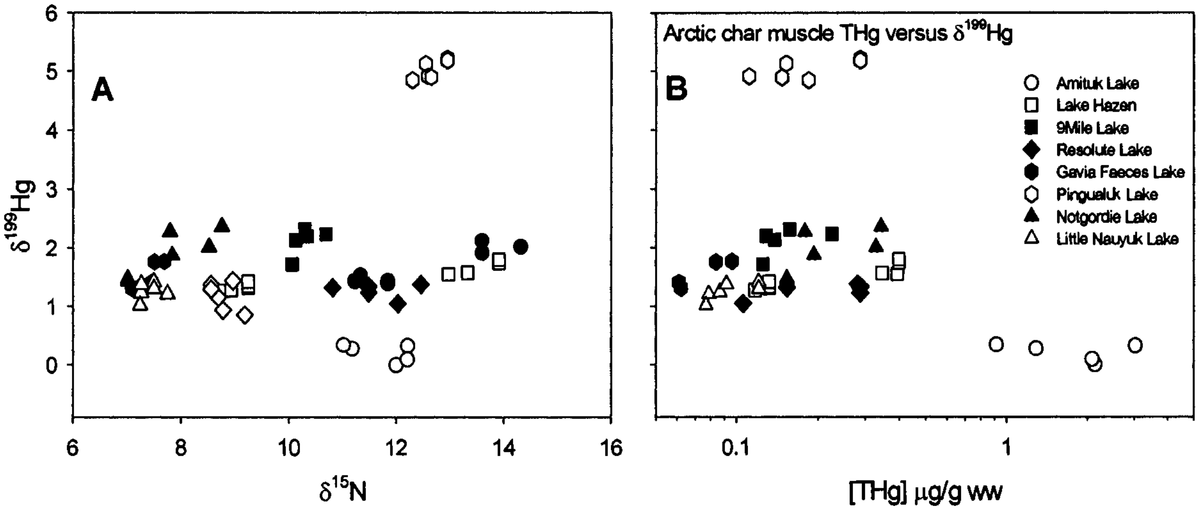
	Linear regression parameters			
FCL versus	r_{adj}^2	p<0.05	slope	intercept
Latitude	-	0.64	-	-
Longitude	-	0.20	-	-
Lake area (LA)	0.15	0.07	0.1814	3.1328
Catchment area (CA)	-	0.72	-	-
CA-LA ratio	-	0.79	-	-
DOC	-	0.90	-	-
Chlorophyll a	-	0.49	-	-



Appx 20: Stomach contents of Arctic char. *Pictures © N. Gantner*



Appx 21: Microscopic photograph of stomach contents of Arctic char. The dark *Chironomidae* pupae (bottom) is approximately 2-3 mm long. Red water mites (*Acaridae*, insert) were present in Arctic char stomachs, but may be co-ingested with chironomids. *Picture provided by W. Michaud (University of Waterloo).*



Appx 22: A. Relationship of δ^{199}Hg and δ^{15}N in Arctic char. B. THg concentrations in Arctic char muscle compared to δ^{199}Hg (legend applies to A and B).

Appx 23: Average Hg ratios, δ and Δ values for all fish, sediment and food web data

Lake	Arctic char	δ^{15}N	[THg]	$^{199/198}$Hg	δ^{199}Hg	Δ^{199}Hg	$^{200/198}$Hg	δ^{200}Hg	Δ^{200}Hg	$^{201/198}$Hg	δ^{201}Hg	Δ^{201}Hg	$^{202/198}$Hg	δ^{202}Hg
		‰	ppm	‰	‰	‰	‰	‰	‰	‰	‰	‰	‰	‰
Lake D	Avg (n=5)	12.5	0.127	1.6935	1.6853	1.8626	2.3091	-0.2888	0.0650	1.3173	0.9696	1.4989	2.9667	-0.7039
	SD	1.4	0.108	0.0010	0.7696	0.7963	0.0003	0.0971	0.0625	0.0007	0.5857	0.6589	0.0005	0.1565
Lake G	Avg (n=5)	8.8	0.202	1.6930	1.1450	1.5184	2.3086	-0.7023	0.0426	1.3164	0.0398	1.1544	2.9645	-1.4824
	SD	0.2	0.024	0.0004	0.2459	0.2106	0.0002	0.0870	0.0245	0.0004	0.2856	0.1778	0.0004	0.1465
Lake Hazen	Avg (n=3) L	13.4	0.380	1.6934	1.6348	1.9077	2.3087	-0.5268	0.0177	1.3170	0.7131	1.5277	2.9655	-1.0835
	SD	0.5	0.030	0.0002	0.1306	0.1175	0.0001	0.0728	0.0618	0.0002	0.1371	0.1004	0.0002	0.0538
	Avg (n=3) S	9.0	0.099	1.6928	1.3036	1.6739	2.3083	-0.6804	0.0583	1.3163	0.2057	1.3110	2.9645	-1.4701
	SD	0.2	0.044	0.0001	0.0492	0.0093	0.0002	0.0801	0.0256	0.0002	0.1308	0.0167	0.0006	0.1922
	Avg (n=6)	11.2	0.240	1.6931	1.4692	1.7908	2.3085	-0.6036	0.0380	1.3167	0.4594	1.4193	2.9650	-1.2768
	SD	2.4	0.157	0.0004	0.2017	0.1482	0.0003	0.1085	0.0478	0.0004	0.3027	0.1350	0.0007	0.2465
Amituk Lake	Avg (n=5)	11.7	1.896	1.6908	0.2111	0.3877	2.3088	-0.3709	-0.0187	1.3154	-0.4437	0.0832	2.9666	-0.7008
	SD	0.6	0.826	0.0003	0.1537	0.1570	0.0002	0.1188	0.0563	0.0002	0.1473	0.1397	0.0003	0.1331
9Mile Lake	Avg (n=5)	10.3	0.155	1.6944	2.1262	2.0980	2.3104	0.1206	0.0643	1.3184	1.5973	1.5131	2.9694	0.1120
	SD	0.2	0.041	0.0005	0.2335	0.2246	0.0002	0.0364	0.0105	0.0003	0.2224	0.1948	0.0002	0.0559
Resolute Lake	Avg (n=4)	11.7	0.207	1.6934	1.2639	1.3227	2.3102	-0.0728	0.0445	1.3175	0.7755	0.9510	2.9680	-0.2334
	SD	0.7	0.091	0.0002	0.1465	0.1318	0.0001	0.0601	0.0568	0.0002	0.1616	0.1177	0.0002	0.0693
Gavia Faeces L.	Avg (n=5)	7.4	0.070	1.6940	1.5233	1.4465	2.3112	0.2522	0.0991	1.3183	1.2867	1.0576	2.9696	0.3047
	SD	0.2	0.020	0.0003	0.2300	0.2286	0.0003	0.0434	0.0400	0.0002	0.1955	0.1866	0.0002	0.0772
Little Nauyuk L.	Avg (n=5)	7.4	0.091	1.6938	1.2439	1.2147	2.3112	0.1445	0.0862	1.3182	1.0210	0.9337	2.9693	0.1161
	SD	0.2	0.018	0.0002	0.1422	0.1395	0.0002	0.0435	0.0167	0.0001	0.1286	0.1222	0.0004	0.0763
Notgordie Lake	Avg (n=5)	8.0	0.240	1.6952	2.0011	1.9801	2.3112	0.1301	0.0883	1.3189	1.5226	1.4601	2.9693	0.0832
	SD	0.7	0.089	0.0006	0.3603	0.3508	0.0001	0.0239	0.0185	0.0004	0.3131	0.2853	0.0002	0.0507
Lake Pingualuk	Avg (n=5)	12.6	0.177	1.7003	4.9947	4.6923	2.3125	0.7047	0.1014	1.3229	4.6033	3.7006	2.9725	1.2006
	SD	0.2	0.067	0.0002	0.1546	0.1485	0.0001	0.0336	0.0443	0.0001	0.1420	0.1166	0.0002	0.0607

Appx 24: Average Hg ratios, δ^{x}Hg and Δ^{x}Hg values for all fish, sediment and food web data

		$^{199/198}$Hg	δ^{199}Hg	Δ^{199}Hg	$^{200/198}$Hg	δ^{200}Hg	Δ^{200}Hg	$^{201/198}$Hg	δ^{201}Hg	Δ^{201}Hg	$^{202/198}$Hg	δ^{202}Hg
Sediments	**Sample ID**	‰	‰	‰	‰	‰	‰	‰	‰	‰	‰	‰
Lake D	Lake D 2007	1.6916	0.0932	0.2032	2.3095	-0.1912	0.0282	1.3159	-0.2087	0.1197	2.9656	-0.4368
Lake G	2007 Avg. (n=2 dig)	1.6911	-0.1946	0.0140	2.3090	-0.4133	0.0027	1.3154	-0.5912	0.0312	2.9644	-0.8279
	SD	0.0001	0.0719	0.0680	0.0000	0.0145	0.0069	0.0000	0.0310	0.0196	0.0000	0.0152
Lake Hazen	2007 - Spl I (n=3 dig)	1.6903	-0.6502	-0.3711	2.3086	-0.5792	-0.0225	1.3147	-1.1768	-0.3439	2.9636	-1.1079
	SD	0.0001	0.0489	0.0595	0.0000	0.0184	0.0391	0.0000	0.0325	0.0673	0.0002	0.0509
	2007 (n=3 samples)	1.6904	-0.5052	-0.2227	2.3090	-0.5780	-0.0144	1.3150	-1.1271	-0.2839	2.9644	-1.1215
	SD	0.0001	0.1262	0.1293	0.0003	0.0108	0.0137	0.0002	0.0434	0.0521	0.0007	0.0120
	06+'07 (n=4, 2 yr)	1.6905	-0.4568	-0.1848	2.3090	-0.5287	0.0139	1.3151	-1.0267	-0.2147	2.9644	-1.0799
	SD	0.0003	0.2216	0.2079	0.0004	0.0864	0.0563	0.0004	0.2184	0.1743	0.0008	0.0606
Amituk Lake	Amituk Lake '06	1.6907	-0.3345	-0.0921	2.3094	-0.4723	0.0112	1.3155	-0.8872	-0.1637	2.9654	-0.9622
9Mile Lake	9Mile Lake '06	1.6924	0.1828	0.3608	2.3104	-0.3903	-0.0351	1.3167	-0.2844	0.2471	2.9663	-0.7069
Resolute Lake	Resolute '06	1.6920	-0.3485	0.1629	2.3094	-0.9956	0.0248	1.3156	-1.4431	0.0836	2.9625	-2.0305
Gavia Faeces	Average (n= 3 dig)	1.6929	0.4092	0.5812	2.3110	-0.1913	0.1519	1.3171	-0.1321	0.3814	2.9664	-0.6829
	SD	0.0001	0.0986	0.0821	0.0001	0.0922	0.0569	0.0001	0.0565	0.0303	0.0003	0.1012
Little Nauyuk	Average (n= 2 dig)	1.6924	-0.1165	0.0704	2.3110	-0.3113	0.0616	1.3168	-0.5176	0.0403	2.9664	-0.7420
	SD	0.0000	0.0238	0.0176	0.0003	0.1209	0.0383	0.0002	0.1306	0.0070	0.0005	0.1644
Notgordie Lake	Notgordie '06	1.6935	0.3225	0.7359	2.3101	-1.0006	-0.1758	1.3167	-0.9752	0.2588	2.9644	-1.6413
Lake Pingualuk	Pingualuk 2007	1.6896	-0.1948	-0.2577	2.3094	0.1687	0.0433	1.3152	-0.3365	-0.5243	2.9678	0.2498
Food web (3 Lakes)												
Zooplankton	Hazen Lake '07	1.6960	2.1817	2.2978	2.3115	-0.0591	0.1725	1.3193	1.5068	1.8533	2.9671	-0.4609
	Resolute L. '05	1.6980	3.3427	3.4014	2.3120	0.1355	0.2526	1.3206	2.4985	2.6737	2.9678	-0.2331
	Resolute L. '06	1.6960	2.1614	2.1952	2.3117	0.0436	0.1111	1.3192	1.4454	1.5464	2.9681	-0.1343
	Resolute L. '07	1.6971	2.8472	2.8838	2.3121	0.1927	0.2658	1.3201	2.0661	2.1755	2.9680	-0.1455
	Av. (3 spl yrs)	1.6970	2.7838	2.8268	2.3119	0.1239	0.2098	1.3200	2.0033	2.1319	2.9680	-0.1710
	SD	0.0010	0.5932	0.6051	0.0002	0.0752	0.0858	0.0007	0.5293	0.5649	0.0002	0.0541
	Gavia Faeces L. '06	1.6948	1.4838	1.5081	2.3126	0.4198	0.4683	1.3187	1.0243	1.0970	2.9682	-0.0967
	Notgordie L. '06	1.6975	3.0791	3.0495	2.3124	0.3057	0.2467	1.3204	2.3590	2.2708	2.9688	0.1173
Insects	Hazen L. '06	1.6924	-0.0844	0.3167	2.3100	-0.7479	0.0522	1.3161	-1.0349	0.1623	2.9638	-1.5923
	Resolute L. '06	1.6946	1.2320	1.3130	2.3116	-0.0773	0.0845	1.3184	0.7461	0.9882	2.9676	-0.3219
SRMs	DORM1 (n=3)	1.6931	1.2455	1.1988	2.3105	0.1416	0.0484	1.3177	1.0795	0.9400	2.9693	0.1855

Appx 24: Average Hg ratios, δ^{x}Hg and Δ^{x}Hg values for all fish, sediment and food web data

SD	0.0003	0.0556	0.0678	0.0002	0.0757	0.0398	0.0001	0.1006	0.0860	0.0008	0.2163
BRS (n=3)	1.6912	-0.1827	-0.0735	2.3101	-0.2388	-0.0208	1.3161	-0.3973	-0.0711	2.9673	-0.4338
SD	0.0000	0.0287	0.0289	0.0001	0.0305	0.0275	0.0000	0.0237	0.0174	0.0000	0.0153

Appx 25: Dates of sampling and personnel present (2005-2007)

	2005		2006		2007	
Lake	**Date**	**Team**	**Date**	**Team**	**Date**	**Team**
Lake D	-	-	-	-	July 28	GP, JDR, MP, NG
Lake G	-	-	-	-	July 27	
Lake F	-	-	-	-	July 28	
Hazen	-	-	July 31- Aug 2	NG, DI, GK	July 29, Aug 2-6	
Amituk	July 5	NG, PI, DI	July 28	NG, PI, DI, GK	July 29	GK, DI, PI
Aquiatusuk	July 21, Aug 08	NG*, PI, DI, DCM*			July 30	
12Mile	July 23	NG, PI, DI, DCM	17-18 July	NG, PI, DI	Aug 4	
9Mile	17,28 July	NG, PI, DI	18,24 July	NG, PI, DI, DCM*	Aug 4	
North	July 19		July 24		Aug 2	
Meretta	July 26		July 26	NG, PI, DI, DCM	Aug 3 + 6	
Char	July 16,17,26-27		July 25		July 22,28,30, Aug 1	
Resolute	July 15		July 23,27	NG, PI, DI, DCM*	July 22,28,30-31, Aug 6	
Small	July 25	NG, PI, DI, DCM	July 17	NG, PI, DI	Aug 1-2	
Teardrop	-	-	July 27	NG, PI, DI, DCM	Aug 1+3	
Boomerang	-	-	-	-	July 31-Aug 1	GK, DI, PI
Keyhole	-	-	July 13	NG, PI, DI	-	-
Gavia						
Faeces	-	-	July 5-6		-	-
Little						
Nauyuk	-	-	July 5-6		-	-
Notgordie	-	-	July 7		-	-

* asterix (*) marks participation in one of multiple trips

Note: NG = Nikolaus Gantner, PI = Pilipoosie Iqaluk, DI = Debbie Iqaluk, DCM = Derek Muir, GK = Günter Köck
JDR = Jim Reist, MP= Mike Power, GP = Geoff Power

Appx 26: List of Abbreviations

Abbreviation	Full Term
ACIA	Arctic Climate Impact Assessment
AMAP	Arctic Monitoring and Assessment Program
ANCOVA	Analysis of Co-Variance
BMF	Biomagnification Factors
CA	Catchment Area
CA/LA	Catchment-to-Lake-Area ratio
CCIW	Canadian Centre for Inland Waters
DDT	Dichloro-Diphenyl-Trichloroethane
DL	Detection Limit
DOC	Dissolved Organic Carbon
FCL	Food Chain Length
HgSI	Mercury Stable Isotope (ratios)
IPCC	Intercontinental Panel on Climate Change
LA	Lake Area
MDE	Mercury Depletion Event
MDF	Mass-Dependent Fractionation
MeHg	methylmercury
MIF	Mass-Independent Fractionation
NCP	Northern Contaminants Program
PBDE	Poly-Brominated Diphely Ethers
PON	Particulate Organic Nitrogen
POC	Particulate Organic Carbon
POP	Persistent Organic Pollutants
THg	Total mercury
TL	Trophic Level
TMF	Trophic Magnification Factor
TNW	Tundra North-West