Controls of Fish Mercury Concentrations in Subarctic Lakes of Canada

by

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Examining Committee Membership

The following served on the Examining Committee for this thesis. The decision of the Examining Committee is by majority vote.

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Author's Declaration

This thesis consists of material all of which I authored or co-authored: see Statement of Contributions included in the thesis. This is a true copy of the thesis, including any required final revisions, as accepted by my examiners.

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

Statement of Contributions

While the research in this thesis is my own, many have contributed to the large and collaborative project on which my PhD is based, and co-authors provided valuable input to data chapters (Chapters 2, 3, and 4)

Samples used in my PhD thesis have been collected since 2013 by Heidi K. Swanson (my supervisor) and her past and present crew, including students, postdocs, colleagues, friends, and partner First Nations. Before I started my PhD in September 2018, ten lakes had been sampled in the study area, the Dehcho Region of Northwest Territories (NT), Canada. I was part of the sample collection and processing crew in 2019 (one lake) and 2021 (two lakes), although 2021 data are not included in the current thesis due to the delays caused by the Covid-19 pandemic. Shelley Lundvall, Amy Nguyen, Ari Yamaguchi, Tara Boag, Isabel Hilgendag, Rachel Broders, Heather Dixon, Leanne Baker, Mike Low, George Low, Barb Low, and Brian Branfireun are among the students, postdocs, colleagues, and friends who contributed to the field and laboratory processes. Partner First Nations in NT, including Łíídlii Kúé First Nation (Fort 611 Simpson), Tthets'éhk'edéli First Nation (Jean Marie River), Sambaa K'e First Nation (Trout Lake), Ka'a'gee 612 Tu First Nation (Kakisa), Deh Gáh Got'ie First Nation (Fort Providence), and Pehdzeh Ki First Nation 613 (Wrigley), provided guidance and support throughout the project.

Analyses of water and sediment parameters included in the current thesis are conducted by partner laboratories, including the University of Waterloo Environmental Isotope Laboratory in Waterloo, the Western University Biotron Center for Experimental Climate Change Research in London, and the University of Alberta Biogeochemical Analytical Service Laboratory in Edmonton. Analyses of geospatial data were performed by

MTE consultants Inc., specifically, Jared Cunningham. Contributions of authors to each data chapter of this thesis are as follows:

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Information regarding research ethics approval for my PhD research is fully provided in Appendix A. Financial support was provided from Canada First Excellence Research Fund (Global Water Futures; Northern Water Futures project), the Cumulative Impacts Monitoring Program (Government of the Northwest Territories), the Northern Contaminants Program, Dehcho First Nations, Dehcho Aboriginal Aquatic Resources and Oceans Management program (Fisheries and Oceans Canada), Discovery Grants to Heidi K. Swanson, Brian A. Branfireun, and Brian D. Laird, and a Northern Research Supplement to Heidi K. Swanson. Significant in-kind support was provided by the Dehcho Aboriginal Aquatic Resources and Oceans Management Program. During my PhD, I personally received scholarships and awards from the Department of Biology and University of Waterloo, including the Brian M. Jessop Graduate Scholarship in Fisheries Science, Dr Noel Hynes Graduate Scholarship, J. Frank Brookfield Memorial Graduate Scholarship, Science Senate Graduate Scholarship, Graduate Scholarship, and Academic Readiness Bursary. I have also been supported by teaching assistantships from Department of Biology, and by financial aids and research assistantships from my supervisor, Dr Heidi K. Swanson.

Abstract

Mercury (Hg) is a global pollutant that in its organic and more toxic form, methyl Hg (MeHg), can cause detrimental health effects on fish, wildlife, and humans. In lake ecosystems, concentrations of Hg ([Hg]) in fish reflect a complex outcome of interactions among variables that act at different ecological and spatial scales. From catchment and lake characteristics to fish and food web ecology, numerous processes interact to influence Hg uptake, methylation, bioaccumulation, and biomagnification, and ultimately, [Hg] in fish. As a result, fish [Hg] often vary among lakes and species, and among individuals of the same population. While factors that affect fish [Hg] in lakes are well studied and generally well understood, the relative importance and influence of these factors can vary among populations, species, ecosystems, and geographical regions, making it difficult to generalize and predict variability in fish [Hg].

Generally, little is known about controls of fish [Hg] in northern regions, where ecosystems and biota can be profoundly influenced by environmental perturbations, and where fish provide a critically important subsistence food source. In the subarctic Dehcho Region, Northwest Territories, Canada, fish [Hg] vary widely among lakes due to largely unknown mechanisms, and have, on occasion, triggered both general and site-specific consumption notices (sometimes referred to as consumption advisories or contaminant advisories). These advisories, and the lack of understanding about sources of among-lake variation in fish [Hg], are of concern for First Nation communities that rely on wild-caught fish as a subsistence food source. This thesis is aimed to address knowledge gaps regarding causes of among-lake variation in fish [Hg] in the Dehcho Region, and to improve the current understanding of what variables and processes control fish [Hg] in remote and understudied subarctic lakes.

In Chapter 1, I provide a detailed overview of freshwater fish and fisheries, Hg cycling in the environment, and controls of [Hg] in fish, with a focus on lakes, especially in the Canadian subarctic. I then describe my study area, the Dehcho Region, and elaborate on the importance of wild-caught freshwater fish for local communities and Hg-related fish safety concerns, which frames my specific objectives for each data chapter (i.e., Chapters 2, 3, and 4). My data chapters, and associated analyses and findings, use comprehensive geospatial, environmental, and biological data that were collected from eleven lakes in the Dehcho Region over the period of seven years, from 2013 to 2019.

In Chapter 2, I conducted an ecosystem-scale study to investigate causes of among-lake variability in fish [Hg] in the study area. I investigated how interacting processes at scales ranging from whole catchments to individual organisms influence [Hg] in Northern Pike (*Esox lucius*), a predatory fish of widespread subsistence and commercial importance. Findings indicated that more than four-fifths of the among-lake variability in [Hg] in Northern Pike was explained by fish growth rates (negative) and concentrations of methyl Hg ([MeHg]) in benthic invertebrates (positive). Fish growth rates and [MeHg] in benthic invertebrates were, in turn, influenced by concentrations of dissolved organic carbon and MeHg in water, and total Hg in sediment. These in-lake variables were ultimately driven by catchment characteristics. Lower-elevation lakes in relatively larger catchments with proportionally more temperate/subpolar needleleaf and mixed forests had Northern Pike with higher [Hg].

In Chapter 3, my goal was to understand causes of among-lake variability in fish growth rates in the study area. I investigated how fish trophic ecology (i.e., trophic level and foraging area) and lake-catchment interactions influence growth rates in Northern Pike. Growth was slower in lakes that were subject to greater inferred catchment influence, which were lakes located at lower-elevation and in relatively larger catchments with proportionally greater forest cover. Northern Pike in lakes subject to greater inferred catchment influence also had more depleted carbon stable isotope ratios. As carbon stable isotope ratios in benthic invertebrates were also more depleted in lakes that were subject to greater inferred catchment influence, slower Northern Pike growth in lakes with greater catchment influence may reflect the fact that terrestrially derived organic matter has lower nutritional value and less bioaccessibility relative to internally derived organic matter.

In Chapter 4, I investigated biomagnification of Hg through lake food webs in the study area. I examined how estimates of Hg biomagnification rates can be affected by the species of mercury (total Hg vs methyl Hg) that is measured in fish and used in biomagnification models. I also investigated relationships between rates of Hg biomagnification and [Hg] in resident fish, and how rates of Hg biomagnification are related to ecosystem characteristics, including in-lake and in-catchment variables. Results showed that uncertainty around estimates of Hg biomagnification rate was lower when estimated using measured concentrations of MeHg, the biomagnifying form of Hg. Rates of Hg biomagnification were positively but not significantly related to [Hg] in resident fish used for subsistence. Rates of Hg biomagnification were on average higher than previously reported for lakes at lower latitudes, and were higher in lakes surrounded by catchments with proportionally greater forest cover.

In Chapter 5, I synthesized findings that were discussed in Chapters 2 through 4. Together, results from this thesis show that mercury levels in resident fish are influenced by biological processes (growth rates in fish, Hg concentrations in primary consumers, and biomagnification of Hg through food webs), that are, in turn, influenced by lake-catchment interactions (higher concentrations of DOC and both total and methyl Hg in

lakes located in lower-elevation catchments with relatively larger size and proportionally more forest cover). Given that catchment physical attributes (elevation, relative size, and proportional forest cover) appeared to result in changes in abiotic and biotic ecosystem compartments in downstream lakes and ultimately influenced fish [Hg], findings provide invaluable direction for prioritizing lakes for monitoring and management, especially because physical characteristics of lakes and catchments can be sensed remotely. Ongoing monitoring and assessment of fish [Hg] are necessary in northern lakes due to profound effects of climate warming on northern latitudes and the heavy reliance of northern communities on wild-caught freshwater fish. This thesis reveals insights that address knowledge gaps regarding causes of among-lake variability in fish [Hg] in the Dehcho Region and improve the current and general understanding of fish [Hg] controls in subarctic lakes.

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Last but not least, I thank my family, especially mom and dad, for their genuine love and unconditional support. I hope that someday I will finally be able to do something for you — it will be incomparable to what you have done for me, but it will make me feel better.

Dedication

تقدیم به پدرم و مادرم؛

باشد که ماحصل تلاشم، نسیم گونه، غبار خستگیتان را بزداید.

To Mom and Dad;

I hope this would break your routine and relieve you, like a morning breeze.

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Chapter 1

General Introduction

1.1 Study Background

Freshwater fish comprise $\sim$40% (c. $15,000$) of all fish species (Helfman et al., 2009), and provide a myriad of ecosystem services, including nutrient cycling, resource connectivity, and provisions to higher trophic levels, including humans (Mota et al., 2014; Weeratunge et al., 2014; Lynch et al., 2016). Only about 10% of the global freshwater fish harvest is destined for non-food purposes (e.g., production of fishmeal and fish oil), and the remaining $\sim$90% is used for human consumption, which highlights the importance of freshwater fish to global food production (Welcomme et al., 2010; FAO, 2020).

Aside from food security, freshwater fish provide economic, recreational, social, and cultural benefits worldwide (see Lynch et al., 2016). Occupationally, freshwater fisheries directly support over 21 million people, and an additional 36 million if post-harvest activities are considered (FAO, 2014; FAO and WFC, 2018). The recreational value (e.g., diving, snorkeling, boating, tourism, restaurants, hotels, etc.) of freshwater fisheries estimated using non-market valuation methods, such as travel cost, contingent valuation, and unit day values (see U.S. ACE, 2000; Poe et al., 2013), outweighs subsistence and commercial values in some developed countries (FAO, 2010). To many, freshwater fish provide an invaluable sense of community identity and occupational attachment (Weeratunge et al., 2014; Lynch et al., 2016; Castañeda et al., 2020).

Compared to marine fish, freshwater fish contribute a proportionally higher biomass to global fisheries and are claimed to have a proportionally greater influence on human livelihood (Lynch et al., 2016; FAO, 2020). Setting an all-time record and reflecting an increase of 4.5% compared to the late 1990s, harvests from freshwater accounted for 12.5% of global fish catch in 2018 (96.4 mt) (FAO, 2020). This is a substantial contribution given that only ~2.5% of the planet's water is freshwater, of which $< 0.3\%$ (~0.01% of all water, fresh and salt) is liquid surface water (Mittermeier et al., 2010). Further, figures of freshwater fish harvests are likely underestimated as small-scale fisheries (i.e., subsistence and local trade) are often unrecorded or underreported (Welcomme et al., 2010). Although direct estimates of fish catches from lakes are not available, a global simulation of lakes greater than 0.1 km^2 estimated an annual yield of 8.9 mt in 2011 (Deines et al., 2017), when total freshwater fish capture was 11.5 mt (FAO, 2012). Lakes thus provided more than three-quarters of the total freshwater fish catch in 2011, which illustrates the importance of lakes to global fisheries and livelihood.

Lake fish and fisheries are integral to the well-being of Canadian communities, especially in the north. A global geo-statistical study on lakes (waterbodies > 0.1 km^2) showed that 62% (c. $879,800$) of the world's lakes are in Canada, covering 9% of the country's vast surface area (~10 million km^2) that spans a large latitudinal gradient, from 42 to 83 °N (Messager et al., 2016). Due to limited infrastructure and resource availability at higher latitudes of Canada, northern residents rely more heavily on wild-harvested freshwater fish than southern residents (see Koch, 2021, and references therein). Among the three northern territories of Canada, Northwest Territories (NT) is the most populated, with ~$45,000$ people (Statistics Canada, 2017). The territory is home to a vast number of lakes, and catch statistics released annually by Fisheries and Oceans Canada show that

628 t of freshwater fish were commercially landed in NT in 2018 (DFO, 2018). The relative ratio of freshwater fish harvest to human population thus appears to be about 14 t per 1000 people in NT. This figure is about 20 times greater than that of, for example, Ontario (the most populated Canadian province, where the same relative ratio of freshwater fish harvest to human population is about 0.71 t per 1000 people), underlying the importance of lake fish and fisheries in NT.

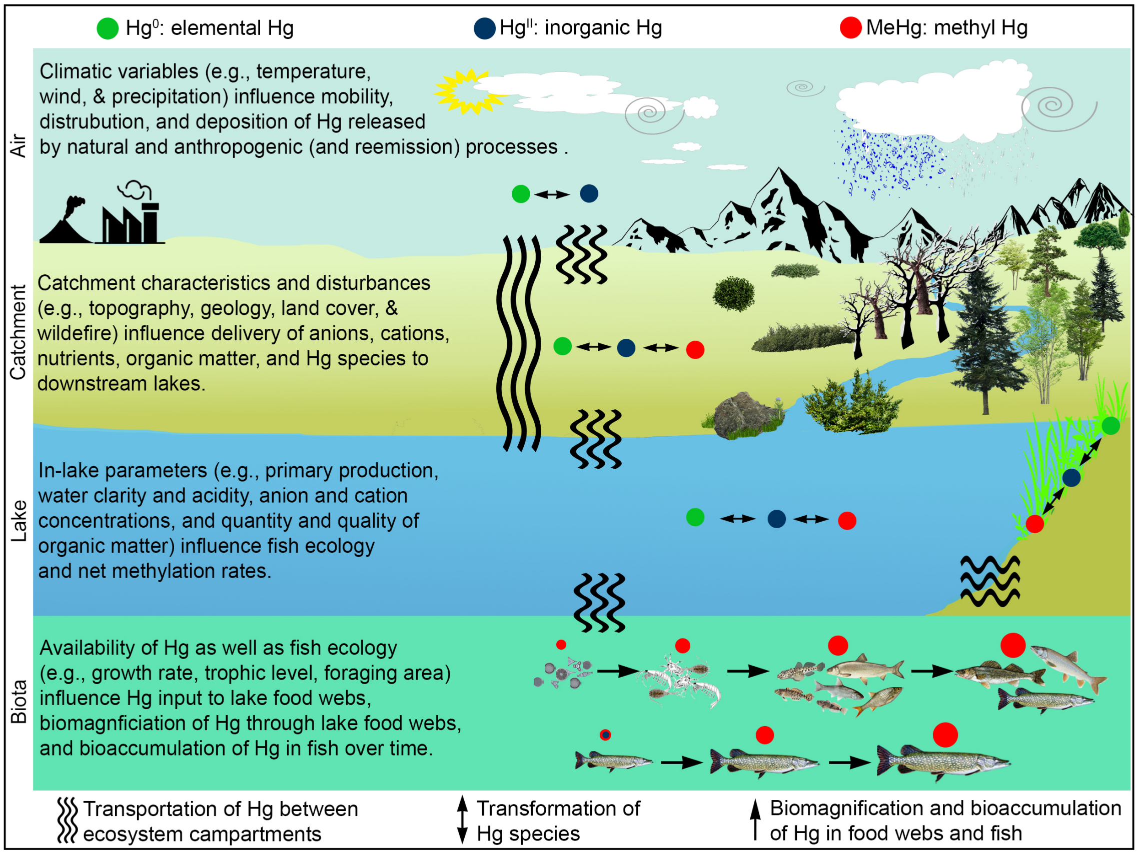
Commercial, subsistence, and recreational fisheries all play an important role in NT. The commercial fishery, which is estimated at $1,000$ t (equivalent to $1.5 million) per year in NT, is believed to occur mostly ($\sim 90\%$) in Great Slave Lake (dating back to 1940s), with the remaining 10% occurring on relatively smaller lakes and the Mackenzie Delta (Kennedy, 1962; Stewart and Low, 2000; DFO, 2010a; Rompkey and Patterson, 2010). Although commercial harvests have generally decreased over time due to overexploitation and lack of management, fish stocks appear to have stabilized following the implementation of management measures by Fisheries and Ocean Canada, such as catch quotas and suspension of fisheries (Keleher, 1972; NWT CIMP and Audit Working Group, 2009; Golder Associates, 2010). While over 20 freshwater fish species (in more than 300 stocks) are harvested throughout NT, Lake Whitefish (*Coregonus clupeaformis*), Lake Trout (*Salvelinus namaycush*), Northern Pike (*Esox lucius*), and Walleye (*Sander vitreus*) are the only species that appear in freshwater landing statistics for the territory (DFO, 1990, 2018).

Recreational fisheries are estimated to generate more than $7 million per year from non-resident anglers in NT (St-Louis, 2006). Although challenging access (most lakes are accessible only by floatplanes in summer and snowmobile in winter) has been a limiting factor for recreational fishing, many operators provide tourism-based fishing activities that include accommodations and guides (St-Louis, 2006; Golder Associates, 2010). Freshwater fish have also provided sustenance for Indigenous people of the territory for millennia. More than half (50.7%) of the NT population is Indigenous, of whom 63.2% are First Nations, 16.3% are Métis, and 19.6% are Inuit (Statistics Canada, 2017). Reports of harvesting activities show that 37% of NT residents participate in subsistence harvesting, such as hunting and fishing (Statistics Canada, 2017), suggesting heavy reliance on wild meat and fish (Golder Associates, 2010). Rich in protein, omega-3 polyunsaturated fatty acids, and other essential micronutrients, freshwater fish are important food alternatives to store-bought items that are limited and costly in the remote north (Kuhnlein and Receveur, 1996; WHO and FAO, 2010; Pirkle et al., 2016). Further, wild-caught freshwater fish are critical components of economy, society, autonomy, sovereignty, and cultural continuity of Indigenous communities (Fieldhouse and Thompson, 2012). Local communities within NT are concerned, however, about mercury-related safety of wild-caught freshwater fish.

Listed among the top ten (groups of) chemicals of major public health concern, mercury (Hg) is a potentially toxic element that can be released into the environment from both natural and anthropogenic sources (WHO, 2017). As a naturally occurring element, Hg can be released from the lithosphere into the atmosphere, hydrosphere, and biosphere through natural processes, such as volcanic activity and weathering of rocks (Selin, 2009; Pirrone et al., 2010). As the only metal that is liquid at room temperature, has high density, and amalgamates easily with other metals, Hg has also been utilized by humans since ancient times (Selin, 2009; Pirrone et al., 2010). Anthropogenic activities are an important source of Hg to the environment, with major sources including artisanal and small-scale gold mining (38%), coal combustion (21%), metal production (15%), and cement production (11%) (AMAP/UN Environment, 2019; Environment, 2019). The smaller sources include waste incineration, oil refinement, and production of an array of electric (e.g., batteries and lamps), cosmetic (e.g., soaps and creams), and medical (e.g., dental amalgams and blood pressure devices) goods (AMAP/UN Environment, 2019; Environment, 2019).

The predominant ($>$ 90%) form of Hg that is released into the atmosphere is in the form of elemental Hg (Hg^0), which can undergo a complex cycling (Figure 1.1). Atmospheric Hg^0 can be exchanged between the atmosphere and terrestrial environments (e.g., vegetation and soil) and between the atmosphere and aquatic environments, although the latter is of less importance in freshwaters (see Selin, 2009; Obrist et al., 2018; Branfireun et al., 2020). In the air, soil, and water, Hg^0 can be oxidized to form inorganic Hg (Hg^{II}) by ozone, hydroxyls, halogens, particulate minerals, and humic substances (see Selin, 2009; Obrist et al., 2018; Branfireun et al., 2020; Saiz-Lopez et al., 2020). If not reduced to reform Hg^0 and if still resident in the atmosphere, resultant Hg^{II}, which is a more water soluble and particle-reactive species, partitions into aerosols and clouds, where it can be deposited to terrestrial and aquatic environments through wet and/or dry deposition (see Selin, 2009; Obrist et al., 2018; Branfireun et al., 2020; Saiz-Lopez et al., 2020). Oxidation of Hg^0 to Hg^{II} is considered a key step in removing Hg^0 from the atmosphere, and while deposition of oxidized Hg^{II} is dominant in oceans, more recent studies show that deposition and/or uptake of Hg^0 (e.g., on or by terrestrial vegetation) can be an important source of Hg to terrestrial environments (see Selin, 2009; Obrist et al., 2018; Branfireun et al., 2020).

In both aquatic and terrestrial environments, Hg^{II} can be methylated to form methyl Hg (MeHg), which is the organic and more toxic form of Hg species (Figure 1.1). Pro-



Climatic variables (e.g., temperature, wind, & precipitation) influence mobility, distrubution, and deposition of Hg released by natural and anthropogenic (and reemission) processes .

Catchment characteristics and disturbances (e.g., topography, geology, land cover, & wildefire) influence delivery of anions, cations, nutrients, organic matter, and Hg species to downstream lakes.

In-lake parameters (e.g., primary production, water clarity and acidity, anion and cation concentrations, and quantity and quality of organic matter) influence fish ecology and net methylation rates.

Availability of Hg as well as fish ecology (e.g., growth rate, trophic level, foraging area) influence Hg input to lake food webs, biomagnficiation of Hg through lake food webs, and bioaccumulation of Hg in fish over time.

Figure 1.1. A schematic diagram of known factors and processes that can influence mercury (Hg) cycling in the environment, and ultimately, Hg concentrations in fish.

duction of MeHg can occur biotically via aerobic and anaerobic microbes in, for example, periphyton and surface sediments, and/or abiotically via light in the water column (see Paranjape and Hall, 2017). Biotic methylation driven by facultative anaerobes (e.g., iron-reducing bacteria, sulfate-reducing bacteria, and methanogenic archaea) in surface sediments is thought to have the greatest contribution to the production of MeHg in most lakes (see Ullrich et al., 2001; Paranjape and Hall, 2017), although terrestrial and aquatic vegetation have recently been shown to contribute more to the production of MeHg in lakes than what was previously believed (see Obrist et al., 2018; Burke et al., 2020; Branfireun et al., 2020). The relative contribution of MeHg sources depends on characteristics of the system and factors that affect the activity of methylators, including pH, temperature, depth of hypolimnion, supply of sulphate and organic matter, and presence of electron donors and receptors (see Ullrich et al., 2001; Paranjape and Hall, 2017; Obrist et al., 2018; Branfireun et al., 2020). MeHg can be demethylated to re-form Hg^{II} through biotic and/or abiotic processes, although photo-demethylation of MeHg in water column is thought to be the dominant demethylation process in most lakes (see Ullrich et al., 2001; Paranjape and Hall, 2017). The net outcome of methylation and demethylation processes determines the amount of MeHg that is available for biological uptake.

Assimilation of MeHg by primary producers introduces MeHg into aquatic food webs, where MeHg can be transferred, through dietary interactions, to biota at higher trophic levels, including fish (Figure 1.1). Primary producers can also take up Hg^{II} and methylation can occur in the gastrointestinal content of biota (Rudd et al., 1980; Mason et al., 1996; Watras et al., 1998; Martín-Doimeadios et al., 2017). Through a process known as "bloom dilution", the density of primary producers can greatly impact the amount of MeHg that is incorporated into the base of the aquatic food webs (Pickhardt et al., 2002). As algal biomass increases, the uptake of MeHg per algal cell decreases, which can subsequently reduce trophic transfer of MeHg to biota at higher trophic levels (Pickhardt et al., 2002). Due to rapid accumulation relative to excretion in tissue proteins, MeHg bioaccumulates in organismal tissue over time (Trudel and Rasmussen, 2006) and biomagnifies with each trophic transfer in aquatic food webs (see Mason et al., 1996; Ward et al., 2010a; Kidd et al., 2011). As a result, the proportion of MeHg to total Hg (%MeHg) can increase from $\sim$15% in primary producers to as much as 95% in predatory fish (Bloom, 1992; Watras et al., 1998; Lescord et al., 2018). While dermal and oral routes as well as inhalation are common exposure pathways of Hg^0 and Hg^{II} to humans, consumption of fish is, globally, the primary exposure pathway of MeHg to humans, although rice consumption could also be a major MeHg exposure pathway for some populations (see Eagles-Smith et al., 2018).

Exposure to Hg can pose serious health hazards to fish, wildlife, and humans. In humans, detrimental health effects of fish-derived Hg have attracted public attention since 1956, when an infant in Minamata, a small town on Kyushu Island in southern Japan, was diagnosed with unusual neurological disorders; this was followed by observations of similar symptoms in more than $2,000$ people (Harada, 1995; Hachiya, 2006; Kessler, 2013). Referred to as "Minamata Disease", severe manifestations of the disease resulted in sensory disturbance, dysarthria, hearing impairment, tremors, and/or death, and in mild cases caused reductions in fine motor skills, stumbling, unsteadiness, insomnia, headache, and/or memory loss (Harada, 1995; Hachiya, 2006; Kessler, 2013). The devastating incident was found to be caused by a poisoning from consumption of large quantities of fish and shellfish that were contaminated with MeHg that had been discharged in wastewater by a local chemical factory (Harada, 1995; Hachiya, 2006; Kessler, 2013). Quite a few studies have shown that exposure to Hg can adversely affect endocrine, immune, digestive, cardiovascular, and neurological systems in humans, and many studies have released evidence regarding adverse effects of Hg on immune, neurological, and reproductive systems in fish and fish-eating wildlife (see Cohen, 2003; Driscoll et al., 2013; Rice et al., 2014).

Much effort has been made to reduce anthropogenic Hg emissions, yet Hg continues to be a global health concern. The Minamata Convention on Mercury, for example, is an international treaty designed to reduce anthropogenic emissions of Hg by providing manufacturing, importing, and exporting controls over an array of commercial products as well as by setting priorities and guidance regarding coal utilization (see Selin et al., 2018). While evidence suggests that declines in anthropogenic Hg emissions by North America and Europe are largely offset by increases in Asia, a recent review of global emissions showed a 30% decline in Hg emissions from anthropogenic sources and a 20% decline in the total Hg emission from 1990 to 2010 (Zhang et al., 2016). Only ∼45% of annual Hg emissions to the atmosphere originate from present-day anthropogenic (39%) and natural (6%) sources, however (AMAP/UN Environment, 2019). More than half (55%) of annual Hg emissions to the atmosphere are from mobilization (i.e., reemissions) of previously-released-and-deposited Hg (AMAP/UN Environment, 2019). This so called "legacy" Hg from antecedent or historical releases has the potential to continue cycling in the environment and cause effects on biota (Pollman and Engstrom, 2020).

From emission to accumulation in fish, Hg cycling involves numerous biogeochemical and ecological processes, making concentrations of Hg ([Hg]) in fish dependent on a wide assortment of factors. Individual- and species-specific characteristics of fish are important determinants of [Hg] in fish (Figure 1.1). Accumulation of Hg in fish muscle tissues can be influenced by a processes known as "growth dilution", where fast-growing individuals tend to accumulate less Hg for a given prey intake than their slow-growing counterparts (e.g., Karimi et al., 2007; Sharma et al., 2008). In addition to growth dilution causing a negative relationship between fish [Hg] and growth rate/efficiency, [Hg] in prey items are often positively related to fish [Hg] because Hg in fish is mostly diet-derived (Hall et al., 1997; Chen and Folt, 2005). Fish age, size, and feeding ecology can also influence fish [Hg]. Due to bioaccumulation, larger and older fish often have higher [Hg] than younger and smaller individuals (e.g., Swanson et al., 2011; Depew et al., 2013). Because of differences in basal [Hg] and rates of biomagnification between profundal (offshore) and littoral (nearshore) food webs, fish that are connected to pelagic food webs and that occupy higher trophic levels in the food web often have higher [Hg] than their counterparts that rely on littoral food webs and prey items from lower trophic levels (e.g., Kidd et al., 1995; Power et al., 2002; Karimi et al., 2016).

In lake ecosystems, fish [Hg] can further be influenced by in-lake environmental conditions (Figure 1.1). Fish generally tend to have higher [Hg] in lakes with higher temperature, and higher concentrations of sulphate, and/or organic matter (e.g., dissolved organic carbon (DOC)), and in lakes with lower pH and/or lower concentrations of dissolved oxygen (e.g., Wyn et al., 2009; Thomas et al., 2020; Burke et al., 2020), as these conditions can promote methylation of Hg to MeHg and enhance availability of MeHg for biological uptake (see Ullrich et al., 2001; Paranjape and Hall, 2017). Another well-studied in-lake determinant of [Hg] in fish is primary productivity. High primary productivity can reduce [Hg] in fish through a) bloom dilution and associated declines in uptake rates of Hg into the bottom of the food web that can decrease trophic transfer and biomagnification of Hg (e.g., Larsson et al., 1992; Pickhardt et al., 2002; Kozak et al., 2021) and/or b) growth dilution associated with higher fish growth/efficiency in more productive systems due to greater prey abundance and availability (e.g., Hammar et al., 1933; Larsson et al., 1992; Thomas et al., 2020). It has also been shown that higher primary productivity can increase [Hg] in aquatic food webs by promoting anoxic conditions and increasing availability of Hg and organic matter for methylating microbes, which increases net methyl Hg production (see Langston and Bebianno, 1998; Mauro et al., 2002; Bravo et al., 2017; Eagles-Smith et al., 2018).

As lakes are not closed systems, catchment characteristics and processes influence fish [Hg] in lakes through various processes and mechanisms (Figure 1.1). Type of vegetative covers, as well as disturbances, including permafrost thaw and fire, can affect inputs of both inorganic and methyl Hg to downstream lakes (e.g., Kainz and Lucotte, 2006; Demers et al., 2007; Eklöf et al., 2016; Denkenberger et al., 2020), and ultimately Hg loading to lake food webs and [Hg] in resident fish (e.g., Eagles-Smith et al., 2016; Sumner et al., 2020; Thomas et al., 2020). Catchment topography, relative size, land cover composition, wildfire, and permafrost thaw can also influence delivery of organic matter (often measured as DOC), nutrients (e.g., phosphorous), and ions (e.g., sulfur) to downstream lakes (e.g., Sobek et al., 2007; Williams et al., 2010; Rydberg et al., 2010; Dranga et al., 2017; Kritzberg et al., 2020), which can, in turn, alter processes of methylation, uptake, bioaccumulation, and biomagnification of Hg, and ultimately, [Hg] in fish (e.g., Hammar et al., 1933; Pickhardt et al., 2002; Ranåker et al., 2014; Benoît et al., 2016; Eagles-Smith et al., 2016; van Dorst et al., 2019; Burke et al., 2020; Thomas et al., 2020).

Atmospheric and climatic factors can also influence fish [Hg] in lakes largely because they impact distribution, mobility, and deposition of Hg (Figure 1.1). Atmospheric Hg^0 has a long residence time (0.5-2 years) and can be transported across the globe via atmospheric processes, such as wind currents (see Selin, 2009). Changes in wind patterns and fluctuations in air circulation can affect transport of atmospheric Hg from distant locations (see Selin et al., 2007). As incorporation of Hg^{II} into rain/snow is a major source of Hg deposition in aquatic and terrestrial environments, increased rates of precipitation generally result in higher inputs of Hg to both aquatic and terrestrial environments, ultimately leading to higher [Hg] in fish (e.g., Lamborg et al., 2002; Pacyna et al., 2006). Temperature can also influence atmospheric transport and deposition of Hg by influencing wind currents and precipitation (e.g., Grimalt et al., 2010; Pack et al., 2014), although temperature may also influence primary production and availability of MeHg in aquatic environments by affecting depth of epilimnion and anoxic conditions (e.g., Hecky et al., 1991; Bloom, 1992; Avramescu et al., 2011), as well as Hg accumulation in fish via effects on metabolic and growth rates (e.g., Brandt et al., 2011).

The complexity of interactions that can take place among diverse biogeochemical and ecological factors to influence Hg cycling makes fish [Hg] widely variable, and challenging to understand and predict, especially in the current era of rapidly changing environmental conditions. Studies have shown that climate- and human-induced changes to lake catchments accelerate permafrost thaw and disturb fire regimes and hydrology (Prowse et al., 2006; Hugelius et al., 2020), which can, in turn, influence lake-catchment interactions, energy dynamics, and contaminant transfer in lakes (Rydberg et al., 2010; Karlsson et al., 2015), and ultimately, [Hg] in fish (e.g., Carrie et al., 2010; Sumner et al., 2020; Thomas et al., 2020; Kozak et al., 2021). In lake ecosystems, [Hg] in fish can vary among systems and species, and among individuals of the same population (e.g., Evans et al., 2005; Lockhart et al., 2005; Depew et al., 2013). Although drivers of among-lake variability in fish [Hg] have been widely studied, patterns cannot be predicted or generalized across systems and species because of the variability in the relative importance of predictor variables (see Chételat et al., 2015; Eagles-Smith et al., 2018). Further whole-ecosystem studies, especially in understudied systems, are thus warranted.

The Dehcho Region of the NT hosts dozens of lakes among which fish [Hg] vary substantially due to largely unknown mechanisms. Fish [Hg] may vary by up to seven times among lakes in the Dehcho Region, and large individuals of apex predators (e.g., Northern Pike) can have [Hg] that exceed the Commercial Sale Fish Guideline of 0.50 mg/kg wet weight in some systems, especially in smaller lakes (Evans et al., 2005; Lockhart et al., 2005; Laird et al., 2018). Causes of among-lake variability of [Hg] in fish have not been comprehensively investigated in the region, and the lack of mechanistic understanding regarding causes of among-lake variability of [Hg] in fish from the Dehcho Region has raised concerns over food safety and security by both local communities and partner scientists (Evans et al., 2005; Lockhart et al., 2005; Laird et al., 2018). To ensure food safety, lake- and species-specific Hg-related fish consumption advisories were established and publicized in the area.

In the Dehcho Region, fish consumption advisories regarding Hg are developed based on consumption frequency (e.g., servings per week or month) of a given species (and size) from a given lake for four different groups (pregnant and breastfeeding women, children 1-5 years old, children 5-11 years old, and the general population). Consumption advisories can be found on notice boards near lakes, as well as on maps and downloadable documents from the portal of the Government of Northwest Territories (see Health and Social Services, 2021). While Hg-related advisories regarding fish consumption are aimed to ensure safety of consumers against potential harmful effects of fish-derived Hg, they can sometimes be overconservative. Nutritional benefits associated with consumption of wild-caught fish can hardly be found in energy-dense, store-bought food items (WHO and FAO, 2010; Hellberg et al., 2012; Hosomi et al., 2012; Seabert et al., 2014). There is also evidence suggesting that omega-3 fatty acids can ameliorate toxicity and/or concentration of Hg in fish, wildlife, and humans (e.g., Nøstbakken et al., 2012; Choi et al., 2014; Laird et al., 2018). Developing and establishing fish consumption advisories solely on assessment of Hg exposure can thus result in overly restrictive advisories that deprive humans of health benefits associated with fish consumption (see Arnold et al., 2005; Hellberg et al., 2012).

Understanding controls of [Hg] in fish is a fundamental component of developing more comprehensive risk-benefit assessments that could eventually lead to more informed and balanced consumption guidance. The sources of among-lake variation of fish [Hg] are of concern for Dehcho First Nations that rely on wild-caught fish as a main subsistence food source. My PhD thus aims to address concerns and knowledge gaps regarding causes of variability in fish [Hg] among lakes in the remote Dehcho Region, and contribute to the current and general understanding of fish [Hg] controls in subarctic lakes.

1.2 Study Area

The Dehcho Region is located along the Mackenzie River Basin in southwest NT (Figure 1.2). The region covers a vast area of $\sim 60,000$ km^2 ($60°25'$ to $63°11'$ N and $117°24'$ to $121°35'$ W) in the Canadian subarctic, where day length varies from $\sim$5 to $\sim$20 h during a 365-day cycle. According to decadal climate data from the Fort Simpson Airport (Figure 1.2), daily average temperature varies from -24 to 17.4 °C (1981-2010 yearly average of -2.8 °C) and daily average total precipitation varies from 151 to 610 mm (1981-2010 yearly average of 388 mm) in the study area (CCN, 2021).

Lakes in Dehcho Region support important commercial and subsistence fisheries, especially the larger Tathlina and Kakisa lakes. Commercial fishing in the remote Tathlina and Kakisa lakes became feasible with the construction of an all-weather road in the area in the 1940s, which led to the first official investigation of the lakes and resident fish (Kennedy, 1962). Commercial fisheries in Tathlina and Kakisa lakes began in the early 1950s with a primary focus on Walleye, although other species (e.g., Lake Whitefish and Northern Pike) were also common in landings (Stewart and Low, 2000). Commercial fisheries have not been sustainable in either lake. During the early years when there was a 20 t quota in Tathlina Lake, catch fluctuated below the quota; the population eventually

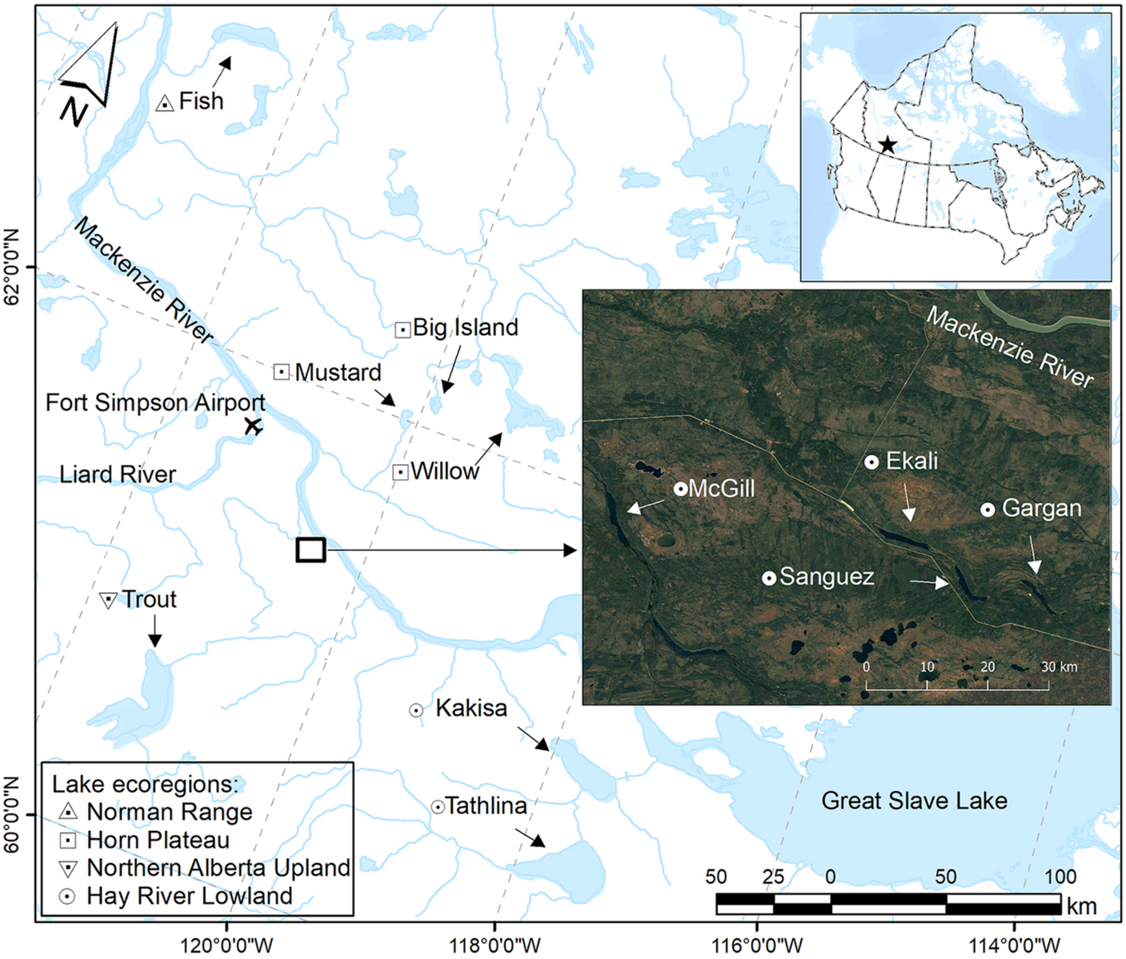


Figure 1.2. Location of study lakes in the Dehcho Region, NT, Canada (base data source: Northwest Territories Centre for Geomatics and Google Earth v7.1.8).

collapsed in 2001, which led to a closure of the fishery for several years and a much more conservative quota of 5 t in 2008 (DFO, 2010a,b; Gallagher et al., 2011; Stewart et al., 2016). Although the catch has generally been more stable in Kakisa Lake, the quota of 20 t has not been met since the 1990s (Stewart and Low, 2000; NWT CIMP and Audit Working Group, 2009). Fisheries in both Tathlina and Kakisa lakes are now monitored and managed by Fisheries and Ocean Canada and appear to be relatively more stable than in the past (Golder Associates, 2010).

Subsistence harvests of freshwater fish in the Dehcho Region are also important, especially for local communities. Dehcho Region is home to First Nations communities who comprise a population of just over $3,000$ people (Statistics Canada, 2017). Written documents dates subsistence harvests of freshwater fish by Dehcho Frist Nations back to mid-1900s (Kennedy, 1962; Stewart and Low, 2000), and local First Nations indicate that they have harvested lakes in Dehcho Region for subsistence since time immemorial. Multiple generations emphasize the importance of wild-caught freshwater fish to their livelihoods and well-being; they share stories that have been passed on for generations and reveal facts about the critical vitality of locally-harvested freshwater fish for the diet, economy, society, independence, and identity of Dehcho First Nations. They also raise serious, Hg-related, safety concerns over wild-caught freshwater fish, which are relied upon as a critical and nutritious subsistence food source.

Studies have shown substantial among-lake variations in fish [Hg] in the Dehcho Region (Evans et al., 2005; Lockhart et al., 2005; Depew et al., 2013; Laird et al., 2018). Most fish in most lakes that have been investigated to date have [Hg] below the commercial sale health guideline (e.g., 0.50 mg/kg wet weight for Commercial Sale of Fish Guideline of Health Canada). For example, Lake Whitefish, which are common in the area and are of high interest for human consumption, have [Hg] well below the 0.50 mg/kg wet weight guideline ((Evans et al., 2005; Lockhart et al., 2005; Depew et al., 2013; Laird et al., 2018). Conversely, some apex predatory species in smaller lakes tend to have relatively higher [Hg]. For example, Northern Pike, another frequently caught species, are sometimes found with [Hg] that exceed the 0.5 mg/kg wet weight guideline (Evans et al., 2005; Lockhart et al., 2005; Depew et al., 2013; Laird et al., 2018).

Little is known about drivers of among-lake variability in fish [Hg] in the Dehcho

Region. In a broadscale study of fish [Hg] in the Canadian north, Evans et al. (2005) noted that [Hg] in fish tended to increase with fish size, age, and trophic level in Dehcho Region, yet stated that the nature of these relationships varies among lakes. Evans et al. (2005) also mentioned that fish [Hg] tended to be higher in smaller compared to larger lakes, likely because of greater input of MeHg from catchments to smaller lakes and/or higher summer epilimnion temperatures and associated promotion of net methylation rates in smaller lakes. Although it was not directly investigated, Laird et al. (2018) highlighted that their ongoing work and growing dataset suggest a complex interplay among fish [Hg] and fish, lake, and catchment characteristics. The current gaps in our understanding of controls of fish [Hg] in the Dehcho Region are likely due to the remote location and associated difficulties in conducting research. Sparse available information suggests that numerous factors and processes that act at different biological and spatial scales interactively control [Hg] in fish in the Dehcho Region, a hypothesis that fits the environmental diversity of lakes in the area.

My PhD research focused on eleven lakes in the Dehcho Region that are of subsistence value for local First Nations (Figure 1.2). The study lakes are all distributed across the Taiga Plains ecozone (ELC, 2017) and span four different ecoregions, including Horn Plateau, Northern Alberta Upland, Hay River Lowland, and Norman Range (Figure 1.2). The study lakes vary substantially in surface area (1-565 km^2), are surrounded by heterogenous catchments (e.g., size: 73-15,808 km^2, altitude: 284-783 m asl, wetland cover: 7-22%, and burned area: 0-23%), and thus vary considerably in water chemistry (e.g., chlorophyll-a: 2.24-15.53 μg/L, DOC: 9.76-21.30 mg/L, water clarity: 0.7-4.0 m, water [Hg]: 0.53-1.64 ng/L, and sediment [Hg]: 45.88-155.68 ng/g). The diversity in lake and catchment characteristics sets the stage for complex interactions among numerous factors and processes that can influence [Hg] in resident fish. Fish species present in the region include Northern Pike, Walleye, Lake Whitefish, Lake Trout, Longnose Sucker (*Catostomus catostomus*), White Sucker (*Catostomus commersoni*), Ninespine Stickleback (*Pungitius pungitius*), Burbot (*Lota lota*), and Cisco (*Coregonus artedi*), although fish diversity varies among lakes, and quantitative relative abundance or presence/absence surveys have not been completed in the area.

1.3 Study Objectives

The principal objective of my PhD research is to investigate controls of [Hg] in subsistence fish species in the remote Dehcho Region, and to understand underlying causes of the long-documented among-lake variability of fish [Hg] in the area. My research goal is addressed in three data chapters.

In my first data chapter (thesis Chapter 2), I report on a whole-ecosystem study that comprehensively examined drivers of among-lake variability of [Hg] in fish by focusing on Northern Pike, a predatory fish species with widespread commercial and subsistence importance throughout the northern hemisphere; [Hg] in Northern Pike have been shown to vary substantially among lakes in the Dehcho Region, and this species is the only one caught in all studied lakes. The specific objectives of this chapter were to:

i. Elucidate drivers of spatial variability in [Hg] in Northern Pike in the Dehcho Region by relating [Hg] in Northern Pike to fish, lake, and catchment characteristics; and,

ii. Provide a plausible mechanistic understanding of how interacting processes at scales ranging from individual organisms to whole catchments influence fish [Hg].

In addition to revealing complex interactions among variables and ecosystem compartments that control fish [Hg] in subarctic lakes, I showed that more than four-fifths of among-lake variability in Northern Pike [Hg] is driven by growth rates in fish and [MeHg] in primary consumers (which represent [Hg] at lower trophic levels in food webs). In my next two data chapters, I thus investigated growth rates in Northern Pike and biomagnification of Hg through lake food webs.

In my second data chapter (thesis Chapter 3), I comprehensively investigated among-lake variability in growth rate of Northern Pike in the Dehcho Region. The specific objectives of this chapter were to:

i. Investigate effects of lake-catchment interactions on growth rate in Northern Pike; and,
ii. Explore relationships between feeding ecology (i.e., trophic levels and foraging area) and growth rate in Northern Pike.

I found that growth rates in Northern Pike were inversely related to inferred catchment influence, which, given a relationship between inferred catchment influence and carbon isotope ratios in Northern Pike, likely reflect effects of allochthonous inputs on quality of basal organic matter that propagates through food webs.

In my third data chapter (thesis Chapter 4), I investigated biomagnification of Hg through lake food webs in the Dehcho Region. The specific objectives of this chapter were to:

i. Explore if using concentrations of total Hg versus methyl Hg in fish in biomagnification models influenced estimates of Hg biomagnification rates, and,

ii. Investigate if lake and catchment characteristics affect rates of Hg biomagnification through lake food webs.

I found that estimates of Hg biomagnification rates were more precise, and significantly lower, when estimated using measured concentrations of MeHg (the biomagnifying form of Hg). I also found that rates of Hg biomagnification were higher in lakes surrounded by catchments with proportionally more forest cover, which results in higher concentrations of DOC and Hg (both total and methyl Hg) in downstream lakes.

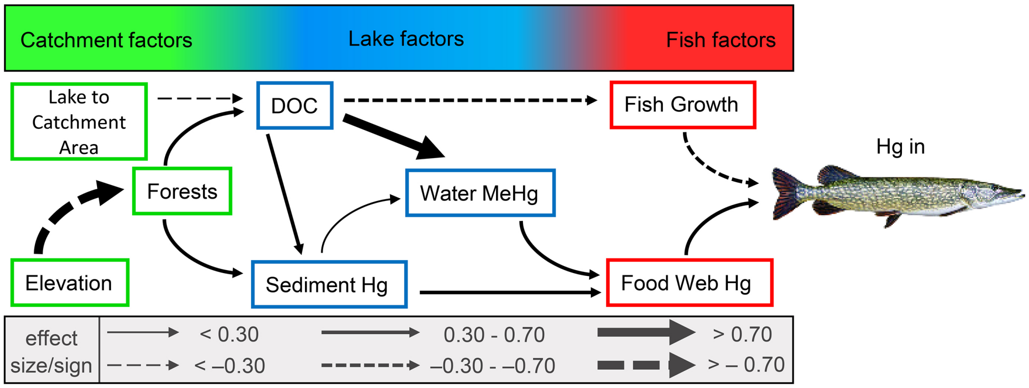
Chapter 2

Understanding among-lake variability of mercury concentrations in Northern Pike (*Esox lucius*): a whole-ecosystem study in subarctic lakes

This chapter is an adapted version of the following published paper[1]:

Moslemi-Aqdam M., Baker L. F., Baltzer J. L., Branfireun B. A.,Evans M. S., Laird B. D., Low G., Low M., and Swanson H. K. 2022. Understanding among-lake variability in mercury concentrations in Northern Pike (*Esox lucius*): a whole-ecosystem study in subarctic lakes. *Science of the Total Environment*, 822, 153430. https://doi.org/10.1016/j.scitotenv.2022.153430

Graphical Abstract



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2.1 Introduction

Mercury (Hg) is a global pollutant. The organic and more toxic form of Hg, methyl Hg (MeHg), can have detrimental health effects on ecosystems, wildlife, and humans (see Driscoll et al., 2013). In lake ecosystems, Hg concentrations ([Hg]) in fish often vary among systems and species (e.g., Evans et al., 2005; Lockhart et al., 2005; Depew et al., 2013). Although many of the factors that affect fish [Hg] are well understood, their relative importance and influence varies with ecological context, making it difficult to generalize and predict variability in fish [Hg], despite a large volume of previous research (see Chételat et al., 2015; Eagles-Smith et al., 2018). The dominant form of Hg in fish tissues is MeHg (Bloom, 1992; Lescord et al., 2018) and, globally, fish consumption is the most important route of Hg exposure in humans (see Sheehan et al., 2014). It is thus crucial to understand and predict how fish [Hg] vary, particularly in remote northern regions, where locally harvested fish are a dietary mainstay and are critical to food security (Pirkle et al., 2016).

Accumulation of Hg in fish is affected by a myriad of factors that influence input of Hg to aquatic systems, net methylation rates, uptake by lower trophic levels, and bioaccumulation and biomagnification rates (see Chételat et al., 2015; Eagles-Smith et al., 2018). For example, primary productivity, oxygen availability, light penetration, pH, and concentrations of dissolved organic carbon (DOC) and sulphate all influence net methylation rates (see Paranjape and Hall, 2017; Branfireun et al., 2020), and thus MeHg concentrations in water, sediment, and biota at the base of food webs (Winfrey and Rudd, 1990; Chen and Folt, 2005; de Klerk et al., 2013). Concentrations of Hg in lower trophic level biota reflect the amount of bioavailable Hg in sediment and water, and rates of bioconcentration, bioaccumulation, and biomagnification (e.g., Pickhardt et al., 2002; Kidd et al., 2012). As Hg in fish is primarily diet-derived (Hall et al., 1997), trophic position, foraging habitat (or carbon source), and [Hg] in diet items are often particularly important variables to consider when seeking to understand variability of [Hg] in fish (e.g., Kidd et al., 1995; Chen and Folt, 2005; Power et al., 2002). Fish [Hg] are also often negatively related to growth efficiency and growth rates, a phenomenon commonly referred to as somatic growth dilution (e.g., Sharma et al., 2008; Thomas et al., 2020). Many of the above variables, including fish trophic level, foraging habitat, and growth rate, as well as [Hg] in lower trophic level biota, are regulated to varying degrees by physicochemical factors in lakes such as nutrient concentrations, primary productivity, water clarity, pH, and dissolved organic matter (DOM) (Pickhardt et al., 2002; Karlsson et al., 2009; Kidd et al., 2012; Ranåker et al., 2014; Benoît et al., 2016; van Dorst et al., 2019). Lake-specific physicochemical conditions can thus influence [Hg] in fish by altering biogeochemical cycling and availability of Hg, as well as uptake, bioaccumulation, and biomagnification in organisms and through food webs.

In-lake physicochemical conditions are influenced by characteristics of lake catchments. Attributes of catchments, including elevation, slope, soil type, vegetation cover (e.g., coniferous vs deciduous forest, grasslands), and disturbance (e.g., forestry, fire frequency) affect loading of both inorganic and methyl Hg to sediments and waters in downstream lakes (e.g., Kainz and Lucotte, 2006; Demers et al., 2007; Eklöf et al., 2016; Denkenberger et al., 2020; Willacker et al., 2020). Catchment area (relative to lake area), elevation, slope, and vegetation/land cover also affect several water chemistry variables (e.g., DOM, water clarity, pH, primary productivity) (e.g., Sobek et al., 2007; Williams et al., 2010; Dranga et al., 2017; Kritzberg et al., 2020) that regulate net rates of in-lake

Hg methylation (see Paranjape and Hall, 2017; Branfireun et al., 2020), rates of Hg uptake and biomagnification (Pickhardt et al., 2002; Kidd et al., 2012), and growth rates and trophic ecology of fish (Ranåker et al., 2014; Benoît et al., 2016; van Dorst et al., 2019). Catchment characteristics thus affect, either directly or indirectly, almost all aspects of Hg cycling in lake ecosystems.

Given that [Hg] in fish reflect complex interactions among biogeochemical and ecological processes that operate at multiple spatial and biological scales, it is perhaps not surprising that a holistic and mechanistic understanding of among-lake variability in fish [Hg] is often elusive, particularly in remote, northern regions where ecosystem-scale data are challenging to obtain. Gaining insights into how ecosystem compartments (e.g., fish populations, food webs, lakes, and catchments) act individually and interactively to affect Hg accumulation has allowed researchers to explain variability of fish [Hg] in more southerly lakes (e.g., Greenfield et al., 2001; Thomas et al., 2020; Willacker et al., 2020). Expanding this approach into remote northern regions, where climate warming is having measurable and profound effects on lake-catchment interactions (e.g., through accelerated permafrost thaw, changes in fire regimes, and hydrology) (Prowse et al., 2006; Hugelius et al., 2020) and where fish are a critically important subsistence food source (Pirkle et al., 2016), will allow more informed predictions of effects of environmental change and its impacts on northern food safety and security.

Using comprehensive data on trophic ecology and growth rates of fish, lake water chemistry, sediment Hg, and catchment characteristics, we investigated among-lake variability of [Hg] in Northern Pike (*Esox lucius*) in subarctic lakes. Our study focused on 11 lakes in the Dehcho Region of Northwest Territories, Canada. Previous studies have shown that [Hg] in Northern Pike vary substantially among lakes in the Dehcho Region;

while Northern Pike have [Hg] below the Commercial Sale of Fish Guideline (0.50 mg/kg wet weight) in most lakes in the area, large individuals of this predatory fish species have elevated [Hg] in some lakes, especially in smaller lakes (Evans et al., 2005; Lockhart et al., 2005; Laird et al., 2018). Consumption of wild-harvested fish, including Northern Pike, provides an array of nutrients (e.g., low-fat protein, minerals, vitamins, and omega-3 polyunsaturated fatty acids) that are essential for human health (WHO and FAO, 2010). A recent study from the Dehcho Region showed a negative relationship between levels of Hg and omega-3 polyunsaturated fatty acids in Northern Pike (Laird et al., 2018). Understanding sources of [Hg] variation in Northern Pike is a priority for Indigenous communities in the region, particularly in the face of climate- and development-induced changes to lands and waters.

Authors of previous research conducted in the Dehcho Region and in broad-scale studies in northern Canada (that included some Dehcho lakes) have stated that [Hg] in resident fish are likely influenced by complex interplay of multiscale factors and processes (Evans et al., 2005; Laird et al., 2018), but underlying mechanisms of [Hg] variability in fish in the region are not fully understood. Our objective was to elucidate drivers of spatial variability in [Hg] in Northern Pike in the Dehcho Region by relating lake-specific, size-standardized [Hg] in Northern Pike to a suite of variables representing fish ecology, water chemistry, and catchment characteristics. We used a combination of linear regression and piecewise structural equation models to address this objective and to explore complex interactions among variables and ecosystem compartments.

2.2 Materials and Methods

2.2.1 Study area

The Dehcho Region is in the Mackenzie River basin in Northwest Territories (NT), Canada (Figure 2.1). Decadal climate data from Fort Simpson Airport indicate a yearly average temperature of -2.8 °C and a yearly average total annual precipitation of 388 mm for the period of 1981 to 2010 in the study area (CCN, 2021). The eleven study lakes span $\sim 60,000$ km^2 ($60°25'$ to $63°11'$ N and $117°24'$ to $121°35'$ W) and four ecoregions (ELC, 2017), including Horn Plateau, Northern Alberta Upland, Hay River Lowland, and Norman Range (Figure 2.1). Surface area of the study lakes ranges from 1.1 to 565.3 km^2, catchment areas range from 73.2 to $15,808.8$ km^2, mean elevation ranges from 274.0 to

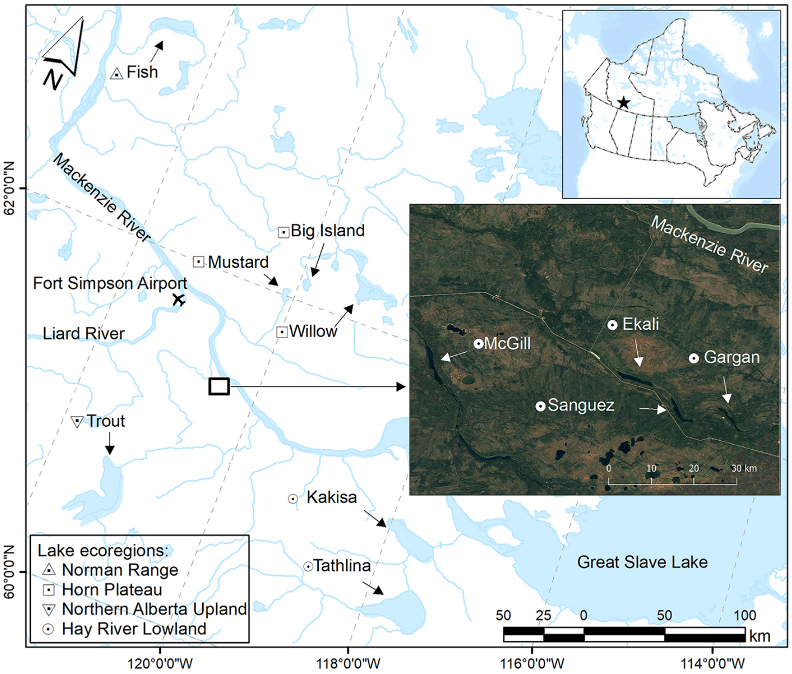


Figure 2.1. Location of study lakes in the Dehcho Region, NT, Canada (base data source: Northwest Territories Centre for Geomatics and Google Earth v7.1.8).

783.2 m asl, and maximum depths range from 3 to 30 m. The study lakes support fish populations that are harvested by First Nations for commercial and/or subsistence purposes. All study lakes contain Northern Pike. Other fish present in the region include Walleye (*Sander vitreus*), Lake Whitefish (*Coregonus clupeaformis*), Longnose Sucker (*Catostomus catostomus*), White Sucker (*Catostomus commersoni*), Burbot (*Lota lota*), and Cisco (*Coregonus artedi*), although fish diversity varies among lakes, and quantitative relative abundance or presence/absence surveys have not been completed.

2.2.2 Field sampling

As part of a large, collaborative, community-driven project on mercury levels in fish in the Dehcho Region, we collected Northern Pike, benthic invertebrates, water, and sediment from the eleven lakes over a period of seven years, from 2013 to 2019 (August-September each year). All lakes were sampled in more than one year to reflect community priorities and/or results from interim analyses, and 1-3 lakes were sampled each year depending on access (e.g., float plane and remote camp vs road access). A full schedule of sampling effort is provided in Table B.1 (Appendix B). The remote nature and logistical challenges associated with comprehensive sampling of the study lakes prevented sampling of all lakes in the same year. As we were particularly concerned with among-lake variation in water parameters being confounded with among-year variation, we launched two additional and dedicated water sampling campaigns in years 2018 and 2019, where water samples were collected from all lakes within a period of one week in late August via floatplane (Table B.1).

Northern Pike were caught by Indigenous guardians/harvesters using gill nets (20-140 mm stretched mesh size). Fish were measured for fork length (mm) and weight (g). A dorsal muscle sample (anterior to dorsal fin) was excised for stable isotope and mercury analyses. Cleithra (a large bone that extends upwards from the base of the pectoral fin and anchors to the cranium above the gills) and otoliths (a calcium carbonate structure in the inner ear) were also collected for age and growth analyses. Large fish were processed carefully to ensure suitability for human consumption after sampling, and were supplied to band members of partner First Nations communities. Tissue and cleithra samples were stored in WhirlpakTM bags, and frozen in the field at $-20\,^{\circ}\mathrm{C}$ for further processing in the laboratory. Otoliths were dried and stored in paper envelopes. Benthic invertebrates were collected from both littoral (nearshore) and profundal (offshore, and from the approximate center of lakes) areas using dip nets (250 μm mesh size) and Ekman dredges, respectively. Benthic invertebrates were coarsely sorted and identified to Family in the field, stored in acid-washed (10% HCl) scintillation vials, and then frozen at $-20\,^{\circ}\mathrm{C}$ in a portable freezer for further analyses.

Lake water samples were collected near the surface ($<$ 30 cm depth) and from the approximate center of each lake. Known volumes of near-surface water samples were grab-collected in new, sterile amber Nalgene bottles and were filtered on-site through ethanol-rinsed 0.42 μm Whatman® GF/F filters before being frozen for analysis of chlorophyll-a (a proxy for primary production). Sixty milliliters of near-surface water was pumped directly from the lake through muffled quartz QMA filters using a peristaltic pump, stored in amber glass bottles, and kept cool for analysis of dissolved organic carbon (DOC). Both filtered (quartz QMA filters and tubes were renewed between samples) and unfiltered near-surface water samples were collected using "clean hands-dirty hands" technique (U.S. EPA, 1996) and sterile double-bagged PETG bottles. Samples were preserved with trace metal-grade HCl (OmniTrace®, Supelco) to 0.5% v/v, and kept cool for analyses of total Hg and MeHg. Additional water samples were collected for analysis of sulphate (SO_4) concentrations. Near surface dissolved oxygen (DO, mg/L) and pH were measured on site using a multi-probe hand-held meter (YSI Professional Plus series, Yellow Springs, USA). When water conductivity was below 100 μS/cm, pH measurements were adjusted with 2.0 M KCl to ensure accuracy (Covington et al., 1983). Water clarity was characterized using a Secchi disk at the time of water sampling. Sediment samples were collected from the central, deep locations in each lake using an Ekman dredge. The top 2-3 cm of sediment in the dredge was collected using an acid-washed Teflon spoon, placed in a Whirl-pak^{TM} bag, and frozen at -20 °C in the field.

2.2.3 Laboratory processes

At the University of Waterloo (ON, Canada), all frozen Northern Pike and benthic invertebrate (shells were removed where applicable and taxonomic assignments were checked for accuracy) tissue samples were freeze-dried at -54 °C and 0.014 mBar for 48 h using a LabConco® FreeZone freeze-drier (USA). Freeze-dried samples were homogenized using ultra-fine scissors and glass rods inside borosilicate scintillation vials. Homogenized Northern Pike samples were weighed (0.30-0.35 mg) into tin cups using an analytical microbalance model XP05DR (Switzerland) and analyzed (n = 535) for stable Nitrogen (δ^{15}N) and Carbon (δ^{13}C) isotope ratios using a Thermo Fisher Scientific® Delta XL continuous-flow isotope ratio mass spectrometer (USA) connected to a Fisons Instruments® 1108 Elemental Analyzer (UK) at the University of Waterloo Environmental Isotope Laboratory. Values of δ^{13}C and δ^{15}N are expressed in parts per mil (‰) relative to international standards of Vienna Pee-Dee Belemnite and N_2 gas, respectively. Precision was estimated using duplicate samples (results in Table B.2).

Freeze-dried and homogenized tissue samples of Northern Pike (n = 535) and benthic invertebrates (n = 461 composite samples; several individuals of the same benthic invertebrate taxa were combined to meet minimum weight requirements) were analyzed for concentrations of total Hg and methyl Hg, respectively. Analyses were conducted at the Western University Biotron Center for Experimental Climate Change Research in London (ON, Canada) in accordance with United States Environmental Protection Agency methods 1630 and 7473 (U.S. EPA, 1998, 2007). Briefly, total Hg analyses were conducted on a Milestone® DMA-80 Direct Mercury Analyzer (Italy), and methyl Hg analyses were conducted on samples that were extracted after addition of potassium hydroxide (KOH) solution and either hot block (Bloom and Fitzgerald, 1988) or microwave digestion. Detection was by cold vapor atomic fluorescence spectroscopy using a Tekran 2700 Automated methyl Hg Analysis System (Canada). Quality control-quality assurance (QA/QC) was completed in accordance with the lab's ISO 17025 accreditation (results in Table B.2).

Water and sediment samples were analyzed for both total and methyl Hg at the Biotron Center in accordance with United States Environmental Protection Agency methods 1630 and 7473, respectively (U.S. EPA, 1998, 2007). Sediment samples were freeze-dried and homogenized via the same procedure described earlier for tissue samples. Overall, 57 water and 50 sediment samples were analyzed for both total and methyl Hg during the study (see Table B.2 for QA/QC). Analysis of chlorophyll-a (a proxy for primary production) was conducted at the University of Alberta Biogeochemical Analytical Service Laboratory in Edmonton (AB, Canada). Briefly, chlorophyll-a was extracted with 95% ethanol on a filter overnight and measured using a Shimadzu® RF-1501 Spectrofluorophotometer (North America Analytical & Measuring Instruments). Analyses of DOC and SO_4 were conducted at the Biotron Center, where concentrations of DOC were determined using phosphoric acid digestion followed by persulphate oxidation on an Aurora® 1030W total organic carbon analyzer (USA), and concentrations of SO_4 were determined using a Dionex ICS-1600 reagent-free ion chromatography system (USA) and a AS-50 autosampler (USA).

2.2.4 Geospatial data

Publicly available geospatial data on the Government of Canada portal were used to quantify catchment characteristics. Data were accessed in May 2019 and analyzed using the coordinate system ESPG:3581 NAD83(CSRS)/NWT Lambert. For each lake, we calculated total catchment area (CA) in km^2 based on the ArcticDEM of the High Resolution Digital Elevation Model v1.3 (HRDEM, 2021). This same source was used to delineate catchments, and to obtain mean elevation and mean slope in most catchments. As the headwaters of Kakisa and Tathlina lakes are not included in the ArcticDEM, the Canadian Digital Elevation Model 1945-2011 (CDEM, 2021) was used for these lakes. Lake surface area (LA) and catchment land cover compositions were calculated (in km^2) based on the 30-m resolution data layer of 2010 Land Cover of Canada (LCC, 2021a). Finally, lake to catchment area ratios (LA:CA) were calculated. Full details of the software and packages used for geospatial analyses are described in Moslemi-Aqdam et al. (2021) or Chapter 3.

2.2.5 Data analysis

Data analysis was conducted in RStudio v1.3.959 (R Studio Team, 2020) integrated with R v4.0.1 (R Core Team, 2020) using core packages, unless otherwise noted. In all statistical analysis, alpha was set at 0.05, and each lake represented a single replicate (n = 11, data were averaged among years; Table B.1 and Table B.3). When appropriate, data were $\log_{10}$ transformed to generate normally distributed data and residuals of statistical models were re-examined to ensure that $\log_{10}$ transformations improved normality.

We structured our analysis and consideration of variables in the context of three categories (Figure 2.2), including i) Fish factors, ii) Lake factors, and iii) Catchment factors, with the ultimate response variable being total Hg concentrations in Northern Pike ($[\mathrm{Hg}]_{\mathrm{NRPK}}$). To account for among-lake variation in mean size of sampled Northern Pike, which can affect Hg concentrations, $[\mathrm{Hg}]_{\mathrm{NRPK}}$ were standardized to a fork length (FL) of 600 mm (Figure 2.2). This size was similar to the median FL of Northern Pike across all lakes (580 mm) and was chosen to avoid extrapolation beyond captured sizes. Standardization of $[\mathrm{Hg}]_{\mathrm{NRPK}}$ was accomplished using an analysis of covariance (ANCOVA) that included $\log_{10}$-$[\mathrm{Hg}]_{\mathrm{NRPK}}$ as the dependent variable, lake as the categorical variable, $\log_{10}$-FL as the continuous covariate, and an interaction term between the categorical lake variable and the continuous $\log_{10}$-FL covariate. The fully saturated model that included the interaction term (i.e., slopes were specific to each lake) was used to generate

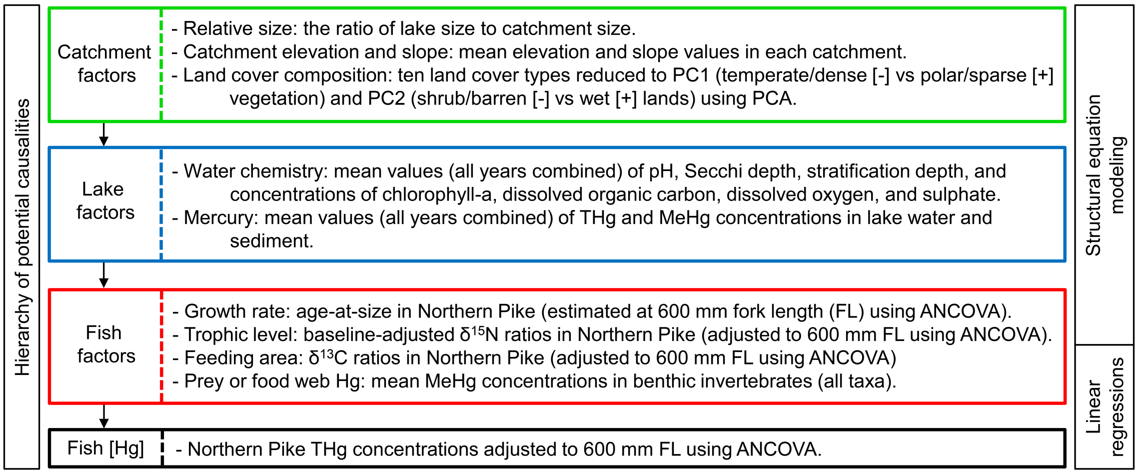


Figure 2.2. A summary of analytical variables and procedures used to investigate among-lake varibility of mercury concentrations in Northern Pike.

lake-specific least squares means of $\log_{10}$-$[\mathrm{Hg}]_{\mathrm{NRPK}}$ (at FL 600 mm) using the R package lsmeans v2.30 – 0 (Lenth, 2016). Lake-specific least squares mean $[\mathrm{Hg}]_{\mathrm{NRPK}}$, and associated ANCOVA results, are provided in Table B.3.

We considered fish factors as variables that operate at the organismal level, including foraging habitat (δ^{13}C), trophic level (δ^{15}N), growth rate of Northern Pike, and [Hg] in lower trophic level biota (Figure 2.2). To adjust δ^{15}N ratios in Northern Pike for among-lake variation in baseline (Post, 2002), lake-specific mean δ^{15}N ratios from Sphaeriidae benthic invertebrates were subtracted from δ^{15}N in individual Northern Pike. Sphaeriidae were collected from each study lake at the same time as Northern Pike were collected (see Chapter 3 or Moslemi-Aqdam et al. (2021) for details). Because fish size can affect trophic ecology, lake-specific least square means were estimated for δ^{13}C and for baseline-adjusted δ^{15}N in Northern Pike ($\delta^{13}\mathrm{C}_{\mathrm{NRPK}}$ and $\delta^{15}\mathrm{N}_{\mathrm{NRPK}}$, respectively) at 600 mm FL using ANCOVAs similar to the one described above for $[\mathrm{Hg}]_{\mathrm{NRPK}}$ (Figure 2.2).

Least square means age at 600 mm FL was also estimated as a proxy for growth (e.g., Burke et al., 2020) in Northern Pike (age-at-size$_{\mathrm{NRPK}}$) using an ANCOVA that included $\log_{10}$-age as the dependent variable, lake as the categorical variable, $\log_{10}$-FL as the continuous covariate, and an interaction term between lake and $\log_{10}$-FL (Figure 2.2). Age was determined from cleithra (in ~80% of cases), or from sectioned otoliths (for methodology, see Faust et al. (2013)) where cleithra were not available. Both cleithra and otoliths are known to provide reliable age estimates in Northern Pike (Casselman, 1990; Faust et al., 2013). We did not find systematic differences between ages determined using otoliths and ages determined using cleithra in a subset of Northern Pike (n = 70) for which both data from both structures were available (pooled McNemar test: $x^2 = 0.429$, df = 1, $p = 0.513$; semi-pooled Evans-Hoeing test: $x^2 = 4.833$, df = 5, $p = 0.437$; and unpooled

Bowker test: $x^2 = 15.667$, df = 16, $p = 0.476$). Finally, to represent [Hg] in the food webs Northern Pike rely on (i.e., MeHg concentrations in basal/lower trophic level biota), arithmetic mean [MeHg] in benthic invertebrates ($[\text{MeHg}]_{\text{invert}}$) was calculated using all taxa combined in each lake (Figure 2.2). Lake-specific data for all fish factors, including results of corresponding ANCOVAs, are provided in Table B.3. Lake-specific information of benthic invertebrate taxa collected during the course of study are provided in Table B.4.

Lake factors included variables that operate within the wetted boundary of lakes, including water chemistry, and total Hg and MeHg levels in lake waters and sediments (Figure 2.2). We used total Hg and MeHg concentrations in unfiltered water samples ($[\text{THg}]_{\text{wat}}$ and $[\text{MeHg}]_{\text{wat}}$) and raw total Hg and MeHg concentrations in sediment samples ($[\text{THg}]_{\text{sed}}$ and $[\text{MeHg}]_{\text{sed}}$) in our analyses. Data for [THg] and [MeHg] in filtered water samples, as well as sediment [THg] and [MeHg] normalized to organic matter content were not used in the analysis but are available in Table B.5 and Table B.6, respectively. The percentage of total Hg in the form of MeHg (%MeHg), a proxy for net methylation (see Paranjape and Hall, 2017; Burke et al., 2020), was calculated for both sediment and water samples, but was not used in analysis due to collinearity with [MeHg] (Table B.5 and Table B.6). We considered and interpreted [MeHg] as an integrated reflection of overall net MeHg production and cycling in the system (Branfireun et al., 2020). For all lake factors (except stratification depth), arithmetic means were calculated from multiple samples (all data included) that were collected over the study period (Table S1). Stratification depth in September (approximate time of sampling) was estimated using a fetch index (i.e., the average of maximum effective lake width and length) that was calculated for each lake and bounded to the maximum depth of the lakes (Pompilio et al., 1996). Lake-specific data for lake factors are provided in Table B.3.

Catchment factors included variables that operate outside lake boundaries, including catchment land cover, elevation, slope, and lake to catchment area ratio (Figure 2.2). Lake-specific data on catchment factors are provided in Table B.3. Land cover in our study area included ten categories (Table B.3), which are described in Natural Resources Canada (2019). Land cover data (expressed as percentages of CA) were z-transformed (mean-subtracted and divided by standard deviation) and then reduced using Principal Component Analysis (PCA) in the package vegan $v2.5-6$ (Oksanen et al., 2019). The first two principal axes (PC1 and PC2) had eigenvalues > 1 and together explained 75% of the variance in catchment land cover (Figure B.1). PC1 indicated a gradient of temperate/subpolar needleleaf and mixed forests (negative loadings) to subpolar/polar grass and shrub lands (positive loadings), and PC2 indicated a gradient of shrub and barren lands (negative loadings) to wetland (positive loadings). Lake-specific PC1 and PC2 scores were extracted for use in further analyses as catchment factors (Figure 2.2).

Our analyses followed a hierarchical procedure (Figure 2.2 and Table 2.1). We first considered $[\mathrm{Hg}]_{\mathrm{NRPK}}$ to be influenced by fish factors, including $\delta^{15}\mathrm{N}_{\mathrm{NRPK}}$, $\delta^{13}\mathrm{C}_{\mathrm{NRPK}}$, age-at-size$_{\mathrm{NRPK}}$, and [MeHg]invert (representing mercury in food webs Northern Pike rely on). At this level, previously elucidated causalities between trophic ecology and growth rate in Northern Pike in our studied systems (see Moslemi-Aqdam et al. (2021) or Chapter 3) were not repeated. At the second level, fish factors were considered to be influenced by lake factors, including water chemistry (DOC, Chlorophyll-a, DO, pH, SO_4, stratification, and water clarity), $[\mathrm{THg}]_{\mathrm{sed}}$, $[\mathrm{MeHg}]_{\mathrm{sed}}$, $[\mathrm{THg}]_{\mathrm{wat}}$, and $[\mathrm{MeHg}]_{\mathrm{wat}}$. Here, potential causalities among lake factors were also evaluated (e.g., effects of DOC and water clarity on primary production, [THg], and [MeHg] in water and sediment). Finally at the third level, lake factors were considered to be influenced by catchment factors, including land cover, elevation, slope, and relative size. Here, potential causalities among catchment factors were also evaluated (e.g., relationships between land cover and elevation or slope). We therefore investigated underlying mechanisms of variables known to influence fish [Hg] through direct and indirect pathways (see Thomas et al., 2020), which was made possible by our comprehensive sampling design that targeted variables from various ecosystem compartments.

We investigated the hierarchy of causal relationships using a combination of simple linear regressions and a piecewise structural equation model (piecewise-SEM) (Figure 2.2

Table 2.1. Causalities between variables known to influence fish mercury.

Response	Predictor	Causality
Fish mercury	Fish factors	Fish ecology, such as growth rate, foraging area, and trophic position, affect Hg levels in fish (e.g., Power et al., 2002; Sharma et al., 2008; Burke et al., 2020). Levels of Hg in fish is influenced by Hg levels in prey (e.g., Hall et al., 1997; Chen and Folt, 2005).
Fish factors	Fish factors	Foraging area and feeding efficiency affect growth rate in fish (van Dorst et al., 2019).
Fish factors	Lake factors	Water chemistry (e.g., primary production, DOC, pH, and DO) affects fish ecology, including growth rate, foraging area, and feeding efficiency (e.g., Benoît et al., 2016; van Dorst et al., 2019). Levels of Hg and MeHg, as well as water chemistry (e.g., primary production) in lakes affect Hg uptake to lake food webs (e.g., Winfrey and Rudd, 1990; Pickhardt et al., 2002).
Lake factors	Lake factors	Water chemistry (e.g., DOC, pH, SO4, DO, light, and chlorophyll-a) affects net methylation rates of Hg as well as Hg and MeHg levels inside lakes (see Paranjape and Hall, 2017; Branfireun et al., 2020). There are also interactions between water chemistry variables, such as among DOC, primary production, and light availability (see Karlsson et al., 2009).
Lake factors	Catchment factors	Catchment factors affect direct input of Hg and MeHg to downstream lakes (e.g., Demers et al., 2007; Eklöf et al., 2016; Denkenberger et al., 2020). Physical attributes of catchments, including relative size and land cover composition, affect in-lake environmental conditions (e.g., Sobek et al., 2007; Williams et al., 2010; Kritzberg et al., 2020).
Catchment factors	Catchment factors	Catchment topography (e.g., elevation and slope) affect land cover composition (Matasci et al., 2018).

and Table 2.1). Briefly, SEM is a useful multivariate approach in investigating causality in complex networks of relationships (Grace et al., 2010). A common concern with SEM is that parameter estimation, which is accomplished using a single variance-covariance matrix, can become unrealistic when the number of variables approaches or exceeds the number of observations (Lefcheck, 2016; Deng et al., 2018). Piecewise-SEM, however, evaluates each path (or sub-model) individually, and is thus extremely flexible and useful in ecological applications when there are often many factors relative to observations (Lefcheck, 2016), such as in this whole-ecosystem study. Goodness-of-fit in piecewise-SEM is evaluated by a Chi-square distributed Fisher's test, where $p > 0.05$ indicates that the model adequately represents patterns in the data (Shipley, 2013; Lefcheck, 2016).

We first investigated relationships among $[\mathrm{Hg}]_{\mathrm{NRPK}}$ and fish factors using simple linear regressions, where $[\mathrm{Hg}]_{\mathrm{NRPK}}$ was the response variable (Figure 2.2). Considering fish factors determined relevant by the linear regressions ($p \leq 0.05$), we then developed a conceptual metamodel of hypothesized relationships among variables known to influence [Hg] in fish and evaluated these causalities in piecewise-SEM (Table 2.1, Figure 2.2, and Figure 2.3). The complexity of causal relationships, as well as the large number of variables involved, necessitated a rigorously simplified analytical procedure to avoid overloading issues. In our piecewise-SEM, the first sub-model included $[\mathrm{Hg}]_{\mathrm{NRPK}}$ as a response variable and fish factors determined relevant by the linear regressions as predictors. The second sub-model included a predictor of the first sub-model as a response variable. As part of the piecewise-SEM, each new response variable was linearly modelled, one at a time, against hypothetical predictors (Figure 2.3). Significant predictors ($p \leq 0.05$) were determined and retained, while considering collinearity. This procedure was continued until a sub-model was developed for each predictor determined relevant in previous steps, and ultimately resulted in a single piecewise-SEM with standardized, and therefore directly comparable, effects sizes of relevant causalities among fish, lake, and catchment factors, and $[Hg]_{NRPK}$ in the study area.

Collinearity was assessed using Pearson's correlation matrices that were developed separately for fish factors, lake factors, and catchment factors, and where applicable, correlations between variables with no known cause-effect mechanisms were accounted for by incorporation of correlated errors (see Lefcheck, 2016). As a measure of collinearity, variation inflation factor (VIF) was also reported for sub-models with > 1 predictors, with VIF < 5 being acceptable and VIF > 5 and 10 being concerning and problematic, respectively (James et al., 2013). Piecewise-SEM was fit using centered data and in the package piecewise-SEM v.2.1.2 (Lefcheck et al., 2020).

2.3 Results and Discussion

Substantial among-lake variability was observed in $[Hg]_{NRPK}$ in the Dehcho Region (Table B.3). Mercury concentrations (in mg/kg wet weight (ww) adjusted to 600 mm fork length) in Northern Pike differed more than fourfold among lakes in the study area (minimum = 0.158 mg/kg ww in Trout Lake, and maximum = 0.678 mg/kg ww in Sanguez and McGill Lakes). Size-standardized $[Hg]_{NRPK}$ were below the Commercial Sale of Fish Guideline (0.50 mg/kg ww) in Trout, Mustard, Willow, Kakisa, Big Island, Tathlina, Fish, and Gargan lakes, were near the guideline in Ekali Lake, and were above the guideline in Sanguez and McGill lakes (Table B.3). Not all of the studied lakes are fished commercially and the Commercial Sale of Fish Guideline (0.50 mg/kg ww) only applies to retail fish; thus, we use the guideline here as a general reference point. Consistent with our findings, results from previous authors showed that Northern Pike had higher [Hg] in McGill and Sanguez lakes compared to Northern Pike in neighboring lakes in the Dehcho Region (Evans et al., 2005; Lockhart et al., 2005; Laird et al., 2018). Lake-specific, Hg-related consumption advisories are currently in place for Northern Pike in some of the study lakes, including Ekali, Sanguez, and McGill (see Health and Social Services, 2021).

The variability (0.16-0.68 mg/kg ww) we observed for size-standardized $[\mathrm{Hg}]_{\mathrm{NRPK}}$ among lakes within the Dehcho Region is only slightly smaller than the range reported for northern populations of Northern Pike across a much larger geographic scale; a range of 0.10-0.80 mg/kg ww adjusted to a similar size (i.e., 622 mm fork length) was reported for populations that spanned all three northern territories of Canada (see Lockhart et al., 2005). High variability in [Hg] in Northern Pike among lakes has previously been reported in the Dehcho Region, but detailed understanding of the causes of this variation have long been unknown (Evans et al., 2005; Lockhart et al., 2005; Laird et al., 2018).

2.3.1 Fish factors that affected $[\mathrm{Hg}]_{\mathrm{NRPK}}$

In our analysis of fish factors on $[\mathrm{Hg}]_{\mathrm{NRPK}}$, we found no relationships between trophic level and $[\mathrm{Hg}]_{\mathrm{NRPK}}$ (Figure 2.3a and Table B.7). While relationships between [Hg] in fish and δ^{15}N-inferred trophic level are common in the literature (e.g., Kidd et al., 1995; Power et al., 2002; Depew et al., 2013), baseline-adjusted $\delta^{15}\mathrm{N}_{\mathrm{NRPK}}$ varied less than three ‰ (~one trophic level) among the study lakes (Table B.3), indicating that Northern Pike feed at approximately the same relative trophic level in each system. Feeding at a similar trophic level does not rule out the possibility of effects of variable Hg biomagnification rates through lake food webs. Rates of Hg biomagnification can vary among lakes and influence [Hg] in resident fish Kidd et al., 2012; Kozak et al., 2021, e.g.,, and are a current knowledge gap that warrant additional study in these lakes. Such additional research should include as comprehensive a sampling of biota (at multiple trophic levels) as possible.

It appeared at first that carbon source (benthic vs pelagic) affected $[\mathrm{Hg}]_{\mathrm{NRPK}}$ (Figure 2.3b and Table B.7), but the relationship between $\delta^{13}\mathrm{C}_{\mathrm{NRPK}}$ and $[\mathrm{Hg}]_{\mathrm{NRPK}}$ was confounded with a relationship between growth rate and mercury concentrations in Northern Pike. Slow-growing Northern Pike had significantly more depleted $\delta^{13}\mathrm{C}$ values than fast-growing Northern Pike (Pearson's $r = 0.82$, $p = 0.002$). In a recent study that included a detailed assessment of the positive relationship between Northern Pike growth and $\delta^{13}\mathrm{C}$ in the study lakes (i.e., Chapter 3), Moslemi-Aqdam et al. (2021) found that $\delta^{13}\mathrm{C}$ values in Northern Pike were positively associated with $\delta^{13}\mathrm{C}$ values in benthic invertebrates collected from littoral areas in the lakes. Using comprehensive lake and catchment data, Moslemi-Aqdam et al. (2021) showed in Chapter 3 of this thesis that the origin of organic matter (internal vs terrestrial primary production) was likely responsible for among-lake variability in $\delta^{13}\mathrm{C}$ in both Northern Pike and benthic invertebrates, and that among-lake variability in $\delta^{13}\mathrm{C}$ in Northern Pike was likely not related to differences in habitat use (i.e., benthic vs pelagic), which can influence [Hg] in fish (e.g., Power et al., 2002). We thus did not explore the relationships between $[\mathrm{Hg}]_{\mathrm{NRPK}}$ and $\delta^{13}\mathrm{C}_{\mathrm{NRPK}}$ further.

Northern Pike [Hg] were significantly and positively related to two other fish factors, age-at-size$_{\mathrm{NRPK}}$ and $[\mathrm{MeHg}]_{\mathrm{invert}}$ (Figure 2.3c, Figure 2.3d, and Table B.7). The relationship between $[\mathrm{Hg}]_{\mathrm{NRPK}}$ and age-at-size$_{\mathrm{NRPK}}$ indicated lower [Hg] in faster-growing Northern Pike compared to slower-growing counterparts, which is consistent with the somatic growth dilution effect that is well-documented for this species (e.g., Sharma et al., 2008; Thomas et al., 2020). The positive relationship between $[\mathrm{Hg}]_{\mathrm{NRPK}}$ and $[\mathrm{MeHg}]_{\mathrm{invert}}$

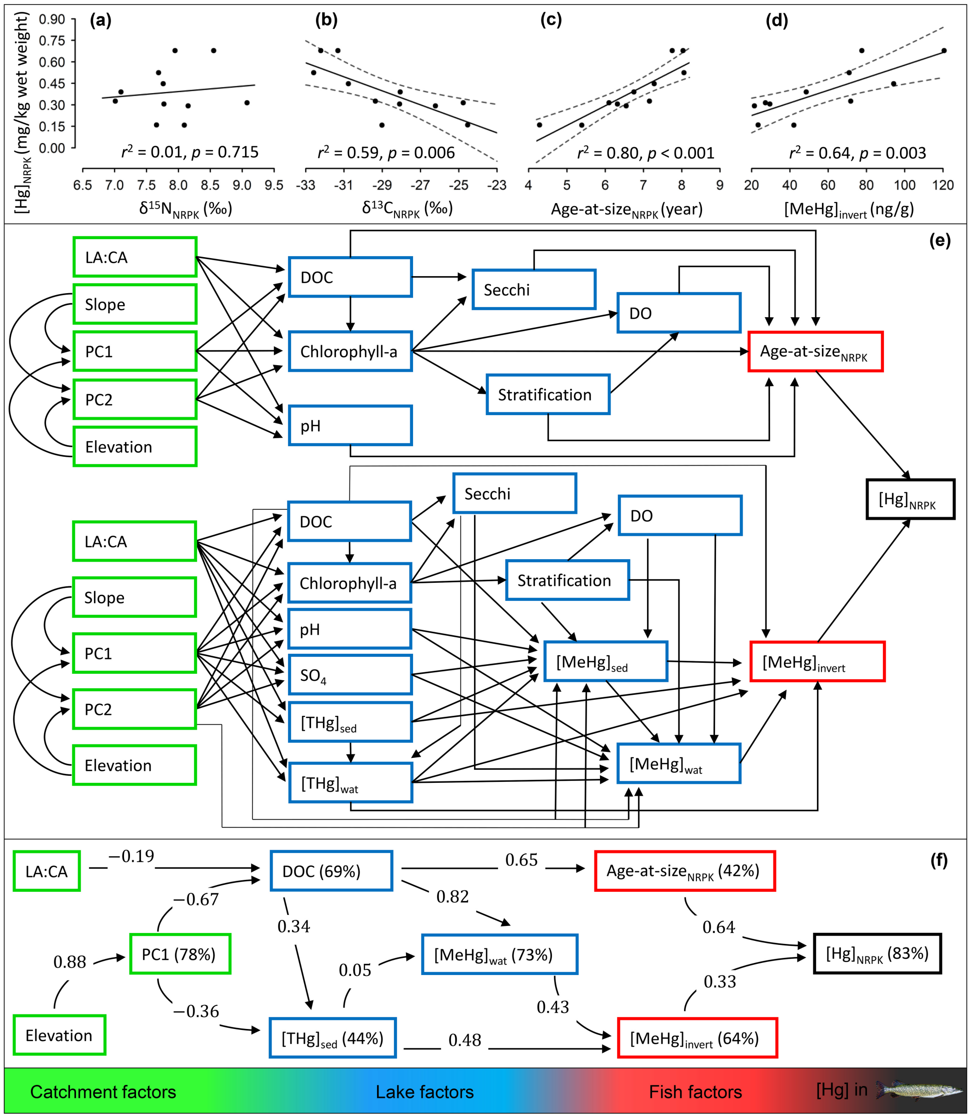


Figure 2.3. Individual linear relationships between Hg levels in Northern Pike and fish factors representing trophic level **(a)**, foraging area **(b)**, growth rate **(c)**, and Hg levels in food webs (benthic invertebrates) **(d)**, the conceptual metamodel of all hypothesized causalities among variables evaluated in piecewise-SEM **(e)**, and final piecewise-SEM depicting supported causalities in the study area **(f)**. In (a) to (d), solid lines represent fitted lines and dashed lines represent ±95 confidence intervals. In (e), the metamodel was developed considering significant predictors of fish [Hg] in (a) to (d) and $\delta^{13}C_{NRPK}$ was excluded due to collinearity with age-at-size$_{NRPK}$. In (f), percentages represent cumulative variation of the dependent variables explained by explanatory variables and numbers represent standardized effect sizes of explanatory variables (numbers are directly comparable; the higher the number, the stronger the effect). PC1 and PC2 are the first two principal components of the land cover PCA and represent temperate/dense [−] vs polar/sparse [+] vegetation and shrub/barren lands [−] vs wetlands [+], respectively. Catchment and lake factors are duplicated in the middle panel to facilitate visualization.

is consistent with the knowledge that [Hg] in fish tissues, which is mostly in the form of MeHg (Bloom, 1992; Lescord et al., 2018), is primarily derived from dietary intake (Hall et al., 1997; Ward et al., 2010b). Our conceptual metamodel (Figure 2.3e) was thus developed for hypothesized relationships between predictor variables operating at the scales of lake and catchment on each of age-at-size$_{\text{NRPK}}$ and [MeHg]$_{\text{invert}}$, which together explained 83% of among-lake variability in [Hg]$_{\text{NRPK}}$ in piecewise-SEM. Detailed statistics of piecewise-SEM analysis and the final model are provided in Table B.8. Collinearity among predictor variables retained in sub-models of piecewise-SEM was < 5 in all cases (Table B.9). A schematic summary of the final piecewise-SEM is depicted in (Figure 2.3f. Statistics indicate that the model adequately represented patterns in the data (Fisher's C $= 28.747$, df $= 40$, $p = 0.913$).

2.3.2 Lake factors that affected fish factors

At the second level of analysis, we investigated effects of lake factors on fish factors that significantly predicted [Hg]$_{\text{NRPK}}$. We found that age-at-size$_{\text{NRPK}}$ was higher in lakes with higher concentrations of DOC (Table B.8), indicating a constraining effect of DOC on growth rate in Northern Pike. There was also a significantly positive relationship between age-at-size$_{\text{NRPK}}$ and pH (Table B.8), but we did not retain pH in the model because DOC and pH were significantly and positively correlated (Pearson $r = 0.74$, $p = 0.009$), and because the observed range of pH among our studied lakes (7.2-8.3) was within the optimum range for Northern Pike growth (6-9). Concentrations of DOC explained 42% of the among-lake variability in age-at-size$_{\text{NRPK}}$ (Figure 2.3f).

Negative relationships between DOC and fish growth rate have been observed for many freshwater fish species and are largely attributed to influences on mediating variables, including light attenuation (impeded visual performance at higher DOC), primary productivity (decreased biomass at lower trophic levels at higher DOC), and concentrations of dissolved oxygen (which affects metabolism and feeding efficiency) (e.g., Ranåker et al., 2014; Benoît et al., 2016; van Dorst et al., 2019). We observed, however, no significant relationships between age-at-size$_{\text{NRPK}}$ and chlorophyll-a (indicator of biomass at lower trophic levels), water clarity (visual performance), or dissolved oxygen (metabolic rates/feeding efficiency) (Table B.8). It is possible that our point measurements of these parameters, while repeated and averaged across several years, do not capture variability at a scale fine enough to detect relationships. In a recent study on growth rates of Northern Pike in this same study area, however, Moslemi-Aqdam et al. (2021) proposed that differences in organic matter sources and quality may explain the negative relationship between Northern Pike growth rate and DOC (see Chapter 3). Among-lake variability in δ^{13}C in Northern Pike mirrored among-lake variability in δ^{13}C in benthic invertebrates, and was related to physicochemical indicators of catchment/terrestrial input (see Chapter 3 or Moslemi-Aqdam et al. (2021)); the findings indicated that lakes with higher DOC are subject to greater catchment influence/input (as indicated by DOC quality measures, catchment attributes, and water chemistry), and that food webs in these lakes are likely fueled by lower-quality, terrestrially derived carbon. Allochthonous organic matter is known to have relatively lower nutritional value (e.g., fatty acid concentrations) than autochthonous organic matter (Lau et al., 2009; Hiltunen et al., 2019).

For the second fish factor that significantly predicted [Hg] in Northern Pike, [MeHg]$_{\text{invert}}$, 64% of among-lake variability was explained by positive relationships with concentrations of MeHg in water and concentrations of total Hg in sediment (Figure 2.3f and Table B.8). This result suggests that MeHg uptake into lower trophic levels, and consequent bottom-up trophic transfer of MeHg to Northern Pike, is higher in lakes with higher concentrations of mercury in abiotic ecosystem compartments. Among factors hypothesized to affect [MeHg] in water, concentrations of DOC in water and total Hg in sediment were the most influential (both with positive effects), and explained 73% of the among-lake variability in water [MeHg] in the study area (Figure 2.3f and Table B.8). DOC can promote production of MeHg in lakes via various pathways, including enhancing the transport of Hg from catchments to downstream lakes (Shanley et al., 2008), and by increasing activity of microbial methylators (Lambertsson and Nilsson, 2006) and rates of biological uptake (Wiener et al., 2006). In addition, high levels of DOC and associated light attenuation can inhibit photo-demethylation of MeHg (Klapstein et al., 2018). While effects of DOC on Hg cycling and methylation in lakes can be influenced by molecular composition and aromaticity of DOC (see Shanley et al., 2012) and recalcitrant and highly aromatic DOC can actually make Hg less bioavailable (e.g., Eagles-Smith et al., 2016), the positive relationship between MeHg and DOC that we observed suggests that DOC promotes production of MeHg in these lakes. The underlying mechanisms and magnitude of effect warrant further research.

Concentrations of MeHg in water are often correlated with: i) water clarity (photo-demethylation) (e.g., Lehnherr and St. Louis, 2009); ii) [MeHg] in sediment (sediment-water exchange of MeHg) (e.g., Guédron et al., 2020); and iii) catchment land cover (e.g. proportional area of wetlands) (e.g., St. Louis et al., 1994). We found, however, that Secchi depth, [MeHg] in profundal sediment, and proportional wetland cover (i.e., PC2) were not significantly related to [MeHg] in water (Table B.8). While the lack of relationship between [MeHg] in profundal sediment and [MeHg] in water might be attributed to among-lake differences in partitioning between aqueous and particulate phases (since unfiltered samples were used in our analyses), and/or to differences in organic matter content of sediments (see Babiarz et al., 2001; Nasr and Arp, 2017; Obrist et al., 2018), we found similar results when we repeated the data analysis using [THg] and [MeHg] in available filtered water samples, and when we normalized [THg] and [MeHg] in sediment samples to organic matter content of sediment (Table B.10). The significant positive relationship between [MeHg] in water and [THg] in profundal sediment (Figure 2.3f and Table B.8) suggests that sediments contribute bioavailable inorganic Hg for methylation, with produced MeHg then subject to sediment-water exchange. Given that most of the study lakes do not stratify, however, it is also likely that [MeHg] in water and [THg] in sediment reflect overall Hg loadings to each of the lakes.

Concentrations of MeHg in water and sediment can vary substantially, even at fine temporal and spatial scales, and often asynchronously (Bachand et al., 2014). Concentrations of aqueous MeHg can vary greatly over the course of season in a single lake (Regnell et al., 1997; Wang et al., 2012). Production of MeHg in sediment is a spatially and temporally variable process that is regulated by an array of variables, including temperature, pH, redox conditions, and the supply of sulphate, bioaccessible organic matter, and bioavailable inorganic Hg (see Paranjape and Hall, 2017; Obrist et al., 2018). These variations in [MeHg] in water and sediment would not have been captured by our spot measurements of water and sediment parameters, and may explain the decoupling we observed between [MeHg] in water and [MeHg] in sediment. In addition, littoral sediments and periphyton can be important sites of methylation (Demers et al., 2007; Branfireun et al., 2020; Burke et al., 2020). In these generally well-mixed lakes, the profundal sediments used in our analyses may, in hindsight, not be the primary sites of methylation. Habitat-specific differences in benthic invertebrates [MeHg] support this contention; benthic invertebrates collected from littoral zones of these lakes had higher [MeHg] than benthic invertebrates collected from profundal zones (Table B.11). Despite the comprehensive nature of the sampling program, dominant sites of within-lake net MeHg production and flux to the water column were likely not fully sampled, and may have also varied in extent and magnitude from catchment to catchment.

2.3.3 Catchment factors that affected lake factors

In the final stage of our hierarchical analysis, we investigated effects of catchment characteristics on lake factors and found that lakes surrounded more by temperate/subpolar needleleaf and mixed forests (negative loadings on land cover PC1; Figure B.1) had higher levels of $[\mathrm{THg}]_{\mathrm{sed}}$ and DOC concentrations, whereas lakes surrounded more by polar/subpolar shrub and grass lands (positive loading on land cover PC2; Figure B.1) had lower levels of $[\mathrm{THg}]_{\mathrm{sed}}$ and DOC concentrations (Table B.8). We infer that forested catchments export more Hg and DOC to downstream lakes in the study area, which is consistent with previous findings (Hurley et al., 1998; Sobek et al., 2007; Shanley et al., 2012; Denkenberger et al., 2020). Indeed, lake-specific PC1 scores explained 41% of among-lake variability in $[\mathrm{THg}]_{\mathrm{sed}}$ (Table B.8). In addition, DOC concentrations in lakes decreased significantly as lake to catchment area ratios increased (Table B.6), indicating greater terrestrial influence on lakes that are small relative to their catchments. Together, lake-specific PC1 scores and LA:CA explained 69% of among-lake variability in DOC concentrations (Figure 2.3f). Not surprisingly, there was a significant negative relationship between elevation and land cover PC1 (Table B.8), indicating that shrub and grass lands were dominant land cover types in higher-elevation catchments, whereas needleleaf and mixed forests were dominant land cover types in lower-elevation catchments; elevation explained 78% of the variability in land cover PC1 (Figure 2.3f). Overall, results indicated that lower elevation catchments with proportionally more forest cover and relatively larger size exported more DOC and Hg to downstream lakes.

2.4 Conclusion

The vast Dehcho Region is characterized by diverse ecoregions and landscape features. A lack of mechanistic understanding regarding sources of among-lake variability in fish [Hg] in the Dehcho Region has raised concerns over food safety and security for local Indigenous people. Here, we present a plausible mechanistic model of how processes operating at scales from whole catchments to individual organisms interact to influence fish [Hg] in subarctic lakes used for subsistence. Our results indicate that catchments with greater relative (to downstream lake) surface area and with greater proportional cover of temperate/subpolar needleleaf and mixed forests export more DOC and Hg to downstream lakes, which subsequently affect MeHg levels in water, MeHg levels in benthic invertebrates, and growth rate in Northern Pike. Together, MeHg concentrations in benthic invertebrates and growth rates in Northern Pike explained 83% of among-lake variability in [Hg] in Northern Pike. Based on the results from this study, lakes most vulnerable to increases in fish mercury concentrations are those that are small and situated in large, low-elevation, and heavily-forested catchments, because these catchment characteristics act to decrease fish growth rate and increase food web mercury concentrations via increases in concentrations of mercury and organic matter in downstream lakes. Particularly in this era of rapid environmental change, these findings provide invaluable guidance for prioritizing lakes for Hg monitoring in a remote subarctic region, especially because many catchment characteristics can be sensed remotely. Our study shows that fish mercury is controlled by complex interactions between biotic and abiotic ecosystem compartments, thereby reaffirming the necessity of considering whole ecosystems for understanding and predicting change.

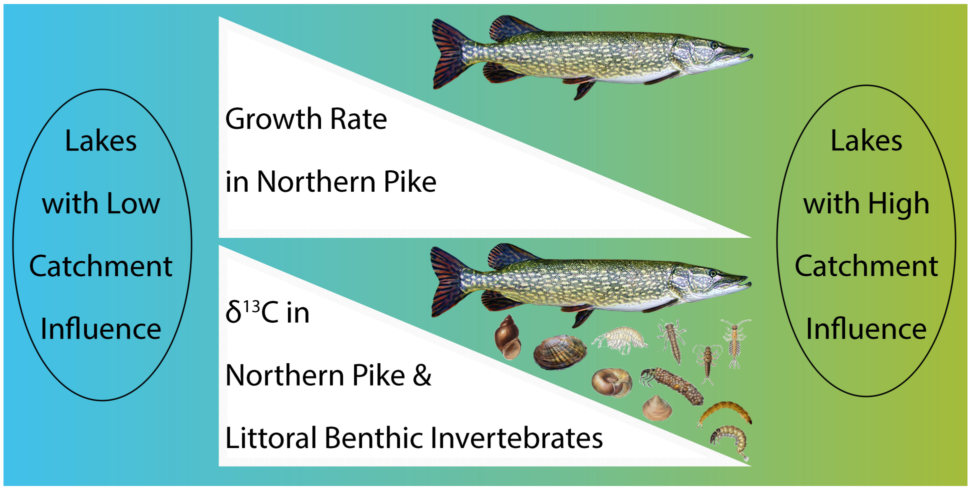
Chapter 3

Catchments affect growth rate of Northern Pike, *Esox lucius*, in subarctic lakes

This chapter is an adapted version of the following published paper[1]:

Moslemi-Aqdam M., Low G., Low M., Branfireun B. A., and Swanson H. K. 2021. Catchments affect growth rate of Northern Pike, *Esox lucius*, in subarctic lakes. *Aquatic Sciences*, 83, 3, 59. https://doi.org/10.1007/s00027-021-00817-4

Graphical Abstract



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[1] *reproduced with permission from Springer Nature.*

3.1 Introduction

Growth rates and growth-associated traits in freshwater fishes can be influenced by a range of biotic and abiotic factors that vary among lake ecosystems (Wilson et al., 2019; Höhne et al., 2020). As growth rates regulate accumulation of contaminants (e.g., mercury) in fish tissues, as well as fish population dynamics and ultimately fisheries sustainability (King, 2007; Sharma et al., 2008), understanding drivers of variability in fish growth rates among systems is of toxicological, conservational, and managerial value, particularly in an era of rapid environmental change.

Northern Pike (*Esox lucius*) is a cool-water, visual, and ambush predator that often occupies an apex trophic position in freshwater food webs (Scott and Crossman, 1973; Vøllestad et al., 1986; Craig, 1996). Northern Pike support important commercial, subsistence, and recreational fisheries throughout the northern hemisphere (Scott and Crossman, 1973), and are increasingly used as a model organism in studies of ecology and evolution due to their wide geographic distribution (Forsman et al., 2015). Growth rates and population productivity of Northern Pike have been documented to vary in response to a number of environmental and biological factors, including water temperature (Casselman, 1978; Margenau et al., 1998), water clarity (Craig and Babaluk, 1989; Margenau et al., 1998), lake primary production (Diana 1987), prey quality and availability (Diana, 1983; Navarro and Johnson, 1992; Margenau et al., 1998; Venturelli and Tonn, 2006), and habitat suitability (Vøllestad et al., 1986; Casselman and Lewis, 1996). Several of these factors are mediated by inputs from the catchment, including terrestrially derived nutrients and dissolved organic matter (DOM).

While studies specific to Northern Pike are lacking, Finstad et al. (2014) found that increases in terrestrially derived dissolved organic carbon (DOC) resulted in a unimodal response in biomass of Brown Trout (*Salmo trutta*) in southern Norway. Fish biomass increased with DOC concentrations to reach a maximum at 3 mg/L; above this concentration of DOC, fish biomass decreased. An initial increase in fish biomass in response to increasing DOC concentrations (up to 4 mg/L) was also observed for Yellow Perch (*Perca flavescens*) in several boreal lakes in north-central Ontario, Canada (Tanentzap et al., 2014). When concentrations of DOC are relatively low ($<$ 4 mg/L), positive relationships between DOC and fish biomass and growth rate have been attributed to energy subsidization, screening of UV-radiation, and increased loading of co-varying nutrients (e.g., phosphorus and nitrogen), which stimulate in-lake primary production (Finstad et al., 2014; Tanentzap et al., 2014). When DOC exceeds 3-4 mg/L (up to 22 mg/L), declines in fish biomass and growth rates have been documented in several freshwater fish species, including Pikeperch (*Sander lucioperca*), Walleye (*Sander vitreus*), Lake Trout (*Salvelinus namaycush*), and European Perch (*Perca fluviatilis*). These declines have been largely attributed to light attenuation and consequent impairing effects on water clarity, primary production, predator-prey interactions, metabolism, habitat suitability, foraging area, and feeding efficiency (e.g., Karlsson et al., 2009, 2015; Finstad et al., 2014; Ranåker et al., 2014; Craig et al., 2017; van Dorst et al., 2019).

Effects of DOC concentrations on lake productivity are affected by the structural and chemical properties of DOM, which regulate its availability for biota at the base of food chains (see Nebbioso and Piccolo, 2013; Creed et al., 2018). Terrestrial DOM that is delivered to freshwaters in runoff is predominantly derived from soil organic matter, which in turn is mainly composed of decomposition products of terrestrial vegetation. Soil-derived DOM exported from catchments tends to be aromatic in character, and of higher-molecular weight (e.g., tannins, phenols), as lower molecular weight compounds have already been metabolized in soils at earlier stages of decomposition (Kalbitz et al., 2003; Yang et al., 2021). Thus, terrestrially derived DOM tends to be less bioaccessible and less energy-rich, whereas DOM derived from in-lake primary production tends to be fresher in character, with more short-chain, low molecular weight compounds (proteins, sugars, amino acids) that may be quickly metabolized (Taipale et al., 2014; Brett et al., 2017). Relative proportions of catchment- versus lake-derived organic matter can therefore affect lake productivity and, ultimately, fish growth. As quantity and quality of DOM exported from terrestrial environments to downstream lakes is influenced by a myriad of catchment attributes, including area, elevation, slope, and land cover composition (e.g., Williams et al., 2010; Dranga et al., 2017; Kritzberg et al., 2020; Toming et al., 2020), gaining a fulsome understanding of among-system variability in fish growth rates requires comprehensive data on catchment and lake physicochemistry, in addition to data on in-lake biota. There is a general paucity of studies that link catchment characteristics with water chemistry and fish growth rates, and this is particularly true in remote northern regions, where food security and local economies are often reliant on healthy fisheries (Islam and Berkes, 2016; Martin et al., 2020), and where climate-induced change to land cover is particularly rapid (e.g., Wang et al., 2020).

In the Dehcho Region, Northwest Territories (NT), Canada, Northern Pike are locally harvested as a subsistence food source for First Nations (Stewart and Low, 2000). Along with other species (e.g., Walleye, Sander vitreus, and Lake Whitefish, Coregonus clupeaformis), Northern Pike also used to support commercial fisheries in some regional lakes during the second half of the 20^{th} century (Stewart and Low, 2000; DFO, 2010a). Commercial harvest has been suspended since the early-2000s due to stock collapses

(Kennedy, 1962; DFO, 2010a), but local subsistence harvests of Northern Pike continue. Harvests of Northern Pike and associated human consumption have been affected by high mercury concentrations in some lakes in the Dehcho Region, and among-lake variability in mercury concentrations has been partially linked to fish size-at-age or growth rates (e.g., Evans et al., 2005; Lockhart et al., 2005). Despite local dietary significance and the known role of growth rate in regulating mercury concentrations in Northern Pike (e.g., Sharma et al., 2008), little is known about what drives variability in growth rates in Northern Pike in this large subarctic region. Local Indigenous fishers have suggested that changes to the land and to inflowing waters are affecting growth and feeding of several fish species in lakes, including Northern Pike. Interactions among fish ecology and lake and catchment physicochemistry thus deserve further investigation.

The objectives of this study were to quantify variability in Northern Pike growth rate among eleven lakes located within a $60,000$ km^2 area in the Dehcho Region, NT, and to investigate factors in lake waters and catchments that may explain this variability. Indigenous knowledge regarding long-term changes in lake catchments and inflows, water color (tannins), amount of algae in nearshore lake waters, and fish numbers and quality, led to several predictions. We tested the prediction that growth rate in Northern Pike would be lower in lakes subject to more catchment influence, which we quantified using measures of water chemistry, lake area, catchment area, lake area to catchment area ratios, catchment elevation, catchment slope, and land cover composition of catchments. We also investigated whether growth rate in Northern Pike was higher in lakes with more in-lake primary production. Finally, relationships between growth and trophic ecology of Northern Pike were investigated by relating growth rates to stable Nitrogen and Carbon isotope ratios (used to infer trophic position and carbon source, respectively).

3.2 Materials and Methods

3.2.1 Field sampling

As part of a large, collaborative, and community-driven study on fish from the Dehcho Region, NT, eleven lakes (Figure 3.1) were comprehensively sampled over a period of seven years, from 2013 to 2019. Sampling was conducted by a joint University-Indigenous guardian crew, and took place each year in mid-August to mid-September. Between one and three lakes were sampled each year, depending on access (e.g., float plane and remote camp vs road access). Some lakes were sampled in more than one year to reflect

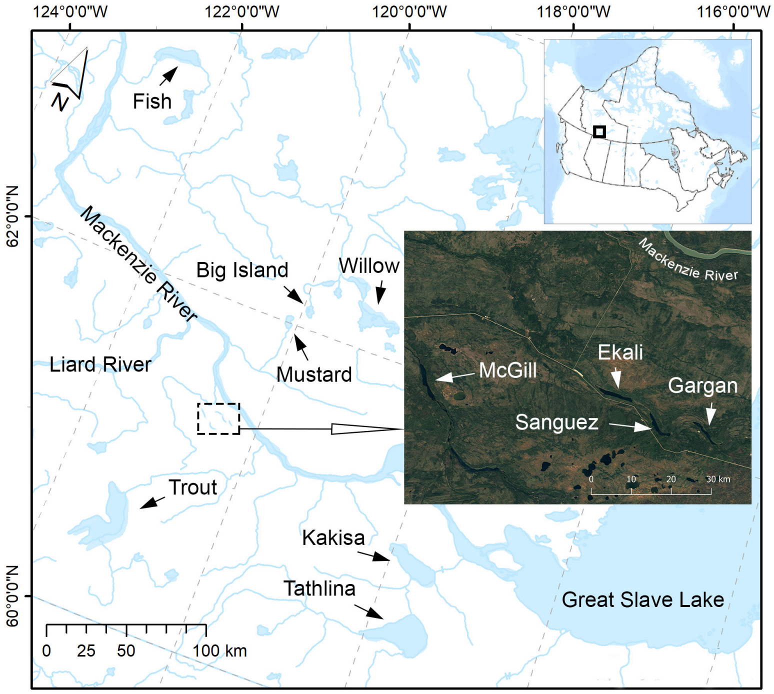


Figure 3.1. Location of the study lakes in the Dehcho Region, NT, Canada. Willow, Big Island, and Mustard lakes are located in the Horn Plateau ecoregion, Trout Lake is located in the Northern Alberta Upland ecoregion, Fish Lake is located in the Norman Range ecoregion, and all other lakes are located in the Hay River Lowland ecoregion. Base data source: Northwest Territories Centre for Geomatics and Google Earth v7.1.8.

community priorities and/or results from interim analyses. A full schedule of sampling efforts is presented in Table C.1 (Appendix C). Four ecoregions were represented by the study lakes, including the Horn Plateau (Willow, Big Island, Mustard lakes), Northern Alberta Upland (Trout Lake), Hay River Lowland (Ekali, Gargan, Sanguez, McGill, Kakisa, Tathlina lakes), and Norman Range (Fish Lake).

In each lake, Northern Pike were caught using multi-mesh gill nets, with stretched mesh size ranging from 20 to 140 mm. Captured Northern Pike were measured on-site for fork length (FL). Northern Pike $>$ 120 mm FL were then weighed (to the nearest g) and processed in the field, whereas individuals $<$ 120 mm FL were frozen whole before being shipped to the laboratory for further processing. Dorsal muscle tissue (skinless) samples were extracted from each large fish for stable isotope analysis, and cleithra were extracted to estimate age, and subsequently growth rate (Casselman, 1990; Faust et al., 2013). All tissues were frozen at -20 °C in the field. Care was taken to ensure that large fish were maintained in a condition that was suitable for human consumption after sampling, and were supplied to band members of partner First Nations communities after sampling was complete. Benthic invertebrates were collected from each lake to facilitate among-lake comparisons of stable isotope ratios in fish (i.e., baseline correction; Post, 2002). Because fractionation can vary between littoral and pelagic primary producers during DIC uptake (France, 1995), and because Northern Pike primarily feed in nearshore habitats (Vøllestad et al., 1986; Craig, 1996), benthic invertebrates were collected from littoral, nearshore areas using dip nets (250 μm mesh size) to represent the isotopic baseline. Benthic invertebrates were sorted and identified to Family in the field, and frozen in the field at -20 °C.

Lake water samples were collected near the surface (approximately 30 cm below the surface) from each lake. The remote nature and challenging logistics associated with comprehensive sampling of the study lakes prevented sampling of all lakes within one year. We were, however, particularly concerned with among-lake variation in water chemistry parameters being confounded with among-year variation. To address this, all study lakes were sampled for water chemistry in two dedicated campaigns in both 2018 and 2019. Samples were collected from each lake via floatplane within a period of one week in late August in each of 2018 and 2019. Water samples were collected from the approximate center of lakes. Known volumes of surface water were grab-collected in new, sterile amber Nalgene bottles, and were filtered on-site through ethanol-rinsed 0.42 μm Whatman® GF/F filters before being frozen for analysis of chlorophyll-a (Chl-a), a proxy for primary production. Sixty ml of surface water was pumped directly from the lake through muffled quartz QMA filters using a peristaltic pump, stored in amber glass bottles, and kept cool for analyses of DOC, DIC, SUVA (specific UV absorbance), abs254 (UV Absorbance at 254 nm), and Freshness Index (a measure of the ratio of more recently produced to more decomposed DOM, Parlanti et al., 2000). Approximately 500 ml of surface water was collected and kept cool for analyses of concentrations of nutrients and ions, including total phosphorus (TP), total nitrogen (TN), sodium (Na), calcium (Ca), manganese (Mg), chloride (Cl), alkalinity (measured as $CaCO_3$), bicarbonate (measured as HCO_3), and conductivity. At the time of water sampling, water clarity was characterized using a Secchi disk.

3.2.2 Laboratory processes

At the University of Waterloo (ON, Canada), frozen Northern Pike < 120 mm FL were thawed to room temperature, blotted to remove excess moisture, and then measured, weighed, and processed. Skinless dorsal muscle tissue samples and cleithra were removed and frozen at -20 °C. All frozen Northern Pike and benthic invertebrate tissues (shells were removed, where applicable) were freeze-dried at -54 °C and 0.014 mBar for 48 h using a LabConco® FreeZone freeze-drier (USA). Freeze-dried samples were homogenized using ultra-fine scissors and glass rods inside borosilicate scintillation vials, before being weighed (0.3-0.35 mg) into tin cups using a Mettler-Toledo® Analytical Microbalance model XP05DR (Switzerland). Prepared samples were analyzed for δ^{13}C and δ^{15}N using a Thermo Fisher Scientific® Delta XL continuous-flow isotope ratio mass spectrometer (USA) connected to a Fisons Instruments® 1108 Elemental Analyzer (UK) at the University of Waterloo Environmental Isotope Laboratory. Values of δ^{13}C and δ^{15}N are expressed in parts per mil (‰) relative to international standards of Vienna Pee-Dee Belemnite and N_2 gas, respectively. Precision was estimated using duplicate samples that were completed for approximately 9% ($n = 38$) and 28% ($n = 160$) of Northern Pike and benthic invertebrate samples, respectively. Mean difference (± one standard deviation) in δ^{15}N and δ^{13}C between duplicate samples was 0.001 (± 0.105) ‰ and 0.079 (± 0.229) ‰, respectively, for Northern Pike, and 0.038 (± 0.438) ‰ and 0.013 (± 0.437) ‰, respectively, for benthic invertebrates.

Growth rates of Northern Pike were estimated using increment widths measured on cleithra. Measuring increment widths of annuli in each individual fish resulted in substantially more lake-specific data than using age and size data of individual fish, and therefore, a more robust analysis of growth rates. Compared to the number of Northern Pike used in Chapter 2 (n = 535), Northern Pike used in this data chapter were individuals for which cleithra were available (n = 432). Whole cleithra were hand-cleaned to remove soft tissue after being immersed in warm water for 10-20 seconds. While placed beside a fixed ruler on a lab bench, air-dried cleithra were then photographed using a Nikon D7500 DSLR digital camera (Japan). Widths of annuli (i.e., yearly growth increments interpreted as alternating translucent and opaque zones under reflected light) were measured (in mm) from the origin to the outermost edge of cleithra along the anterior length (Craig, 1996), with age calculated as the number of annuli. The outer limit of each translucent zone was considered as the formation point of the next annulus. For quality control, images were examined blind by an experienced reader twice, with a 3-week time interval between reads. Samples were examined for a third time if age estimates differed between the first two reads, and samples were not used in further analyses if the third reading did not agree with one of the first two.

Analyses of Chl-a, TP, and TN were conducted at the University of Alberta Biogeochemical Analytical Service Laboratory in Edmonton (AB, Canada). Briefly, Chl-a was extracted with 95% ethanol on a filter overnight and measured using a Shimadzu® RF-1501 Spectrofluorophotometer (North America Analytical & Measuring Instruments), and TP and TN analyses were performed by Flow Injection Analysis using a Lachat® QuikChem 8500 FIA automated analyzer (Canada). Analyses of DIC, DOC, SUVA, abs254, Freshness Index, Na, Ca, Mg, Cl, alkalinity, bicarbonate, and conductivity were conducted at the Western University Biotron Center for Experimental Climate Change Research in London (ON, Canada). Concentrations of DIC and DOC were determined using phosphoric acid digestion followed by persulphate oxidation on an Aurora® 1030 W total organic carbon analyzer (USA). Fluorescence and absorbance measures were made with appropriate corrections on a Horiba Aqualog® spectrofluorometer (Japan). Absorbance at 254 nm (abs254) is reported directly as an absolute measure of total absorbance characteristics of higher molecular weight DOM. Freshness Index was calculated as the ratio of emission at 380 nm and the emission maximum between 420-235 nm at an excitation wavelength of 310 nm. Specific absorbance at 254 nm (SUVA), a measure of the degree of aromaticity of DOM, was calculated as abs254 divided by DOC concentrations (Weishaar et al., 2003). Analyses of Na, Ca, Mg, Cl, alkalinity, bicarbonate, and conductivity were performed using a Dionex ICS-1600 reagent-free ion chromatography system (USA) and a AS-50 autosampler (USA).

3.2.3 Lake trophic states

Trophic state was quantified for each lake using Carlson's Trophic State Index (CTSI; Carlson 1977) and measures of Chl-a, TP, and Secchi depth (Secchi) with the following equations:

$$\mathrm{CTSI} = \frac{\mathrm{TSI}_{\text{Chl-a}} + \mathrm{TSI}_{\mathrm{TP}} + \mathrm{TSI}_{\mathrm{Secchi}}}{3}$$

$$\mathrm{TSI}_{\text{Chl-a}} = 9.81\ ln\ \text{Chl-a}\ (\mu g/L) + 30.60$$

$$\mathrm{TSI}_{\mathrm{TP}} = 14.42\ ln\ \mathrm{TP}\ (\mu g/L) + 4.15$$

$$\mathrm{TSI}_{\mathrm{Secchi}} = 60.00 - 14.41\ \text{secchi depth}\ (m)$$

where ln was natural logarithm. Following Fuller and Jodoin (2016), lakes were then classified into one of four categories: oligotrophic (i.e., low productivity) for CTSI $<$ 38, mesotrophic (i.e., moderate productivity) for 38 $<$ CTSI $>$ 48, eutrophic (i.e., high productivity) for 48 $<$ CTSI $>$ 61, and hypereutrophic (i.e., extreme productivity) for CTSI $>$ 61. The ratio of TN to TP (TN:TP) was used to assess whether nitrogen or phosphorus limited primary production in each lake (e.g., Hecky et al., 1993).

3.2.4 Geospatial analysis

We used publicly available geospatial data on the Government of Canada portal to identify ecoregions and quantify lake and catchment characteristics. Data were accessed in May 2019 and analyzed using the coordinate system ESPG:3581 NAD83(CSRS)/NWT Lambert. For each lake, we calculated total catchment area (CA) based on the HRDEM v1.3 (HRDEM, 2021). This same source was used to delineate watershed divides, and obtain mean elevation and mean slope in most catchments, but the headwaters of Kakisa and Tathlina lakes are not included in this data layer; thus, the Canadian Digital Elevation Model 1945-2011 (CDEM, 2021) was used for these lakes. Lake surface areas (LA) and land cover compositions for each catchment were calculated based on the 30-m resolution 2015 Land Cover of Canada data layer (LCC, 2021b). Land cover in our study area included 10 categories: Wetland, Water, Temperate or Subpolar Shrubland, Temperate or Subpolar Needleleaf Forest, Temperate or Subpolar Broadleaf Deciduous Forest, Subpolar Taiga Needleleaf Forest, Subpolar or Polar Shrubland, Subpolar or Polar Grassland, Mixed Forest, and Barren Land. Detailed information about each category is available in (Natural Resources Canada, 2019). Land cover data were expressed as a percentage of CA. Finally, the ratio of lake area to catchment area (LA:CA) was calculated by dividing LA to CA. Analyses of lake and catchment characteristics were done using QGIS v3.12.2 (QGIS Development Team, 2020), GDAL/OGR (GDAL/OGR Contributors, 2020), PostgreSQL/PostGIS v2.5.4 (PostGIS Development Group, 2020), and WhiteboxTools (Lindsay, 2016), as well as with R packages raster v3.3-13 (Hijmans, 2020) and rgdal v1.5-16 (Bivand, 2020).

3.2.5 Data analysis

Statistical analyses were completed using RStudio v1.3.959 (R Studio Team, 2020) integrated with R v4.0.1 (R Core Team, 2020) and core packages, unless otherwise noted. Residuals of all linear models were visually assessed using residual plots, and tested for normality using Shapiro-Wilk tests. Data were $\log_{10}$-transformed where necessary to meet assumptions.

Lake-specific growth rates in Northern Pike were quantified using the following equation proposed by Gallucci and Quinn (1979):

$$\text{Size} = \frac{\omega}{k} \times (1 - e^{\,k \,\times\, \text{age}})$$

where k is the growth coefficient parameter of the von Bertalanffy growth equation (von Bertalanffy, 1938), and ω is an estimate of early growth rate, in a length-per-time unit. The ω growth parameter was chosen for subsequent comparisons among lakes because: i) it is statistically robust, and its use in comparisons avoids problems of analyzing and interpreting the interdependent growth coefficient and asymptotic length parameters estimated by the von Bertalanffy growth equation (Gallucci and Quinn 1979); ii) it facilitates comparisons with similar studies (e.g., Benoît et al., 2016); and, iii) ω provides an estimate of growth rate in early life, when environmental conditions are particularly influential on fish growth rates (Pankhurst and Munday, 2011). To fit growth models, we used incremental width data, which were measurements of individual annuli (in mm) made on images of cleithra using the package RFishBC v0.2.3 (Ogle, 2019). This avoided the bias that can be introduced when fish size at previous ages is estimated through back-calculation

(Francis, 1990; Höhne et al., 2020). Fish size at a given age was represented by the size of cleithra at a given annulus (i.e., distance from the origin to ith annulus of cleithra along the anterior length). To check the assumption that size of cleithra represented the size of fish, distance from the origin to the outermost edge of cleithra along the anterior length was regressed against FL of corresponding individuals. A significant and positive linear relationship was found ($y = 24.292 + 9.83x$, adjusted $r^2 = 0.983$). Incremental data did not appear to suffer from the Rosa Lee phenomenon (i.e., smaller estimated sizes for a given age in older fish compared to younger fish), according to inspection following Duncan (1980). Age 0^+ Northern Pike were excluded from growth analyses, as were annuli that represented growth (since winter annulus formation) in the year of capture. Lake-specific growth models were fit with the package FSA v0.8.30 (Ogle et al., 2020) using incremental data for all fish in a given lake.

Relationships among the physiochemical variables for all lakes and their catchments were investigated using two principal component analyses (PCA) that were performed in the package vegan v2.5-6 (Oksanen et al., 2019). First, a PCA was completed on land cover data. A second PCA was performed using lake chemistry variables, including Chl-a, DOC, DIC, abs254, Freshness Index, SUVA, TP, TN, Na, Ca, Mg, Cl, alkalinity, bicarbonate, and conductivity. Lake area to catchment area ratios, mean catchment elevation, mean catchment slope, and lake-specific PC1 and PC2 scores of the land cover PCA were also included as variables in this second PCA. All data were z-transformed (i.e., mean-subtracted and divided by standard deviation) before PCAs were completed.

To investigate whether among-lake variation in growth rate of Northern Pike could be explained by variability among lakes in primary productivity, water clarity, inferred terrestrial influence, or fish trophic ecology, lake-specific estimates of growth rate (ω) in

Northern Pike were related to Chl-a, Secchi depth, DOC concentrations, PC1 and PC2 scores from the second PCA (inferred catchment influence and in-lake productivity, respectively; see results), and Northern Pike trophic level (δ^{15}N) and dietary carbon source (δ^{13}C). Simple linear regressions (LR) were used, and alpha was set at 0.05. Each lake was treated as a replicate ($n = 11$). Compared to median FL of Northern Pike in Chapter 2 (580 mm for n = 535 individuals), median FL of all (n = 432) Northern Pike individuals included in this chapter was 562 mm (mean = 565.5 mm), a size that was observed in all of the studied lakes. To avoid extrapolation of stable isopote ratios beyond size of captured Northern Pike, and because fish size can affect trophic ecology, ratios of δ^{15}N and δ^{13}C in Northern Pike were thus estimated at 560 mm FL using analyses of covariance (i.e., ANCOVA) in the package lsmeans v2.30-0 (Lenth, 2016). Ratios of δ^{15}N for individual Northern Pike were first adjusted for among-lake variation in baseline ($\delta^{15}\text{N}_{\text{base}}$) (Post, 2002) by using the following equation:

$$\delta^{15}\text{N}_{\text{adj}} = \delta^{15}\text{N} - \text{lake mean } \delta^{15}\text{N}_{\text{base}}$$

Ratios of δ^{15}N in samples from the Sphaeriidae family were used for baseline adjustments (Post, 2002). Following initial analyses focused on Northern Pike, linear regressions were also used to relate δ^{13}C ratios in benthic invertebrates to PC1 and PC2 scores from the second PCA.

3.3 Results and Discussion

3.3.1 Characteristics of lakes and inferred catchment influence

There was considerable variability in physical and chemical characteristics among the studied lakes and catchments (Table C.2 and Figure 3.2). Lake and catchment areas varied by more than two orders of magnitude; LA ranged from 1 to 565 km^2, and CA ranged from 113 to $15,809$ km^2 (Table C.2). The LA:CA varied from 0.002 to 0.161, with extremes represented by McGill Lake (small lake with large catchment) and Big Island Lake (medium lake with small catchment). Lakes located in the Horn Plateau ecoregion were an order of magnitude larger relative to their catchments than lakes located in other ecoregions (Table C.2 and Figure 3.2).

Lakes located on the Horn Plateau, as well as Fish Lake, located in the Norman Range ecoregion, had different catchment land cover compared to lakes located in the other ecoregions (Figure 3.2). These higher-elevation/latitude lakes (i.e., Big Island, Mustard, Willow, and Fish) were clearly separated from lakes located in the Hay River Lowland (i.e., Ekali, Sanguez, Gargan, and McGill, Trout, Kakisa, and Tathlina) along the horizontal axis (i.e., PC1) of the land cover PCA (Figure 3.3a). Subpolar/polar land cover types were negatively associated with PC1 whereas temperate/subpolar land cover types were positively associated with PC1, and this axis explained 65% of total variance in land cover composition (Figure 3.3a). For example, while subpolar taiga Needleleaf forest covered 10-20% of catchment areas of lakes located at higher elevations or at higher latitudes (Big Island, Willow, Mustard, Fish), this forest type covered only 0.01-1% of the catchment areas for lakes located in the Hay River Lowland (i.e., Ekali, Sanguez, Gargan, and McGill, Trout, Kakisa, and Tathlina) (Figure 3.2). Variability in the proportion of wetland area

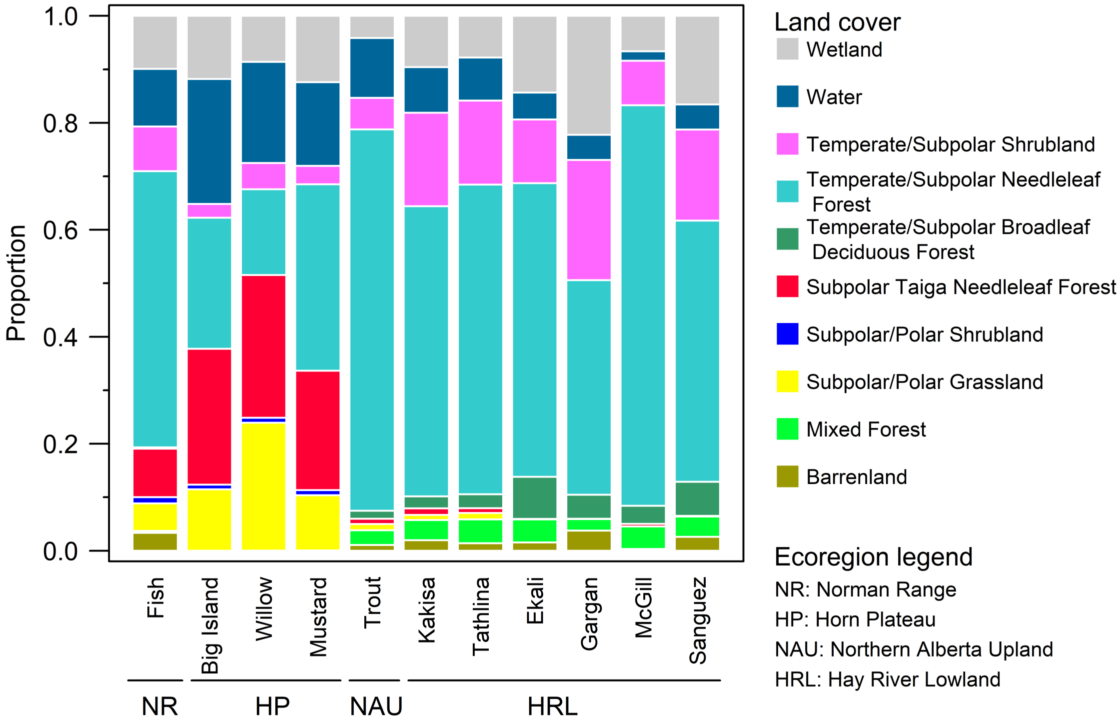


Figure 3.2. Proportion of land cover composition relative to catchment area for each study lake in the Dehcho Region, NT, Canada. Lakes are grouped by ecoregions; ecoregions are indicated beneath the X-axis.

primarily drove separation along PC2, which explained another 20% of variance in catchment land cover (Figure 3.3a). Lake-specific PC1 and PC2 scores of the land cover PCA were extracted and renamed "region" and "wetland", respectively (Figure 3.3a), for use in the second PCA (Figure 3.3b).

Patterns among lakes and ecoregions in catchment size and composition were reflected in water chemistry, and results from the second PCA reflect a gradient of catchment influence along PC1 (Figure 3.3b). Lakes located in larger catchments (relative to lake area) and with steeper-sloped catchments were characterized by higher levels of tracers that reflect terrestrial inputs, including higher levels of ions, DOC, DIC, alkalinity, conductivity, and abs254 (Table C.2 and Figure 3.3b). Variables such as conductivity,

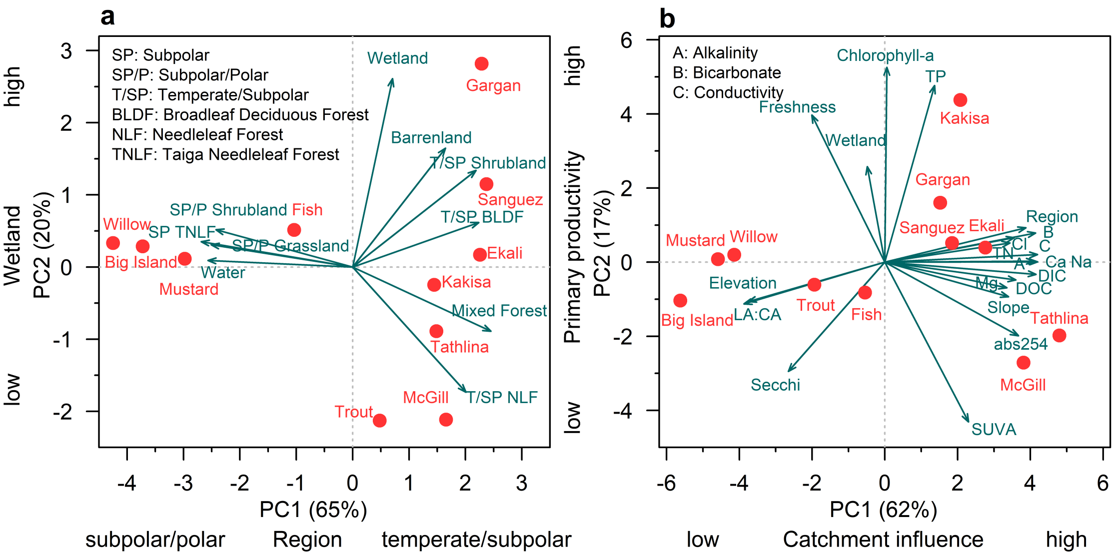


Figure 3.3. Biplots of Principal Component Analyses (PCA). Left panel **(a)** depicts PCA performed using proportional land cover data (i.e., Figure 3.2). Right panel **(b)** depicts PCA performed using all variables presented in Table C.2, as well as PC1 and PC2 scores from the analyses depicted in (a). Axes of the land cover PCA (a) were inferred to represent variability among ecoregions (PC1) and proportional cover of wetlands (PC2). Axes of the PCA that included all variables (b) were inferred to represent a gradient of catchment influence (PC1) and primary productivity (PC2).

alkalinity, bicarbonate, Ca, Mg, and Cl are reflective of mineral weathering in the catchment (e.g., Kamenik et al., 2001; Singh et al., 2008), and have previously been used as indicators of catchment influence on lake ecosystems in studies of fish ecology (e.g., Burke et al., 2020). The horizontal axis (PC1) of the second PCA explained 62% of total variance in the dataset, and while elevation and LA:CA were negatively associated with this axis, mean catchment slope, TN, DOC, DIC, abs254, alkalinity, bicarbonate, conductivity, Na, Ca, Mg, and Cl were all positively associated with this axis (Figure 3.3b). Higher-elevation/latitude lakes (i.e., Big Island, Mustard, Willow, and Fish) were negatively associated with PC1 whereas all lakes located in the Hay River Lowland (i.e., Ekali, Sanguez, Gargan, and McGill, Kakisa, and Tathlina) were positively associated with PC1. We thus infer that PC1 reflects a gradient of catchment influence from low (negative scores) for lakes in the Horn Plateau and Norman Range ecoregions, to high (positive scores) for lakes in the Hay River Lowland. Trout Lake, located in the Northern Alberta Upland ecoregion, grouped with the higher elevation/latitude lakes, which likely reflects its relatively large LA to CA ratio (Table C.2 and Figure 3.3b).

The vertical axis (PC2) of the second PCA explained 17% of variation in this analysis, and was driven by proxies and indicators of in-lake primary production, including Chl-a concentrations and Freshness Index (a measure of the ratio of more recently produced to more decomposed DOM) (Figure 3.3b). Primary production varied independently of the patterns observed among ecoregions for catchment influence and LA:CA; both the lowest and highest Chl-a values were observed for lakes in the Hay River Lowland (McGill and Kakisa, respectively). Positive and close associations of TP and Chl-a along PC2 (Figure 3.3b) suggested that primary productivity in most of the studied lakes was limited by phosphorus, an observation that was indeed supported by results of TN:TP calculations. With TN:TP $>$ 20, ten of eleven studied lakes were found to be phosphorus-limited (Hecky et al. 1993), and mesotrophic or moderately productive (Table C.3). Kakisa Lake, however, was nitrogen-limited (i.e., TN:TP $<$ 20; Hecky et al., 1993), and eutrophic or highly productive (Table C.3).

3.3.2 Northern Pike growth rates

A total number of 2953 annuli widths were measured on Northern Pike cleithra (n $=$ 432) and used to model growth rates in Northern Pike. Detailed descriptive statistics of incremental width data for annuli are presented in Table C.4. Northern Pike growth rate (i.e., ω) varied twofold among lakes, and was lowest (10.3) in Tathlina Lake and highest (20.9) in Mustard Lake (Table C.2). Growth rates of Northern Pike, estimated using

Table 3.1. Estimates and ±95% confidence intervals of early growth rate (ω) for Northern Pike in the study lakes. Ecoregions include Norman Range (NR), Horn Plateau (HP), Northern Alberta Upland (NAU), and Hay River Lowland (HRL).

Lake (ecoregion)	Early growth rate (ω)		
	Estimate	LCI	UCI
Fish (NR)	13.36	12.17	14.57
Big Island (HP)	17.84	15.04	20.88
Willow (HP)	14.48	13.14	15.96
Mustard (HP)	20.86	16.68	25.62
Trout (NAU)	15.72	13.66	18.12
Kakisa (HRL)	13.29	12.34	14.35
Tathlina (HRL)	10.29	9.54	11.02
Ekali (HRL)	10.19	9.23	11.24
Gargan (HRL)	11.26	9.86	12.57
McGill (HRL)	11.08	10.09	12.06
Sanguez (HRL)	10.37	9.92	10.81

increment width data in this data chapter, were significantly correlated to the age-at-size estimations that were used as a proxy for Northern Pike growth in Chapter 2 (Pearson $r = -0.949$, t = −6.521, df = 9, $p < 0.005$).

While there was no clear relationship between Northern Pike growth rates and either in-lake primary production or water clarity, there was a statistically significant relationship between growth rates in Northern Pike and the inferred catchment influence. Log_{10}-transformed Northern Pike growth rate was significantly and negatively (LR: $F_{1,\,9} = 36.80$, $p < 0.001$, adj. $r^2 = 0.78$; Figure 3.4a) related to PC1 scores from the second PCA (Figure 3.3b), which we inferred to reflect catchment influence; growth rates in Northern Pike were higher in lakes with lower inferred catchment influences. Consistent with this, we also found a significant negative relationship ($F_{1,\,9} = 18.44$, $p = 0.002$, adj. $r^2 = 0.64$; Figure 3.4b) between ω ($\log_{10}$) and DOC ($\log_{10}$) concentrations. While previous researchers have found that catchment influence or DOC concentrations have affected growth rates in other fish species, such as Pikeperch (Ranåker et al., 2014), Walleye (Stasko et al., 2015; Benoît et al., 2016), Lake Trout (King, 2007), Bluegill (*Lepomis macrochirus*) (Craig et al., 2017), and European Perch (van Dorst et al., 2019) through effects on primary productivity and/or water clarity, our results suggest that this was not the case in our study. There were no significant relationships between Northern Pike growth rates ($\log_{10}$) and Chl-a ($\log_{10}$), Carlson's trophic state index, PC2 (Chl-a, Freshness In-

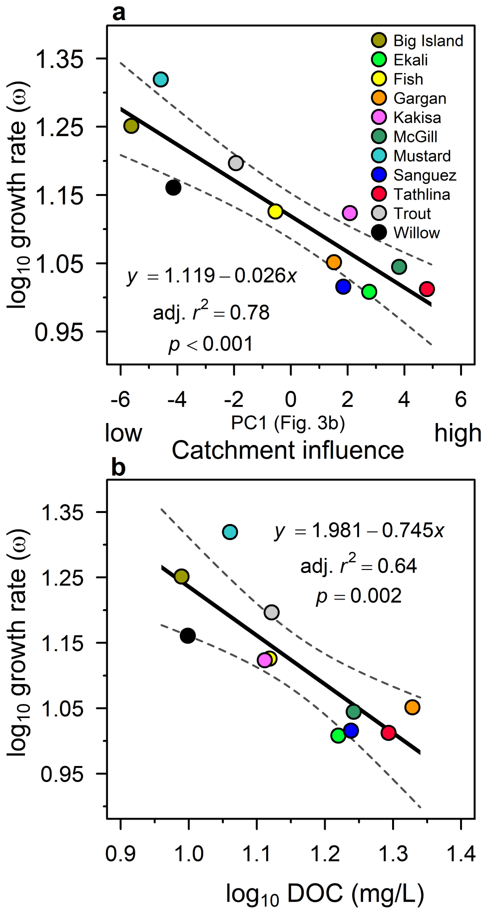


Figure 3.4. Linear regressions between lake-specific estimates of Northern Pike growth rate (ω) and **(a)** inferred catchment influence (PC1 scores in Figure 3.3b), and **(b)** concentrations of dissolved organic carbon in lake water. Solid lines represent fitted regressions and dashed lines represent ±95% confidence intervals.

dex, TP, SUVA; Figure 3.3b), or secchi depth (LR, $F_{1,\,9} = \leq 0.08$, $p \geq 0.269$, adj. $r^2 \leq 0.04$), and there were also no significant relationships between DOC ($\log_{10}$) and Chl-a ($\log_{10}$) (LR: $F_{1,\,9} = 0.20, p = 0.663$), or between DOC ($\log_{10}$) and water clarity (i.e., Secchi) (LR: $F_{1,\,9} = 1.64$, $p = 0.232$). In these generally well-mixed and relatively shallow lakes, it is likely that water clarity is influenced more by suspended sediments than DOC concentrations, although data that span more of the open-water season would be required to test this assertion.

One possible explanation for the negative relationship between Northern Pike growth rates and inferred catchment influence is the quality of terrestrially derived basal food resources. There was a strong positive correlation (Pearson $r = 0.93$, $p < 0.001$) between DOC ($\log_{10}$) and abs254 ($\log_{10}$), and between DOC and other variables indicative of catchment weathering (e.g., conductivity, alkalinity, bicarbonate, Ca, Mg, and Cl; Kamenik et al., 2001; Singh et al., 2008) along PC1 (Figure 3.3b), which indicates that the greater the catchment influence, the greater the inputs of more aromatic, high molecular weight terrestrial organic matter. Organic matter derived from terrestrial environments is often of relatively lower nutritional quality compared to DOM derived from internal production, which provide relatively higher levels of essential amino acids and macronutrients (Taipale et al., 2014; Brett et al., 2017). Further, elevated quantities of DOC from terrestrially derived sources do not compensate for the poorer quality of these resources (Hiltunen et al., 2019).

Our proposed mechanism for the negative relationship between Northern Pike growth rates and inferred catchment influence (i.e., lower-quality DOM from the catchment) is consistent with results of our analysis on Northern Pike growth rates and trophic ecology. Lake-specific growth rate in Northern Pike (i.e., $\log_{10}\,\omega$) was not related to lake-specific least squares mean of either adjusted (LR: $F_{1,\,9} = 0.79$, $p = 0.398$) or unadjusted (LR: $F_{1,\,9} = 0.074$, $p = 0.792$) δ^{15}N. Both lake-specific least squares mean $\delta^{15}N_{adj}$ and δ^{15}N in Northern Pike varied less than three ‰(∼one trophic level), indicating that Northern Pike feed at approximately the same trophic level in all studied lakes. There was, however, a significant and positive relationship between lake-specific $\log_{10}\,\omega$ and least squares mean δ^{13}C (LR: $F_{1,\,9} = 31.08$, $p < 0.001$, adj. $r^2 = 0.75$; Figure 3.5a). To determine if the positive relationship between Northern Pike growth and δ^{13}C reflected among-lake differences in habitat use of Northern Pike (i.e., the more conventional interpretation) or among-lake differences in sources of DIC, we related δ^{13}C of a representative baseline benthic invertebrate family (lake-specific arithmetic mean δ^{13}C of Sphaeriidae) to inferred catchment influence (i.e., PC1 scores of the second PCA), with which DIC was strongly and positively associated (Figure 3.3b).

There was a significant negative relationship between δ^{13}C of Sphaeriidae and inferred catchment influence (LR: $F_{1,\,9} = 33.19$, $p < 0.001$, adj. $r^2 = 0.77$; Figure 3.5b). Negative relationships between inferred catchment influence and δ^{13}C were also observed when this analysis was conducted on other benthic invertebrate taxa that were captured in most or all lakes (Chironomidae, Dytiscidae, Corixidae, and Lymnaeidae), as well as when all benthic invertebrate taxa were combined (Table C.5 and Table C.6). Relationships were stronger when analyses were restricted to individual taxonomic families, which is not surprising given the diversity of feeding tactics among analyzed taxa, and the fact that fractionation of carbon isotopes (^{13}C and ^{12}C) can vary among functional feeding groups (Woodland et al., 2012; Anas et al., 2019b). Overall, the results indicate that benthic invertebrates had more depleted δ^{13}C ratios in lakes subject to more catchment influence, which is in consistent with the findings of Anas et al. (2019a). Given that δ^{13}C

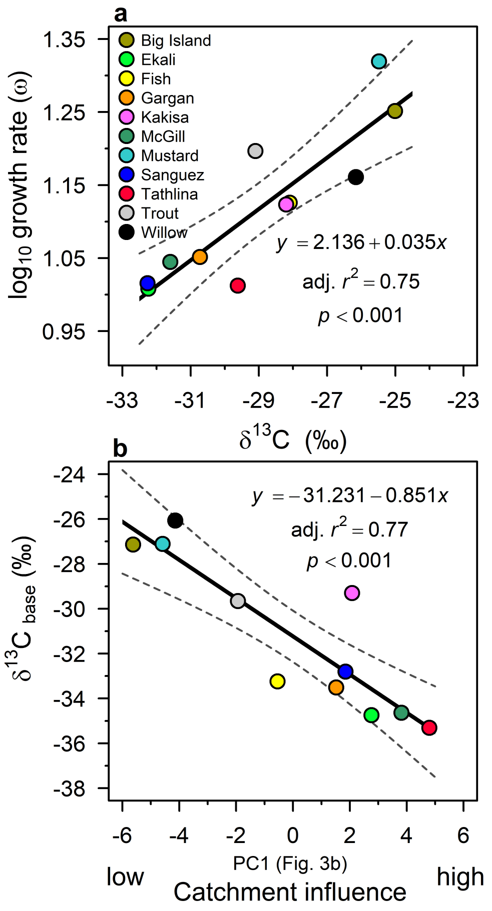


Figure 3.5. Results of linear regressions between **(a)** Northern Pike growth rate and least squares mean δ^{13}C (calculated at 560 mm FL), and **(b)** mean δ^{13}C of baseline benthic invertebrates (Sphaeriidae) and inferred catchment influence (PC1 scores from in Figure 3.3b). Solid lines represent fitted regressions and dashed lines represent ±95% confidence intervals.

ratios of benthic invertebrates varied among lakes in a pattern similar to that of Northern Pike, and that a large proportion of this variability was explained by inferred catchment influence (e.g., 77% for Sphaeriidae), we infer that among-lake variability in δ^{13}C ratios in Northern Pike reflected variability in the sources of DIC, rather than variability in habitat use. This inference is supported by a significant, strong, and positive (Pearson $r = 0.93$, df $= 9$, $p < 0.001$) relationship between lake-specific δ^{13}C ratios in Northern Pike and lake-specific δ^{13}C ratios in littoral benthic invertebrates (all taxa combined).

Benthic consumers reliant on terrestrially derived organic matter tend to be relatively less nutritious (in terms of fatty acids and other macro nutrients) than those assimilating internally produced resources (e.g., Lau et al., 2009), although terrestrially derived organic matter can support consumer growth and production to some extent (e.g., Kelly et al., 2014). Given the low nutritional value and depleted δ^{13}C ratios previously observed in benthic consumers that rely more on terrestrial resources (Lau et al., 2009; Anas et al., 2019a), we suggest that our observation of slower growth of Northern Pike in lakes with greater inferred catchment influence reflects relatively lower food quality that propagates through trophic levels. Further research, ideally studies that include fatty acid concentrations, would provide an independent test of our inference of lower nutritional quality in basal resources of lakes that are more influenced by their catchments. Further, we suggest that future research into relationships between terrestrial influence and δ^{13}C ratios in fishes should ideally include species that are known to feed mostly in littoral habitats, (e.g., pike), mostly in pelagic habitats (e.g., adult cisco), and in both pelagic and littoral habitats (e.g., lake trout); as pike are known to occupy and feed mostly in littoral habitats and have relatively small home ranges, effects of whole-lake drivers (such as terrestrial influence) — especially those that affect nutrient cycling in the littoral zone — may be easier to detect.

Population density, prey-predator interactions, and presence of submerged aquatic plants are all known to strongly influence growth rate in Northern Pike as an ambush predator (Vøllestad et al., 1986; Diana, 1987; Casselman and Lewis, 1996; Craig, 1996; Margenau et al., 1998). Our sampling relied heavily on subsistence harvest practices, and was not designed to allow quantification of fish community composition or relative abundance. Qualitative information retrieved from our gill nets (Table C.7), however, indicate that Lake Whitefish are present in the majority of study lakes (n lake = 10 of 11); juvenile Lake Whitefish are a preferred prey item for Northern Pike (Scott and Crossman, 1973). Other commonly-observed small-bodied fish species (potential prey items for Northern Pike) include Cisco (*Culaea inconstons*) and Ninespine Stickleback (*Pungitius pungitius*). Diversity of forage fish species appears to vary among lakes (Table C.7), however, and this likely affects growth of Northern Pike in a way that we cannot currently quantify. Presence of potential competitors also varied among lakes; we observed Lake Trout (*Salvelinus namaycush*) in lakes where Northern Pike grew relatively faster (Big Island, Willow, and Mustard lakes), and Walleye (*Sander vitreus*) in lakes where Northern grew relatively slower (e.g., Sanguez, Tathlina, Ekali, and McGill lakes), but additional sampling would be required to test predictions related to this observation. Future research on presence/absence and relative abundance of competitor fishes and prey fishes, as well as coverage of submerged aquatic vegetation, could yield additional insights into among-lake variability that we cannot yet explain in Northern Pike growth rates in these remote and understudied lakes that support food security and overall well-being of northerners in the region.

3.4 Conclusion

Growth rates of Northern Pike were lower in lakes with greater inferred catchment influence, which were smaller lakes surrounded by relatively larger, lower-elevation, and more-forested catchments, likely because of lower nutritional value and less bioaccessibility of catchment-derived organic matter relative to internally-produced organic matter. The Dehcho Region is located at northern latitudes, where climate warming is having profound effects on lake-catchment interactions via accelerated permafrost thaw, and changes in fire regimes and hydrology (Prowse et al., 2006; Hugelius et al., 2020). To our knowledge, this is the first study that integrates food web, water, lake, and catchment data to explore causes of among-lake variability in growth of Northern Pike, an important subsistence and commercial species across northern latitudes. Our results revealed that growth rate in Northern Pike decreased with increasing catchment influence on lake water chemistry, which occured in smaller lakes surrounded by relatively larger, lower-elevation, and more-forested catchments. The negative relationship between growth rate of Northern Pike and inferred catchment influence may be driven by the lower nutritional value and less bioaccessibility of terrestrially derived organic matter compared to more readily bioaccessible organic matter that is produced within the lake. Further research that includes fatty acid concentrations in muscle tissues and prey-predator interactions could lend further insight into among-lake variability in growth rates of Northern Pike. As many lake and catchment attributes included in our study can be quantified and monitored remotely (e.g., remote sensing), our findings may accelerate development of models for growth rate, especially in remote regions where field data are difficult to obtain and where fish are a primary subsistence food source. Our findings also aid in identifying and predicting faster and slower growing populations of Northern Pike in subarctic lakes, which is of significance for toxicologists and fisheries managers, given the vital regulatory role of growth rate in fish population dynamics, fisheries sustainability, and contaminant accumulation in fish tissues.

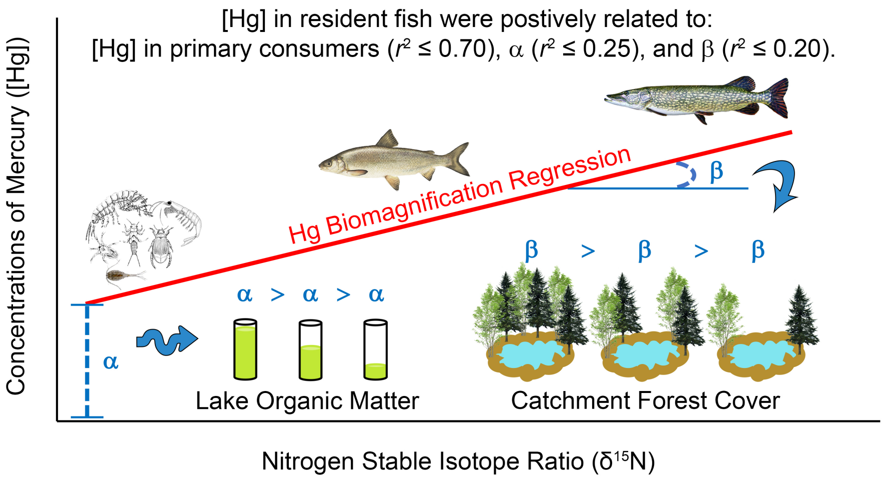
Chapter 4

Biomagnification of mercury through lake food webs in the Canadian subarctic

This chapter is an adapted version of the following in-review manuscript[1]:

Moslemi-Aqdam M., Low M., Low G., Laird B. D., Branfireun B. A., and Swanson H. K. Biomagnification of mercury through lake food webs in the Canadian subarctic. *Science of the Total Environment*, submitted on 31 May 2022.

Graphical Abstract



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[1] *under review in Elsevier.*

4.1 Introduction

Mercury (Hg) in aquatic food webs is a global concern because the organic and more toxic form of Hg, methylmercury (MeHg), can biomagnify to potentially harmful levels in fish, fish-eating wildlife, and humans (Driscoll et al., 2013). In lakes, accumulation of Hg in fish varies among systems in response to a variety of ecosystem characteristics. For example, in-lake variables, such as primary productivity, light penetration, pH, and concentrations of dissolved organic matter (often measured as dissolved organic carbon (DOC)) and inorganic Hg affect net methylation rates (Ullrich et al., 2001; Paranjape and Hall, 2017), uptake into lower-level biota (Pickhardt et al., 2002; Chen and Folt, 2005), and ultimately accumulation of Hg in higher trophic level fish (Eagles-Smith et al., 2016; Eklöf et al., 2016; Thomas et al., 2020; Moslemi-Aqdam et al., 2022). These in-lake variables are influenced, in turn, by characteristics of surrounding catchments, such as topography, relative surface area, and land cover composition (Kamenik et al., 2001; Sobek et al., 2007; Williams et al., 2010; Bravo et al., 2017; Denkenberger et al., 2020). Catchments with higher proportional areas of forest and/or wetland have higher Hg concentrations in resident fish, and that these relationships are driven by catchment influences on Hg loading, net methylation, uptake into biota, and accumulation through food webs (Eagles-Smith et al., 2016; Eklöf et al., 2016; Thomas et al., 2020; Moslemi-Aqdam et al., 2022). Whereas factors and mechanisms regulating Hg concentrations in fish have been extensively studied and reviewed (Chételat et al., 2015; Eagles-Smith et al., 2018), studies of the factors that control or affect rates of Hg biomagnification through lake food webs are more limited. This is particularly true in northern regions, where environmental perturbations are having measurable effects on ecosystem characteristics and processes (Rydberg et al., 2010; Hugelius et al., 2020), and where locally-harvested fish are a dietary mainstay

(Donaldson et al., 2010; Pirkle et al., 2016).

Rates of Hg biomagnification through lake food webs are often investigated in one or a few lakes per study, and findings from broader comparison studies that have been conducted are inconsistent. In a global meta-analysis, rates of Hg biomagnification through lake food webs were positively related to concentrations of chlorophyll-a and dissolved organic carbon (DOC), but unrelated to concentrations of total phosphorus, Hg in water and sediment, and lake area, catchment area, or percent wetland area in catchments (Lavoie et al., 2013). In another study among temperate lakes in central Canada, positive relationships were found between rates of Hg biomagnification and both total phosphorus concentration and lake area (Kidd et al., 2012). Whereas Kozak et al. (2021) reported higher Hg biomagnification rates with increasing productivity along a climate-productivity gradient in lakes in Fennoscandia, Clayden et al. (2013) observed lower rates of Hg biomagnification in more productive lakes in eastern Canada. In a study of lakes in Arctic Canada, Gantner et al. (2010) reported no relationships between rates of Hg biomagnification and ecosystem characteristics, including lake area to catchment area ratios, and concentrations of DOC and chlorophyll-a. While broadly generalizable patterns on controls of Hg biomagnification rates through lake food webs cannot be gleaned from these studies, findings to date suggest that rates of Hg biomagnification through lake food webs are controlled by complex mechanisms that operate both inside and outside lake boundaries. Dominant drivers likely vary among regions, with the spatial scale investigated, and with the magnitude of variability in possible predictor variables. Understanding controls of Hg biomagnification through lake food webs thus requires comprehensive data along known or suspected gradients in lake and catchment physicochemistry; these data are challenging to obtain, particularly in remote locations.

One of the sources of uncertainty in estimates and comparisons of Hg biomagnification rates is the species of Hg that is measured and used in analyses. The rate at which Hg biomagnifies through aquatic food webs is often quantified by a simple linear regression between Hg concentrations ($\log_{10}$-transformed) and stable nitrogen isotope ratios (δ^{15}N; indicative of relative trophic position of consumers in food webs (Minagawa and Wada 1984) in tissues of biota (Kidd et al., 2012; Clayden et al., 2013; Kozak et al., 2021). Relying on an assumption that approximately 95% of total Hg in fish muscle tissue is in the form of MeHg (Bloom, 1992), one common convention in quantifying rates of Hg biomagnification is to use concentrations of total mercury ([THg]; an analytical reporting convention for the sum of all readily digested and oxidized Hg species) in fish and concentrations of MeHg ([MeHg]) in lower trophic level biota (e.g., benthic invertebrates) (Kidd et al., 2012; Clayden et al., 2013; Kozak et al., 2021). Recent studies have shown, however, that the percent MeHg of total Hg (%MeHg) in fish can vary among water bodies, and among individuals and species of fish (Stefansson et al., 2013; Peng et al., 2016; Arcagni et al., 2018; Lescord et al., 2018). For example, a 2018 study on several freshwater fish species from subarctic lakes in Canada showed considerable inter- and intra-specific variability in %MeHg; %MeHg was generally higher in larger, older, and pelagic feeding individuals/species (Lescord et al., 2018). Despite the growing evidence of variability in %MeHg in fish, the consequences of using fish [THg] versus fish [MeHg] in generating estimates of Hg biomagnification rates are poorly understood and have received relatively little research attention (but see Wyn et al., 2009).

Locally harvested fish from lakes in the Canadian subarctic are critical to culture, economy, and food security of Indigenous communities (Kuhnlein and Receveur, 1996; Fieldhouse and Thompson, 2012). To date, studies on rates and controls of Hg biomagnification through food webs in lakes of the Canadian subarctic are lacking. Accordingly, critical knowledge gaps exist regarding magnitude of variability in Hg biomagnification rates among lakes, causes of the variability among lakes, and the degree to which this variability explains Hg concentrations in resident subsistence fish species (see Moslemi-Aqdam et al., 2022). Equipped with comprehensive geospatial, environmental, and biological data, we investigated rate of Hg biomagnification through food webs in ten remote subarctic lakes across a wide geographic area ($\sim 60,000$ km^2) in Northwest Territories, Canada. Lake and catchment characteristics, as well as [THg] and [MeHg] in biotic and abiotic ecosystem compartments, vary widely in the Dehcho Region (see Moslemi-Aqdam et al., 2021, 2022). We investigated whether using [THg] versus [MeHg] measured in fish influenced estimates of Hg biomagnification rates, and if rates of Hg biomagnification explained among-lake variability in [THg] in resident fish. We then assessed if among-lake variation in rates of Hg biomagnification could be explained by lake and/or catchment characteristics. We expected that using measured concentrations of MeHg (the biomagnifying form of Hg) in all biota would result in less uncertainty around estimates of Hg biomagnification rates, and that among-lake variability in [THg] in resident fishes would be explained, at least partially, by lake-specific rates of Hg biomagnification. Finally, we expected to observe higher rates of Hg biomagnification in lakes with higher levels of DOC, Hg, and MeHg in water, and that variability in these water chemistry variables would be related to catchment factors, such as proportion of forest land cover and relative catchment size.

4.2 Materials and Methods

4.2.1 Field sampling

As part of a large, collaborative, community-driven project on Hg concentrations in fish, ten subarctic lakes were sampled in the Dehcho Region, Northwest Territories, Canada (Figure 4.1). The studied lakes are of vital subsistence significance for First Nations communities in the area. We collected fish, primary consumer, water, and sediment samples over seven years, from 2013 to 2019 (mid-August to mid-September each year) from each of the lakes, and quantified characteristics of each lake catchment using geospatial analyses. Lakes sampled in a given year reflected community priorities and results from interim analyses. Between one and three lakes were sampled each year depending on

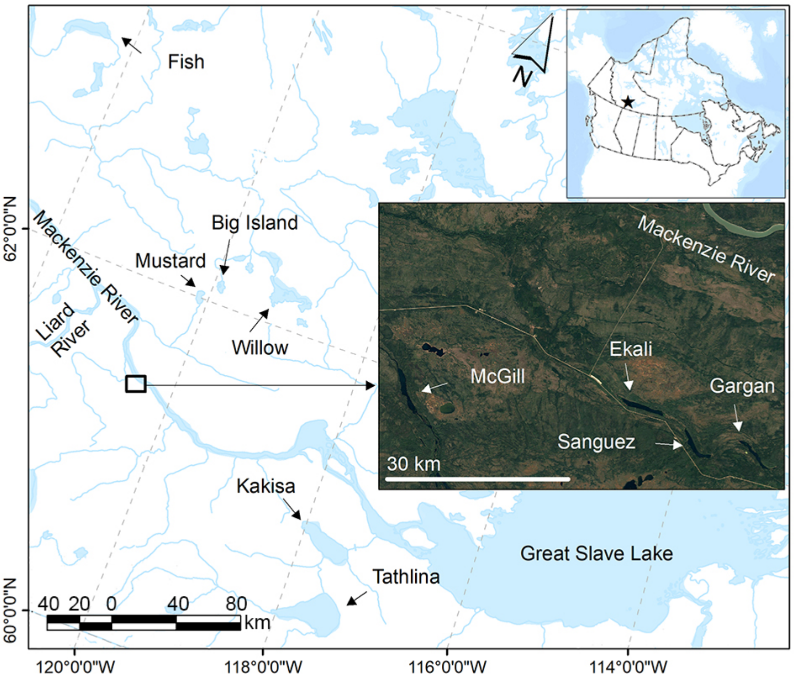

Figure 4.1. Location of study lakes in Northwest Territories, Canada (base data source: Northwest Territories Centre for Geomatics and Google earth v7.1.8).

access (i.e., float plane and remote camping vs. road access). A full schedule of sampling efforts, along with lake coordinates and modes of access are provided in Table D.1 (Appendix D).

Fish were collected by Indigenous guardians/harvesters using gill nets (stretched mesh size: 20-140 mm), and measured for fork length to the nearest mm. Skinless dorsal muscle tissue samples were extracted from individual fish, and frozen at -20 °C in the field for later analyses of THg, MeHg, and δ^{15}N. Fish were processed carefully to maintain suitability for human consumption, and were supplied, upon completion of sampling, to members of partner First Nations according to lake-specific fish consumption advisories related to Hg in the study area (Health and Social Services, 2021). Primary consumers (benthic invertebrates and zooplankton) were also collected. Benthic invertebrates were collected from both offshore (profundal; from approximate center of lakes) and nearshore (littoral) areas using Ekman dredges and dip nets (250 μm mesh size), respectively. A 63 μm mesh Wisconsin net was towed horizontally through the water column to collect bulk zooplankton; where applicable, large algal taxa were removed from samples after visual inspection. Benthic invertebrates were sorted and identified to taxonomic Family in the field. All primary consumers were stored in acid-washed (10% HCl) scintillation vials and frozen at -20 °C in the field for later analyses of MeHg and δ^{15}N. Sediment samples were collected from the approximate center of each lake using an Ekman dredge. The top 2-3 cm of sediment in the dredge was collected using an acid-washed Teflon spoon, placed in a Whirl-pak™ bag, and frozen at -20 °C for THg, MeHg, and loss on ignition (LOI) analyses.

Water samples were collected near the surface (< 30 cm depth) from the approximate center of lakes. The remote nature and logistic challenges associated with comprehensive sampling prevented sampling of all lakes within one year. We were concerned with among-lake variation in water parameters being confounded with among-year variation, however, and thus launched two additional and dedicated water sampling campaigns. In both 2018 and 2019, water samples were collected from all lakes within a period of one week in late August via floatplane (Table D.1). Both filtered and unfiltered water samples were collected (and preserved, when applicable) using standard methods that are fully described in Moslemi-Aqdam et al. (2022) or Chapter 2. Samples were kept cool for analyses of THg, MeHg, and water chemistry variables, including concentrations of chlorophyll-a (a proxy for primary production), dissolved organic carbon (DOC), and sulphate (SO_4). Water clarity was characterized using a Secchi disk at the time of water sampling. Dissolved oxygen (DO) and pH were measured on site using a multi-probe sonde (YSI 6000 series, Yellow Springs, USA). To ensure accuracy, pH measurements were adjusted with 2.0 M KCL when water conductivity was below 100 uS/cm (Covington et al., 1983).

4.2.2 Laboratory processes

At the University of Waterloo (ON, Canada), frozen samples of fish, primary consumers (benthic invertebrate and zooplankton), and sediments were freeze-dried at -54 °C and 0.014 mBar for 48 h using a LabConco® FreeZone freeze-drier (USA). When applicable, shells were removed from benthic invertebrates prior to freeze-drying. Freeze-dried samples were homogenized using ultra-fine scissors and glass rods inside borosilicate scintillation vials. Freeze-dried fish and primary consumer samples were then weighed (0.30-0.35 mg) using a Mettler-Toledo® Analytical Microbalance model XP05DR (Switzerland) for analysis of δ^{15}N, which was conducted at the University of Waterloo Environmental Isotope Laboratory using a Thermo Fisher Scientific® Delta XL continuous-flow isotope ratio mass spectrometer (USA) connected to a Fisons Instruments® 1108 Elemental Analyzer (UK). Values of δ^{15}N are expressed in parts per mil (‰) in relation to the international standard of N_2 gas. Precision of δ^{15}N analysis was estimated using duplicate samples (results in Table D.2).

Analyses of concentrations of THg and MeHg were performed on samples of fish, primary consumers (MeHg only), water, and sediment at the Western University Biotron Center for Experimental Climate Change Research Analytical Laboratory in London (ON, Canada) in accordance with United States Environmental Protection Agency methods 1630, 1631, and 7473 (U.S. EPA, 1998, 2002, 2007) under strict ISO 17025 protocols. Briefly, THg analyses for biological matrices were conducted on a Milestone® DMA-80 Direct Mercury Analyzer (Italy). Analyses of THg in water were achieved using a Tekran® 2700 Automated THg Analysis System (Canada). Analyses of MeHg were conducted on biological samples that were extracted after addition of potassium hydroxide (KOH) solution and either hot block or microwave digestion. Water samples that were analyzed for MeHg were first subjected to a sub-boiling distillation to remove matrix interferences. Detection of MeHg was achieved via cold vapor atomic fluorescence spectroscopy using a Tekran® 2700 Automated MeHg Analysis System (Canada). Tissues of several primary consumers from the same taxa in each lake were combined for MeHg analysis to meet minimum weight requirements. Quality control-quality assurance (QA/QC) was completed in strict accordance with the lab's ISO 17025 accreditation (results in Table D.2).

Analysis of chlorophyll-a in water was conducted at the University of Alberta Biogeochemical Analytical Service Laboratory in Edmonton (AB, Canada). Briefly, chlorophyll-a was extracted with 95% ethanol on a filter overnight and measured using a Shimadzu®

RF-1501 Spectrofluorophotometer (North America Analytical & Measuring Instruments). Analyses of DOC and SO_4 in water and LOI in sediment were conducted at the Biotron Center for Experimental Climate Change Research. Concentrations of DOC were determined using phosphoric acid digestion followed by persulphate oxidation on an Aurora® 1030W total organic carbon analyzer (USA). Concentrations of SO_4 were determined using a Dionex ICS-1600 reagent-free ion chromatography system (USA) and a AS-50 autosampler (USA). Sediment LOI was determined as percent weight loss during ignition of samples in a muffle furnace for 2 hours at 360 °C (ASTM 2021). Lake-specific descriptive statistics of in-lake variables obtained from all sediment and water samples collected and analyzed over the study period are provided in Table D.3.

4.2.3 Geospatial analysis

Publicly available geospatial data on the Government of Canada portal were used to characterize catchment attributes. Data were accessed in May 2019 and analyzed using the coordinate system ESPG:3581 NAD83(CSRS)/NWT Lambert. The ArcticDEM of the High Resolution Digital Elevation Model v1.3 (HRDEM, 2021) and the Canadian Digital Elevation Model 1945-2011 (CDEM, 2021) were used to calculate total catchment area (CA), and to obtain mean elevation and slope in the catchments. Lake surface areas (LA) and land cover compositions (expressed as percentages of catchment area) were estimated using the 30-m resolution data layer of 2015 Land Cover of Canada (LCC, 2021b). Land cover composition of catchments was characterized by estimating proportional contributions of 10 categories (Table D.3): wetland, water, forest vegetation classes (e.g., mixed, needleleaf, and broadleaf forests), and non-forest vegetation classes (e.g., grass, shrub, and barren lands). Land cover classes are described by Natural Resources Canada (Natural Resources Canada 2019). Finally, the ratio of lake to catchment area (LA:CA) was calculated by dividing lake area by catchment area. Software and packages used for geospatial analyses are fully described in Moslemi-Aqdam et al. (2021) or Chapter 3, and lake-specific catchment characteristics are presented in Table D.3.

4.2.4 Data analysis

Statistical analyses were performed in RStudio v1.3.959 (R Studio Team, 2020) integrated with R v4.0.1 (R Core Team, 2020) using core packages unless otherwise noted. When appropriate, data were $\log_{10}$ transformed to generate normally distributed data and residuals of statistical models were re-examined to ensure that $\log_{10}$ transformations improved normality. Alpha was set at 0.05 for all analyses, unless otherwise noted. Analyses that aimed to quantify and compare rates of Hg biomagnification among lakes were based on a dataset that included δ^{15}N ratios, [THg] and [MeHg] from 332 individual fish (see Table D.4) and [MeHg] from 370 primary consumer (see Table D.5) samples collected from the ten studied lakes.

To assess how using [THg] versus [MeHg] data for fish affected estimates of Hg biomagnification rates, two sets of simple linear regression (LR) analyses were run. In the first set of regressions, $\log_{10}$ [THg] data for fish and $\log_{10}$ [MeHg] data for primary consumers were used to represent the dependent variable (mercury concentration). In the second set of regressions, $\log_{10}$ [MeHg] data for both fish and primary consumers were used to represent the dependent mercury concentration variable. In both analyses, the independent variable was represented by consumer δ^{15}N values, that were adjusted to lake-specific baseline values of δ^{15}N to account for among-lake variation at the base of food webs (Post, 2002). Baseline variation in δ^{15}N can affect among-lake comparisons of y-intercepts, which are believed to reflect Hg concentrations at the base of the food web (Kidd et al., 2012; Clayden et al., 2013; Kozak et al., 2021). Consumer δ^{15}N ratios were adjusted to lake-specific baseline values of δ^{15}N by substracting δ^{15}N ratios in Lymnaeidae snails (arithmetic mean if n per lake > 1; Table D.5).

Long-lived primary consumers, such as snails and clams, are often used to represent isotopic baselines becasue they integrte spatial and temporal variations of the isotopic signature at the base of aquatic food webs (Post, 2002). When consumers rely on both benthic and pelagic food webs in lakes, two organisms may be chosen to represent the base of benthic and pelagic food webs using a two-source baseline model (Post, 2002). Whereas Sphaeriidae clams collected from lake profundal areas were used to represent lake-specific baseline values in Chapter 2 and Chapter 3, Lymnaeidae snails collected from lake littoral areas were used to represent baseline values in this data chapter. This change in baseline organism was unavoidable because in this chapter, matched [MeHg] and δ^{15}N data were required for representative baseline taxa, and for several lakes we were unable to collect enough mass for both isotope and MeHg analyses. We did assess the effects of using different baseline organisms and models, and concluded that our choice did not affect general interprations or inference. For example, the range of δ^{15}N in Northern Pike (the focal species in both Chapter 2 and Chapter 3) varied less than three ‰ ($\sim$one trophic level) when estimated using Lymnaeidae in a one-source model (4.1-5.1‰), when estimated using Sphaeriidae in a one-source model (4.1-4.9‰), and when estimated using both Lymnaeidae and Sphaeriidae in a two-source model (4.6-6.1‰). In addition, statistically insignificant relationships between δ^{15}N and [THg] in Northern Pike (observed in Chapter 2) and between δ^{15}N and growth rate in Northern Pike (observed in Chapter 3) remained consistent (i.e., $p > 0.05$) across baseline adjustment methods.

Rates of Hg biomagnification (slopes) were compared between the two sets of mercury-δ^{15}N regressions using a paired t-test. In addition, rates of Hg biomagnification were compared among lakes for each set of regressions using an analysis of covariance (ANCOVA). In this analysis, the dependent variable was $\log_{10}$ [THg] or $\log_{10}$ [MeHg], the continuous covariate was baseline-adjusted δ^{15}N ratios, and the categorical independent variable was lake. An interaction term between lake and δ^{15}N ratio was included, which allowed models to fit lake-specific slopes. ANCOVA analyses were run in the package lsmean v2.30-0 (Lenth, 2016). Pairwise comparisons of slopes, where alpha was set to 0.001 to account for multiple comparisons (Kidd et al. 2012), were conducted in the package emmeans v1.5.4 (Lenth, 2021).

To assess whether among-lake variability in [THg] in resident fish could be explained by among-lake variability in rates of Hg biomagnification, [THg] in two commonly-consumed resident fish species were related to slopes of mercury-δ^{15}N regressions. Fish species that were captured in the majority and/or all of the ten study lakes were chosen to allow robust statistical analyses. [THg] was used as the dependent variable (rather than [MeHg]) because while matched [THg] and [MeHg] data were used in the analysis of biomagnification (i.e., we compared slopes using similar sample sizes), considerably more species- and lake-specific [THg] data exist for fish than was used in the biomagnification analysis (Table D.4 and Table D.6), which allowed for more robust consideration of effects of fish size on [THg] (see below). The two fish species investigated were Lake Whitefish (*Coregonus clupeaformis*) and Northern Pike (*Esox Lucius*); these species were captured in 9 and 10 lakes, respectively (Table D.4).

To account for effects of among-lake variability in size on [THg] and to avoid pseudoreplication in analyses investigating relationships between [THg] in the resident fish and rates of Hg biomagnification (one estimate of slope/intercept per lake), lake-specific least squares mean [THg] for Lake Whitefish and Northern Pike were estimated at fork lengths of 400 and 600 mm, respectively. These sizes represented species-specific median values of fork length (all lakes combined), and were observed in all lakes, thereby avoiding extrapolation beyond captured sizes. Least-squares means were estimated using species-specific ANCOVAs that included $\log_{10}$ [THg] as the dependent variable, $\log_{10}$ fork length as the continuous covariate, lake as the categorical independent variable, and an interaction term between lake and $\log_{10}$ fork length that allowed for estimation of lake-specific slopes. Tukey post hoc multiple comparison tests were performed to determine if least square mean [THg] in fish differed significantly among lakes.

Using simple LR, species-specific least-squares mean [THg] in fish were then related to lake-specific slopes of mercury-δ^{15}N regressions, and also to intercepts of mercury-δ^{15}N regressions; intercepts are often used to represent Hg concentrations at the base of the food webs, especially when intercepts and [MeHg] in primary consumers are positively related (Kidd et al., 2012; Clayden et al., 2013; Kozak et al., 2021). To test whether intercepts of mercury-δ^{15}N relationships and [MeHg] in primary consumers were positively related in this study, we related intercepts to [MeHg] in primary consumers using simple LR. Because fish Hg is primarily diet-derived and studies have shown that Hg levels in fish at upper trophic levels can be influenced by factors that affect [Hg] at the bottom of food webs (e.g., absolute inputs of MeHg to lakes, and density, community structure, and nutritional value of primary consumers) (Wong et al., 1997; Chen and Folt, 2005; Orihel et al., 2007; Karimi et al., 2016), we also explored relationships between least-squares mean [THg] in fish and [MeHg] in primary consumers using simple LR. The slopes and intercepts used in these analyses were those estimated by regressing $\log_{10}$ [THg] data for fish and $\log_{10}$ [MeHg] data for primary consumers against baseline-adjusted δ^{15}N values, because this is the most common method employed in the literature (Kidd et al., 2012; Clayden et al., 2013; Kozak et al., 2021).

To examine whether slopes of mercury-δ^{15}N regressions were related to ecosystem characteristics, stepwise multiple regression (MR) using p value criterion (inclusion $p = 0.01$ and exclusion $p = 0.10$) was performed in the package olsrr v0.5.3 (Hebbali, 2020). Slopes and intercepts of mercury-δ^{15}N regressions, fit using $\log_{10}$ [THg] in fish and $\log_{10}$ [MeHg] in primary consumers vs baseline-adjusted δ^{15}N, were related to a suite of lake and catchment variables (Table D.3); the independent variables included [THg] and [MeHg] in sediment, [THg] and [MeHg] in water, concentrations of chlorophyll-a, DOC, dissolved oxygen, and sulphate in water, pH and Secchi depth, catchment elevation and slope, lake to catchment area ratio (LA:CA), and land cover composition.

To avoid issues associated with a large number of explanatory variables relative to the number of replicates (n = 10 lakes), land cover categories that reflected forest types (Table D.3) were summed (hereinafter "forest"), and land cover categories that reflected non-forest types (Table D.3) were summed (hereinafter "non-forest"). Landcover categories included in the analysis that investigated ecosystem drivers of slopes were thus: wetland, water, forest, and non-forest. Due to effects of aqueous inorganic and methyl Hg partitioning to/from particles and effects of organic matter content on inorganic and methyl Hg in sediments, we used [THg] and [MeHg] in filtered water samples and adjusted [THg] and [MeHg] in sediments by dividing them by lake-specific sediment LOI values (Babiarz et al., 2001; Scudder et al., 2009; Obrist et al., 2018; Nasr and Arp, 2017). We also investigated whether intercepts of mercury-δ^{15}N regressions were related to ecosystem characteristics using stepwise MR. Finally, we performed Pearson correlation analyses among ecosystem characteristics to shed light on possible underlying mechanisms of variables determined relevant (by stepwise MR) in explaining among-lake variation in slopes and intercepts of mercury-δ^{15}N regressions.

4.3 Results and Discussion

4.3.1 Hg biomagnification and Hg species

Rates of Hg biomagnification, as estimated from slopes of mercury-δ^{15}N regressions, were significantly different when quantified using [THg] versus [MeHg] data for fish (paired t-test: $t_9 = 13.603$, $p < 0.001$). Using [MeHg] for both fish and primary consumers in mercury-δ^{15}N regressions resulted in estimates of slope that were $\sim$10% lower than when slopes were quantified using [THg] data for fish and [MeHg] data for primary consumers (Table 4.1). As reflected in the confidence intervals of the slope estimates (Table 4.1), uncertainty around rate estimates was generally lower when measured [MeHg] in both fish and primary consumers were used in biomagnification models, relative to when [THg] in fish and [MeHg] in primary consumers were used. The only study we are aware of that makes a similar comparison was conducted on lakes in eastern Canada by Wyn et al. (2009). In this study, the authors analyzed a subsample of fish individuals and species for [MeHg], and reported that rates of Hg biomagnification were similar when quantified using [THg] vs [MeHg] in fish (Wyn et al., 2009). Our analyses included analyzing both [THg] and [MeHg] in all fish included in the analysis, and results indicated that estimates of Hg biomagnification rates can be more uncertain and significantly higher when [THg] in fish were used, relative to when [MeHg] in fish were used. While not surprising given that the inorganic fraction of THg in fish is not subject to biomagnification, our result suggests that caution is warranted when biomagnification rates are quantified

Table 4.1. Descriptive statistics of mercury-δ^{15}N regressions. Baseline-adjusted δ^{15}N values were regressed against **(a)** THg concentrations in fish and MeHg concentrations in primary consumers, and **(b)** MeHg concentrations in both fish and primary consumers. Letters in the group column represent pairwise differences in slopes and are identical for both panels.

Lake	**(a)** Estimates (95% LCI-UCI)		r^2	**(b)** Estimates (95% LCI-UCI)		r^2	Group
	Intercept	Slope		Intercept	Slope		
Big Island	0.545 (0.29-0.79)	0.161 (0.13-0.18)	0.784	0.624 (0.37-0.87)	0.142 (0.11-0.17)	0.741	a
Mustard	0.477 (0.20-0.75)	0.191 (0.15-0.22)	0.851	0.535 (0.27-0.79)	0.173 (0.13-0.20)	0.842	ab
Willow	0.246 (0.00-0.57)	0.193 (0.15-0.23)	0.726	0.292 (0.00-0.61)	0.184 (0.14-0.22)	0.706	ab
Fish	0.576 (0.41-0.73)	0.192 (0.17-0.21)	0.818	0.624 (0.46-0.78)	0.181 (0.16-0.20)	0.799	ab
Gargan	1.241 (0.96-1.51)	0.171 (0.13-0.20)	0.805	1.291 (0.99-1.58)	0.153 (0.11-0.19)	0.741	ab
Ekali	0.938 (0.70-1.17)	0.188 (0.15-0.22)	0.582	0.986 (0.75-1.21)	0.174 (0.14-0.20)	0.555	ab
Sanguez	0.812 (0.57-1.05)	0.194 (0.17-0.21)	0.698	0.859 (0.62-1.09)	0.183 (0.16-0.20)	0.673	ab
Kakisa	0.368 (0.13-0.60)	0.207 (0.18-0.23)	0.741	0.427 (0.19-0.66)	0.194 (0.16-0.21)	0.714	ab
Tathlina	0.751 (0.53-0.97)	0.216 (0.18-0.24)	0.758	0.809 (0.59-1.02)	0.200 (0.17-0.22)	0.734	ab
McGill	0.562 (0.42-0.70)	0.253 (0.23-0.27)	0.876	0.609 (0.46-0.75)	0.240 (0.22-0.26)	0.853	b

using [THg] in fish and then compared among systems; some of the among-system variability may in fact reflect differences in %MeHg in fish rather than differences in rates of biomagnification.

When %MeHg varies widely among individual fish or species within a food web, it is more likely that rates of Hg biomagnification will be different when quantified using [THg] versus [MeHg]. In our study, %MeHg in fish ranged from ∼45 to 120% (Figure D.1; estimates of 120% are reasonable, factoring in method-acceptable reporting error for both THg and MeHg), indicating variability in the proportion of MeHg to THg in fish muscle tissues. Lescord et al. (2018) observed a similar range (39-124%) for %MeHg in a study of several fish species collected from subarctic lakes of Canada. Large variability in %MeHg in fish (as low as 5%) has also been observed in fish collected from lakes, rivers, and estuaries around the world (Stefansson et al., 2013; Peng et al., 2016; Arcagni et al., 2018), and it is likely that this variability affects estimates of Hg biomagnification rates when [THg] data for fish are used in mercury-δ^{15}N regressions. Fish age, size, trophic ecology, proximate composition of muscle tissues, and assimilation efficiency of inorganic and methyl Hg in muscle tissues have all been shown to influence [MeHg], and thus %MeHg, in fish, although relationships vary among species (Stefansson et al., 2013; Peng et al., 2016; Bradley et al., 2017; Lescord et al., 2018; Arcagni et al., 2018). Investigating causes of %MeHg variability in fish is beyond the scope of this study, but %MeHg in our samples was related to both fish size and δ^{15}N, and the nature and magnitude of effects differed among species (Figure D.1). Uncertainty around estimates of Hg biomagnification rates would be expected to increase as variability in %MeHg in fish tissues increases. Thus, when using [THg] data in estimates of Hg biomagnification rates through food webs, the following should be considered: i) what is the range or likely range of %MeHg in biota for which THg is to be used as a proxy for MeHg; and, ii) how precise do the estimates of biomagnification need to be to effectively test the hypotheses of interest?

4.3.2 Hg biomagnification and Hg in resident fish

Lake-specific descriptive statistics of [THg] in Lake Whitefish and Northern Pike, as well as results of ANCOVAs and pairwise tests, are presented in Table D.6. There was greater among-lake variability in size-standardized [THg] in the apex and piscivorous predator Northern Pike than in the lower-trophic level and benthivorous Lake Whitefish. Size-standardized [THg] in Lake Whitefish varied ~2.5-fold among lakes whereas size-standardized [Hg] in Northern Pike varied ~4.5-fold among lakes (Table D.6). Although positive, tionships between lake-specific slopes of mercury-δ^{15}N regressions and lake-specific, size-standardized [THg] were not significant for Lake Whitefish (simple LR:

$_{1,\,7} = 1.705$, $p = 0.232$, $r^2 = 0.20$) and Northern Pike (simple LR: $F_{1,\,8} = 0.890$, $p = 0.373$, $r^2 = 0.10$). Similarly, relationships between intercepts of mercury-δ^{15}N regressions and size-standardized [THg] were postive but not signficant for Lake Whitefish (simple LR: $F_{1,\,8} = 2.143$, $p = 0.186$, $r^2 = 0.24$) and Northern Pike (simple LR: $F_{1,\,8} = 1.970$, $p = 0.198$, $r^2 = 0.20$).

Intercepts of mercury-δ^{15}N regressions are commonly accepted to represent concentrations of Hg at the bottom of food webs (Kidd et al., 2012; Clayden et al., 2013; Kozak et al., 2021), although this inference has never been tested in a controlled experiment. Similarly, our results from a linear regression between intercepts of mercury-δ^{15}N regressions and [MeHg] in primary consumers indicated that there was a significant and positive relationship between intercepts of mercury-δ^{15}N regressions and [MeHg] in biota at lower trophic levels (Figure 4.2a). Although [THg] in resident fish species were not significantly related to intercepts of mercury-δ^{15}N regressions, lake-specific, size-standardized [THg] in fish were significantly and positively related to lake-specific [MeHg] in primary

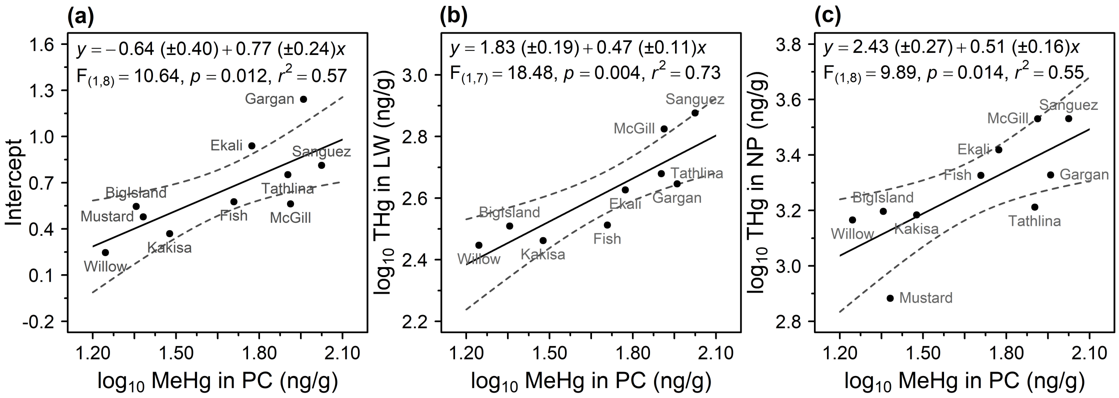

Figure 4.2. Linear relationships between concentrations of MeHg in primary consumers (PC) and **(a)** intercepts of mercury-δ^{15}N regressions ([total Hg] in fish and [methyl Hg] in primary consumers versus baseline adjusted δ^{15}N), **(b)** size-standardized concentrations of total Hg in Lake Whitefish (LW), and **(c)** size-standardized concentrations of total Hg in Northern Pike (NP). Solid lines represent fitted regressions and dashed lines represent ±95% confidence intervals.

consumers; linear regressions indicated that more than 70% of among among-lake variability in size-standardized [THg] in Lake Whitefish (Figure 4.2b) and more than 50% of among-lake variability in size-standardized [THg] in Northern Pike (Figure 4.2c) were explained by [MeHg] in primary consumers. The higher r^2 of the relationship for Lake Whitefish compared to that for Northern Pike may reflect the fact that Lake Whitefish feed directly on primary consumers whereas the more predatory Northern Pike are often one trophic level removed from primary consumers (Scott and Crossman, 1973; Cott et al., 2011). These results, when coupled with recent findings that demonstrated the importance of growth rate in predicting [THg] in Northern Pike in these study lakes (see Moslemi-Aqdam et al. (2022) or Chapter 2), suggest that mercury levels at the bottom of lake food webs explain [THg] in lower trophic level fish species better then [THg] in higher trophic level fish species.

4.3.3 Hg biomagnification and ecosystem characteristics

Biomagnification of mercury through the food webs of the study lakes was indicated by significant and positive linear regressions between mercury concentrations and δ^{15}N values in all lake-specific models (simple LR: $F_{1,21-127} = 95.16\text{-}687.50$, $p < 0.001$). Delta^{15}N explained up to 88% of the variability in mercury concentrations within lake food webs. (Table 4.1 and Figure D.2). The average rate of Hg biomagnification (i.e., slopes of regressions between [THg] in fish and [MeHg] in primary consumers versus baseline-adjusted δ^{15}N) for these subarctic lakes (0.197) is higher than the average reported for lakes at temperate (0.16) and tropical (0.12) latitudes and similar to the value reported for a polar lake (0.19) in a worldwide meta-analysis (Lavoie et al., 2013). It thus appears that Hg biomagnifies at higher rates through lake food webs in the Canadian subarctic than through lake food webs at lower latitudes, which is consistent with previous assertions that biomagnification of Hg through lake food webs is higher in the colder, low productivity systems that are often found at more northern latitudes (Lavoie et al., 2013; Kozak et al., 2021).

Rates of Hg biomagnification ranged from 0.161 to 0.253, and varied significantly among lakes (ANCOVA, interaction term: $F_9 = 3.09$, $p < 0.001$; Table 4.1 and Table D.7). Our observed values are within the range of those reported in previous studies for lake systems (0.16-28) (see Kidd et al., 2011). Among-lake variability in mercury-δ^{15}N slopes was best explained by a model that included percent forest cover (stepwise MR: $F_{1,\ 8} = 11.59$, $p = 0.009$). Explaining 59% of the among-lake variability, percent forest cover was significantly and positively related to mercury-δ^{15}N slopes (Figure 4.3a), indicating that rates of Hg biomagnification were higher in lakes surrounded by catchments with proportionally more forested land cover. Consistent with findings of previous research that has reported higher concentrations of ions, mercury, and dissolved organic matter in lakes downstream of forested catchments (Kamenik et al., 2001; Sobek et al., 2007; Williams et al., 2010; Bravo et al., 2017; Denkenberger et al., 2020), our Pearson correlation analyses revealed that lakes located in more forested catchments had higher concentrations of THg, MeHg, DOC, and SO_4 (Figure D.3). These water chemistry conditions could lead to higher rates of Hg biomagnification through lake food webs by elevating [MeHg] and %MeHg in resident biota (Tsui and Finlay, 2011; Braaten et al., 2019; Riva-Murray et al., 2020).

Water chemistry can affect concentrations and speciation of Hg in lake biota, and thus rates of biomagnification, through a variety of mechanisms. For example, relatively higher levels of DOC (an analytical measure for concentrations of dissolved organic matter) in lakes can result in higher %MeHg in fish via effects on growth efficiency. Previous

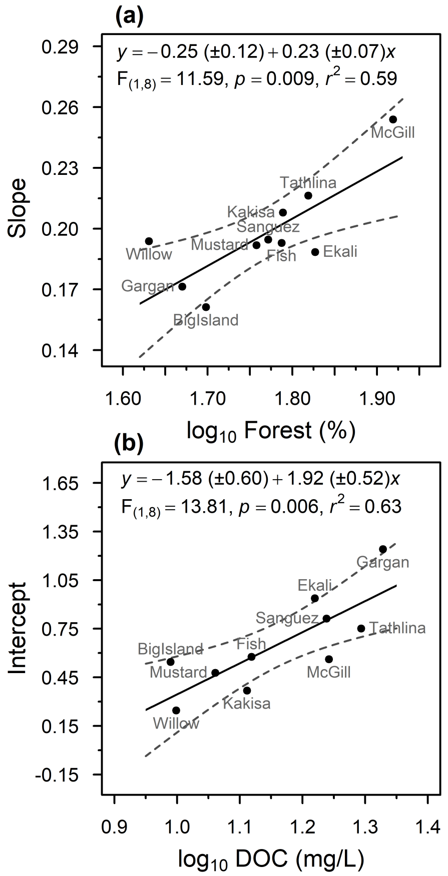

Figure 4.3. Linear relationships between slopes **(a)** and intercepts **(b)** of mercury-δ^{15}N regressions ([total Hg] in fish and [methyl Hg] in primary consumers versus baseline adjusted δ^{15}N) and best ecosystem predictors determined by stepwise multiple regressions. Solid lines represent fitted regressions and dashed lines represent ±95% confidence intervals.

studies in the study lakes and in other lakes have shown that higher concentrations of DOC can reduce growth rates in fish via effects on quality of basal organic matter, as well as light attenuation, primary productivity, and availability of dissolved oxygen (Ranåker et al., 2014; Benoît et al., 2016; van Dorst et al., 2019; Moslemi-Aqdam et al., 2021). As MeHg binds to thiols in proteins in fish tissues (Trudel and Rasmussen, 2006; Niu et al., 2017), reduced growth rates and growth efficiency in fish can result in higher %MeHg in fish by lowering the amount of lipid relative to protein (often assessed via a C:N proxy variable) in muscle tissues (Lescord et al., 2018). Furthermore, higher in-lake levels of total Hg, MeHg, DOC, and SO_4 have all been linked to higher net MeHg production (Ullrich et al., 2001; Pickhardt et al., 2002; Chen and Folt, 2005; Paranjape and Hall, 2017; Burke et al., 2020; Moslemi-Aqdam et al., 2022), and we observed significant and positive relationships between [MeHg] concentrations in primary consumers and fish. Indeed, [MeHg] in primary consumers were higher in lakes located in catchments with proportionally more forest cover, and where concentrations of total Hg, MeHg, DOC, and SO_4 were also higher (Figure D.3). Higher rates of Hg biomagnification observed in lakes surrounded by proportionally more forested catchments could thus reflect effects of forest cover on water chemistry in downstream lakes and resultant higher %MeHg in resident biota.

Among-lake variability in intercepts of mercury-δ^{15}N regressions was best explained by DOC concentrations in water (stepwise MR: $F_{1,\,8} = 13.81$, $p = 0.006$). Concentrations of DOC were significantly and positively related to intercepts, and explained 63% of the among-lake variability in intercept estimates (Figure 4.3b). Assuming that intercepts reflect [MeHg] at the bottom of food webs, which is supported by the observed significant and positive relationship between [MeHg] in primary consumers and intercepts (Figure 4.2) and previous literature (Kidd et al., 2012; Clayden et al., 2013; Kozak et al., 2021), the observed significant and positive relationship between intercepts of mercury-δ^{15}N regressions and DOC concentrations suggests that [MeHg] in basal biota are higher in lakes with higher concentrations of dissolved organic matter. Although effects of DOC on MeHg levels in lakes and resident biota can be complicated by the structural complexity of DOC (i.e., molecular composition and aromaticity of organic matter) (see Eagles-Smith et al., 2016), higher concentrations of DOC can lead to higher [MeHg] at the bottom of food webs through a variety of mechanisms, including enhanced mercury transport from catchments, activity of bacterial methylators, and/or rates of biological uptake (Lambertsson and Nilsson, 2006; Wiener et al., 2006; Shanley et al., 2008).

4.4 Conclusion

Biomagnification rates of mercury through food webs in subarctic lakes of Canada were estimated with better confidence intervals using measured concentrations of methylmercury in all biota. Rates of biomagnification were positively but not significantly related to mercury concentrations in resident fish used for subsistence. Rates of biomagnification were significantly and positively related to proportional forest cover in catchments, with which in-lake concentrations of mercury and organic matter were positively correlated. Identifying factors and mechanisms that drive spatial variability in the biomagnification of Hg through aquatic food webs is important for understanding variability in accumulation and concentrations of Hg in fish. This is particularly true in the north, where fish are a critical food source for local communities (Donaldson et al. 2010; Pirkle et al. 2016) and where ecosystem processes can be profoundly influenced by climate and development-induced changes in land cover, fire regime, permafrost thaw, and hydrology (Rydberg et al. 2010; Hugelius et al. 2020). This research is the first comprehensive study on biomagnification of Hg through lake food webs in the Canadian subarctic. Our findings indicate that while among-lake differences in Hg biomagnification rates remain similar when rates are estimated using THg vs MeHg concentrations in fish, using THg concentrations in fish as a proxy for concentrations of MeHg could lead to overestimation of Hg biomagnification rates, likely due to variability of %MeHg in fish tissues. Whilst rates of Hg biomagnification were not related to among-lake variability of THg concentrations in resident fish, MeHg concentrations in lower trophic level biota were positively related to THg concentrations in resident fish. The positive relationship between mercury concentrations in fish and mercury concentrations in primary consumers was stronger for the benthivore Lake Whitefish than the predator Northern Pike, likely highlighting larger effects of food web mercury concentrations on mercury levels in fish species connected more closely to lower food level biota. Both lake (DOC concentrations) and catchment (forest cover) factors, and likely their interactions, influenced biomagnification of Hg through lake food webs, and biomagnification rates were higher in food webs of lakes surrounded by proportionally more forested catchments. These results help advance a holistic understanding of causes of spatial variability in Hg biomagnification in lake systems in the subarctic, provide insights that help better understand variability in Hg levels in northern food fish, and guide future research and monitoring programs in remote subarctic lakes.

Chapter 5

General Discussion

Through research conducted and presented in this thesis, I sought to understand causes of long documented spatial variability in fish [Hg] among lakes in the Dehcho Region. Revealing some of the mechanisms through which fish, lake, and catchment characteristics may interact to influence fish [Hg], findings improve our understanding of controls of [Hg] in subarctic freshwater fish and our ability to make informed predictions. Results also reveal additional knowledge gaps and lay the groundwork for future research.

5.1 Research Synthesis

When considered as a whole (Figure 5.1), results of the body of research in this thesis indicate that the concentrations of mercury in fish in the Dehcho Region are influenced by biological processes (i.e., fish growth rates and food web Hg, including biomagnification rates), that are, in turn, influenced by lake-catchment interactions. In brief, higher concentrations of ions (e.g., SO_4), organic matter (i.e., DOC), and mercury (i.e., total and methyl Hg) occur in low-elevation lakes with relatively larger, more forested catchments (Chapters 2, 3, and 4). Such lakes had higher concentrations of mercury in primary consumers (Chapters 2 and 4), higher rates of mercury biomagnification through food webs (Chapter 4), and lower growth rates in Northern Pike (Chapter 3). Together, these conditions lead to higher concentrations of mercury in Northern Pike (Chapters 2 and 4). Evidently, numerous variables at different levels of biological organization (i.e., fish, lake, and catchment) interact to ultimately influence [Hg] in Northern Pike in the Dehcho Region.

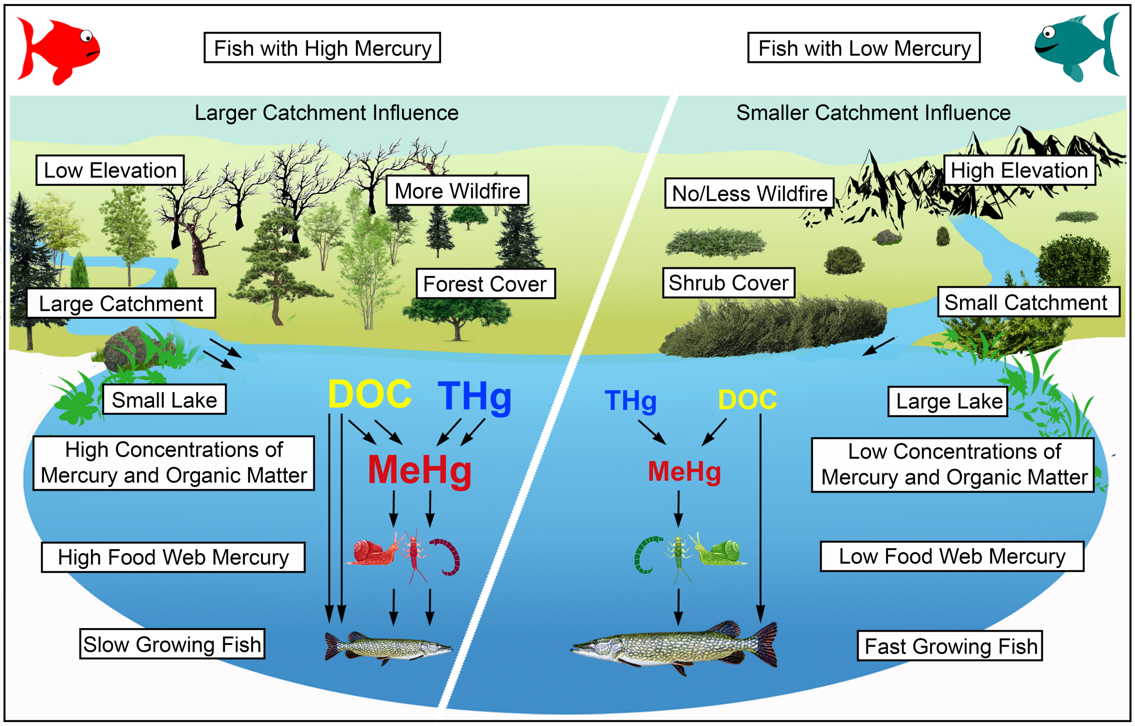

Figure 5.1. A schematic diagram depicting a synthesis of the findings. Lower-elevation lakes that were surrounded by relatively larger and more forested catchments (that likely experience more wildfire) had higher concentrations of total mercury (THg), dissolved organic carbon (DOC), and methyl mercury (MeHg) in lake water [Chapters 2, 3, and 4], which were associated with lower growth rates in fish [Chapter 3], higher concentrations of mercury in primary consumers [Chapters 2 and 4], and higher biomagnification rates of mercury through food webs [Chapter 4], ultimately leading to higher concentrations of mercury in resident fish [Chapters 2 and 4].

Consistent with the findings of the few studies that have been conducted in the region or included some lakes within the region (e.g., Evans et al., 2005; Lockhart et al., 2005; Laird et al., 2018), results of this thesis indicated higher [Hg] in fish from small lakes located in the Hay River Lowland ecoregion (e.g., Sanguez and McGill). Elevated fish [Hg] in these lakes appear to be driven by several biological processes in response to lake-catchment interactions. Biological drivers of [Hg] in fish (i.e., lower fish growth rates, higher [MeHg] in primary consumers, and higher rates of Hg biomagnification) were all present in Sanguez and McGill lakes, where concentrations of mercury and organic matter were relatively high in association with relatively larger size, higher elevation, and more forest cover of catchments. Relatively lower fish [Hg] were observed in other small (e.g., Ekali and Gargan) and large (e.g., Kakisa and Tathlina) lakes in the Hay River Lowland ecoregion, and also in lakes located in other ecoregions, including the Norman Range (Fish Lake), the Northern Alberta Upland (Trout Lake), and the Horn Plateau (Willow, Mustard, and Big Island lakes). In Gargan Lake, for example, fish growth rate was relatively high and rate of Hg biomagnification was relatively low, even though there were relatively high [MeHg] in primary consumers. Fish are thus likely to have higher [Hg] where lake-catchment interactions result in several biological responses that synergistically act to increase fish [Hg].

Attributes of catchments (i.e., elevation, relative size, and proportional forest cover) were related to abiotic (in-lake levels of DOC, SO4, and total and methyl Hg) and biotic (Hg concentrations in primary consumers, fish growth rates, and biomagnification rates of Hg) conditions in downstream lakes, and ultimately influenced fish [Hg]. As authors of previous research around the world have suggested, forest cover may directly influence dynamics of mercury and organic matter in downstream lake via delivery of more mercury and carbon, although results are variable among forest types (e.g., Sobek et al., 2007; Eklöf et al., 2016; Eagles-Smith et al., 2016). Proportional forest cover, forest type, and relative size of catchments may also affect how wildfire impacts dynamics of mercury and carbon in downstream lakes; catchment wildfire has been shown to influence [Hg] in resident biota, including fish (Garcia and Carignan, 2005; Kelly et al., 2006; Rydberg et al., 2010). The effects of wildfire on [Hg] in abiotic and biotic ecosystem compartments in downstream lakes can be mediated through changes in mercury and/or carbon input from catchments that may reflect effects of fire on hydrology, land cover, and permafrost (Garcia and Carignan, 2005; Kelly et al., 2006; Rydberg et al., 2010; Riggs et al., 2017;

Wulder et al., 2020).

According to the Canadian National Fire Database, most lakes in Dehcho Region (i.e., all lakes except Willow, Big Island, and Mustard in the Horn Plateau) have experienced some degree of wildfire in the last decade (CNFD, 2021). Relative to Fish Lake in the Norman Range and Trout Lake in Northern Alberta Upland, lakes located in the Hay River Lowland (e.g., Sanguez, McGill, Ekali, Tathlina, and Kakisa) are sur-

(a)

Lakes	Ecoregions	Fire
Gargan	HRL	22.83
Sanguez	HRL	15.63
Ekali	HRL	9.28
McGill	HRL	3.67
Kakisa	HRL	9.71
Tathlina	HRL	7.24
Fish	NR	5.47
Trout	NAU	4.05
Big Island	HP	0.00
Mustard	HP	0.00
Willow	HP	0.00

(b)

Variables	Pearson statistics against Fire	
	r	*p*
Chlorophyll-a	0.32	0.338
Dissolved oxygen	−0.25	0.466
Secchi depth	−0.36	0.274
DOC	**0.78**	**0.005**
SO_4	0.49	0.128
Sediment total Hg	**0.59**	**0.054**
Sediment methyl Hg	0.22	0.507
Sediment %MeHg	−0.29	0.38
Water total Hg not-filtered	0.26	0.445
Water methyl Hg not-filtered	0.44	0.179
Water %methyl Hg not-filtered	0.46	0.152

(c)

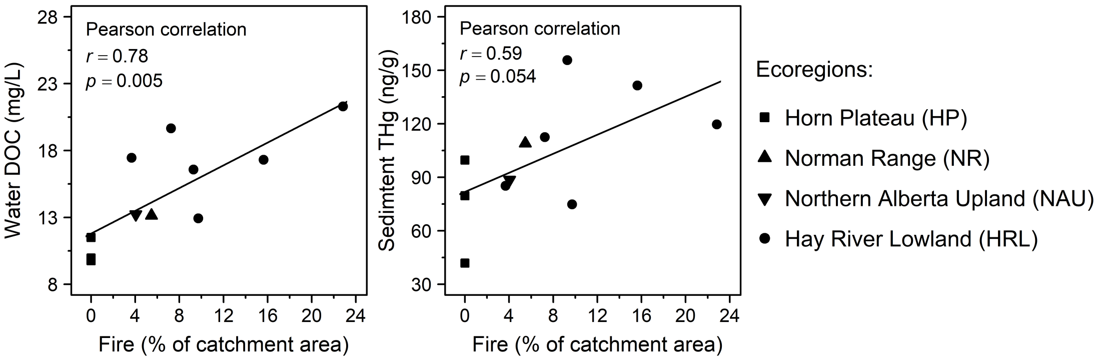

Figure 5.2. Data relating to fire in catchments of the study lakes. Statistics of catchment fire for the studied lakes. Percent catchment area burned by fire from 2010 to 2015 **(a)**, pearson correlation statistics [$r > 0.50$ are in bold] between catchment fire and select in-lake parameters **(b)**, and biplots depicting correlations between catchment are burned (between 2010 and 2015) and each of concentrations of dissolved organic carbon [DOC] in water and concentrations of total Hg [THg] in sediment. **(c)**

rounded by catchments with proportionally greater areas burned by wildfire in the period 2010-2015 (Figure 5.2). I initially investigated effects of wildfire using a cumulative fire variable (percentage of catchment area burned over 2010-2015; Figures 5.2) in Chapter 2, the whole-ecosystem study on among-lake variability of fish [Hg]. Results from these analyses showed that there were positive correlations between catchment fire and in-lake parameters, including concentrations of mercury and organic matter (i.e., DOC) (Figure 5.2). The influence of wildfire on downstream lakes can vary, however, with post-fire recovery or other landscape changes, such as land cover types and permafrost thaw (see Rothenberg et al., 2010; Santos et al., 2019; Li et al., 2021). Unfortunately, the available Dehcho data did not allow assessment of the potential temporal variability of post-fire effects, which is why results associated with wildfire were excluded during the peer-reviewed publication process. Given that fish [Hg] in the Dehcho Region were found to be influenced by concentrations of both organic matter and mercury within lakes, the preliminary results regarding associations between catchment wildfire and in-lake concentrations of mercury and organic matter may suggest that catchment wildfire is a broad and important driver of in-lake conditions (that is, [Hg] in abiotic and biotic ecosystems compartments) in the area, an idea that is worth further and more comprehensive investigation.

Results presented in this thesis may also enhance our understanding of carbon flux in lake ecosystems, which are important components of landscape and global carbon cycles (McCullough et al., 2018). In Chapter 3, where I investigated the effects of lake-catchment interactions on carbon stable isotopes in resident biota, I found that greater inferred influence from catchments resulted in more depleted (negative) δ^{13}C signature in resident biota, including primary consumers and Northern Pike. Assuming similar taxonomical (Anas et al., 2019b) and spatial (France, 1995) fractionation during uptake of dissolved organic carbon (DIC), variation in δ^{13}C of primary consumers is driven by how limiting DIC is and/or δ^{13}C of the DIC source (Raven et al., 1994; France et al., 1997). Sources of DIC include respired organic matter, the atmosphere, and/or products of weathering in the catchment (del Giorgio and France, 1996; Grey et al., 2000). Whereas DIC derived from atmospheric and geogenic sources exhibits δ^{13}C values ranging from -7 to 0 ‰, DIC derived from respiration (of both autochthonous and allochthonous organic matter) exhibits δ^{13}C values ranging from -20 to -35 ‰(LaZerte, 1983; Rounick and Winterbourn, 1986; Hecky and Hesslein, 1995; Wachniew and Różański, 1997).

In Chapter 3, I observed δ^{13}C within the range of -20 to -35 ‰in primary consumers, and that the higher the inferred catchment influence, the more depleted the δ^{13}C values in primary consumers. It may thus be likely that δ^{13}C of DIC derived from allochthonous organic matter are more depleted than δ^{13}C of DIC derived from autochthonous organic matter (see also Anas et al., 2019a). These findings further our understanding of the underlying energetics in these subarctic lakes, and may provide evidence that can help address the challenge of differentiating the sources (i.e., autochthonous vs allochthonous) of DIC that is originated from respired organic matter (Post, 2002). Indeed, further comprehensive research is required to investigate the observed phenomenon; lakes in Dehcho Region have widely different surface areas and productivity levels, which can influence the δ^{13}C signature of the source DIC, and many physicochemical processes that affect sources of DIC to lakes are being affected by climate change.

Reaffirming the complexity of controls on fish [Hg] in lake ecosystems, findings from this thesis also provide insights that can be used for prioritizing monitoring and management practices in remote northern regions, where conducting research is challenging and costly. According to findings from this thesis, research and management priorities should focus on low-elevation lakes that are surrounded by relatively larger catchments with proportionally more forest cover (and likely, more wildfire). These catchment characteristics appear to be associated with higher concentrations of DOC and Hg (both total and methyl Hg) in downstream lakes, resulting in biological mechanisms with synergetic effects on fish [Hg], including lower growth rates in fish, higher [MeHg] in primary consumers, and higher rates of Hg biomagnification through lake food webs. Particularly because lake and catchment physical attributes (elevation, relative size, forest cover, and likely, wildfire) can be sensed remotely, results provide invaluable guidance for mangers and decision-makers regarding fish [Hg] monitoring programs and associated consumption advisories at subarctic latitudes. Continuous monitoring and assessment of fish [Hg] are necessary in remote northern lakes due to the profound effects of climate warming on northern regions (Rydberg et al., 2010; Hugelius et al., 2020), as well as the heavy reliance of northern communities on wild-caught freshwater fish (Donaldson et al., 2010; Pirkle et al., 2016).

5.2 Study Limitations

While findings from this thesis revealed numerous mechanisms through which [Hg] in fish may be influenced in subarctic lakes, there are several limitations associated that should be acknowledged and deserve further studies.

One key limitation is the availability of data and generalizability of findings, especially with respect to results associated with among-lake variability of [Hg] and growth rates in Northern Pike. Chapters 2 and 3 focused on Northern Pike, largely because this was the only species captured from all the studied lakes over the course of sampling, thereby maximizing statistical power and modeling ability. Additionally, Northern Pike exhibit the widest variability of [Hg] among lakes in the Dehcho Region and can also be found with [Hg] that exceed health guidelines in some lakes (e.g., Evans et al., 2005; Lockhart et al., 2005; Laird et al., 2018). Species-specific differences in habitat, feeding, and behavioral ecology (see Scott and Crossman, 1973) lead to variable responses of fish [Hg] and growth to environmental factors, such as lake and catchment characteristics (e.g., Benoît et al., 2016; Thomas et al., 2020). Even though Northern Pike is a model organism that has increasingly helped researchers to better understand aquatic ecology and biology (Forsman et al., 2015), further research on other resident fish species in the study area, once more data are available, is warranted and necessary to increase generalizability of the findings.

My analyses and findings are also limited by the quantity data analyzed. Analyses in this thesis were mostly restricted to eleven lakes. While whole ecosystem data on this number of remote lakes are rare, it is important to highlight that a greater number of lakes would allow for more robust parameter estimations. More data would also help elucidate additional (i.e., second- and third-order) effects or interactions that are difficult to capture with small sample sizes (Anderson, 2008). Relationships that are commonly observed in literature, such as between lake productivity and fish growth rates (e.g., Wilson et al., 2019) and/or between lake productivity and biomagnification rates (e.g., Kozak et al., 2021), were statistically insignificant in this thesis, which may have reflected low statistical power. Furthermore, most in-lake variables used this thesis (i.e., water and sediment parameter) are based on samples that were collected once per year from each lake in a consistent, but brief, time period (late August to early September). Although in-lake variables were sampled for more than one year in each study lake, which increased robustness of the findings, and consistency in sampling time avoided possible bias associated with temporal variation, fully capturing interactions among in-lake environmental variables (which may vary widely at fine temporal and/or spatial scales) is challenging with the point measurements used in this thesis. It would be more possible to capture, for example, interactions of Hg concentrations between sediment and water samples (e.g., Guédron et al., 2020) or interactions between lake productivity and DOC concentrations (e.g., Seekell et al., 2015) with greater temporal and spatial replication.

5.3 Remaining Questions

Important insights regarding mechanisms through which the lithosphere, hydrosphere, and biosphere interact to influence fish [Hg] were revealed through the research conducted and presented in this thesis, yet there are many remaining unknowns, some of which are outlined below.

The role of catchments in direct inputs of MeHg to downstream lakes is unknown. Findings from Chapters 2 and 4 revealed that lakes in relatively larger, lower-elevation, and more forested catchments (and likely more wildfire) had higher concentrations of ions, organic matter, and both total Hg and methyl Hg. While higher in-lake levels of ions (e.g., sulphate) and organic matter (e.g., DOC) can certainly promote in-lake production of MeHg (see Paranjape and Hall, 2017), lakes can also receive direct input of MeHg that is produced in catchments (e.g., Bravo et al., 2017). In addition to indirect pathways (e.g., more delivery of DOC that could promote methylation of Hg to MeHg), catchments are likely influencing MeHg concentrations in lakes directly by delivering MeHg that is produced in catchments, a hypothesis that warrants further research (see also Branfireun et al., 2020).

The exact mechanism through which DOC influences Hg cycling in the study area is also unknown. Findings from Chapters 2, 3, and 4 revealed that MeHg levels were higher in lakes with higher DOC concentrations, and that DOC concentrations were higher in lakes with larger inferred catchment influence (i.e., catchments at lower elevations and with larger relative size and more proportional forest cover (and likely greater wildfire). Higher concentrations of DOC have been shown to promote production of MeHg in lakes via several mechanisms, including enhancing delivery of Hg from catchments, microbial activity, and/or biological uptake (Lambertsson and Nilsson, 2006; Wiener et al., 2006; Shanley et al., 2008). Catchment-derived DOC is known, however, to be highly aromatic in character and of greater molecular weight (Kalbitz et al., 2003; Yang et al., 2021), which can actually limit bioavailability of Hg and results in lower concentrations of MeHg in lakes and resident biota (e.g., Wren et al., 1991; Eagles-Smith et al., 2016). Underlying mechanisms by which DOC concentrations influence MeHg concentrations in lakes in the study area, especially in association with the structural complexity of DOC (i.e., molecular composition and aromaticity), warrant further research.

There are many more factors that could potentially drive among-lake variability of [Hg] in fish in the Dehcho Region. Permafrost thaw and peatland cover are catchment characteristics that have been observed elsewhere to influence input of both mercury and organic matter to downstream lakes (e.g., Rydberg et al., 2010), yet were not included in this research and associated analyses. Climatic factors, that have been shown to induce biotic and abiotic responses in lake ecosystems (Carrie et al., 2010; Rydberg et al., 2010; Kirk et al., 2011; Korosi et al., 2015; Gandhi et al., 2015), were also not included in the analyses completed for this thesis. As lake and catchment characteristics are diverse in the study area, it is worth investigating if long-term influences of climatic variables on in-lake parameters might have contributed to the current spatial variability of mercury in the region and resident biota. Fish [Hg] can also be influenced by effects mediated through changes in fish growth rates and trophic transfer of Hg that are associated with community composition of fish (see Ward et al., 2010a; Schartup et al., 2019); comprehensive assessments of community composition and prey-predator interactions have not been conducted for lakes in the Dehcho Region. Fish [Hg] can also be influenced by the presence and quantity of other elements, such as selenium and fatty acid concentrations (Laird et al., 2018; Gochfeld and Burger, 2021), which were not measured and included in the present research, and deserve further study. Overall, there are many aspects of fish [Hg] that warrant further investigation in the Dehcho Region, and deserve further research, particularly because fish are of vital importance for local communities.

5.4 Future Directions

The study limitations and remaining questions that were outlined above provide directions for future research. Outlined below are ideas that have emerged directly from the development of this thesis.

Analyses in Chapter 2 revealed statistically insignificant relationships between [Hg] (both total and methyl Hg) in unfiltered water samples and [Hg] in sediment samples. Using Hg data from filtered water samples and correcting sediment [Hg] for organic matter content revealed that total Hg concentrations in water and sediment samples were positively related, yet no relationship was found for concentrations of methyl Hg between water and sediment samples. Although it is likely that fine scale temporal variations in the production of methyl Hg (see Regnell et al., 1997; Wang et al., 2012; Bachand et al., 2014; Paranjape and Hall, 2017; Obrist et al., 2018) were not captured by the point measurements of water and sediment variables used in the analyses of this thesis, an alternative hypothesis for the lack of the relationship could be that littoral areas are playing a more important role than profundal areas in the production of methyl Hg in the studied lakes.

Sediment and water samples used in this thesis were collected from the approximate middle and deepest location of lakes, yet littoral production of methyl Hg in lakes has recently been shown to be more important for production of MeHg in lakes than previously thought (see Obrist et al., 2018; Burke et al., 2020; Branfireun et al., 2020). Concentrations of methyl Hg in primary consumers collected in the study area provide some evidence for the alternative hypothesis regarding the importance of littoral production of methyl Hg. Benthic invertebrates collected from littoral zones had higher concentrations of methyl Hg than benthic invertebrates collected from profundal zones; this observation was not only true when comparisons used all benthic invertebrates combined, but also when comparisons used similar taxa observed in both littoral and profundal zones. It is thus likely that littoral areas are contributing substantially to the production of methyl Hg in the study area. More samples (e.g., sediment and periphyton samples from littoral areas and more diverse benthic invertebrate taxa from profundal areas) will help provide more and robust empirical evidence regarding the importance of littoral zones in the production of methyl Hg in the study area, which, if supported, will be a substantial contribution to the field given the recent evidence regarding the importance of littoral production of MeHg in lakes (see Branfireun et al., 2020).

Results from Chapter 3 showed that growth rates in Northern Pike were lower in lakes subject to greater inferred catchment influence. In lakes with higher catchment influence, there were higher DOC concentrations; DOC concentrations have been shown to be negatively related to growth rates in many freshwater fish species through influences on primary production (which affects prey availability), water clarity (which affects visual performance and feeding), and dissolved oxygen (which affects metabolism) (e.g., Ranåker et al., 2014; Benoît et al., 2016; van Dorst et al., 2019). I found no associations between DOC concentrations and primary production (i.e., chlorophyll-a concentrations), water clarity (i.e., Secchi depth), and/or dissolved oxygen concentrations in the study area, even though the range of DOC concentrations in the studied lakes (9.8-21.3 mg/L) was similar to studies that observed these interactions (e.g., Ranåker et al., 2014; Benoît et al., 2016; van Dorst et al., 2019). Although the lack of relationships between these water parameters could be related to the point measurements used in this thesis, I proposed that the negative relationship between DOC concentrations in water and growth rates in Northern Pike might be related to DOC quality.

Findings indicated that DOC in the study lakes is primarily catchment-derived. Compared to DOC derived from internal sources (i.e., autochthonous production), DOC derived from external sources (i.e., allochthonous production) is known to be not only highly aromatic in character and of greater molecular weight (Kalbitz et al., 2003; Yang et al., 2021), but also of less nutritional value (e.g., fatty acid concentrations) (Lau et al., 2009; Hiltunen et al., 2019). Lower growth rates of Northern Pike in lakes with higher DOC concentrations could thus be attributed to less bioaccessibility and lower nutritional value of catchment-derived DOC that propagates through lake food webs, a hypothesis that can be independently tested. One approach to test this proposed hypothesis could be a study of concentrations of nutritional elements (e.g., fatty acids) in fish muscle tissues. If the hypothesis is supported, it would be the first observation of this phenomenon in an apex fish species. Given that Northern Pike are known to be a lie-in-wait predator with a preference for littoral habitats, it would also be valuable to study inter-specific variation of both growth rates and fatty acid concentrations in the study area using species with different habitat preferences (e.g., adult Cisco with pelagic preferences and Lake Trout with littoral-pelagic preferences).

Results from Chapter 4 revealed that Hg biomagnification rates can vary depending on the species of Hg (i.e., total Hg vs MeHg) used to model biomagnification. The percent of MeHg to total Hg (i.e., %MeHg) in fish data varied approximately three-fold, and there was inter-specific variability in relationships between %MeHg and size of fish, as well as between %MeHg and trophic level of fish. While there have been scientific attempts to link variability in fish %MeHg to fish ecology, such as size, age, carbon source, or trophic positions (e.g., Lescord et al., 2018), the possible relationships between %MeHg in fish and ecosystem characteristics remain understudied. With lake and catchment properties being important factors influencing fish ecology, Hg cycling, and [Hg] in fish in the Dehcho Region, it is likely that there are relationships between %MeHg in fish and ecosystem variables, although my preliminary analyses indicate that larger sample sizes (i.e., a greater number of lakes as the experimental unit) are needed to investigate among-lake drivers of %MeHg in fish. Understanding the variation of %MeHg in response to fish ecological factors will also be valuable, and sample sizes will likely be easier to achieve as individual fish will be the experimental unit in the statistical analyses.

5.5 Concluding Remarks

Findings of the present thesis provide the first comprehensive understanding of causes of among-lake variability in fish mercury concentrations in the Dehcho Region, that addresses questions raised by local First Nations and improves the current and general understanding about fish mercury controls in subarctic lakes.

I showed that fish mercury concentrations in subarctic lakes of the Dehcho Region are influenced by biological processes that are in turn influenced by lake-catchment interactions. Smaller and lower-elevation lakes surrounded by relatively larger and more forested catchments (that have also experienced more wildfire) had higher concentrations of total mercury, dissolved organic carbon, and methylmercury, which resulted in lower growth rates in resident fish, higher concentrations of mercury in primary consumers, and higher biomagnification rates of mercury through food webs, together leading to higher concentrations of mercury in fish. Highlighting catchment characteristics that initiate numerous biotic and abiotic responses in downstream lakes that ultimately influence mercury concentrations in resident fish, my results provide direction for prioritizing lakes for monitoring and management in remote northern regions. This is particularly importnat in this current era of a rapidly changing environment, especially because physical attributes of lakes and catchments can be sensed remotely. Reaffirming the complexity of interactions that occur between various ecosystem compartments (i.e., the biosphere, hydrosphere, and lithosphere) to ultimately influence mercury levels in fish, insights revealed in this thesis underscore the necessity of considering whole ecosystems for understanding and predicting change in fish mercury concentrations.

My PhD journey has been extremely educational, yet the more I know, the more I realize how much I do not know. The observed phenomena throughout the current thesis are based on empirical evidence from the data analyzed. Given the complexity of interactions that can take place among variables to influence mercury concentrations in fish, and the fact that relative influence of fish mercury controls can vary with ecological context, I will not be surprised if inclusion of more spatial and temporal data in the future either supported my findings and associated interpretation, ideas, and hypotheses, or rejected them. I personally appreciate the rejection of my ideas, because it at least means that my hypotheses were testable. Science evolves with progressive refinement, and there is no personal preference in science. The wonder of science is that we can follow nature beyond evidence of our senses in an open-minded fashion, where we are constantly open to the possibility of having to change our minds. The knowledge gained through this work, however, was not only gained using western science methods and approaches.

As part of an academic degree in Doctor of Philosophy in Science, this thesis is composed of Western Science, where knowledge is broken into disciplines, logics, rationality, and objectivity (Mazzocchi, 2006; Langer, 2011; Levac et al., 2018). Findings revealed here are based on measurement of observables — the philosophical tradition of deduction and positivism that overlooks metaphysical and spiritual realms as a source of knowledge. Yet, several of the hypotheses tested and much of the apporaches used and knowlege gained in this thesis reflect Traditional Knowledge — the type of knowledge that is metaphysical, holistic, symbolic, relational, and intergenerational (Mazzocchi, 2006; Langer, 2011; Levac et al., 2018). Actions, relationships, personal experiences, and generational observations have all made critical contributions to the insights revealed in this research. Built on a paradigm that considers land as a relation rather than an object, Traditional Knowledge shared with the research team by the Indigenous peoples of the region has profoundly influenced the questions, methods, and findings of the research presented here. Grounded in active collaboration between academic and Indigenous people in co-learning, this work could be a step forward in reconciliation.

The larger project on which my research is based tries to address the practical and locally identified concerns of local communities in Canada. First Nations of within the Dehcho Region are involved in the research, from question formulation to data collection and interpretation. According to their needs and concerns, local communities define where (e.g., lakes), what (e.g., fish species), and how (e.g., fishing sites and times) to collect samples. They also participate in field and laboratory processes. As eyes on the land, the elderly and adult members of the communities continually guide the sample collection and processing in the field. Younger members pay visits to youth camps and laboratory facilities, where they can take part in various activities of the project and learn about its methods and objectives. In periodic meetings that are held locally within the communities every year, results are shared with First Nations to integrate their field observations and historical knowledge and aid in interpretation of the findings, which are also shared with public health collaborators to inform both general and local mercury-related fish consumption advisories. Finally, research outcomes and future goals are discussed in wide consultations between academic and Indigenous communities to ensure that the project continues to address local needs and concerns.

Although reconciling what took years to destroy will not happen instantly, I hope that my co-operative and co-learning research in the Canadian north would help lay the foundation for a relationship that is based on mutual respect and understanding.

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Appendices

A Research Ethics

This research is conducted under Aurora Research Institute Scientific Research License #16046 and #16633 and University of Waterloo Animal Care Committee Animal Use Protocol 40576 and A-18-04.

B Supplementary Information for Chapter 2

Table B.1. Latitude and longitude (of approximate centre) of the study lakes, mode of access, and sample collection years for sediment (S), water (W), Northern Pike (F), and benthic invertebrates (B). All sampling took place from mid-August to mid-September each year. Additional fish were also collected by First Nations community members in March (F*) and May (F**). Ecoregions represented by the study lakes include the Norman Range (NR), Horn Plateau (HP), Northern Alberta Upland (NAU), and Hay River Lowland (HRL).

Lake (ecoregion)	Latitude (N)	Longtitdue (W)	Access	Year						
				2013	2014	2015	2016	2017	2018	2019
Fish (NR)	63°10′56.56″	122°32′37.79″	Floatplane	-	-	-	-	-	S, W	S, W, F, B
Big Islalnd (HP)	62°05′12.78″	119°52′08.04″	Floatplane	W, F, B	-	-	S, W	S, W, F, B	S, W	S, W
Willow (HP)	62°10′09.82″	119°11′05.20″	Floatplane	-	-	-	S, W, F, B	-	S, W	S, W
Mustard (HP)	62°00′08.29″	120°05′37.90″	Floatplane	-	-	S, W, F, B	S, W	S, W	S, W	S, W
Trout (NAU)	60°36′51.74″	121°10′09.56″	Road	W, F, B	-	-	-	-	S, W	W
Kakisa (HRL)	60°55′23.91″	117°39′56.10″	Road	-	-	S, W, F, B	-	S, W, F, B	S, W, F*	S, W
Tathlina (HRL)	60°32′19.81″	117°30′29.04″	Road	-	S, W, F, B	-	F*	-	S, W, F, B	S, W
Ekali (HRL)	61°17′22.68″	120°34′58.33″	Trail	W, F, B	-	W	S, W	-	S, W	S, W
Gargan (HRL)	61°14′56.40″	120°22′55.53″	Trail	-	S, W, F, B	-	-	-	S, W	S, W
McGill (HRL)	61°18′12.42″	121°00′49.96″	Floatplane	-	S, W, F, B	-	-	-	S, W, F, B	S, W
Sanguez (HRL)	61°15′14.51″	120°29′32.06″	Trail	W, F, B	-	-	S, W, F, B	S, W, F, B	S, W, F, F**	S, W, F, F**

Table B.2. Mean absolute differences ± standard deviation (count) between duplicates (DUP) in analyses of carbon isotope ratios (δ^{13}C), nitrogen isotope ratios (δ^{15}N), total Hg (Hg) concentrations, and methyl Hg (MeHg) concentrations for fish, benthic invertebrates, water, and sediment samples. Detection limits (DL), recoveries of Dorm-4 reference materials (RM), and spike recoveries (SR) are also given for Hg and MeHg analyses.

Analysis	Parameter	Fish	Invertebrate	Water	Sediment
δ^{13}C	DUP	AD: 0.006 ± 0.117 RPD: 4.38 ± 28.53 ‰ (n = 49)	-	-	-
δ^{15}N	DUP	AD: 0.063 ± 0.212 RPD: 5.57 ± 28.39 ‰ (n = 49)	-	-	-
Hg	DUP	AD: 0.057 ± 0.089 RPD: 2.14 ± 1.30 ng/g ww (n = 5)	-	AD: 0.118 ± 0.171 RPD: 12.83 ± 17.49 ng/L (n = 7)	AD: 0.919 ± 9.016 RPD: 11.90 ± 9.21 ng/g (n = 6)
MeHg	DUP	-	AD: 13.170 ± 17.110 RPD: 52.88 ± 20.12 ng/g dw (n = 10)	AD: 0.002 ± 0.003 RPD: 2.63 ± 3.73 ng/L (n = 7)	AD: 0.124 ± 0.168 RPD: 56.93 ± 40.76 ng/g (n = 6)
Hg	DL	0.067 ± 0.027 ng	-	0.003 ± < 0.001 ng	0.073 ± 0.005 ng
	RM	99.8 ± 3.4 % (n = 64)	-	102.7 ± 7.6 % (n = 10)	105.0 ± 8.6 % (n = 6)
	SR	98.0 ± 9.5 % (n = 64)	-	101.3 ± 1.5 % (n = 10)	102.5 ± 1.9 % (n = 6)
MeHg	DL	-	028 ± 0.026 ng	0.002 ± 0.001 ng	0.004 ± 0.002 ng
	RM	-	93.1 ± 6.0 % (n = 53)	90.7 ± 6.0 % (n = 10)	100.5 ± 3.1 % (n = 6)
	SR	-	100.3 ± 6.3 % (n = 53)	97.3 ± 4.9 % (n = 10)	96.8 ± 6.3 % (n = 6)

Table B.3. Lake-specific total mercury concentrations in Northern Pike, along with fish, lake, and catchment factors in the Dehcho Region, NT, Canada. See legend in Figure B.1 for abbreviations of land cover categories. Ecoregions represented by the study lakes include the Norman Range (NR), Horn Plateau (HP), Northern Alberta Upland (NAU), and Hay River Lowland (HRL).

	Variable (unit): abbreviation, if used	Lake (ecoregions)				
		Fish (NR)	Big Island (HP)	Willow (HP)	Mustard (HP)	Trout (NAU)
	Hg in Northern Pike (mg/kg ww): $[\mathrm{Hg}]_{\mathrm{NRPK}}$	0.39 (0.02)	0.31 (0.04)	0.29 (0.02)	0.16 (0.05)	0.16 (0.02)
	Fish Factors					
	Carbon source in Northern Pike (‰): $\delta^{13}\mathrm{C}_{\mathrm{NRPK}}$	0.39 (0.02)	0.31 (0.04)	0.29 (0.02)	0.16 (0.05)	0.16 (0.02)
	Trophic level of Northern Pike (‰): $\delta^{15}\mathrm{N}_{\mathrm{NRPK}}$	7.10 (0.12)	9.08 (0.21)	8.15 (0.16)	7.66 (0.53)	8.09 (0.20)
	Growth of Northern Pike (year): Age-at-size$_{\mathrm{NRPK}}$	6.748 (0.29)	6.098 (0.48)	6.549 (0.30)	4.284 (0.85)	5.386 (0.40)
	Hg in benthic invertebrates (ng/g): $[\mathrm{MeHg}]_{\mathrm{invert}}$	48.52 (8.25)	27.20 (3.24)	21.45 (3.66)	23.40 (4.49)	42.08 (6.77)
	Lake Factors					
	Total Hg in unfiltered water (ng/L): $[\mathrm{THg}]_{\mathrm{wat}}$	0.82 (0.33)	0.58 (0.05)	0.55 (0.07)	0.70 (0.08)	0.53 (0.10)
	MeHg in unfiltered water (ng/L×100): $[\mathrm{MeHg}]_{\mathrm{wat}}$	2.87 (0.43)	1.97 (0.37)	1.11 (0.33)	1.14 (0.23)	1.83 (0.66)
	Total Hg in sediment (ng/g): $[\mathrm{THg}]_{\mathrm{sed}}$	108.98 (26.80)	79.64 (23.70)	41.88 (13.21)	99.65 (15.99)	
	MeHg in sediment (ng/g): $[\mathrm{MeHg}]_{\mathrm{sed}}$	0.33 (0.10)	0.46 (0.11)	0.36 (0.06)	0.31 (0.07)	0.17 (NA)
	Chlorophyll-a (μg/L)	3.44 (0.86)	5.15 (0.68)	5.26 (0.60)	7.23 (1.52)	6.93 (1.26)
	Secchi depth (m)	3.00 (NA)	4.00 (NA)	2.80 (NA)	2.75 (0.25)	1.65 (0.85)
	Dissolved organic carbon (mg/L): DOC	13.14 (0.66)	9.76 (0.27)	9.97 (0.53)	11.50 (0.430	13.23 (0.91)
	Dissolved oxygen (mg/L): DO	9.60 (NA)	10.60 (NA)	9.60 (NA)	9.30 (NA)	10.8 (NA)
	Sulphate (mg/L): SO_4	14.48 (1.85)	3.56 (0.04)	6.34 (0.17)	8.83 (0.68)	5.46 (0.35)
	pH	7.65 (NA)	7.17 (NA)	7.58 (NA)	7.28 (0.56)	7.85 (0.05)
	Stratification depth (m)	10.2	21.3	18.1	0.9	5.9
	Catchment Factors					
	Lake area (km^2): LA	117.07	18.22	128.58	23.31	500.01
	Catchment area (km^2): CA	2616.86	112.96	1248.81	198.68	5.893.46
	LA:CA (ratio×100)	4.47	16.13	10.29	11.73	8.48
	Catchment elevation (m asl)	428.25	783.22	704.82	750.83	584.69
	Catchment slope (degree)	2.57	2.49	1.79	2.22	3.34
Land cover classes (% of CA)	Barren land	0.179	0	0	0	0
	Mixed forest	0.359	0	0.004	0.055	2.755
	SP/P Grass land	5.21	11.491	23.904	10.323	1.182
	SP/P Shrub land	1.151	0.859	0.93	0.951	0
	SP Taiga needleleaf forest	9.43	25.398	26.689	22.322	1.02
	T/SP Broadleaf deciduous forest	0.265	0	0	0	1.518
	T/SP needleleaf forest	58.075	24.504	16.037	34.865	75.751
	T/SP Shrub land	3.931	2.585	4.923	3.433	2.074
	Wetland	10.612	11.818	8.604	12.407	4.494
	Water	10.787	23.345	18.908	15.643	11.205

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Table B.3; continued from previous page

Variable	Lake (ecoregions)					
	Kakisa (HRL)	Tathlina (HRL)	Ekali (HRL)	Gargan (HRL)	McGill (HRL)	Sanguez (HRL)
$[\mathrm{Hg}]_{\mathrm{NRPK}}$ (mg/kg ww)	0.31 (0.01)	0.33 (0.01)	0.52 (0.03)	0.45 (0.04)	0.69 (0.06)	0.69 (0.01)
Fish Factors						
$\delta^{13}\mathrm{C}_{\mathrm{NRPK}}$ (‰)	0.31 (0.01)	0.33 (0.01)	0.52 (0.03)	0.45 (0.04)	0.69 (0.06)	0.69 (0.01)
$\delta^{15}\mathrm{N}_{\mathrm{NRPK}}$ (‰)	7.77 (0.09)	7.01 (0.8)	7.69 (0.11)	7.76 (0.17)	8.55 (0.15)	7.95 (0.04)
Age-at-size$_{\mathrm{NRPK}}$ (year)	6.319 (0.21)	7.156 (0.22)	8.059 (0.32)	7.279 (0.46)	8.034 (0.46)	7.751 (0.11)
$[\mathrm{MeHg}]_{\mathrm{invert}}$ (ng/g)	29.53 (2.70)	71.78 (8.95)	70.98 (5.79)	94.22 (12.07)	77.58 (9.52)	120.60 (8.55)
Lake Factors						
$[\mathrm{THg}]_{\mathrm{wat}}$ (ng/L)	1.64 (0.64)	1.22 (0.27)	0.70 (0.06)	0.80 (0.13)	0.90 (0.22)	0.74 (0.11)
$[\mathrm{MeHg}]_{\mathrm{wat}}$ (ng/L)	2.56 (1.27)	8.90 (0.87)	7.12 (3.33)	5.53 (0.82)	7.40 (91.72)	4.74 (0.78)
$[\mathrm{THg}]_{\mathrm{sed}}$ (ng/g)	74.87 (14.23)	112.58 (10.29)	155.68 (11.30)	119.70 (9.50)	85.27 (5.78)	141.52 (9.86)
$[\mathrm{MeHg}]_{\mathrm{sed}}$ (ng/g)	0.22 (0.04)	0.33 (0.09)	0.09 (0.03)	0.35 (0.21)	0.33 (0.11)	0.77 (0.24)
Chlorophyll-a (μg/L)	15.53 (7.39)	3.51 (1.57)	7.12 (0.80)	8.58 (2.91)	2.24 (0.16)	5.62 (0.29)
Secchi depth (m)	0.75 (0.35)	1.75 (NA)	2.90 (0.30)	2.38 (0.38)	2.45 (0.05)	2.58 (0.39)
DOC (mg/L)	12.93 (1.34)	19.66 (0.86)	16.58 (1.02)	21.30 (3.48)	17.46 (1.34)	17.31 (1.80)
DO (mg/L)	11.00 (NA)	11.40 (NA)	9.40 (NA)	9.00 (NA)	9.70 (NA)	9.71 (0.38)
SO_4 (mg/L)	20.06 (1.53)	13.18 (0.94)	11.27 (0.69)	14.08 (1.15)	14.21 (1.19)	10.18 (2.14)
pH	7.74 (0.22)	7.86 (NA)	8.17 (0.16)	7.95 (0.24)	7.90 (0.04)	8.25 (0.08)
Stratification depth (m)	0	0	2.1	0	2.8	13
Catchment Factors						
LA (km^2)	335.76	565.29	1.88	1.1	2.32	1.59
CA (km^2)	15808.82	11606.31	179.8	73.16	982.6	106.86
LA:CA (ratio×100)	2.12	4.87	1.05	1.5	0.24	1.49
Catchment elevation (m asl)	408.64	456.68	274.04	283.45	354.56	277.99
Catchment slope (degree)	4.17	4.27	4.36	2.33	5.22	3.19
Land cover classes (% of CA)						
Barren land	0.025	0.027	0.006	0	0	0
Mixed forest	4.088	4.677	4.46	2.46	4.514	4.033
SP/P Grass land	0.985	1.207	0.028	0.041	0.005	0.047
SP/P Shrub land	0	0	0	0	0	0
SP Taiga needleleaf forest	1.399	0.958	0.067	0.027	0.505	0.066
T/SP Broadleaf deciduous forest	2.354	2.665	8.075	4.975	3.562	6.709
T/SP needleleaf forest	63.134	65.305	63.425	61.154	80.516	63.226
T/SP Shrub land	8.159	7.948	3.364	1.422	2.199	2.601
Wetland	11.337	9.18	15.554	25.205	6.935	18.602
Water	8.52	8.033	5.022	4.716	1.765	4.716

Mercury concentrations, carbon source, trophic level, and size-at-age in Northern Pike were standardized at fork length = 600 mm (ANCOVA results: $F_{21,\,513} = 145.5$, $p < 0.001$ for $[\mathrm{Hg}]_{\mathrm{NRPK}}$, $F_{21,\,513} = 199.8$, $p < 0.001$ for $\delta^{13}\mathrm{C}_{\mathrm{NRPK}}$, $F_{21,\,513} = 41.9$, $p < 0.001$ for $\delta^{15}\mathrm{N}_{\mathrm{NRPK}}$, and $F_{21,\,513} = 179.9$, $p < 0.001$ for Age-at-size$_{\mathrm{NRPK}}$). Values of δ^{15}N were adjusted for baseline variation among lakes (see the main text for details).

Table B.4. Lake-specific information on presence (⋆) of benthic invertebrate taxa collected from the study lakes in the Dehcho Region, NT, Canada. Ecoregions represented by the study lakes include the Norman Range (NR), Horn Plateau (HP), Northern Alberta Upland (NAU), and Hay River Lowland (HRL).

Taxa	Lake										
	Fish (NR)	Big Island (HP)	Willow (HP)	Mustard (HP)	Trout (HRL)	Kakisa (HRL)	Tathlina (HRL)	Ekali (HRL)	Gargan (HRL)	McGill (HRL)	Sanguez (HRL)
Acari (mites)	⋆										
Aeshnidae	⋆				⋆	⋆	⋆	⋆	⋆	⋆	⋆
Apananiidae				⋆							
Baetidae	⋆	⋆		⋆		⋆		⋆			
Baetiscidae						⋆					
Caenidae	⋆		⋆			⋆	⋆				
Ceratopogonidae	⋆						⋆				
Chironomidae	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆		⋆
Coenagrionidae					⋆	⋆	⋆	⋆	⋆	⋆	⋆
Coenogridae	⋆										
Corduliidae	⋆				⋆						
Corixidae	⋆	⋆	⋆	⋆		⋆	⋆	⋆	⋆	⋆	⋆
Crangonyctidae	⋆										
Cyclopoida									⋆		
Dytiscidae	⋆	⋆	⋆	⋆		⋆	⋆		⋆	⋆	⋆
Ephemeridae	⋆		⋆			⋆	⋆			⋆	
Erpobdellidae	⋆	⋆		⋆		⋆	⋆				
Gammaridae	⋆	⋆	⋆	⋆		⋆	⋆	⋆		⋆	⋆
Glossiphoniidae	⋆					⋆		⋆		⋆	⋆
Gomphidae							⋆				
Gyrinidae						⋆		⋆		⋆	
Haemopidae	⋆										
Haliplidae	⋆							⋆			
Heptageniidae	⋆	⋆				⋆	⋆				
Hyalellidae	⋆	⋆	⋆	⋆		⋆	⋆	⋆			⋆
Hydracarina			⋆	⋆							
Hydrophilidae	⋆										
Hydropsychidae						⋆					

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Table B.4; continued from previous page

Taxa	Lake										
	Fish (NR)	Big Island (HP)	Willow (HP)	Mustard (HP)	FTrout (HRL)	Kakisa (HRL)	Tathlina (HRL)	Ekali (HRL)	Gargan (HRL)	McGill (HRL)	Sanguez (HRL)
Laevicaudata									⋆		
Lepidostomatidae						⋆					
Leptoceridae						⋆					
Leptohyphidae						⋆					
Leptophlebiidae						⋆					
Libellulidae								⋆			⋆
Limnephilidae	⋆			⋆				⋆			
Lymnaeidae	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆
Molannidae							⋆				
Neophemeridae	⋆										
Notonectidae	⋆							⋆	⋆	⋆	
Oligochaeta		⋆		⋆							
Perlidae	⋆		⋆								
Phryganeidae	⋆			⋆		⋆	⋆				⋆
Physidae		⋆					⋆	⋆	⋆	⋆	⋆
Piscicolidae	⋆	⋆				⋆					
Planorbidae	⋆					⋆	⋆	⋆	⋆		⋆
Polycentropodidae						⋆					
Sialidae								⋆			
Sphaeriidae	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆
Tipulidae	⋆			⋆							⋆
Unionidae							⋆	⋆	⋆	⋆	⋆
Valvatidae	⋆		⋆	⋆		⋆	⋆				

Table B.5. Lake-specific arithmetic mean (± standard devidation) concentrations of total and methyl Hg (in ng/L), and percentages of methyl Hg to total Hg concentrations (%MeHg), in unfiltered and filtered water samples. There were statistically signficiant correlations between i) concentrations of THg in unfiltered and filtered water samples (Pearson $r = 0.0.665$, $p = 0.026$), ii) concentrations of MeHg in unfiltered and filtered water samples (Pearson $r = 0.994$, $p < 0.001$), iii) %MeHg in unfiltered and filtered water samples (Pearson $r = 0.964$, $p < 0.001$), and iv) MeHg concentrations and %MeHg in unfiltered water samples (Pearson $r = 0.902$, $p < 0.001$). We used concentrations of THg and MeHg in unfiltered water samples in our analyses, and interpreted concentrations of MeHg as an integrated reflection of overall net MeHg production and cycling in lake systems. Ecoregions represented by the study lakes include the Norman Range (NR), Horn Plateau (HP), Northern Alberta Upland (NAU), and Hay River Lowland (HRL).

Lake (ecoregion)	Unfiltered water			Filtered water		
	THg	MeHg	%MeHg	THg	MeHg	%MeHg
Fish (NR)	0.820 (±0.47)	0.029 (±0.01)	0.035	0.673 (±0.39)	0.030 (±0.01)	0.044
BigIsland (HP)	0.585 (±0.08)	0.020 (±0.01)	0.034	0.473 (±0.16)	0.012 (±0.00)	0.024
Willow (HP)	0.546 (±0.15)	0.011 (±0.01)	0.020	0.511 (±0.12)	0.010 (±0.01)	0.019
Mustard (HP)	0.701 (±0.15)	0.011 (±0.00)	0.016	0.529 (±0.10)	0.008 (±0.00)	0.014
Trout (NAU)	0.530 (±0.18)	0.018 (±0.01)	0.035	0.653 (±0.58)	0.014 (±0.00)	0.022
Kakisa (HRL)	1.636 (±1.44)	0.026 (±0.03)	0.016	0.823 (±0.34)	0.018 (±0.02)	0.022
Tathlina (HRL)	1.217 (±0.46)	0.089 (±0.02)	0.073	1.324 (±0.57)	0.079 (±0.01)	0.060
Ekali (HRL)	0.696 (±0.15)	0.071 (±0.07)	0.102	0.588 (±0.21)	0.064 (±0.07)	0.109
Gargan (HRL)	0.805 (±0.25)	0.055 (±0.02)	0.069	0.735 (±0.18)	0.044 (±0.01)	0.060
McGill (HRL)	0.905 (±0.44)	0.074 (±0.03)	0.082	0.844 (±0.40)	0.068 (±0.03)	0.081
Sanguez (HRL)	0.736 (±0.27)	0.047 (±0.02)	0.064	0.581 (±0.21)	0.041 (±0.02)	0.070

Table B.6. Lake-specific arithmetic mean (± standard devidation) concentrations of total and methy Hg (in ng/g), and percentages of methyl Hg to total Hg concentrations (%MeHg), in sediment samples. There was a statistically signficiant correlation between [MeHg] and %MeHg in sediment samples (Pearson $r = 0.603$, $p = 0.048$). We used concentrations of total Hg and methyl Hg in sediments in our analyses. Lake-specific arithmetic mean (± standard devidation) loss-on-ignition (LOI; a proxy for organic matter content of the sediment in %) data are also shown. LOI was used to normalize concentrations of total and methy Hg in sediments (using [THg]/LOI and [MeHg]/LOI, respectively) (refrence: U.S. Geological Survey, Scientific Investigations Report 2009-5109). Sediment LOI analyses were conducted at the Western University Biotron Center for Experimental Climate Change Research in London (ON, Canada). Sediment LOI data were not available for Trout Lake; the sediment LOI in Trout Lake was thus predicted using $\text{LOI} = 0.0089\,[\text{THg}]_{\text{sed}}{}^{2} - 1.4685\,[\text{THg}]_{\text{sed}} + 79.08$ ($r^2 = 0.73$) obtained from observations of THg and LOI in the remaining ten study lakes. Ecoregions represented by the study lakes include the Norman Range (NR), Horn Plateau (HP), Northern Alberta Upland (NAU), and Hay River Lowland (HRL).

Lake(ecoregion)	Unadjusted values			LOI	Values adjusted to LOI		
	THg	MeHg	%MeHg		THg	MeHg	%MeHg
Fish (NR)	108.977 (±46.41)	0.327 (±0.18)	0.003	24.316 (±2.52)	4.482	0.130	0.029
Big Island (HP)	79.642 (±47.41)	0.455 (±0.22)	0.006	20.351 (±9.14)	3.913	0.050	0.013
Willow (HP)	41.879 (±32.37)	0.359 (±0.13)	0.009	26.732 (±31.61)	1.567	0.011	0.007
Mustard (HP)	99.650 (±31.99)	0.311 (±0.14)	0.003	18.841 (±5.22)	5.289	0.060	0.011
Trout (NAU)	88.606 (±0.00)	0.174 (±0.00)	0.002	14.532 (±NA)	6.097	0.012	0.002
Kakisa (HRL)	74.870 (±40.24)	0.221 (±0.12)	0.003	9.424 (±7.32)	7.944	0.030	0.004
Tathlina (HRL)	112.583 (±23.02)	0.331 (±0.21)	0.003	8.512 (±4.23)	13.227	0.078	0.006
Ekali (HRL)	155.675 (±22.60)	0.088 (±0.07)	0.001	57.067 (±7.70)	2.728	0.011	0.004
Gargan (HRL)	119.701 (±19.00)	0.351 (±0.42)	0.003	36.081 (±10.63)	3.318	0.033	0.010
McGill (HRL)	85.271 (±14.15)	0.329 (±0.26)	0.004	13.120 (±0.11)	6.499	3.089	0.475
Sanguez (HRL)	141.523 (±22.04)	0.770 (±0.54)	0.005	30.336 (±0.05)	4.665	15.892	3.406

Table B.7. Results of simple linear regressions between total mercury concentrations ([Hg]) in Northern Pike (NRPK) and fish factors (see Table B.3) for the 11 study lakes in the Dehcho Region, NT, Canada.

Parameters	$\delta^{15}N_{\mathrm{NRPK}}$ (‰)	$\delta^{13}C_{\mathrm{NRPK}}$ (‰)	Age-at-size$_{\mathrm{NRPK}}$ (years)	[MeHg]$_{\mathrm{invert}}$ (ng/g)
Intercept	0.090	−1.019	−0.527	0.137
Slope	0.037	−0.049	0.137	0.004
F	0.142	12.690	31.730	15.840
df	1, 9	1, 9	1, 9	1, 9
p	0.715	0.006	< 0.001	0.003
r^2	0.015	0.585	0.779	0.638

Table B.8. Full statistics of piecewise structural equation models (piecewise-SEM) used to evaluate causalities among drivers of mercury concentrations in Northern Pike in the 11 study lakes in the Dehcho Region, NT, Canada. Variables are defined in Table B.3 and Figure B.1.

Response		Predictor						Model		
Variable	r^2	Variable	Std. Est.	Std. Err.	df	Crit. Val.	p	Fisher's C	p	df
$[\mathrm{Hg}]_{\mathrm{NRPK}}$	0.83	$[\mathrm{MeHg}]_{\mathrm{invert}}$	0.328	0.215	8	1.525	0.166			
		Age-at-size$_{\mathrm{NRPK}}$	0.642	0.215	8	2.989	0.017			
$[\mathrm{MeHg}]_{\mathrm{invert}}$	0	$[\mathrm{THg}]_{\mathrm{wat}}$	0.025	0.333	9	0.074	0.943	0.675	0.714	2
$\sim\sim$ Age-at-size$_{\mathrm{NRPK}}$		$\sim\sim$ $[\mathrm{MeHg}]_{\mathrm{invert}}$	0.729	-	11	3.014	0.008			
$[\mathrm{MeHg}]_{\mathrm{invert}}$	0.49	$[\mathrm{MeHg}]_{\mathrm{wat}}$	0.699	0.238	9	2.935	0.017	2.356	0.308	2
$\sim\sim$ Age-at-size$_{\mathrm{NRPK}}$		$\sim\sim$ $[\mathrm{MeHg}]_{\mathrm{invert}}$	0.274	-	11	0.807	0.222			
$\sim\sim$ Age-at-size$_{\mathrm{NRPK}}$		$\sim\sim$ $[\mathrm{MeHg}]_{\mathrm{wat}}$	0.767	-	9	3.588	0.006			
$[\mathrm{MeHg}]_{\mathrm{invert}}$	0.52	$[\mathrm{THg}]_{\mathrm{sed}}$	0.721	0.231	9	3.123	0.012	0.657	0.72	2
$\sim\sim$ Age-at-size$_{\mathrm{NRPK}}$		$\sim\sim$ $[\mathrm{MeHg}]_{\mathrm{invert}}$	0.596	-	11	2.102	0.034			
$[\mathrm{MeHg}]_{\mathrm{invert}}$	0.22	$[\mathrm{MeHg}]_{\mathrm{sed}}$	0.466	0.295	9	1.582	0.148	2.963	0.227	2
$\sim\sim$ Age-at-size$_{\mathrm{NRPK}}$		$\sim\sim$ $[\mathrm{MeHg}]_{\mathrm{invert}}$	0.735	-	11	3.069	0.008			
$[\mathrm{MeHg}]_{\mathrm{invert}}$	0.05	Chlorophyll-a	−0.233	0.324	9	−0.717	0.492	0.609	0.737	2
$\sim\sim$ Age-at-size$_{\mathrm{NRPK}}$		$\sim\sim$ $[\mathrm{MeHg}]_{\mathrm{invert}}$	0.691	-	11	2.7	0.014			
$[\mathrm{Hg}]_{\mathrm{NRPK}}$	0.83	$[\mathrm{MeHg}]_{\mathrm{invert}}$	0.328	0.215	8	1.525	0.166	3.013	0.556	4
		Age-at-size$_{\mathrm{NRPK}}$	0.642	0.215	8	2.989	0.017			
$[\mathrm{MeHg}]_{\mathrm{invert}}$	0.64	$[\mathrm{MeHg}]_{\mathrm{wat}}$	0.426	0.258	8	1.655	0.137			
$\sim\sim$ Age-at-size$_{\mathrm{NRPK}}$		$[\mathrm{THg}]_{\mathrm{sed}}$	0.477	0.258	8	1.853	0.101			
$\sim\sim$ Age-at-size$_{\mathrm{NRPK}}$		$\sim\sim$ $[\mathrm{MeHg}]_{\mathrm{invert}}$	0.325	-	11	0.97	0.18			
$\sim\sim$ $[\mathrm{Hg}]_{\mathrm{NRPK}}$		$\sim\sim$ $[\mathrm{MeHg}]_{\mathrm{wat}}$	0.767	-	9	3.588	0.006			
Age-at-size$_{\mathrm{NRPK}}$	0.07	Chlorophyll-a	−0.263	0.322	9	−0.817	0.435	7.272	0.7	10
$\sim\sim$ Age-at-size$_{\mathrm{NRPK}}$		$\sim\sim$ $[\mathrm{MeHg}]_{\mathrm{invert}}$	0.309	-	11	0.919	0.193			
$\sim\sim$ Age-at-size$_{\mathrm{NRPK}}$		$\sim\sim$ $[\mathrm{MeHg}]_{\mathrm{wat}}$	0.71	-	11	2.855	0.011			
Age-at-size$_{\mathrm{NRPK}}$	0.03	DO	−0.166	0.329	9	−0.504	0.627	10.198	0.423	10
$\sim\sim$ Age-at-size$_{\mathrm{NRPK}}$		$\sim\sim$ $[\mathrm{MeHg}]_{\mathrm{invert}}$	0.287	-	11	0.847	0.211			
$\sim\sim$ Age-at-size$_{\mathrm{NRPK}}$		$\sim\sim$ $[\mathrm{MeHg}]_{\mathrm{wat}}$	0.789	-	11	3.63	0.003			
Age-at-size$_{\mathrm{NRPK}}$	0.42	DOC	0.647	0.254	9	2.542	0.032	8.559	0.381	8
$\sim\sim$ Age-at-size$_{\mathrm{NRPK}}$		$\sim\sim$ $[\mathrm{MeHg}]_{\mathrm{invert}}$	0.166	-	11	0.476	0.324			
$\sim\sim$ Age-at-size$_{\mathrm{NRPK}}$		$\sim\sim$ $[\mathrm{MeHg}]_{\mathrm{wat}}$	0.282	-	11	0.831	0.215			
$\sim\sim$ $[\mathrm{Hg}]_{\mathrm{NRPK}}$		$\sim\sim$DOC	−0.314	-	11	−0.935	0.189			
Age-at-size$_{\mathrm{NRPK}}$	0.01	Secchi	−0.098	0.332	9	−0.296	0.774	7.931	0.636	10
$\sim\sim$ Age-at-size$_{\mathrm{NRPK}}$		$\sim\sim$ $[\mathrm{MeHg}]_{\mathrm{invert}}$	0.323	-	11	0.964	0.182			
$\sim\sim$ Age-at-size$_{\mathrm{NRPK}}$		$\sim\sim$ $[\mathrm{MeHg}]_{\mathrm{wat}}$	0.745	-	11	3.161	0.007			

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Response		Predictor						Model		
Variable	r^2	Variable	Std. Est.	Std. Err.	df	Crit. Val.	p	Fisher's C	p	df
Age-at-size$_{\text{NRPK}}$	0.55	pH	0.738	0.225	9	3.285	0.01	10.022	0.439	10
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{invert}}$	0.042	-	11	0.119	0.454			
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{wat}}$	0.431	-	11	1.351	0.107			
Age-at-size$_{\text{NRPK}}$	0	Stratification	−0.050	0.333	9	−0.151	0.883	9.518	0.484	10
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{invert}}$	0.338	-	11	1.016	0.17			
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{wat}}$	0.744	-	11	3.145	0.007			
[Hg]$_{\text{NRPK}}$	0.83	[MeHg]$_{\text{invert}}$	0.328	0.215	8	1.525	0.166	8.559	0.381	8
		Age-at-size$_{\text{NRPK}}$	0.642	0.215	8	2.989	0.017			
[MeHg]$_{\text{invert}}$	0.64	[MeHg]$_{\text{wat}}$	0.426	0.258	8	1.655	0.137			
		[THg]$_{\text{sed}}$	0.477	0.258	8	1.853	0.101			
Age-at-size$_{\text{NRPK}}$	0.42	DOC	0.647	0.254	9	2.542	0.032			
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{invert}}$	0.166	-	11	0.476	0.324			
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{wat}}$	0.282	-	11	0.831	0.215			
$\sim\sim$ [Hg]$_{\text{NRPK}}$		$\sim\sim$ DOC	−0.314	-	11	−0.935	0.189			
[MeHg]$_{\text{wat}}$	0.1	[THg]$_{\text{wat}}$	0.309	0.317	9	0.974	0.356	15.395	0.496	16
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{invert}}$	0.166	-	11	0.476	0.324			
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{wat}}$	0.307	-	11	0.913	0.194			
$\sim\sim$ [Hg]$_{\text{NRPK}}$		$\sim\sim$ DOC	−0.314	-	11	−0.935	0.189			
$\sim\sim$ [MeHg]$_{\text{wat}}$		$\sim\sim$ DOC	0.81	-	11	3.904	0.002			
[MeHg]$_{\text{wat}}$	0.37	[THg]$_{\text{sed}}$	0.572	0.274	9	2.091	0.046	8.153	0.419	8
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{invert}}$	0.166	-	11	0.476	0.324			
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{wat}}$	0.314	-	11	0.936	0.188			
$\sim\sim$ [Hg]$_{\text{NRPK}}$		$\sim\sim$ DOC	−0.314	-	11	−0.935	0.189			
$\sim\sim$ [MeHg]$_{\text{wat}}$		$\sim\sim$ DOC	0.598	-	11	2.111	0.034			
[MeHg]$_{\text{wat}}$	0	[MeHg]$_{\text{sed}}$	−0.041	0.333	9	−0.122	0.906	23.543	0.1	16
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{invert}}$	0.166	-	11	0.476	0.324			
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{wat}}$	0.289	-	11	0.853	0.209			
$\sim\sim$ [Hg]$_{\text{NRPK}}$		$\sim\sim$ DOC	−0.314	-	11	−0.935	0.189			
$\sim\sim$ [MeHg]$_{\text{wat}}$		$\sim\sim$ DOC	0.858	-	11	4.721	0.001			
[MeHg]$_{\text{wat}}$	0.73	DOC	0.854	0.174	9	4.921	0.001	7.199	0.706	10
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{invert}}$	0.166	-	11	0.476	0.324			
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{wat}}$	0.542	-	11	1.823	0.053			
$\sim\sim$ [Hg]$_{\text{NRPK}}$		$\sim\sim$ DOC	−0.314	-	11	−0.935	0.189			
[MeHg]$_{\text{wat}}$	0.42	pH	0.645	0.255	9	2.535	0.032	21	0.179	16
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{invert}}$	0.166	-	11	0.476	0.324			

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Response		Predictor						Model		
Variable	r^2	Variable	Std. Est.	Std. Err.	df	Crit. Val.	p	Fisher's C	p	df
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ $[\text{MeHg}]_{\text{wat}}$	0.082	-	11	0.233	0.411			
$\sim\sim$ $[\text{Hg}]_{\text{NRPK}}$		$\sim\sim$ DOC	−0.314	-	11	−0.935	0.189			
$\sim\sim$ $[\text{MeHg}]_{\text{wat}}$		$\sim\sim$ DOC	0.492	-	11	1.597	0.075			
$[\text{MeHg}]_{\text{wat}}$	0	DO	0.065	0.333	9	0.196	0.849	17.958	0.326	16
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ $[\text{MeHg}]_{\text{invert}}$	0.166	-	11	0.476	0.324			
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ $[\text{MeHg}]_{\text{wat}}$	0.29	-	11	0.859	0.208			
$\sim\sim$ $[\text{Hg}]_{\text{NRPK}}$		$\sim\sim$ DOC	−0.314	-	11	−0.935	0.189			
$\sim\sim$ $[\text{MeHg}]_{\text{wat}}$		$\sim\sim$ DOC	0.863	-	11	4.834	0.001			
$[\text{MeHg}]_{\text{wat}}$	0.18	SO4	0.424	0.302	9	1.404	0.194	13.537	0.633	16
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ $[\text{MeHg}]_{\text{invert}}$	0.166	-	11	0.476	0.324			
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ $[\text{MeHg}]_{\text{wat}}$	0.27	-	11	0.792	0.226			
$\sim\sim$ $[\text{Hg}]_{\text{NRPK}}$		$\sim\sim$ DOC	−0.314	-	11	−0.935	0.189			
$\sim\sim$ $[\text{MeHg}]_{\text{wat}}$		$\sim\sim$ DOC	0.716	-	11	2.902	0.01			
$[\text{MeHg}]_{\text{wat}}$	0.24	Stratification	−0.488	0.291	9	−1.678	0.128	19.111	0.263	16
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ $[\text{MeHg}]_{\text{invert}}$	0.166	-	11	0.476	0.324			
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ $[\text{MeHg}]_{\text{wat}}$	0.57	-	11	1.964	0.043			
$\sim\sim$ $[\text{Hg}]_{\text{NRPK}}$		$\sim\sim$ DOC	−0.314	-	11	−0.935	0.189			
$\sim\sim$ $[\text{MeHg}]_{\text{wat}}$		$\sim\sim$ DOC	0.643	-	11	2.374	0.023			
$[\text{MeHg}]_{\text{wat}}$	0.1	Chlorophyll-a	−0.311	0.317	9	−0.982	0.352	16.132	0.444	16
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ $[\text{MeHg}]_{\text{invert}}$	0.166	-	11	0.476	0.324			
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ $[\text{MeHg}]_{\text{wat}}$	0.215	-	11	0.623	0.275			
$\sim\sim$ $[\text{Hg}]_{\text{NRPK}}$		$\sim\sim$ DOC	−0.314	-	11	−0.935	0.189			
$\sim\sim$ $[\text{MeHg}]_{\text{wat}}$		$\sim\sim$ DOC	0.862	-	11	4.804	0.001			
$[\text{MeHg}]_{\text{wat}}$	0.07	Secchi	−0.260	0.322	9	−0.808	0.44	16.096	0.446	16
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ $[\text{MeHg}]_{\text{invert}}$	0.166	-	11	0.476	0.324			
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ $[\text{MeHg}]_{\text{wat}}$	0.34	-	11	1.023	0.168			
$\sim\sim$ $[\text{Hg}]_{\text{NRPK}}$		$\sim\sim$ DOC	−0.314	-	11	−0.935	0.189			
$\sim\sim$ $[\text{MeHg}]_{\text{wat}}$		$\sim\sim$ DOC	0.787	-	11	3.605	0.004			
$[\text{MeHg}]_{\text{wat}}$	0	PC2	−0.051	0.333	9	−0.153	0.882	17.314	0.366	16
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ $[\text{MeHg}]_{\text{invert}}$	0.166	-	11	0.476	0.324			
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ $[\text{MeHg}]_{\text{wat}}$	0.283	-	11	0.835	0.214			
$\sim\sim$ $[\text{Hg}]_{\text{NRPK}}$		$\sim\sim$ DOC	−0.314	-	11	−0.935	0.189			
$\sim\sim$ $[\text{MeHg}]_{\text{wat}}$		$\sim\sim$ DOC	0.863	-	11	4.824	0.001			
$[\text{Hg}]_{\text{NRPK}}$	0.83	$[\text{MeHg}]_{\text{invert}}$	0.328	0.215	8	1.525	0.166	7.038	0.533	8
		Age-at-size$_{\text{NRPK}}$	0.642	0.215	8	2.989	0.017			
$[\text{MeHg}]_{\text{invert}}$	0.64	$[\text{MeHg}]_{\text{wat}}$	0.426	0.258	8	1.655	0.137			

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Table B.8; continued from previous page

Response		Predictor						Model		
Variable	r^2	Variable	Std. Est.	Std. Err.	df	Crit. Val.	p	Fisher's C	p	df
		[THg]$_{\text{sed}}$	0.477	0.258	8	1.853	0.101			
Age-at-size$_{\text{NRPK}}$	0.42	DOC	0.647	0.254	9	2.542	0.032			
[MeHg]$_{\text{wat}}$	0.73	DOC	0.822	0.238	8	3.461	0.009			
		[THg]$_{\text{sed}}$	0.05	0.238	8	0.208	0.84			
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{invert}}$	0.166	-	11	0.476	0.324			
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{wat}}$	0.539	-	11	1.811	0.054			
$\sim\sim$ [Hg]$_{\text{NRPK}}$		$\sim\sim$ DOC	−0.314	-	11	−0.935	0.189			
[THg]$_{\text{sed}}$	0	[THg]$_{\text{wat}}$	−0.033	0.333	9	−0.100	0.922	16.242	0.576	18
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{invert}}$	0.166	-	11	0.476	0.324			
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{wat}}$	0.539	-	11	1.811	0.054			
$\sim\sim$ [Hg]$_{\text{NRPK}}$		$\sim\sim$ DOC	−0.314	-	11	−0.935	0.189			
[THg]$_{\text{sed}}$	0.4	DOC	0.635	0.258	9	2.466	0.036	6.886	0.549	8
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{invert}}$	0.166	-	11	0.476	0.324			
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{wat}}$	0.539	-	11	1.811	0.054			
$\sim\sim$ [Hg]$_{\text{NRPK}}$		$\sim\sim$ DOC	−0.314	-	11	−0.935	0.189			
[THg]$_{\text{sed}}$	0.41	PC1	−0.638	0.257	9	−2.487	0.035	13.176	0.781	18
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{invert}}$	0.166	-	11	0.476	0.324			
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{wat}}$	0.539	-	11	1.811	0.054			
$\sim\sim$ [Hg]$_{\text{NRPK}}$		$\sim\sim$ DOC	−0.314	-	11	−0.935	0.189			
[THg]$_{\text{sed}}$	0.08	PC2	0.279	0.32	9	0.872	0.406	18.935	0.396	18
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{invert}}$	0.166	-	11	0.476	0.324			
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{wat}}$	0.539	-	11	1.811	0.054			
$\sim\sim$ [Hg]$_{\text{NRPK}}$		$\sim\sim$ DOC	−0.314	-	11	−0.935	0.189			
[THg]$_{\text{sed}}$	0.28	LA:CA	−0.525	0.284	9	−1.849	0.098	16.746	0.541	18
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{invert}}$	0.166	-	11	0.476	0.324			
$\sim\sim$ Age-at-size$_{\text{NRPK}}$		$\sim\sim$ [MeHg]$_{\text{wat}}$	0.539	-	11	1.811	0.054			
$\sim\sim$ [Hg]$_{\text{NRPK}}$		$\sim\sim$ DOC	−0.314	-	11	−0.935	0.189			
[Hg]$_{\text{NRPK}}$	0.83	[MeHg]$_{\text{invert}}$	0.328	0.215	8	1.525	0.166	11.939	0.748	16
		Age-at-size$_{\text{NRPK}}$	0.642	0.215	8	2.989	0.017			
[MeHg]$_{\text{invert}}$	0.64	[MeHg]$_{\text{wat}}$	0.426	0.258	8	1.655	0.137			
		[THg]$_{\text{sed}}$	0.477	0.258	8	1.853	0.101			
Age-at-size$_{\text{NRPK}}$	0.42	DOC	0.647	0.254	9	2.542	0.032			
[MeHg]$_{\text{wat}}$	0.73	DOC	0.822	0.238	8	3.461	0.009			
		[THg]$_{\text{sed}}$	0.05	0.238	8	0.208	0.84			
[THg]$_{\text{sed}}$	0.44	DOC	0.339	0.468	8	0.725	0.489			

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Table B.8; continued from previous page

Response		Predictor						Model		
Variable	r^2	Variable	Std. Est.	Std. Err.	df	Crit. Val.	p	Fisher's C	p	df
		PC1	−0.358	0.468	8	−0.765	0.466			
∼∼ Age-at-size$_{\text{NRPK}}$		∼∼ [MeHg]$_{\text{invert}}$	0.166	-	11	0.476	0.324			
∼∼ Age-at-size$_{\text{NRPK}}$		∼∼ [MeHg]$_{\text{wat}}$	0.539	-	11	1.811	0.054			
∼∼ [Hg]$_{\text{NRPK}}$		∼∼ DOC	−0.314	-	11	−0.935	0.189			
DOC	0.01	Chlorophyll-a	−0.112	0.331	9	−0.338	0.743	31.342	0.302	28
∼∼ Age-at-size$_{\text{NRPK}}$		∼∼ [MeHg]$_{\text{invert}}$	0.166	-	11	0.476	0.324			
∼∼ Age-at-size$_{\text{NRPK}}$		∼∼ [MeHg]$_{\text{wat}}$	0.539	-	11	1.811	0.054			
∼∼ [Hg]$_{\text{NRPK}}$		∼∼ DOC	−0.330	-	11	−0.989	0.176			
DOC	0.58	LA:CA	−0.760	0.217	9	−3.509	0.007	21.878	0.787	28
∼∼ Age-at-size$_{\text{NRPK}}$		∼∼ [MeHg]$_{\text{invert}}$	0.166	-	11	0.476	0.324			
∼∼ Age-at-size$_{\text{NRPK}}$		∼∼ [MeHg]$_{\text{wat}}$	0.539	-	11	1.811	0.054			
∼∼ [Hg]$_{\text{NRPK}}$		∼∼ DOC	−0.455	-	11	−1.446	0.093			
DOC	0.68	PC1	−0.826	0.188	9	−4.399	0.002	10.261	0.853	16
∼∼ Age-at-size$_{\text{NRPK}}$		∼∼ [MeHg]$_{\text{invert}}$	0.166	-	11	0.476	0.324			
∼∼ Age-at-size$_{\text{NRPK}}$		∼∼ [MeHg]$_{\text{wat}}$	0.539	-	11	1.811	0.054			
∼∼ [Hg]$_{\text{NRPK}}$		∼∼ DOC	−0.334	-	11	−1.002	0.173			
DOC	0.02	PC2	0.151	0.33	9	0.458	0.658	30.556	0.337	28
∼∼ Age-at-size$_{\text{NRPK}}$		∼∼ [MeHg]$_{\text{invert}}$	0.166	-	11	0.476	0.324			
∼∼ Age-at-size$_{\text{NRPK}}$		∼∼ [MeHg]$_{\text{wat}}$	0.539	-	11	1.811	0.054			
∼∼ [Hg]$_{\text{NRPK}}$		∼∼ DOC	−0.349	-	11	−1.053	0.162			
[Hg]$_{\text{NRPK}}$	0.83	[MeHg]$_{\text{invert}}$	0.328	0.215	8	1.525	0.166	17.106	0.906	26
		Age-at-size$_{\text{NRPK}}$	0.642	0.215	8	2.989	0.017			
[MeHg]$_{\text{invert}}$	0.64	[MeHg]$_{\text{wat}}$	0.426	0.258	8	1.655	0.137			
		[THg]$_{\text{sed}}$	0.477	0.258	8	1.853	0.101			
Age-at-size$_{\text{NRPK}}$	0.42	DOC	0.647	0.254	9	2.542	0.032			
[MeHg]$_{\text{wat}}$	0.73	DOC	0.822	0.238	8	3.461	0.009			
		[THg]$_{\text{sed}}$	0.05	0.238	8	0.208	0.84			
[THg]$_{\text{sed}}$	0.44	DOC	0.339	0.468	8	0.725	0.489			
		PC1	−0.358	0.468	8	−0.765	0.466			
DOC	0.69	PC1	−0.667	0.389	8	−1.715	0.125			
		LA:CA	−0.185	0.389	8	−0.475	0.648			
∼∼ Age-at-size$_{\text{NRPK}}$		∼∼ [MeHg]$_{\text{invert}}$	0.166	-	11	0.476	0.324			
∼∼ Age-at-size$_{\text{NRPK}}$		∼∼ [MeHg]$_{\text{wat}}$	0.539	-	11	1.811	0.054			
∼∼ [Hg]$_{\text{NRPK}}$		∼∼ DOC	−0.374	-	11	−1.142	0.143			

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Table B.8; continued from previous page

Response		Predictor						Model		
Variable	r^2	Variable	Std. Est.	Std. Err.	df	Crit. Val.	p	Fisher's C	p	df
PC1	0.78	Elevation	0.884	0.156	9	5.658	0	28.474	0.913	40
$\sim\sim$ Age-at-size$_{NRPK}$		$\sim\sim$ [MeHg]$_{invert}$	0.166	-	11	0.476	0.324			
$\sim\sim$ Age-at-size$_{NRPK}$		$\sim\sim$ [MeHg]$_{wat}$	0.539	-	11	1.811	0.054			
$\sim\sim$ [Hg]$_{NRPK}$		$\sim\sim$ DOC	−0.374	-	11	−1.142	0.143			
PC1	0.55	Slope	−0.740	0.224	9	−3.297	0.009	55.603	0.041	40
$\sim\sim$ Age-at-size$_{NRPK}$		$\sim\sim$ [MeHg]$_{invert}$	0.166	-	11	0.476	0.324			
$\sim\sim$ Age-at-size$_{NRPK}$		$\sim\sim$ [MeHg]$_{wat}$	0.539	-	11	1.811	0.054			
$\sim\sim$ [Hg]$_{NRPK}$		$\sim\sim$ DOC	−0.374	-	11	−1.142	0.143			
			Final Piecewise-SEM							
[Hg]$_{NRPK}$	0.829	[MeHg]$_{invert}$	0.328	0.215	8	1.525	0.166	28.474	0.913	40
		Age-at-size$_{NRPK}$	0.642	0.215	8	2.989	0.017			
[MeHg]$_{invert}$	0.640	[MeHg]$_{wat}$	0.426	0.258	8	1.655	0.137			
		[THg]$_{sed}$	0.477	0.258	8	1.853	0.101			
Age-at-size$_{NRPK}$	0.418	DOC	0.647	0.254	9	2.542	0.032			
[MeHg]$_{wat}$	0.730	DOC	0.822	0.238	8	3.461	0.009			
		[THg]$_{sed}$	0.05	0.238	8	0.208	0.84			
[THg]$_{sed}$	0.444	DOC	0.339	0.468	8	0.725	0.489			
		PC1	−0.358	0.468	8	−0.765	0.466			
DOC	0.691	PC1	−0.667	0.389	8	−1.715	0.125			
		LA:CA	−0.185	0.389	8	−0.475	0.648			
PC1	0.781	Elevation	0.884	0.156	9	5.658	< 0.001			
$\sim\sim$ Age-at-size$_{NRPK}$		$\sim\sim$ [MeHg]$_{invert}$	0.166	-	11	0.476	0.324			
$\sim\sim$ Age-at-size$_{NRPK}$		$\sim\sim$ [MeHg]$_{wat}$	0.539	-	11	1.811	0.054			
$\sim\sim$ [Hg]$_{NRPK}$		$\sim\sim$ DOC	−0.374	-	11	−1.142	0.143			

Table B.9. Variation inflation factor (VIF) for sub-models with $>$ one predictors in piecewise-SEM (see Table B.8).

Response	Predictors	VIF
$[\mathrm{Hg}]_{\mathrm{NRPK}}$	$[\mathrm{MeHg}]_{\mathrm{invert}}$ + Age-at-size$_{\mathrm{NRPK}}$	2.157
$[\mathrm{MeHg}]_{\mathrm{invert}}$	$[\mathrm{MeHg}]_{\mathrm{wat}}$ + $[\mathrm{THg}]_{\mathrm{sed}}$	1.587
$[\mathrm{MeHg}]_{\mathrm{wat}}$	DOC + $[\mathrm{THg}]_{\mathrm{sed}}$	1.140
DOC	PC1 + LA:CA	3.917

Table B.10. Results of piecewise-SEM sub-models investigating hypothesized pathways that influence concentrations of MeHg in water. Concentrations of total Hg and MeHg in water are from filtered samples (see Table B.5) and concentrations of total Hg and MeHg in sediments are normalized to organic matter content of sediments (see Table B.6). Overall, results are similar to findings obtained when concentrations of total Hg and MeHg in unfiltered water samples and non-normalized total Hg and MeHg concentrations in sediment smaples are used (see corresponding relationships in Table B.8).

Response		Predictor					
Variable	r^2	Variable	Std. Est.	Std. Err.	df	Crit. Val.	p
$[\text{MeHg}]_{\text{wat}}$	0.00	$[\text{MeHg}]_{\text{sed}}$	−0.056	0.333	9	−0.168	0.871
$[\text{MeHg}]_{\text{wat}}$	0.48	$[\text{THg}]_{\text{wat}}$	0.691	0.241	9	2.865	0.019
$[\text{MeHg}]_{\text{wat}}$	0.32	[THg]sed	0.563	0.276	9	2.041	0.072
$[\text{MeHg}]_{\text{wat}}$	0.69	DOC	0.83	0.186	9	4.461	0.002
$[\text{MeHg}]_{\text{wat}}$	0.42	pH	0.612	0.253	9	2.576	0.03
$[\text{MeHg}]_{\text{wat}}$	0.00	DO	0.04	0.333	9	0.119	0.908
$[\text{MeHg}]_{\text{wat}}$	0.18	SO_4	0.43	0.301	9	1.427	0.187
$[\text{MeHg}]_{\text{wat}}$	0.22	Stratification	−0.474	0.291	9	−1.615	0.141
$[\text{MeHg}]_{\text{wat}}$	0.14	Chlorophyll-a	−0.368	0.31	9	−1.186	0.266
$[\text{MeHg}]_{\text{wat}}$	0.06	Secchi	−0.244	0.322	9	−0.755	0.469
$[\text{MeHg}]_{\text{wat}}$	0.01	PC2	−0.1080	0.331	9	−0.3259	0.752

Table B.11. Average concentrations of MeHg (ng/g) in benthic invertebrates collected from littoral (near shore) and profundal (off shore) areas in the 11 study lakes (all lakes combined)..

Taxa	Littoral	Profundal
All (taxa shown in Table B.4)	67.53	16.30
Bivalvia (Veneroida)	54.64	42.89
Clitellata (Hirudinida)	24.31	8.94
Gastropoda (Basommatophora & Heterostropha)	21.27	17.71
Malacostraca (Amphipoda)	29.05	23.45
Insecta (Diptera)	16.94	7.83
Insecta (Ephemeroptera)	20.72	20.20

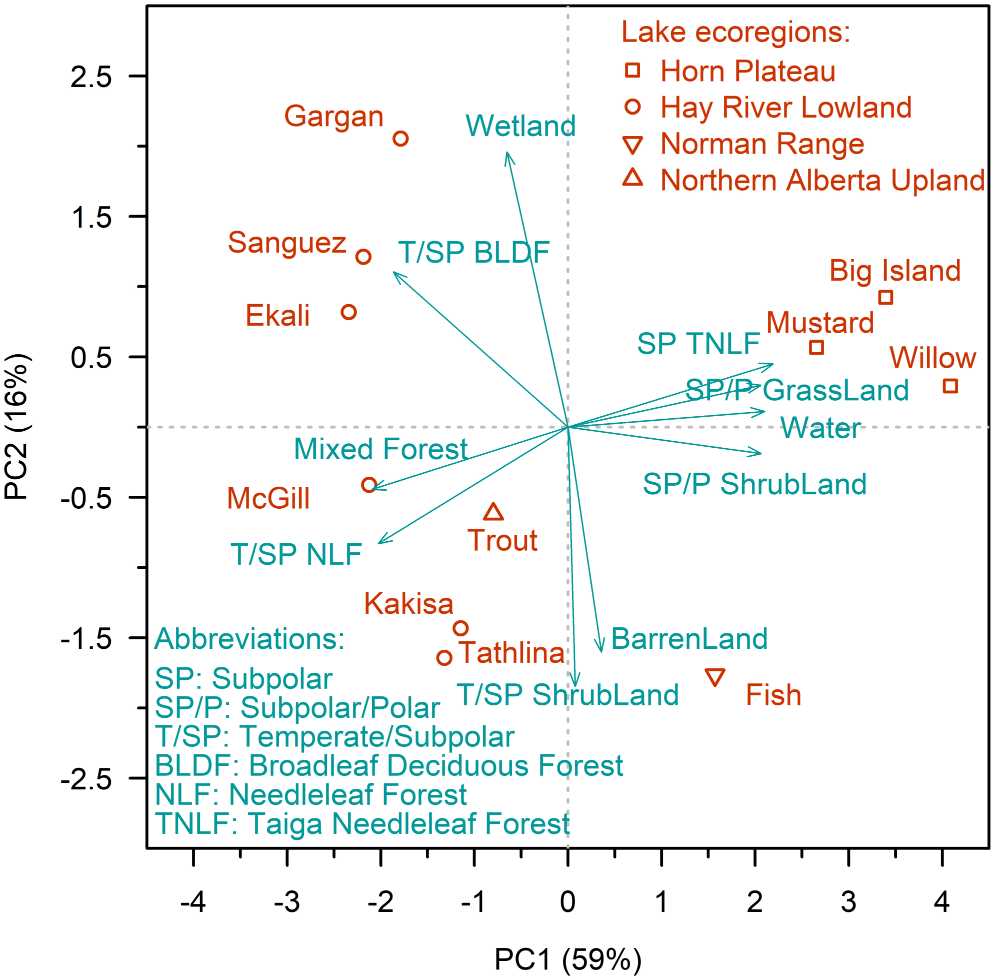

Figure B.1. Results of Principal Component Analysis on proportional land cover data (arrows) in catchments of the study lakes (symbols) in the Dehcho Region, NT, Canada. Results indicated that lakes in the Hay River Lowland ecoregion (i.e., Ekali, Sanguez, Gargan, and McGill, Kakisa, and Tathlina) were clearly separated from lakes in the Horn Plateau ecoregion (i.e., Big Island, Mustard, and Willow) along PC1, with Fish Lake (Norman Range ecoregion) and Trout Lake (Northern Alberta Upland ecoregion) in intermediate positions. Temperate/subpolar needleleaf and mixed forests were negatively associated with PC1, whereas subpolar/polar grass and shrub lands were positively associated with PC1. Catchment composition was dominated by mixed and broadleaf deciduous forests for lakes located in the Hay River Lowland ecoregion, and by shrub and grass lands for lakes located in the Horn Plateau ecoregion. Separation of lakes along PC2 was not associated with ecoregions; PC2 represented gradients of proportional land cover from barren and temperate shrub lands (negative loadings) to wetland (positive loadings). Together, the first two axes explained 75% of the variance in catchment land cover.

C Supplementary Information for Chapter 3

Table C.1. Latitude and longitude, mode of access, and collection years for water (W), Northern Pike (F), and benthic invertebrate (B) samples in the study lakes. Sampling took place from mid-August to mid-September in each year. Additional fish were also collected by Indigenous community members in March (F^{*}) and May (F^{**}).

Lake	Latitude (N)	Longtitdue (W)	Access	Year						
				2013	2014	2015	2016	2017	2018	2019
Fish	63°10’56.56″	122°32’37.79″	Floatplane						W	W, F, B
Big Islalnd	62°05′12.78″	119°52′08.04″	Floatplane	W, F, B			W	W, F, B	W	W
Willow	62°10′09.82″	119°11′05.20″	Floatplane				W, F, B		W	W
Mustard	62°00′08.29″	120°05′37.90″	Floatplane			W, F, B	W	W	W	W
Trout	60°36′51.74″	121°10′09.56″	Road	W, F, B					W	W
Kakisa	60°55′23.91″	117°39′56.10″	Road			W, F, B		W, F, B	W, F^{*}	W
Tathlina	60°32′19.81″	117°30′29.04″	Road		W, F, B		F^{*}		W, F, B	W
Ekali	61°17′22.68″	120°34′58.33″	Trail	W, F, B		W	W		W	W
Gargan	61°14′56.40″	120°22′55.53″	Trail		W, F, B				W	W
McGill	61°18′12.42″	121°00′49.96″	Floatplane		W, F, B				W	W
Sanguez	61°15′14.51″	120°29′32.06″	Trail	W, F, B			W, F, B	W, F, B	W, F, F^{**}	W, F, F^{**}

Table C.2. Physical and chemical characteristics of the study lakes. Data for water chemistry variables are presented as arithmetic means (using all available data; see Table C.1) with standard deviations in (). Ecoregions represented by the study lakes include the Norman Range (NR), Horn Plateau (HP), Northern Alberta Upland (NAU), and Hay River Lowland (HRL).

Variable (unit)	Lake (ecoregion)				
	Fish (NR)	Big Island (HP)	Willow (HP)	Mustard (HP)	Trout (NAU)
LA (km^2)	117.1	18.2	128.6	23.3	500
CA (km^2))	2616.9	113	1248.8	198.7	5893.5
LA:CA (ratio)	0.045	0.161	0.103	0.117	0.085
Elevation (m)	428.25	783.23	704.83	750.83	584.69
Slope (m/s)	2.57	2.49	1.79	2.22	3.34
CTSI	40.3	39.6	41.4	44.7	44.5
TP (μg/L)	8.0 (1.41)	7.0 (2.16)	7.0 (2.83)	11.0 (0.82)	10.3 (2.52)
TN (µg/L)	357.0 (56.57)	320.8 (26.60)	342.0 (31.11)	435.5 (39.95)	378.3 (54.50)
Chl-a (µg/L)	3.4 (1.22)	5.2 (1.36)	5.3 (0.84)	7.2 (3.39)	6.9 (1.79)
Secchi (m)	3.0 (NA)	4.0 (NA)	2.8 (NA)	2.7 (0.35)	2.7 (1.20)
DOC (mg/L)	13.1 (0.93)	9.8 (0.54)	10.0 (1.18)	11.5 (0.85)	13.2 (1.58)
DIC (mg/L)	18.3 (0.95)	9.8 (1.29)	14.3 (1.02)	9.6 (1.24)	18.7 (6.29)
abs254 (mg/L)	0.4 (0.00)	0.2 (0.02)	0.2 (0.03)	0.3 (0.02)	0.4 (0.05)
SUVA (mg DOC/L/m)	2.7 (0.17)	2.4 (0.11)	2.4 (0.03)	2.3 (0.18)	2.6 (0.57)
Freshness (mg/L)	0.66 (0.01)	0.66 (0.02)	0.69 (0.01)	0.65 (0.03)	0.66 (0.03)
Alkalinity (mg/L)	81.8 (5.02)	47.5 (3.59)	65.6 (0.59)	45.8 (0.93)	67.0 (4.35)
Bicarbonate (mg/L)	99.8 (6.11)	58.0 (4.39)	80.0 (0.73)	55.9 (1.13)	84.8 (5.30)
Conductivity (µS/cm)	198.1 (NA)	99.8 (NA)	144.4 (NA)	112.9 (6.86)	139.5 (16.19)
Na (mg/L)	2.8 (NA)	1.0 (0.13)	1.5 (0.11)	1.3 (0.30)	2.3 (0.41)
Ca (mg/L)	29.5 (NA)	16.7 (2.10)	23.0 (2.18)	18.1 (2.41)	25.0 (5.92)
Mg (mg/L)	10.0 (NA)	4.3 (0.66)	6.3 (0.62)	4.2 (0.63)	4.9 (0.76)
Cl (mg/L)	1.3 (0.76)	0.1 (0.04)	0.1 (0.03)	0.1 (0.04)	0.4 (0.08)

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Table C.2; continued from previous page

Variable (unit)	Lake (ecoregion)					
	Kakisa (HRL)	Tathlina (HRL)	Ekali (HRL)	Gargan (HRL)	McGill (HRL)	Sanguez (HRL)
LA (km^2)	335.8	565.3	1.9	1.1	2.3	1.6
CA (km^2)	15808.8	11606.3	179.8	73.2	982.6	106.9
LA:CA (ratio)	0.021	0.049	0.01	0.015	0.002	0.015
Elevation (m)	408.64	456.69	274.04	283.46	354.56	277.99
Slope (m/s)	4.17	4.27	4.36	2.33	5.22	3.19
CTSI	58.4	44.2	45.7	47.1	39.4	43.6
TP (μg/L)	30.5 (20.09)	10.3 (3.06)	14.4 (5.98)	14.0 (3.46)	7.25 (1.26)	9.7 (1.97)
TN (µg/L)	474.8 (95.25)	663.3 (71.84)	618.4 (97.02)	682.5 (41.58)	514.0 (49.20)	541.3 (64.26)
Chl-a (µg/L)	15.5 (14.78)	3.5 (2.73)	7.1 (1.39)	8.6 (4.12)	2.2 (0.27)	5.6 (0.50)
Secchi (m)	0.8 (0.49)	1.8 (1.10)	2.9 (0.42)	2.4 (0.53)	2.5 (0.07)	2.6 (0.87)
DOC (mg/L)	12.9 (2.68)	19.7 (1.22)	16.6 (2.51)	21.3 (6.95)	17.5 (2.69)	17.3 (4.41)
DIC (mg/L)	22.9 (2.95)	29.6 (1.47)	22.6 (2.26)	19.9 (2.08)	25.0 (3.37)	21.3 (2.23)
abs254 (mg/L)	0.3 (0.11)	0.7 (0.03)	0.5 (0.08)	0.5 (0.13)	0.5 (0.08)	0.4 (0.06)
SUVA (mg DOC/L/m)	2.3 (0.39)	3.1 (0.44)	2.7 (0.41)	2.4 (0.14)	3.0 (0.36)	2.4 (0.22)
Freshness (mg/L)	0.69 (0.04)	0.63 (0.04)	0.66 (0.03)	0.66 (0.02)	0.61 (0.03)	0.65 (0.03)
Alkalinity (mg/L)	110.6 (6.87)	137.3 (6.15)	109.9 (9.73)	89.5 (5.37)	112.9 (5.24)	103.1 (7.61)
Bicarbonate (mg/L)	134.9 (8.42)	128.3 (62.37)	127.9 (12.70)	112.1 (17.18)	137.7 (6.42)	134.7 (30.27)
Conductivity (µS/cm)	233.3 (26.59)	283.0 (25.46)	219.3 (18.74)	212.9 (11.10)	239.2 (57.77)	224.8 (35.71)
Na (mg/L)	5.2 (0.48)	5.8 (1.04)	4.0 (0.72)	2.9 (0.15)	5.6 (1.27)	4.0 (0.53)
Ca (mg/L)	40.7 (4.05)	52.9 (9.61)	40.9 (5.18)	33.9 (3.47)	41.5 (3.24)	38.6 (5.44)
Mg (mg/L)	8.0 (0.99)	9.8 (1.29)	7.6 (0.87)	6.6 (0.61)	8.7 (0.98)	7.1 (0.83)
Cl (mg/L)	1.4 (0.31)	1.3 (0.41)	2.7 (0.65)	1.6 (0.36)	1.9 (0.68)	2.4 (1.24)

Table C.3. Trophic state indices based on chlorophyll-a concentrations ($TSI_{Chl\text{-}a}$), total phosphorus concentrations (TSI_{TP}), and secchi depth (TSI_{Secchi}), as well as Carlson's trophic state index (CTSI) and nitrogen to phosphorus ratios (TN:TP) for the study lakes in the Dehcho region, NT, Canada.

Lake	$TSI_{Chl\text{-}a}$	TSI_{TP}	TSI_{Secchi}	CTSI	State	TN:TP
Fish	44.17	42.71	34.14	40.34	mesotrophic	44.63
Big Island	40.02	46.68	32.21	39.64	mesotrophic	45.82
Willow	45.16	46.88	32.21	41.42	mesotrophic	48.86
Mustard	45.42	50.00	38.73	44.72	mesotrophic	39.59
Trout	45.96	49.6	37.83	44.46	mesotrophic	36.61
Kakisa	64.15	57.50	53.43	58.36	eutrophic	15.57
Tathlina	51.94	42.92	37.83	44.23	mesotrophic	64.19
Ekali	44.66	49.85	42.61	45.71	mesotrophic	42.94
Gargan	47.54	51.69	42.21	47.14	mesotrophic	48.75
McGill	47.09	38.50	32.72	39.43	mesotrophic	70.90
Sanguez	46.34	47.54	36.86	43.58	mesotrophic	56.00

Table C.4. Descriptive statistics for widths (in mm) of 2,953 annuli measured on 432 Northern Pike cleithra sampled from lakes in Dehcho Region, NT, Canada. Annuli of the current years' growth and 0^+ individuals are not included.

Age-at-capture	n fish	Annuli															
		1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
1	12	15.8															
2	30	14.6	25.3														
3	37	14.3	25.2	33.9													
4	54	13.7	25.1	33.6	40.2												
5	53	14.3	25.7	34.2	40.9	46.1											
6	46	14	25.3	34.1	41	46.5	51.2										
7	32	13.7	24.8	33.3	40	45.9	50.9	55.3									
8	29	13.7	24.5	32.8	39.4	45	50.5	55.5	59.9								
9	41	13.7	24.8	33.2	40.2	45.6	50.5	55.1	59.4	63							
10	22	13.8	23.7	31.6	38.1	43.4	47.8	52	56.2	60.1	63.4						
11	19	13.4	23.8	31.5	38.3	43.7	48.4	52.8	56.9	60.7	64.7	68					
12	16	15	26.2	34.6	41.2	47	52.1	57.5	62.2	66.5	70.7	74.6	78.2				
13	21	14.7	25.6	34	40	45.2	50.1	55.4	60.1	64.6	68.7	72.6	76.3	79.4			
14	10	14.2	24.8	33.4	40.4	46	50.5	54.9	58.9	62.9	66.9	70.4	73.9	77.4	80.4		
15	6	14.7	25.7	33.8	40.3	45.3	49.8	54.1	58.7	63.2	67.4	71.3	74.9	78.4	81.4	84.2	
16	4	14.1	25	33.4	41.2	48.5	55.2	61.8	68.3	73	78	82.5	86	89.9	93	95.2	97.6
Mean		14.1	25.1	33.5	40.1	45.6	50.4	54.9	59.3	63.2	67.2	72	76.9	79.8	83.2	88.6	97.6
SE		0.11	0.16	0.2	0.23	0.26	0.31	0.4	0.5	0.6	0.8	0.99	1.19	1.49	2.35	3.55	2.32
n annuli		432	420	390	353	299	246	200	168	139	98	76	57	41	20	10	4

Table C.5. Results of linear regressions between mean δ^{13}C in littoral benthic invertebrates (all taxa combined as well as individual taxonomic families commonly observed in the studied lakes; see Table C.6) and inferred catchment influence (i.e., PC1 of the second PCA; Figure 3.3b) using data from all study lakes (n = 11) in the Dehcho Region, NT, Canada.

Family	Statistics of linear models				
	intercept	Slope	adj. r^2	F (df)	p
All taxa combined	−28.98	−1.01	0.68	21.82 (1,9)	0.001
Sphaeriidae	−31.23	−0.85	0.77	33.19 (1,9)	< 0.001
Dytiscidae	−29.32	−1.03	0.83	49.98 (1,9)	< 0.001
Corixidae	−30.35	−0.98	0.65	17.40 (1,8)	0.003
Lymnaeidae	−26.16	−1.17	0.72	23.41 (1,8)	0.001
Chironomidae	−30.61	−0.95	0.36	06.61 (1,9)	0.031

Table C.6. Presence (⋆) of benthic invertebrate families collected from littoral zone of the study lakes in the Dehcho Region, NT, Canada. Commonly observed (n lake $\geq$ 9) taxa are in bold.

Family	Lake											
	Fish	Big Island	Willow	Mustard	Trout	Kakisa	Tathlina	Ekali	Gargan	McGill	Sanguez	n lake
Erpobdellidae	⋆	⋆				⋆	⋆	⋆	⋆	⋆		7
Glossiphoniidae	⋆	⋆				⋆		⋆	⋆	⋆		6
Piscicolidae	⋆	⋆										2
Hydrachnidiae		⋆		⋆		⋆				⋆		4
Laevicaudata									⋆			1
Daphniidae								⋆		⋆		2
Dytiscidae	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆	11
Gyrinidae									⋆	⋆		2
Haliplidae	⋆					⋆		⋆			⋆	4
Ceratopogonidae	⋆									⋆		2
Chironomidae	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆	11
Tabanidae		⋆								⋆		2
Tipulidae	⋆			⋆							⋆	3
Baetidae	⋆	⋆		⋆		⋆	⋆	⋆		⋆	⋆	8
Caenidae	⋆	⋆	⋆	⋆		⋆	⋆	⋆			⋆	8
Ephemeridae	⋆					⋆	⋆			⋆	⋆	5
Heptageniidae	⋆	⋆				⋆	⋆	⋆	⋆	⋆		7
Leptohyphidae						⋆						1
Leptophlebiidae				⋆		⋆		⋆		⋆		4
Chironomidae	⋆	⋆	⋆	⋆		⋆	⋆	⋆	⋆	⋆	⋆	10
Notonectidae	⋆				⋆				⋆	⋆		4
Sialidae								⋆				1
Aeshnidae	⋆				⋆	⋆	⋆	⋆	⋆	⋆	⋆	8
Coenagrionidae					⋆	⋆	⋆	⋆	⋆	⋆	⋆	7
Corduliidae	⋆				⋆							2
Gomphidae							⋆					1
Libellulidae								⋆			⋆	2
Nemouridae		⋆										1
Perlidae	⋆		⋆									2
Apataniidae				⋆								1
Helicopsychidae						⋆						1
Lepidostomatidae						⋆						1

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Table C.6; continued from previous page

Family	Lake											
	Fish	Big Island	Willow	Mustard	Trout	Kakisa	Tathlina	Ekali	Gargan	McGill	Sanguez	n lake
Leptoceridae				⋆		⋆		⋆		⋆		4
Limnephilidae						⋆		⋆	⋆	⋆		4
Molannidae								⋆		⋆		2
Phryganeidae	⋆			⋆	⋆	⋆	⋆			⋆	⋆	7
Polycentropodidae						⋆	⋆					2
Unionidae				⋆		⋆	⋆	⋆	⋆	⋆	⋆	7
Sphaeriidae	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆	11
Lymnaeidae	⋆	⋆	⋆	⋆		⋆	⋆	⋆	⋆	⋆	⋆	10
Physidae		⋆				⋆	⋆	⋆	⋆	⋆	⋆	7
Planorbidae	⋆					⋆	⋆		⋆	⋆		5
Valvatidae	⋆	⋆	⋆	⋆		⋆				⋆		6
Gammaridae	⋆	⋆				⋆	⋆	⋆		⋆		6
Crangonyctidae	⋆											1
Hyalellidae	⋆	⋆				⋆	⋆	⋆				5

Table C.7. Presence (⋆) of fish species retrieved from gill nets deployed in the study lakes in the Dehcho Region, NT, Canada.

Species	Lake										
	Fish (NR)	Big Island (HP)	Willow (HP)	Mustard (HP)	Trout (HRL)	Kakisa (HRL)	Tathlina (HRL)	Ekali (HRL)	Gargan (HRL)	McGill (HRL)	Sanguez (HRL)
Barbot (*Lota lota*)		⋆		⋆	⋆			⋆	⋆	⋆	
Brook Stickleback (*Culaea inconstans*)									⋆	⋆	⋆
Cisco (*Coregonus artedii*)	⋆	⋆	⋆		⋆	⋆		⋆	⋆	⋆	⋆
Longnose Sucker (*Catostomus catostomus*)	⋆	⋆		⋆	⋆	⋆	⋆			⋆	
Lake Trout (*Salvelinus namaycush*)		⋆	⋆	⋆	⋆						
Lake Whitefish (*Coregonus clupeaformis*)	⋆	⋆	⋆		⋆	⋆	⋆	⋆	⋆	⋆	⋆
Northern Pike (*Esox lucius*)	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆	⋆
Ninespine Stickleback (*Pungitius pungitius*)		⋆	⋆	⋆	⋆				⋆	⋆	⋆
Walleye (*Sander vitreus*)	⋆				⋆	⋆	⋆	⋆		⋆	⋆
White Sucker (*Catostomus commersoni*)	⋆					⋆	⋆			⋆	

D Supplementary Information for Chapter 4

Table D.1. Latitudes and longitudes (approximate centre), modes of access, and collection years for sediment (s), water (w), fish (f), and primary consumer (p) samples in the study lakes. Sampling took place from mid-August to mid-September in each year. Additional fish were also collected by Indigenous community members in March (f*) and May (f**).

Lake	Latitude (N)	Longtitdue (W)	Access	Year						
				2013	2014	2015	2016	2017	2018	2019
Big Island	62°05′12″	119°52′08″	Floatplane	s, w, f, p	-	-	s, w	s, w, f, p	s, w	s, w
Mustard	62°00′08″	120°05′37″	Floatplane	-	-	s, w, f, p	s, w	W	s, w	s, w
Willow	62°10′09″	119°11′05″	Floatplane	-	-	-	s, w, f, p	-	s, w	s, w
Fish	63°10′56″	122°32′37″	Floatplane	-	-	-	-	-	s, w	s, w, f, p
Gargan	61°14′56″	120°22′55″	Trail	-	s, w, f, p	-	-	-	s, w	s, w
Ekali	61°17′22″	120°34′58″	Trail	s, w, f, p	-	s, w	s, w	-	s, w	s, w
Sanguez	61°15′14″	120°29′32″	Trail	s, w, f, p	-	-	s, w, f, p	s, w, f, p	s, w, f, f**	s, w, f, f**
Kakisa	60°55′23″	117°39′56″	Road	-	-	s, w, f, p	-	s, w, f, p	s, w, f*	s, w
Tathlina	60°32′19″	117°30′29″	Road	-	s, w, f, p	-	f*	-	s, w, f, p	s, w
McGill	61°18′12″	121°00′49″	Floatplane	-	s, w, f, p	-	-	-	s, w, f, p	s, w

Table D.2. Quality control-quality assurance (QA/QC) data for analyses of δ^{15}N ratios, total Hg, and MeHg. The number (n) of duplicates (DUP) analyzed is included, and both values of mean absolute difference (AD) ± standard deviation and mean relative percent difference (RPD) ± standard deviation are presented for duplicate sample analyses. Mean ± standard deviation of detection limits (DL), recoveries of Dorm-4 reference material (RM), and spike recoveries (SR) are also presented for total Hg and MeHg analyses.

Analyses	Parameter	Fish	Primary consumer	Water	Sediment
δ^{15}N	DUP	AD: 0.025 ± 0.281 RPD: 2.13 ± 1.78 ‰ (n = 169)	AD: 0.009 ± 0.279 RPD: 9.82 ± 37.75 ‰ (n = 122)	-	-
Hg	DUP	AD: 0.034 ± 0.060 RPD: 5.61 ± 7.65 ng/g dry (n = 6)	-	AD: 0.118 ± 0.171 RPD: 12.83 ± 17.49 ng/L (n = 7)	AD: 0.919 ± 9.016 RPD: 11.90 ± 9.21 ng/g (n = 6)
MeHg	DUP	-	AD: 13.170 ± 17.110 RPD: 52.88 ± 20.12 ng/g dry (n = 10)	AD: 0.002 ± 0.003 RPD: 2.63 ± 3.73 ng/L (n = 7)	AD: 0.124 ± 0.168 RPD: 56.93 ± 40.76 ng/g (n = 6)
Hg	DL	0.067 ± 0.027 ng	-	0.003 ± <0.001 ng	0.073 ± 0.005 ng
	RM	99.8 ± 3.4 % (n = 64)	-	102.7 ± 7.6 % (n = 10)	105.0 ± 8.6 % (n = 6)
	SR	98.0 ± 9.5 % (n = 64)	-	101.3 ± 1.5 % (n = 10)	102.5 ± 1.9 % (n = 6)
MeHg	DL	0.005 ± <0.001 ng	0.028 ± 0.026 ng	0.002 ± 0.001 ng	0.004 ± 0.002 ng
	RM	85.3 ± 5.0 % (n = 74)	93.1 ± 6.0 % (n = 53)	90.7 ± 6.0 % (n = 10)	100.5 ± 3.1 % (n = 6)
	SR	99.7 ± 8.0 % (n = 74)	100.3 ± 6.3 % (n = 53)	97.3 ± 4.9 % (n = 10)	96.8 ± 6.3 % (n = 6)

Table D.3. Characteristics of each lake and catchment studied in the Dehcho Region, NT, Canada. Values of in-lake variables (water and sediment) are presented as mean ± standard deviation calculated from all samples collected over the study period (see Table D.1).

Variable		Lake				
		Big Island	Mustard	Willow	Fish	Gargan
Unfiltered Water Total Hg (ng/L)		0.58 (0.08)	0.70 (0.15)	0.55 (0.15)	0.82 (0.47)	0.80 (0.25)
Unfiltered Water MeHg (ng/L ×100)		1.97 (0.63)	1.14 (0.45)	1.11 (0.74)	2.87 (0.61)	5.53 (1.64)
Filtered Water Total Hg (ng/L)		0.47 (0.16)	0.53 (0.10)	0.51 (0.12)	0.67 (0.39)	0.74 (0.18)
Filtered Water MeHg (ng/L ×100)		1.16 (0.38)	0.76 (0.46)	0.99 (0.62)	2.96 (0.51)	4.44 (1.27)
Sediment Total Hg (ng/g)		79.64 (47.41)	99.65 (31.99)	41.88 (32.37)	108.98 (46.41)	119.70 (19.00)
Sediment MeHg (ng/g)		0.46 (0.22)	0.31 (0.14)	0.36 (0.13)	0.33 (0.18)	0.35 (0.42)
Sediment Loss of Ignition (LOI %)		20.35 (9.14)	18.84 (5.22)	26.73 (31.61)	24.32 (2.52)	36.08 (10.63)
Chlorophyll-a (μg/L)		5.15 (1.36)	7.23 (3.39)	5.26 (0.84)	3.44 (1.22)	8.58 (4.12)
Secchi Depth (m):		4.00 (NA)	2.75 (0.35)	2.80 (NA)	3.00 (NA)	2.38 (0.53)
Dissolved Organic Carbon (mg/L)		9.76 (0.54)	11.50 (0.85)	9.97 (1.18)	13.14 (0.93)	21.30 (6.95)
Dissolved Oxygen (mg/L)		10.60 (NA)	9.30 (NA)	9.60 (NA)	9.60 (NA)	9.00 (NA)
Sulphate (mg SO_4/L)		3.56 (0.07)	8.83 (1.51)	6.34 (0.37)	14.48 (2.62)	14.08 (2.29)
pH		7.17 (0.00)	7.28 (0.79)	7.58 (0.00)	7.65 (0.00)	7.95 (0.34)
Lake Area (LA km^2)		18.22	23.31	128.58	117.07	1.1
Catchment Area (CA km^2)		112.96	198.68	1248.81	2616.86	73.16
LA to CA (ratio ×100)		16.13	11.73	10.29	4.47	1.5
Mean Catchment Elevation (m asl)		783.22	750.83	704.82	428.25	283.45
Mean Catchment Slope (degree)		2.49	2.22	1.79	2.57	2.33
Land cover classes (% of CA)	Barren Land	0	0.01	0	3.34	3.77
	Mixed Forest	0	0.06	0	0.35	2.15
	Subpolar/Polar Grass Land	11.49	10.32	23.91	5.16	0.04
	Subpolar/Polar Shrub Land	0.86	0.95	0.93	1.15	0
	Subpolar Taiga Needleleaf Forest	25.4	22.33	26.69	9.06	0.01
	Temperate/Subpolar Broadleaf Deciduous Forest	0	0	0	0.26	4.48
	Temperate/Subpolar Needleleaf Forest	24.5	34.85	16.03	51.62	40.13
	Temperate/Subpolar Shrub Land	2.58	3.43	4.93	8.35	22.46
	Wetland	11.82	12.41	8.6	9.92	22.24
	Water	23.34	15.64	18.91	10.79	4.72

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Table D.3; continued from previous page

Variable		Lake				
		Ekali	Sanguez	Kakisa	Tathlina	McGill
Unfiltered Water Total Hg (ng/L)		0.70 (0.15)	0.74 (0.27)	1.64 (1.44)	1.22 (0.46)	0.90 (0.44)
Unfiltered Water MeHg (ng/L $\times 100$)		7.12 (7.44)	4.74 (1.90)	2.56 (2.54)	8.90 (1.51)	7.40 (3.45)
Filtered Water Total Hg (ng/L)		0.59 (0.21)	0.58 (0.21)	0.82 (0.34)	1.32 (0.57)	0.84 (0.40)
Filtered Water MeHg (ng/L $\times 100$)		6.39 (6.91)	4.05 (1.52)	1.85 (1.97)	7.94 (0.90)	6.79 (3.05)
Sediment Total Hg (ng/g)		155.68 (22.60)	141.52 (22.04)	74.87 (40.24)	112.58 (23.02)	85.27 (14.15)
Sediment MeHg (ng/g)		0.09 (0.07)	0.77 (0.54)	0.22 (0.12)	0.33 (0.21)	0.33 (0.26)
Sediment Loss of Ignition (LOI %)		57.07 (7.70)	30.34 (0.05)	9.42 (7.32)	8.51 (4.23)	13.12 (0.11)
Chlorophyll-a (μg/L)		7.12 (1.39)	5.62 (0.50)	15.53 (14.78)	3.51 (2.73)	2.24 (0.27)
Secchi Depth (m):		2.90 (0.42)	2.58 (0.87)	0.75 (0.49)	1.75 (1.10)	2.45 (0.07)
Dissolved Organic Carbon (mg/L)		16.58 (2.51)	17.31 (4.41)	12.93 (2.68)	19.66 (1.22)	17.46 (2.69)
Dissolved Oxygen (mg/L)		9.40 (NA)	9.71 (NA)	11.00 (NA)	11.40 (NA)	9.70 (NA)
Sulphate (mg SO_4/L)		11.27 (1.70)	10.18 (5.25)	20.06 (3.05)	13.18 (1.63)	14.21 (2.39)
pH		8.17 (0.23)	8.25 (0.11)	7.74 (0.38)	7.86 (0.00)	7.90 (0.05)
Lake Area (LA km^2)		1.88	1.59	335.76	565.29	2.32
Catchment Area (CA km^2)		179.8	106.86	15808.82	11606.31	982.6
LA to CA (ratio $\times 100$)		1.05	1.49	2.12	4.87	0.24
Mean Catchment Elevation (m asl)		274.04	277.99	408.64	456.68	354.56
Mean Catchment Slope (degree)		4.36	3.19	4.17	4.27	5.22
Land cover classes (% of CA)	Barren Land	1.54	2.58	1.96	1.39	0.31
	Mixed Forest	4.33	3.82	3.78	4.48	4.23
	Subpolar/Polar Grass Land	0.03	0.05	0.94	1.17	0.01
	Subpolar/Polar Shrub Land	0	0	0	0	0
	Subpolar Taiga Needleleaf Forest	0.06	0.07	1.26	0.92	0.46
	Temperate/Subpolar Broadleaf Deciduous Forest	7.86	6.37	2.23	2.6	3.39
	Temperate/Subpolar Needleleaf Forest	54.89	48.84	54.25	57.89	74.87
	Temperate/Subpolar Shrub Land	11.92	17.01	17.49	15.72	8.32
	Wetland	14.35	16.55	9.59	7.8	6.64
	Water	5.02	4.72	8.52	8.03	1.76

Table D.4. Lake-specific descriptive statistics [mean (min-max), if $n > 1$] for fork length, δ^{15}N, and concentrations of total Hg and MeHg in fish species (total $n = 332$).

Lake	Species	n	Fork length (mm)	δ^{15}N (‰)	Hg (ng/g dw)	MeHg (ng/g dw)
Big Island	BB	2	455.0 (350-560)	11.9 (11.7-12.0)	412.1 (334.8-489.4)	268.2 (197.6-338.8)
	CC	3	121.3 (111-130)	10.7 (10.1-11.7)	272.5 (230.1-321)	184.3 (139.6-237.5)
	LS	2	405.5 (372-439)	8.4 (8.3-8.5)	255.8 (246.9-264.7)	232.4 (224.2-240.5)
	LW	5	360.6 (210-498)	10.3 (8.9-11.5)	304.9 (91.2-868.9)	178.3 (59.6-453.7)
	NP	3	638.3 (530-765)	12.0 (11.5-12.3)	1594.4 (1460.8-1688.2)	1228.8 (940.8-1464.1)
	NS	3	39.0 (33-49)	9.3 (8.5-10.1)	154.1 (114.1-201.7)	108.9 (72.3-133.7)
Mustard	BB	3	497.3 (475-522)	11.8 (11.5-12.1)	551.7 (449.9-671.5)	415.7 (317.6-518.7)
	LS	1	553.0	8.6	733.5	733.5
	NP	3	444.0 (108-654)	9.4 (7.1-11.1)	686.8 (231.3-1302.1)	584.5 (147.9-1133.5)
	NS	1	34.0	6.8	100.0	100.0
Willow	CC	3	98.0 (69-120)	9.7 (9.5-9.9)	188.3 (172.1-212.6)	146.1 (108.1-208.8)
	LS	2	60.5 (60-61)	7.1 (7-7.2)	35 (31.2-38.8)	56.7 (56.4-56.9)
	LW	4	388.3 (248-506)	9.7 (8.9-11.3)	453.6 (111.6-1205.9)	386.2 (62.7-1101.9)
	NP	5	566.8 (165-888)	11.4 (9.7-13.0)	1925.8 (152.3-4467.8)	1715.1 (119.6-3759.2)
	NS	5	39.8 (30-49)	9.9 (9.4-10.4)	137.6 (92.4-166.4)	122.8 (89.4-151.0)
Fish	CC	3	127.7 (105-153)	9.2 (8.6-10.0)	274 (137.9-354.5)	229.8 (140.1-281.8)
	LS	1	468.0	8.3	574.2	574.2
	LW	6	295.7 (123-515)	9.7 (9-10.7)	246.8 (135.8-516.8)	217.4 (101.9-513.5)
	NP	6	574.3 (74-972)	10.2 (7.0-12.0)	2180.4 (166.3-5847.9)	1888.1 (146.7-5124.5)
	WE	11	334.5 (154-576)	11.3 (9.8-12.8)	1587.2 (275.1-4513.8)	1319.2 (124.6-4066.5)
	WS	3	526 (499-543)	8.6 (8.2-8.8)	1118.2 (773.3-1386.7)	844.2 (638.2-1110.1)
Gargan	BB	1	115.0	8.9	305.0	305.0
	BS	1	42.0	8.8	206.0	206.0
	CC	1	361.0	11.1	389.0	389.0
	LW	4	446.3 (300-580)	9.2 (8.8-9.5)	775.0 (267.0-1260.0)	533.3 (202.4-1024.6)
	NP	6	495.2 (353-841)	11.1 (10.4-12.6)	1966.2 (786.0-6180.0)	1649.7 (541.9-5418.2)
	NS	1	30.0	8.6	172.0	172.0
Ekali	BS	3	38.7 (37-40)	8.0 (7.0-8.5)	182.7 (171.6-195.1)	135.0 (112.1-158.7)
	CC	3	265.3 (155-337)	8.8 (8.1-9.3)	726.5 (359.9-969.7)	481.7 (208.7-733.6)
	LW	9	408.0 (185-585)	9.1 (6.5-10)	465.5 (271.0-691.5)	396 (237.4-565.5)
	NP	10	510.1 (222-879)	10.7 (8.3-12.6)	2717.7 (372.4-9848.4)	2318.6 (236.5-8801.2)
	WE	7	387.9 (222-535)	11.2 (9.8-12.8)	1713.2 (829.8-2899.7)	1297.6 (654.0-1667.2)
Sanguez	BS	3	37.3 (33-43)	8.7 (7.6-9.8)	408.3 (343-485.6)	278.2 (244.5-335)
	CC	6	208.7 (121-272)	9.6 (9.0-9.8)	910.1 (539.0-1609.2)	761.5 (409.5-1490.0)

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Table D.4; continued from previous page

Lake	Species	n	Fork length (mm)	δ^{15}N (‰)	Hg (ng/g dw)	MeHg (ng/g dw)
	LW	18	491.1 (357-593)	9.6 (8.2-10.9)	789.6 (418.2-1701.8)	615.1 (306.7-1217.0)
	NP	33	642.1 (230-1004)	11.7 (9.8-13.0)	4845.8 (757.4-15605.8)	3904.4 (538.4-10440.4)
	NS	3	41.7 (40-43)	10 (9.8-10.1)	370.9 (330.5-426.5)	249.8 (205.9-271.9)
	WE	21	460.6 (102-632)	12.2 (10.7-13.3)	2975.9 (512.6-6066.1)	2492.0 (291.8-5304.8)
Kakisa	CC	3	144.3 (135-152)	8.9 (8.7-9.2)	208.2 (187.5-221.6)	155.0 (106.4-217.6)
	LS	2	459.5 (448-471)	9.4 (8.8-10.1)	534.6 (287.9-781.2)	485.7 (239.3-732.0)
	LW	12	367.3 (150-462)	9.3 (7.9-10.6)	334.9 (143.7-808)	275.1 (100.4-713.8)
	NP	15	560.0 (205-885)	11.5 (9.3-13.4)	1999.5 (177.6-4531.7)	1676.5 (108.0-3515.3)
	WE	13	399.2 (185-549)	11.6 (10.6-12.9)	1625.8 (478.4-5638.5)	1301.1 (411.7-5160.2)
	WS	3	431.7 (355-50)	9.9 (8.5-10.8)	303.2 (107.7-482.8)	238.8 (74.7-369.5)
Tathlina	LS	2	474.5 (423-526)	9.0 (9.0-9.1)	1132.6 (963.0-1302.2)	804.9 (635.6-974.1)
	LW	11	373.3 (255-493)	8.5 (7.7-9.3)	424.4 (233-813.2)	315.9 (182.9-640.9)
	NP	15	654.3 (252-1042)	10.6 (8.0-11.9)	1975.7 (376.9-5014.9)	1567.2 (261.8-4043.4)
	WE	10	419.9 (276-536)	11.2 (9.6-12.2)	2790.7 (871.3-4950.0)	2182.8 (898.9-3595.4)
	WS	3	475.7 (440-512)	8.3 (7.8-9.1)	989.3 (548.0-1220.0)	732.0 (364.6-958.0)
McGill	BB	1	75	6.6	134.8	134.8
	BS	2	37.5 (34-41)	8.5 (8.1-8.9)	365.0 (331.0-399.0)	273.0 (222.0-324.0)
	CC	1	75.0	7.1	508.0	508.0
	LS	3	517.0 (482-540)	8.8 (7.7-9.5)	1406.3 (599-1870)	918.9 (306.8-1448.8)
	LW	8	401.4 (100-561)	9 (7.4-10.2)	738.8 (388.5-1514.7)	639.8 (351.1-1372.7)
	NP	9	511.9 (256-937)	10.6 (8.9-11.7)	2975.9 (723.0-7150.0)	2593.3 (458.9-6054.9)
	NS	2	40.5 (37-44)	8.8 (8.4-9.2)	306.5 (299.0-314.0)	210.5 (201.0-220.0)
	WE	9	387.9 (174-560)	10.7 (10.2-11.3)	3229.8 (1218.8-5450.0)	2648.1 (1120.0-4109.3)
	WS	3	476.3 (449-498)	9.1 (8.9-9.4)	808.3 (670.0-928.0)	484.2 (460.5-510.3)

Abbreviations: BB (Burbot, *Lota lota*), BS (Brook Stickleback, *Culaea inconstans*), CC (Cisco, *Coregonus artedii*), LS (Longnose Sucker, *Catostomus catostomus*), LW (Lake Whitefish, *Coregonus clupeaformis*), NP (Northern Pike, *Esox lucius*), NS (Nine-spine Stickleback, *Pungitius pungitius*), WE (Walleye, *Sander vitreus*), and WS White Sucker, *Catostomus commersoni*).

Table D.5. Lake-specific descriptive statistics [mean (min-max), if n > 1] for δ^{15}N and concentrations of MeHg in primary consumers (total n = 370).

Lake	Taxa	n	δ^{15}N (‰)	MeHg (ng/g dw)	Taxa	n	δ^{15}N (‰)	MeHg (ng/g ww)
Big Island	ba	1	1.3	22.1	he	2	1.5 (1.5-1.5)	18.3 (13.3-23.4)
	ch	2	3.3 (2.4-4.2)	17 (8.1-25.9)	hy	1	2.3	18.3
	cl	1	6.2	25.2	ly	2	1.6 (1.5-1.8)	17.9 (17.9-17.9)
	co	2	4.3 (4.1-4.5)	48.3 (35.9-60.6)	ol	1	2.6	12.3
	er	1	5.4	24.3	pi	1	9.3	40.3
	fy	1	3.5	29.7	sf	2	3.2 (3-3.4.0)	31.4 (23.0-39.9)
	ga	1	3.2	23.9	zo	3	3.5 (2.9-4.0)	5.1 (4.4-6.1)
Mustard	ap	1	1.6	2.5	ga	1	2.7	14.4
	ba	1	2.3	7.8	hy	1	2.7	16.6
	ch	2	3.6 (3.2-3.9)	20.7 (19.9-21.4)	ly	1	1.8	14.3
	co	1	3.6	72.5	ol	1	2.8	11.2
	dy	1	2.8	42.4	sf	1	2.9	27.8
	er	1	5.8	25.5	tp	1	3.9	18.2
	fr	1	3.3	51.7	vv	1	2	15.2
Willow	ca	1	1.6	9.4	ly	2	1.9 (0.9-3)	12.2 (8.7-15.6)
	ch	2	4.1 (3.9-4.4)	19.1 (13.9-24.2)	os	1	2.2	12.8
	co	1	3.9	61	pe	1	1	8.5
	dy	1	3.6	43.3	sf	1	3.3	38.7
	ga	1	2.9	18.1	tr	1	3.7	37.4
	hi	1	6.3	15.4	vv	1	3	19.5
	hy	1	1.6	16	zo	6	4.4 (2.3-5.1)	4.5 (2.2-7.8)
Fish	ac	1	6.3	248.6	ga	4	2.2 (1.6-3.2)	14.7 (10.9-17.7)
	ae	3	4.6 (4.4-5.0)	59.2 (58.0-61.3)	he	1	2.3	10
	ca	1	2.3	9.7	hm	2	5.9 (5.8-6.0)	33.0 (32-34.1)
	cd	3	3.8 (3.1-4.3)	64.1 (55.5-70.3)	hr	1	5.2	32.6
	ch	2	4.7 (4.2-5.2)	11.5 (5-18.1)	hy	3	2.5 (2.1-3.2)	23.8 (22.1-26.5)
	cn	1	5.8	81.5	ly	10	1.6 (0.7-2.4)	26.9 (20.6-40.6)
	co	6	5.1 (4.7-5.6)	81.8 (58.8-91.8)	no	2	5.8 (5.7-5.9)	116.3 (115.4-117.2)
	cr	2	2.3 (1.9-2.7)	17.2 (15.7-18.7)	pl	3	2.9 (2.1-3.8)	24.3 (22.1-27.3)
	dy	1	7.2	25.5	sf	1	3.7	43.4
	ef	2	5.5 (5-6)	18.4 (18.2-18.7)	vv	2	1.7 (1.6-1.8)	12.5 (6.6-18.5)
	fr	2	4.3 (4.3-4.3)	44.5 (36.8-52.3)				
Gargan	ae	1	5.9	113	ln	1	4.1	109.5
	am	1	1.1	49	ly	1	3.7	51

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Table D.5; continued from previous page

Lake	Taxa	n	δ^{15}N (‰)	MeHg (ng/g dw)	Taxa	n	δ^{15}N (‰)	MeHg (ng/g ww)
	cn	1	5	133	no	1	5.5	176
	co	1	5	44	pl	1	3.2	28
	hi	1	5.6	65.1	uo	2	5.1 (4.7-5.4)	117.0 (93.0-141.0)
Ekali	ae	8	4.2 (3.5-5)	102.8 (86.6-112.7)	hy	1	3.1	41.1
	ba	1	4	20.8	li	1	4	42.8
	ch	3	3.9 (3.5-4.4)	10.5 (4.7-15.7)	lm	1	1.3	12.7
	cl	1	4.4	30.4	ly	9	3.3 (2-5.5)	80.5 (24.4-132.9)
	cn	2	5 (4.2-5.9)	78 (77.9-78.2)	no	1	5	109.6
	co	2	5.8 (5.5-6.1)	120.9 (87-154.8)	os	1	5	5.6
	cp	2	8.6 (8.5-8.7)	5.6 (5.1-6.1)	pl	1	3.3	19.5
	fy	1	2.7	29.8	sf	3	4.2 (3.3-5.1)	51.5 (45.7-59.9)
	ga	1	3.2	40.8	si	2	4.9 (4.8-4.9)	31.3 (15.9-46.7)
	gl	1	4.1	10.5	uo	7	4.7 (4.2-5.3)	70.7 (59.1-79.7)
	gy	1	4.9	27.4	zo	5	7.3 (7.1-7.5)	23.4 (20.4-29.4)
	ha	1	2.7	114.9				
Sanguez	ae	8	4.5 (3.2-6.2)	173 (97.4-264.4)	gl	1	4.3	32.7
	ba	1	2.3	19.8	hy	1	1	46.5
	ch	4	5.2 (3.3-6.3)	7.3 (3.6-13.6)	li	2	2.4 (2.2-2.6)	118.9 (97.4-140.4)
	cn	2	4.6 (4.4-4.8)	152.2 (148.6-155.8)	ly	2	1.7 (1.7-1.7)	70.7 (69.3-72.1)
	co	2	3.3 (2.7-4.0)	172.1 (146.5-197.7)	ro	1	7	32.1
	cp	2	7.9 (7.7-8.1)	42.1 (36.4-47.9)	sf	1	4.5	132.5
	dy	1	4.4	154.4	tp	1	4.4	65.7
	fr	1	5.4	103.5	uo	10	4.2 (3.7-4.6)	150.5 (104.5-216.3)
	fy	1	2.1	47.4	zo	3	6.4 (6.0-6.7)	6.7 (6.0-8.0)
	ga	1	1.8	78.7	gy	1	5.1	61
Kakisa	ae	1	6.2	77.3	he	2	2.1 (2-2.2)	11.8 (10.2-13.3)
	ba	3	4.5 (3.4-5.7)	15.7 (9.8-19.5)	hr	1	5.6	20.1
	ca	1	2.4	34.4	hy	3	3.8 (3-4.8)	29.7 (28-30.8)
	ch	2	5.0 (4.9-5)	6.0 (2.9-9.0)	lc	2	4.9 (4.5-5.3)	32.5 (18.6-46.3)
	cn	1	5.3	32.1	ld	1	1.2	14.1
	co	5	5.3 (4.8-5.5)	61.9 (52.9-75.6)	le	1	2.9	16.6
	dy	2	3.5 (2.6-4.5)	53.4 (49.2-57.5)	lp	1	1.8	12
	ef	4	5.7 (3.7-7.5)	24.3 (7.2-46.3)	ly	3	2.3 (1.4-3)	19.5 (15.2-22.8)
	er	1	7.5	21.7	pi	1	9.9	6.8
	fr	2	4.6 (4.3-4.9)	28.1 (24.3-31.9)	pl	1	2.8	19.1
	ga	3	5.1 (4.5-5.8)	31 (25.5-36.8)	po	2	6.6 (6.2-7.1)	27.5 (14.9-40.1)

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Table D.5; continued from previous page

Lake	Taxa	n	δ^{15}N (‰)	MeHg (ng/g dw)	Taxa	n	δ^{15}N (‰)	MeHg (ng/g ww)
	gl	1	7.3	22.4	ga	1	2.9	39.4
Tathlina	ae	2	4.8 (3.4-6.2)	113.2 (84-142.3)	gl	1	5.8	8.6
	ca	1	2.4	21.6	he	1	2.7	25.1
	ch	3	3.1 (1.8-3.8)	15.3 (10.0-23.4)	hy	2	2.2 (1.8-2.5)	41.4 (30.9-52)
	cn	2	4.9 (4.4-5.3)	128.9 (128.8-129)	ly	1	2.6	24.4
	co	2	4.4 (4.2-4.6)	174 (127.9-220)	mo	1	5.7	31.1
	cp	1	8.4	15.7	os	2	4.3 (4.2-4.3)	54.7 (37.8-71.6)
	dy	11	5.1 (4-5.9)	155.4 (81.1-196.2)	pl	1	3.7	21
	ef	3	3.4 (2.4-4.6)	20.3 (12.7-30.8)	sf	1	3.7	62.8
	er	2	6.8 (6.5-7.1)	17.3 (9.3-25.4)	uo	1	3.9	120.9
	fr	1	3.1	24	vv	1	4.3	24.5
	fy	1	2.5	65.7				
McGill	ae	1	4	77	ga	1	3.6	36.9
	am	1	3.1	36	gl	1	4.9	39.8
	cn	2	5.8 (5.5-6.1)	95.9 (82.8-109)	ly	24	2.6 (1-4.5)	18.2 (2.5-56.3)
	co	3	4.3 (4.1-4.4)	142.4 (134.3-154.1)	no	2	4.6 (3.8-5.3)	97.7 (74-121.3)
	dy	11	4.6 (3.2-6.3)	197.8 (78-337.5)	sf	1	2.6	55
	ef	2	3.0 (2.1-4)	17.4 (13.7-21.2)	uo	11	5.3 (4.9-5.5)	119.9 (101.3-135.6)
	fy	1	3.6	36.4				

Abbreviations: ac (Acari), ae (Aeshnidae), am (Amphipoda), ap (Apananiidae), ba (Baetidae), ca (Caenidae), cd (Corduliidae), ch (Chironomidae), cl (Cladocera), cn (Coenagrionidae), co (Corixidae), cp (Copepoda), cr (Crangonyctidae), dy (Dytiscidae), ef (Ephemeridae), er (Erpobdellidae), fr (Phryganeidae), fy (Physidae), ga (Gammaridae), gl (Glossiphoniidae), gy (Gyrinidae), ha (Haliplidae), he (Heptageniidae), hi (Hirudinea), hm (Haemopidae), hr (Hydrophilidae), hy (Hyalellidae), lc (Leptoceridae), ld (Lepidostomatidae), le (Leptophlebiidae), li (Libellulidae), lm (Limnephilidae), ln (Lynceidae), lp (Leptohyphidae), ly (Lymnaeidae), mo (Molannidae), no (Notonectidae), ol (Oligochaeta), os (Ostracoda), pe (Perlidae), pi (Piscicolidae), pl (Planorbidae), po (Polycentropodidae), ro (Rotifer), sf (Sphaeriidae), si (Sialidae), tp (Tipulidae), tr (Trichoptera), uo (Unionidae), vv (Valvatidae), zo (Bulk zooplankton).

Table D.6. Size-standardized total Hg concentrations in Lake Whitefish and Northern Pike. Lake-specific sample size (n), mean ± standard error for fork length, total Hg concentrations (ng/g dry weight), and size-standardized total Hg concentrations are presented in panel **(a)**. Size-standardized total Hg concentrations are least squares mean (LSM) generated using ANCOVAs; results of ANCOVAs are presented in panel **(b)**. Samples included in this table include those that were analyzed for both total Hg and methyl Hg (see Table D.4), as well as samples (number in parentheses) that were analyzed for total Hg only (not methyl Hg). Letters depict among-lake pairwise differences in LSM total Hg for each species.

(a)

	Lake	n total (additional samples)	Fork length (mm)	THg (ng/g dw)	LSM and pairwise differences	
Lake Whitefish (LSM at 400 mm fork length)	Big Island	40 (35)	347.9 ± 13.4	302.43 ± 247.4	322.98 ± 24.1	ab
	Willow	42 (38)	407.62 ± 11.1	423.42 ± 390.9	279.88 ± 16.7	a
	Fish	62 (58)	332.52 ± 13.2	294.07 ± 145.6	325.52 ± 19.2	ab
	Gargan	23 (19)	449.04 ± 14.3	637.39 ± 289.5	477.79 ± 28.2	c
	Ekali	41 (32)	436.39 ± 14.5	464.58 ± 171.4	442.59 ± 42.7	bc
	Sanguez	106 (88)	498.8 ± 7.2	883.8 ± 296.9	422.96 ± 26.4	bc
	Kakisa	82 (70)	391.76 ± 5.9	321.72 ± 191.4	752.13 ± 34.2	d
	Tathlina	75 (64)	361.56 ± 5.2	428.48 ± 137.5	289.81 ± 12.6	a
	McGill	34 (26)	335.44 ± 32.4	667.37 ± 320.4	667.20 ± 50.6	d
Northern Pike (LSM at 600 mm fork length)	Big Island	8(5)	609.63 ± 28.9	1617.91 ± 384.5	1572.49 ± 177.7	bcd
	Mustard	6 (3)	490.33 ± 79.6	690.28 ± 389.8	763.369 ± 113.0	a
	Willow	23 (18)	589.09 ± 24.1	1606.43 ± 1006.1	1462.80 ± 98.6	b
	Fish	31 (25)	615.42 ± 32.8	2552.45 ± 1832.6	2119.82 ± 122.2	de
	Gargan	19 (13)	458.42 ± 48.1	1747.95 ± 1543.0	1628.55 ± 72.3	bc
	Ekali	51 (41)	490.82 ± 20.8	1993.87 ± 1618.4	2126.67 ± 190.9	cde
	Sanguez	256 (223)	616.48 ± 11.0	3945.71 ± 2370.7	2621.49 ± 151.0	ef
	Kakisa	58 (43)	525.29 ± 18.4	1398.53 ± 1199.8	3395.72 ± 67.7	g
	Tathlina	52 (38)	665.4 ± 31.2	2174.77 ± 1444.6	1525.96 ± 73.8	b
	McGill	28 (19)	471.11 ± 29.1	2424.12 ± 1575.4	3390.63 ± 282.1	fg

(b)

	Variable	Sum Sq.	df	F	*p*
Lake Whitefish	Intercept	0.563	1	19.868	< 0.001
	Log$_{10}$ fork length	1.874	1	66.192	< 0.001
	Lake	5.93	8	26.182	< 0.001
	Log$_{10}$ fork length × lake	5.209	8	23	< 0.001
	Residuals	13.789	487	-	-
Northern Pike	Intercept	0.004	1	0.188	0.665
	Log$_{10}$ fork length	0.012	1	0.638	0.425
	Lake	1.472	9	8.53	< 0.001
	Log$_{10}$ fork length × lake	1.516	9	8.784	< 0.001
	Residuals	9.818	512	-	-

Table D.7. Results of ANCOVA analyses depicting among-lake variability in slopes of mercury-δ^{15}N regressions where baseline-adjusted δ_{15}N were fit against total Hg concentrations in fish and MeHg concentrations in benthic invertebrates **(a)** and MeHg concentrations in both fish and primary consumers **(b)**. Pairwise differences in slopes are shown in Table 4.1 in the main document.

Variable	**(a)**				**(b)**			
	Sum sq.	df	F	*p*	Sum sq.	df	F	*p*
Intercept	2.18	1	13.84	< 0.001	2.856	1	18.194	< 0.001
δ^{15}N	15.136	1	96.096	< 0.001	11.813	1	75.244	< 0.001
Lake	6.012	9	4.241	< 0.001	5.925	9	4.193	< 0.001
δ^{15}N × Lake	4.383	9	3.092	< 0.001	4.601	9	3.256	< 0.001
Residuals	107.418	682	-	-	107.074	682	-	-

(a)

Species	n fish	n lake	Minimum	Maximum	Median	Mean	Standard Deviation
Brook Stickleback	9	4	52.515	97.67	67.069	71.927	14.1
Ninespine Stickleback	15	6	48.271	112.277	70.064	76.175	21.023
Cisco	23	8	51.851	101.616	75.449	76.298	14.751
Burbot	7	4	54.411	77.24	70.588	68.655	8.785
Longnose Sucker	10	5	51.227	93.707	76.141	74.478	15.357
White Sucker	12	4	51.906	85.285	71.056	71.629	9.593
Lake Whitefish	77	9	49.152	101.829	80.753	79.052	11.876
Northern Pike	105	10	49.556	120.702	84.684	81.357	13.112
Walleye	71	6	45.293	115.239	81.542	81.111	12.681

(b)

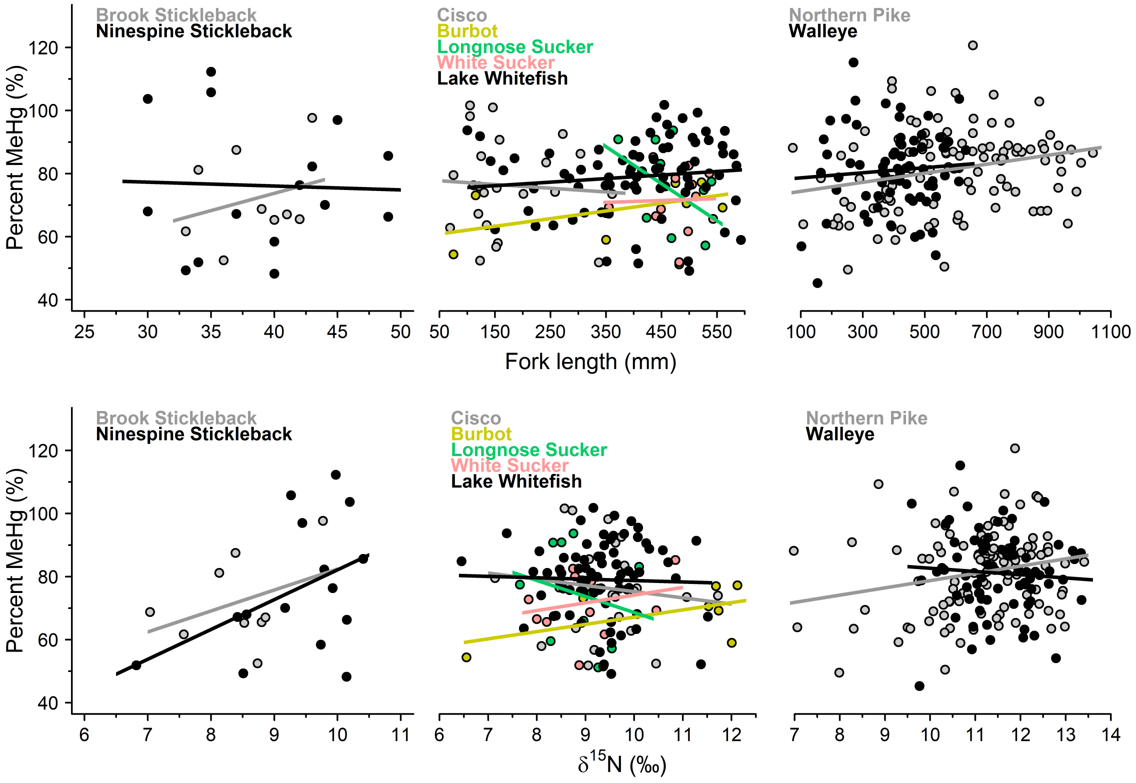

Figure D.1. Percent MeHg concentrations of total mercury concentrations (%MeHg) in fish species, and its relationships with fish size and fish trophic level, in the study area. Descriptive statistics of species-specific %MeHg calculated using data combined in all study lakes are presented in panel **(a)** and relationships between %MeHg and fish size and fish trophic level are presented in panel **(b)**.

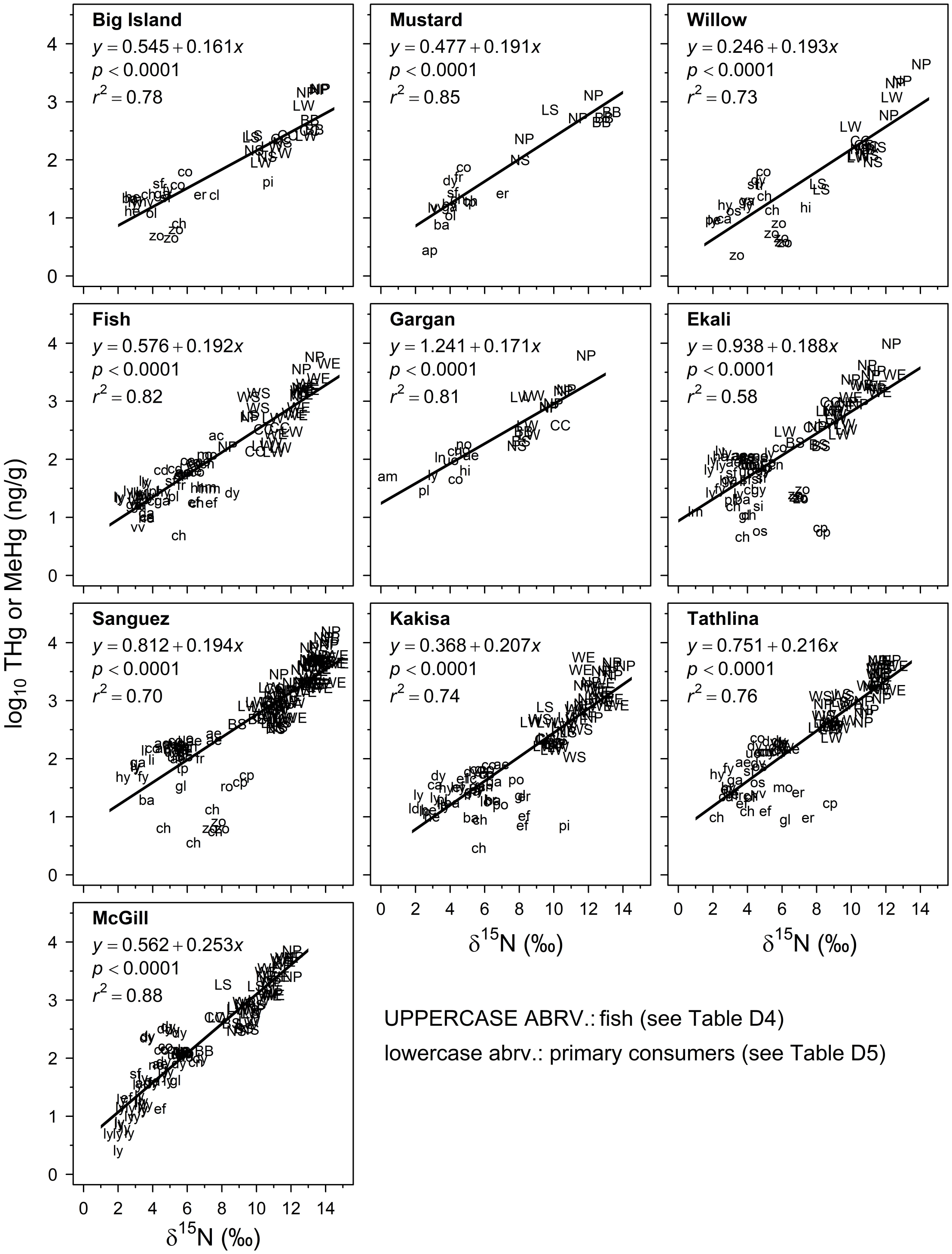

Figure D.2. Lake-specific mercury-δ^{15}N (i.e., Hg biomagnification) regressions generated using concentrations of total Hg in fish and concentrations of MeHg in primary consumers (dependent variable) versus δ^{15}N ratios adjusted for baseline δ^{15}N variations (independent variable). Solid lines represent fitted regressions.

	(a)	(b)										
	% Forest	Primary consumer MeHg	DOI-adjusted sediment THg	DOI-adjusted sediment MeHg	Filtered water THg	Filtered water MeHg	Chlorophyll-a	DO	Secchi	DOC	SO_4	Stratification
Primary consumer MeHg	0.5											
DOI-adjusted sediment THg	0.62	0.36										
DOI-adjusted sediment MeHg	0.04	−0.01	0.60									
Filtered water THg	0.52	0.54	0.77	0.37								
Filtered water MeHg	0.62	0.9	0.37	−0.14	0.67							
Chlorophyll-a	−0.41	−0.30	−0.13	−0.26	−0.22	−0.37						
DO	0.21	−0.12	0.66	0.56	0.53	0.07	0.01					
Secchi	−0.23	−0.05	−0.54	−0.28	−0.59	−0.12	−0.52	−0.52				
DOC	0.44	0.94	0.38	−0.08	0.66	0.89	−0.14	−0.09	−0.21			
SO_4	0.53	0.55	0.48	−0.02	0.68	0.56	0.14	0.02	−0.69	0.64		
Stratification	−0.51	−0.43	−0.53	0.14	−0.64	−0.46	−0.23	0.02	0.56	−0.64	−0.79	
pH	0.39	0.81	0.06	−0.30	0.38	0.81	−0.02	−0.17	−0.23	0.80	0.59	−0.38

Pearson $r > +0.40$

Pearson $r < -0.40$

Figure D.3. Pearson correlation coefficients among studied variables. Panel **(a)** shows Pearson *r* vales for correlations between percent forest cover in catchments and in-lake variables, including concentrations of MeHg in primary consumers. Panel **(b)** shows Pearson *r* values for correlations between in-lake variables, including concentrations of MeHg in primary consumers. Except LA:CA and stratification depth, all other variables are $\log_{10}$ transformed.