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# ABIOTIC CONDITIONS, ALGAL BIOMASS & FISH GROWTH RATES AFFECT FISH MERCURY CONCENTRATIONS IN TWO SUBARCTIC LAKES

by

Ari Yamaguchi

BS, Environmental Science, Drexel University, 2017

# THESIS

Submitted to the Department of Biology Faculty of Science in partial fulfillment of the requirements for the Master of Science in Integrative Biology Wilfrid Laurier University

2020

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#### ABSTRACT

Kakisa Lake and Tathlina Lake, located in the Dehcho Region of the Northwest Territories, support important fisheries for the local Ka'a'gee Tu First Nation (KTFN). Recently, Walleye (Sander vitreus) of typical catch size in Tathlina Lake were found to have mercury concentrations above Health Canada's commercial sale guideline of 0.5 ppm. Wild foods with elevated mercury concentrations can pose health risks to the humans who consume them, depending on consumption amounts and vulnerability factors such as age and pregnancy. Because wild fish can accumulate relatively high mercury levels and subsistence fishing contributes greatly to food security in northern regions, mercury-related health risks to people are greater in the north than in the south, where wild fish are not as frequently consumed. Here, I examine and compare known drivers of fish mercury concentrations in two aquatic food webs to investigate causes of between-lake variation in mercury concentrations in food fishes. I relate analyses of food web structure, fish growth, and lake physicochemistry to mercury concentrations, and attempt to determine why fish mercury concentrations differ between Kakisa Lake and Tathlina Lake. Sediment and water methylmercury availability and primary producer abundance appear to be major factors influencing bioaccumulation of mercury in the food webs of each lake. Concentrations of methylmercury in sediment and water were higher in Tathlina Lake than in Kakisa Lake, and % methylmercury (of total mercury) in these ecosystem components indicate that the net mercury methylation rate is higher in Tathlina Lake than in Kakisa Lake. Kakisa Lake also had higher concentrations of chlorophyll a, indicating relatively higher rates of primary production and possible bloom dilution of mercury, which was further confirmed by trophic biomagnification modeling; these factors appear to have bottom-up impacts on the food webs of both lakes, including other food fishes. Walleye mercury concentrations also

appeared to be affected by growth rates and perhaps growth efficiency, as suggested by evaluations of growth rates. This research is part of a larger project that seeks to assess the risks and benefits of fish consumption in the Northwest Territories, especially by Indigenous communities, who rely on natural fisheries for subsistence and for whom wild foods hold significant cultural and spiritual value.

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# LIST OF ABBREVIATIONS

AMDE	Atmospheric mercury depletion event
ANCOVA	Analysis of covariance
ANOVA	Analysis of variance
Biotron	Biotron Experimental Climate Change Research Centre (Western
	University)
EIL	Environmental Isotope Laboratory (University of Waterloo)
Hg	Mercury
$\mathrm{Hg}^{0}$	Elemental mercury
Hg(I)	Organic (monovalent) mercury
Hg(II)	Inorganic (divalent) mercury
MeHg	Methylmercury
SSHg	Size-standardized mercury concentration
THg	Total mercury
kg	kilograms
k <sub>meth</sub>	Methylation rate
KTFN	Ka'a'gee Tu First Nation
Mg	Megagrams
mg	milligrams
mL	milliliters
mm	millimeters
ng	nanograms
ppm	Parts per million $(= mg/kg)$
SIA	Stable isotope analysis
SRB	Sulfate-reducing bacteria
UW	University of Waterloo
[]	Concentration of

#### **CHAPTER 1: GENERAL INTRODUCTION**

#### 1.1 Study Area

Tathlina Lake (60.555086°N, 117.531469°W; Figure 1.1) is a wide and shallow lake with a surface area of 573 km<sup>2</sup> and an average depth of 1.5 m (maximum 2.7 m) (Kennedy, 1962; DFO, 2010b; Stewart et al., 2016). It is highly turbid as a result of wind mixing. Tathlina Lake is upstream of Kakisa Lake (60.928438°N, 117.716127°W; Figure 1.1), and receives water from the Kakisa River, which runs from upstream (south) of Tathlina Lake and through Kakisa Lake before emptying into the Mackenzie River at Beaver Lake (Kennedy, 1962; Figure 1.1). Kakisa Lake has a surface area of 331 km<sup>2</sup> and an average depth of 3.8 m (maximum 7 m; Roberge et al., 1986; Steward & Low, 2000). Both Kakisa and Tathlina Lakes are relied upon by the Ka'a'gee Tu First Nation (KTFN; located at "Kakisa" in Figure 1.1) for commercial and subsistence fishing. Lady Evelyn Falls lies downstream of Kakisa Lake along the Kakisa River and, standing at 14.6 m tall, prevents upstream movement of fish to Kakisa and Tathlina Lakes from the Mackenzie River (Kennedy, 1962). A series of intense rapids shortly upstream of Kakisa Lake are believed to prevent fish movement between the lakes, as tagged fish have not been observed to make the journey (Roberge et al., 1988; Stewart & Low, 2000). The Kakisa River watershed (Figure 1.1) spans a total of 14,900 km<sup>2</sup> (Roberge et al., 1988).

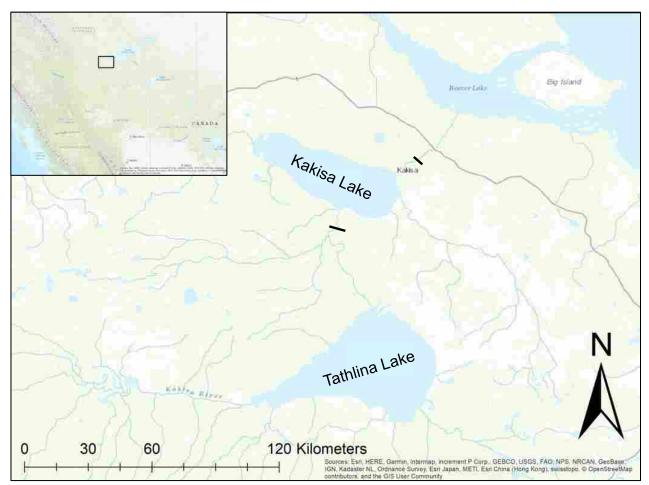


Figure 1.1: Locations of Tathlina and Kakisa Lakes and Ka'a'gee Tu First Nation (denoted "Kakisa"; Tom Pretty, 2019). Black lines indicate rapids and falls that are believed to prevent fish movement into and out of Kakisa Lake.

Since current monitoring efforts began in 2014, the gill net catch in Tathlina Lake has included Walleye (*Sander vitreus*, 23%), Lake Whitefish (*Coregonus clupeaformis*, 38%), and Northern Pike (*Esox lucius*, 30%), with the remaining 9% being suckers (*Catostomus* spp.). Species composition of catches from Kakisa Lake have been similar: Walleye (30%), Lake Whitefish (29%), Northern Pike (21%), suckers (13%) and Cisco (other *Coregonus* spp., 7%). Walleye and Lake Whitefish are the more commonly harvested species, and the only species harvested for commercial purposes, with Walleye dominating the majority of sale; Northern Pike remains a subsistence species (Stewart & Low, 2000; T. Chicot, KTFN, personal

communication). Since Tathlina Lake opened as a commercial fishery in 1953, catches of Walleye have had major declines in every decade, due in large part to overfishing (DFO, 2010b). These declines may have been exacerbated by environmental conditions, such as recurring hypoxia in the winter (Stewart et al., 2016). Following a population crash of Walleye in Tathlina Lake in 2001, which was attributed to an unsustainably high catch quota of 20,000 kg, the fishery has only been opened in 2003 (quota 5,000 kg, actual catch  $\leq$ 500 kg) and 2008 (quota 2,000 kg, actual catch 620 kg; Figure 1.2; DFO, 2010b). At the time of writing, commercial fishing in Tathlina Lake is not believed to be economically viable (G. Low, personal communication, 2018).

The fishery in Tathlina Lake is further undermined by high mercury (Hg) concentrations in Walleye. Mercury concentrations in some large Walleye exceed Health Canada's (2011) commercial sale guideline of 0.5 ppm (Laird et al., 2018). Most of the existing literature on these lakes is comprised of assessments of catch quotas and commercial viability of the fisheries (Kennedy, 1962; Roberge et al., 1986; Roberge et al., 1988; Stewart & Low, 2000; DFO, 2010a&b), and has not addressed Hg.

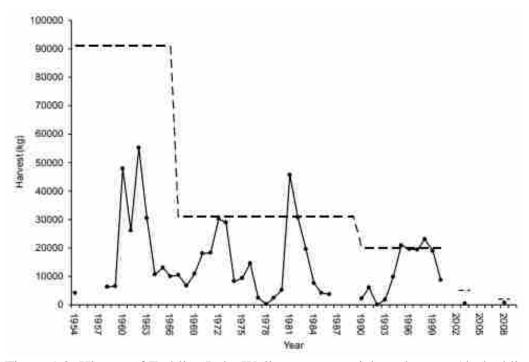


Figure 1.2: History of Tathlina Lake Walleye commercial catch quota (dashed line) and actual commercial catch (solid line) from 1953 to 2008 (DFO, 2010a, contacted for permission for republication).

# 1.2 Food, Human Health, and Mercury in Northern Regions

Northern communities are uniquely at risk of Hg exposure because of the contaminant's disproportionate prevalence in northern wild foods compared to most food available in the south; this includes the food fishes examined in this study (Pirkle et al., 2016). Commercial food options are not as abundantly available in northern areas as in more southerly areas, and can be up to 2.5 times more expensive (Gionet & Roshanafshar, 2013). When commercial foods are available, they are typically low in vital micronutrients and are generally of low nutritional quality, causing obesity and other chronic diseases related to micronutrient deficiencies (Huet et al., 2012; Gionet & Roshanafshar, 2013; Pirkle et al., 2016). Wild-harvested foods, including fish, are often a viable alternative and have higher nutritional value. They also tend to hold significant cultural and spiritual value for Indigenous peoples as food, medicine, and as a source

of connection to the land (Lemire et al., 2015; Pirkle et al., 2016). Notwithstanding the risk of chronic and acute health effects related to exposure to environmental contaminants such as Hg, wild foods are an important and often necessary source of nutrition (Lemire et al., 2015). Without wild foods or feasible commercial alternatives, northern communities face food insecurity (Kuhnlein et al., 2004). There are many wild foods that pose low contaminant exposure risks to people, but there has been difficulty in translating scientific knowledge of what is and is not safe to eat, and in what quantities, into terms that may be understood by the general public (Pirkle et al., 2016). Failures in effective risk communication have caused people to fear and avoid wild foods altogether, driving them toward the health risks that are associated with diets heavy in those commercial foods that are available (Pirkle et al., 2016).

When exposure to mercury is high enough, human health effects include damage to the nervous, neurological, visual, auditory, and immune systems, such as impairment of any or all sensory functions (e.g., constriction of visual field or blindness, loss of tactile sensitivity, reduced or lost hearing), muscle weakness and/or tremors, cognitive impairment (developmental or onset), and death (Wolfe, 1998; Pirkle, et al., 2016). Depending on consumption levels and age at exposure (e.g., prenatal or early childhood compared to adulthood), these effects may be quite severe. Age is inversely proportional to the risk of Hg toxicity, with fetuses more at risk than adults (Pirkle et al., 2016). Understanding the drivers and resulting variability of fish Hg concentrations ([Hg]) in northern aquatic ecosystems allows for informed and targeted approaches to risk communication and development of consumption guidance.

## **1.3 Mercury Cycling & Contamination**

#### 1.3.1 Atmospheric mercury: sources and deposition

Mercury in aquatic ecosystems is derived from both anthropogenic and natural sources (UNEP, 2019). Over the first part of the 21<sup>st</sup> century, anthropogenic Hg has been released to the atmosphere at a rate of 2,000-2,500 Mg/yr (2,220 Mg in 2015), comprising 30% of atmospheric Hg emissions (UNEP, 2019). A further 60% of atmospheric Hg emission comes from previously deposited "legacy" Hg that is recycled from land and water by natural processes, but which can originate from either natural or anthropogenic sources, although there remains much uncertainty around these estimates (UNEP, 2019). The remaining 10% of atmospheric Hg emissions comes from natural geological processes such as volcanic activity (UNEP, 2019). The largest contemporary anthropogenic contributors of Hg to the atmosphere are artisanal and small-scale gold mining/purification (38%, mostly from South America and Sub-Saharan Africa), industry sectors (28%, e.g., chlor-alkali production, industrial waste), and the refinement and burning of coal and other fossil fuels (24%) (UNEP, 2019). There is considerably more atmospheric Hg in the northern hemisphere than in the southern hemisphere, reflecting the relative abundance of natural and anthropogenic source areas in each hemisphere (UNEP, 2019). Rising temperatures due to climate change are expected to exacerbate Hg-related issues by increasing mobilization of long-deposited Hg from the lithosphere into the atmosphere, hydrosphere, and biosphere (Rydberg et al., 2010; AMAP, 2011).

Atmospheric Hg primarily consists of elemental Hg (Hg<sup>0</sup>) (Lindqvist & Rodhe, 1985; Kumari et al., 2015). Upon release to the atmosphere or dissolution into oceans, wind and water currents transport Hg<sup>0</sup>, mostly from East Asia and Africa (UNEP, 2019), to subarctic and Arctic areas, where lower temperatures and atmospheric mercury depletion events (AMDE) favor deposition (Figure 1.3.1.1; AMAP, 2011; Steffen et al., 2008). AMDEs are a seasonal phenomenon, occurring in the Arctic and subarctic spring, when Hg<sup>0</sup> oxidizes with halogen species emitted by freezing sea water, especially Br and BrO, to form HgBr<sub>2</sub>, a reactive form of inorganic Hg that deposits easily (Steffen et al., 2008; AMAP, 2011). Although AMDEs deposit a significant amount of Hg, only 20-40% is retained by the landscape while the rest is reemitted to the atmosphere; this results in a net increase of bioavailable Hg (Douglas et al., 2012). Dry deposition makes up 70% of deposition to Arctic tundra environments (UNEP, 2019).

The traditional understanding of the Hg cycle (Figure 1.3) is that Hg<sup>0</sup> is oxidized in the atmosphere to form divalent inorganic mercury (Hg(II)). This Hg(II) deposits into water and soil, where it is either reduced back to Hg<sup>0</sup> or methylated into the organic (Hg(I)) form monomethylmercury ([HgCH<sub>3</sub>]<sup>+</sup>), or simply methylmercury (MeHg) (Lindqvist & Rodhe, 1985; Kumari et al., 2015). Some MeHg in the water column can be demethylated before being taken up by biota. The largest demethylation process in lakes is photodemethylation, wherein ultraviolet (UV) -A, UV-B, and visible light oxidize Hg(I) in MeHg to inorganic Hg(II) (Lehnherr & St. Louis, 2009). Current global deposition of Hg from the atmosphere onto land and fresh water is estimated at 3600 Mg/yr (UNEP, 2019).

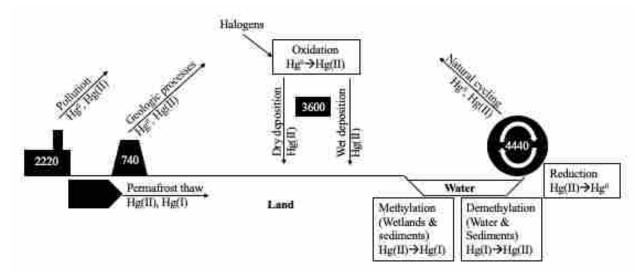


Figure 1.3: The abiotic mercury cycle. Numbers indicate Hg movement in Mg/yr (Lindqvist & Rodhe, 1985; Rydberg et al., 2010; AMAP, 2011; UNEP, 2019).

## 1.3.2 Novel loading pathways

Obrist et al. (2017) recently found that atmospheric  $Hg^0$  can be taken up directly by vegetation. Upon senescence, the Hg contained in this vegetation is released into the soil. In above-permafrost Arctic soils, peak Hg concentrations reach 138 µg/kg (compared to 20-50 µg/kg in tropical and temperate soils; Obrist et al., 2017), and thus soils can be a significant source of mercury to downstream aquatic ecosystems.

While deposition of Hg into building permafrost layers over geologic timescales has resulted in an overall sink effect in the Arctic and subarctic, global climate change is leading to permafrost net thaw, releasing long-deposited Hg to the water table and down-gradient aquatic habitats. Given previous deposition rates and the unrelenting rate of permafrost thaw, loading of Hg from thawed permafrost to lakes may increase to equal the loading from anthropogenic atmospheric emissions (Rydberg et al., 2010). The study area is located within the sporadic discontinuous permafrost zone (10-50% permafrost by area; NRC, 1995), so thawing of permafrost will likely affect Hg loading in the future.

#### 1.3.3 Mercury in biota

In soils and fresh water, Hg exists mostly as Hg(II) or Hg<sup>0</sup>. These forms of mercury have low water solubility on their own, but remain in-matrix as components of larger metal-centric ligands (Lindqvist & Rodhe, 1985; Kidd & Batchelar, 2012). In aquatic systems, Hg(II) is methylated mostly by sulfate-reducing bacteria (SRB) in anoxic sediment (Compeau & Bartha, 1985; Korthals & Winfrey, 1987). MeHg is the form of Hg that bioaccumulates and biomagnifies through aquatic food webs. MeHg typically ranges between 0.1-5% of total Hg (THg) in fresh water; percent methylmercury of total mercury is generally considered an indicator of net methylation rate (e.g., Gilmour & Henry, 1991; Kidd & Batchelar, 2012; Paranjape & Hall, 2017).

MeHg enters food webs from the water column via uptake by phytoplankton, after which it biomagnifies with each trophic transfer (Atwell et al., 1998; Pickhardt et al., 2002; Kidd & Batchelar, 2012). [Hg] increases ~10x per trophic level after bioconcentrating in algae; algal [Hg] can be 10,000-100,000x that of the surrounding water column (Hill & Larsen, 2005). Most Hg in fish is derived from dietary uptake (Hall et al., 1997), and, once absorbed, passes through the phospholipid bilayer of exposed cells and reduces sulfhydryl bonds of thiol ligands. At high enough concentrations, this can lead to disruption of cellular function, sometimes to the point of tissue damage (Stohs & Bagchi, 1995; Clarkson, 2002). Hg has a high affinity for amino acids, especially the sulfhydryl groups of histidine chains and cysteine. The cell membrane itself can then be compromised due to removal by Hg of phosphoryl groups necessary for cell membrane structure (Clarkson, 2002; Kidd & Batchelar, 2012).

## 1.4 Factors Affecting Biomagnification and Accumulation of Hg

#### 1.4.1 Bloom dilution

MeHg enters aquatic food webs when absorbed by primary producers (Pickhardt et al., 2002), and the abundance of primary producers can alter bioaccumulation of Hg through the food web. The bloom dilution hypothesis posits that, for biomagnifying contaminants such as Hg, an increase in biomass of primary producers yields a smaller amount of contaminant per unit mass of producers, as the contaminant load is distributed across a larger biomass of algal cells; this lower concentration then perpetuates into higher trophic levels. In simple terms, if the food web starts at a low concentration, it will end at a lower concentration (Figure 1.4; e.g., Pickhardt et al., 2002; Walters, 2015). Bloom dilution has been demonstrated for a number of contaminants, such as organochlorine pesticides (Berglund, 2003) and several metals including Hg (Hill & Larsen, 2005).

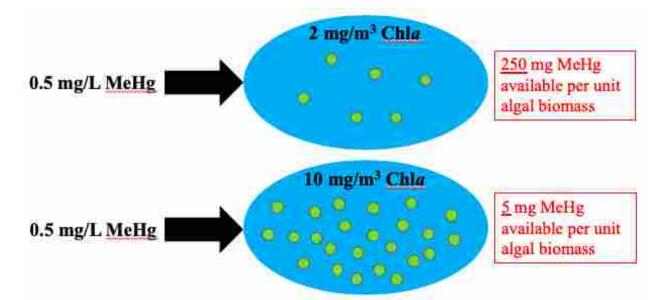


Figure 1.4: Conceptual illustration of bloom dilution. Two lakes have the same water [Hg], but differ in algal biomass, measured by proxy in mg/m<sup>3</sup> of chlorophyll *a* (Chl*a*). Assuming that all available MeHg in the water is absorbed by the primary producers, the ratio of available Hg to algal biomass (numbers in red) is lower in the lake with higher algal biomass. Lower [Hg] in algae lead to lower [Hg] at higher trophic levels.

Previous bloom dilution literature has focused on the negative correlation between biotic [Hg] and was chlorophyll *a* concentrations, but there has not been sufficient study of the coupling mechanisms between pelagic algal biomass and [Hg] in benthos in the context of bloom dilution. Chlorophyll *a* is a proxy measure for pelagic algal biomass, but food fishes rely on prey from both the pelagic and benthic food webs Although the effects of increased algal biomass on [MeHg] have not been directly studied for attached algae, pelagic and benthic food webs are coupled in these shallow lakes, and thus effects of bloom dilution may be expected even in consumers that rely primarily on benthic prey. Benthic detritivores derive nutrients from sinking pelagic detritus, much of which is dead phytoplankton or dead zooplankton, which fed directly on phytoplankton. Other benthic macroinvertebrates consume live plankton, especially when plankton performs diel migrations (Baustian et al., 2014).

#### 1.4.2 Growth dilution

Dilution of Hg can occur at trophic levels above phytoplankton, including in higher trophic level fishes. The growth dilution effect is a result of digestive efficiency, and has been documented to occur for several contaminants, including Hg (Verta, 1990; Karimi et al., 2007; Ward et al., 2010). Consider two individuals of the same population, age, and size at hatching, but one grows more efficiently than the other. The more efficient individual will grow by some weight for each unit of prey consumed, whereas the less efficient individual will grow by a lesser amount for the same unit of prey consumed. Although both fish have the same Hg burden, the faster-growing (i.e., more efficient) individual has added more biomass per unit prey intake, and therefore will have less Hg contained per unit biomass (Karimi et al., 2007; Ward et al., 2010). Growth efficiency can be affected by a number of biotic factors, including nutrient absorption efficiency, prey nutritional quality, and inter- or intra-specific competition; abiotic factors such as hypoxia or duration of seasonal ice-over (i.e., long duration of low-temperature conditions) can have negative effects on growth efficiency, as they can divert energy investment away from growth (Verta, 1990; Karimi et al., 2007; Ward et al., 2017; Ward et al., 2016).

Age-at-size is a simple yet useful method for comparing growth rates between systems. In this method, a log-linear regression is used to quantify the relationship between fish age and fork length, and this relationship is used to predict the time required for an average fish of the sample population to reach a given fork length.

# 1.5 Stable Isotope Analysis & Food Web Construction

Analysis of stable isotope ratios of carbon and nitrogen can be used to quantitatively describe food web structure and trophic ecology of organisms. Gut content analysis, the more traditional method of investigating food web structure and organismal diet, is biased toward

hard-bodied species and shows only a snapshot of an organism's diet (Nielsen et al., 2018), while stable isotope analysis provides a more time-integrated signal and gives a more accurate representation of an organism's integrated diet; however, this method lacks the taxonomic resolution of prey items afforded by gut content analysis (Fry, 2006; Hayden, 2018). As a result of differing fractionation rates of biochemical pathways, as well as specific growth rates of varying tissue types and species, each tissue of a single individual will have different stable isotope ratios that represent the timescale of that tissue's turnover rate (Boecklen et al., 2011; Ben-David & Flaherty, 2012).

 $\delta^{15}$ N ratios increase by approximately 3.0-3.4‰ with each trophic transfer (Figure 1.5; DeNiro & Epstein, 1981), and are used as an indicator of trophic position (Fry, 2006). Because  $\delta^{13}$ C fractionates minimally (0-0.2‰) with trophic transfers, it is useful for differentiating between littoral and pelagic sources in aquatic ecosystems (Figure 1.5; Fry, 2006). Food items with more negative  $\delta^{13}$ C values indicate carbon input from pelagic food webs, whereas less negative  $\delta^{13}$ C values indicate input from littoral food webs. Mercury methylation tends to occur at a higher rate in pelagic areas and, as such, pelagic food webs tend to be associated with higher [Hg] (Figure 1.5; e.g., Fry, 2006; Eloranta, 2013; Hayden, 2018). France & Peters (1997) found that freshwater ecosystems show an average  $\delta^{13}$ C enrichment of 0.2‰ per trophic transfer, though with low predictive capability (R<sup>2</sup>=0.10).

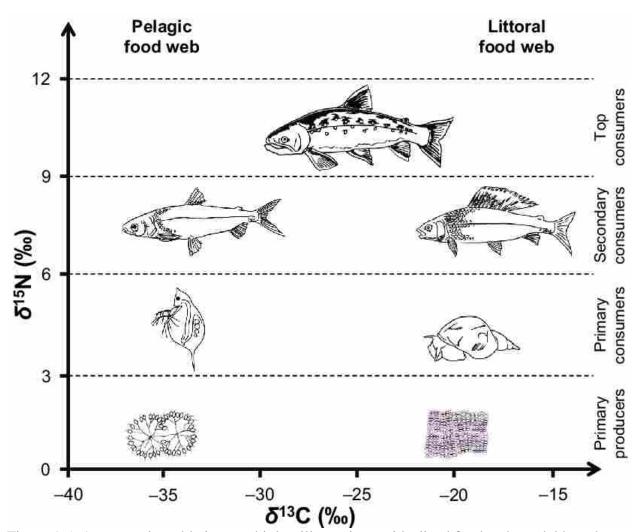


Figure 1.5: An example stable isotope biplot, illustrating an idealized food web model based on isotopic signatures of each organism (Eloranta, 2013, reproduced with permission from the author).

Knowing how isotope ratios fractionate across trophic levels enables construction of food web models. There is often a linear relationship between  $log_{10}([Hg])$  and  $\delta^{15}N$  in food webs; this relationship can vary depending on the ecosystem type (e.g., marine, freshwater, terrestrial), with freshwater systems having a slope of ~0.2 (Lavoie et al., 2013). Using  $\delta^{15}N$  to compare the trophic positions of organisms from different systems requires adjustment for organism size and system- (e.g., lake) specific trophic baseline. As long-lived primary consumers, clams of the Family Unionidae typically serve as ideal baseline organisms. To adjust for among-system differences in trophic baseline, the mean value of lake-specific unionid  $\delta^{15}N$  is subtracted from the  $\delta^{15}N$  value of higher-level consumers of interest to obtain the baseline-adjusted  $\delta^{15}N$  ( $\delta^{15}N_{adj}$ ) (Jardine et al., 2003; Vuorio et al., 2007). Because  $\delta^{15}N$  is colinear with fish size (Jardine et al., 2003), fish size must also be accounted for in comparing  $\delta^{15}N$  between populations.  $\delta^{15}N_{adj}$  is compared at a predetermined, standardized fork length using a least-squares means estimate. The relationship between  $\log_{10}([Hg])$  and  $\delta^{13}C$  is much more variable, but tends to be a negative relationship, indicating that fish [Hg] is higher for individuals whose diets are dominated by pelagic food webs, and fish [Hg] is lower for individuals feeding from more littoral food webs (Power et al., 2002).

Stable isotope ratios are reported as a delta ( $\delta$ ) value, which is a ratio of ratios expressed in permille units (Peterson & Fry, 1987; Fry, 2006; Hayden, 2018). Delta values are calculated using the following equation:

$$\delta^{H} \mathbf{X} = \left(\frac{R_{Sample}}{R_{Standard}} - 1\right) \times 1000$$

Where:

X is the element being analyzed; H is the mass of the heavier isotope; and R is the ratio of heavy/light isotopes.

For example:

$$\delta^{13}C = \left(\frac{\left(\frac{\delta^{13}C}{\delta^{12}C}\right)_{sample}}{\left(\frac{\delta^{13}C}{\delta^{12}C}\right)_{standard}} - 1\right) \times 1000$$

 $R_{Standard}$  for  $\delta^{13}C$  is 0.01118‰, taken from the PeeDee Belemnite rock formation in South Carolina, typically denoted "PDB."  $R_{Standard}$  for  $\delta^{15}N$  is 0.0036765‰, which is the naturally

occurring ratio of  ${}^{15}$ N/ ${}^{14}$ N in the atmosphere, typically denoted as "AIR" (Jardine et al., 2003; Fry, 2006).

## **1.6 Objectives & Hypotheses**

### 1.6.1 Abiotic environment

While effects of bloom dilution, growth dilution, and trophic ecology on fish [Hg] are the focus of this work, these biotic processes only concentrate or dilute Hg that already exists in the system. In order to account for differences in [Hg] at the base of the Kakisa Lake and Tathlina Lake food webs, I will assess [Hg] in the water and sediment of each lake. All other factors being equal, because biomagnification proceeds on an exponential scale, seemingly small differences in environmental [Hg] will result in drastic differences in [Hg] at higher trophic levels. As a more turbid lake, Tathlina Lake is likely to undergo a lower rate of photodemethylation than Kakisa Lake.

H<sub>A-1</sub>: [MeHg] in abiotic environmental compartments (water and sediment) differ between Tathlina Lake and Kakisa Lake; sediments and water in Tathlina Lake are higher in [MeHg] and %MeHg than in Kakisa Lake. H<sub>0-1</sub>: There is no significant difference in MeHg in sediment and water samples between the lakes.

# 1.6.2 Biotic variables affecting [Hg]

#### 1.6.2.1 Bloom dilution

Tathlina Lake's high turbidity should result in lower light availability for primary producers. This would result in higher [Hg] in Tathlina Lake's primary producers, creating a higher "starting" concentration for in-lake biomagnification of Hg. Stewart et al. (2016)

designated Tathlina Lake as mesotrophic, citing its total phosphorus concentration (unfiltered, 10-20  $\mu$ g/L TP), while Stewart & Low (2000) described Kakisa Lake as almost eutrophic (60  $\mu$ g/L TP). **H**<sub>A-2a</sub>: Tathlina Lake has lower algal biomass than Kakisa Lake, as indicated by water [chlorophyll *a*]. **H**<sub>0-2a</sub>: There is no significant difference in water [chlorophyll *a*] between the lakes.

Trophic biomagnification intercepts are interpreted as indicators of [Hg] incorporated at the lowest trophic positions (Lavoie et al., 2013). **H**<sub>A-2b</sub>: The intercept of a linear regression between [Hg] and  $\delta^{15}$ N, which includes all sampled taxa, is higher in Tathlina Lake than in Kakisa Lake. **H**<sub>0-2b</sub>: The intercepts of trophic biomagnification regressions are not significantly different between lakes.

# 1.6.2.2 Trophic ecology, trophic biomagnification & fish growth rates

Fish [Hg] in general increases with trophic position. Calculating least-squares means of size-standardized  $\delta^{15}_{adj}$  allows for the direct comparison of trophic positions between lakes. H<sub>A</sub>. <sub>3a</sub>: Fish feed at a higher trophic position in Tathlina Lake than in Kakisa Lake. Species-specific, size-standardized  $\delta^{15}N_{adj}$  values are significantly higher in Tathlina Lake than in Kakisa Lake. H0-3b: There is no significant difference in species-specific, size-standardized  $\delta^{15}N_{adj}$  between lakes.

Rates of Hg biomagnification within the Tathlina Lake and Kakisa Lake food webs can be quantified by regressing [Hg] against  $\delta^{15}$ N. Log<sub>10</sub>([Hg]) of all sampled biota will be regressed as a function of trophic position (‰  $\delta^{15}$ N) in order to test the hypothesis that the rate of Hg biomagnification is different between the two lakes. **H**<sub>A-3b</sub>: The trophic biomagnification slope, as modeled by  $\log_{10}(\text{Hg})$  and  $\delta^{15}$ N, is higher in Tathlina Lake than in Kakisa Lake. **H**<sub>0-3b</sub>: There is no statistical difference between lakes in trophic biomagnification slopes.

To differentiate between effects of bloom dilution and growth dilution on fish [Hg], fish growth rates must be estimated and compared between lakes. Using age-at-size, fish growth rates are compared between lakes within species to test the hypothesis that differences in [Hg] between lakes are related to differences in fish growth rates between lakes.

H<sub>A-3c</sub>: Differing fish growth rates provide a partial explanation for differences in fish [Hg]. Tathlina Lake has slower-growing fish populations, which leads to higher [Hg]. H<sub>0-3c</sub>: Species-specific age-at-size does not differ significantly between lakes.

# **1.7 Species of Focus**

Because this project aims to inform KTFN community health advisories, all three food fish species in Tathlina and Kakisa Lakes were targeted: Northern Pike, Lake Whitefish, and Walleye. Lake Whitefish occupy a relatively low trophic position compared to other fish species, feeding on plankton in early stages and ontogenetically shifting to benthic macroinvertebrates for the remainder of the lifespan (Coad et al., 1995). Walleye experience similar ontogenetic shifts, feeding on zooplankton and benthic macroinvertebrates in larval and juvenile stages, but then shift again to piscivory in adulthood, including a significant amount of cannibalism (Hartman, 2009). Northern Pike are opportunistic ambush predators and therefore tend to have diets representative of the littoral habitats in which they are well camouflaged. Their diets are flexible and they will consume benthic macroinvertebrates when other prey is scarce, but they strongly prefer fish, including other Northern Pike; it has also been documented that they may sometimes eat rodents or birds (Harvey, 2009). Collection of flesh and aging structures was crucial for both continued monitoring of [Hg] and the present attempt to model its drivers. Benthic macroinvertebrates and small fish were sampled for stable isotope ratios and mercury concentrations in order to construct a food web model for both lakes. The limited existing literature (Kennedy, 1962; Roberge et al., 1986; Roberge et al., 1988; Stewart & Low, 2000; DFO, 2010a&b) has explored only the commercial viability of these fisheries by biomass of food fishes; they did not attempt to investigate any species or groups that were not food fishes.

# CHAPTER 2: ABIOTIC CONDITIONS, ALGAL BIOMASS & FISH GROWTH RATES AFFECT FISH MERCURY CONCENTRATIONS IN TWO SUBARCTIC LAKES

## 2.1 Abstract

In the Dehcho Region of the Northwest Territories, Kakisa and Tathlina Lakes support commercial and subsistence fisheries for the local Ka'a'gee Tu First Nation (KTFN). Analyses of mercury (Hg) in sediment, water, fish, and macroinvertebrates, as well as analyses of water chemistry, fish growth, and stable isotope ratios in biota, were undertaken to determine why Walleye (Sander vitreus) from Tathlina Lake have mercury concentrations ([Hg]) that are often higher than Health Canada's commercial sale guideline (0.5 ppm wet weight), and that are higher than Walleye in downstream Kakisa Lake. In collaboration with Indigenous guardians, Tathlina Lake was sampled in the summers of 2014 and 2018, and Kakisa Lake was sampled in the summers of 2015, 2017, and 2018. Methylmercury (MeHg) concentrations and %MeHg (of total Hg) in unfiltered water, filtered water, and sediment samples were higher in Tathlina Lake (0.094 ng/L—10.737% MeHg, 0.088 ng/L—9.611%, 0.335 ng/g—0.456% respectively) than in Kakisa Lake (0.038 ng/L-3.883%, 0.031 ng/L-4.662%, 0.228 ng/g-0.233%, respectively), indicating that there is a higher net methylation rate and more MeHg available for uptake into the food web in Tathlina Lake than in Kakisa Lake. In addition, both bloom dilution and growth dilution may result in lower [Hg] in Kakisa Lake Walleye; chlorophyll a concentrations in water were higher in Kakisa Lake than Tathlina Lake (mean 4.16 and 0.42 µg/L chlorophyll a, respectively), and analyses of fish age-at-size showed that Walleye grow faster in Kakisa Lake than in Tathlina Lake (10.4 and 14.6 yrs at 450 mm, respectively). Lake Whitefish (Coregonus *clupeaformis*) [Hg] is also higher in Tathlina Lake than in Kakisa Lake but both populations remain below the Health Canada guideline; differences in Lake Whitefish [Hg] are driven by abiotic availability and bloom dilution as described above, but not growth dilution. Northern

Pike (*Esox Lucius*) [Hg] did not differ significantly between the two lakes, which could be related to differing habitat or high diet variability compared to Walleye and Lake Whitefish. Further research would be necessary to explore this question.

## **2.2 Introduction**

In many northern Indigenous communities, wild-caught fish are a significant source of subsistence food and support cultural wellbeing, but also present a source of mercury (Hg) exposure (AMAP, 2011; Pirkle et al., 2016; UNEP, 2019). Several northern jurisdictions have issued either general or site-specific advisories regarding consumption of wild-caught fish. In this context, it is critical to understand sources of variation in fish Hg concentrations ([Hg]) between populations; not only does this help communities decide from where to harvest fish, it also helps scientists, managers, and regulators to predict effects of continued environmental change on fish [Hg].

Spurred by concerns voiced by members of the Ka'a'gee Tu First Nation (KTFN) regarding the safety and security of their subsistence and commercial fisheries on nearby Kakisa and Tathlina Lakes, [Hg] of three food fish species were investigated: Lake Whitefish, (*Coregonus clupeaformis*), Northern Pike (*Esox Lucius*), and Walleye (*Sander vitreus*). Tathlina Lake Walleye and Lake Whitefish (*Coregonus clupeaformis*) both have significantly higher [Hg] than their Kakisa Lake counterparts. Furthermore, some Tathlina Lake Walleye exceeded Health Canada's (2010) commercial sale guideline of 0.5 ppm THg (wet weight). In both lakes, Northern Pike and Lake Whitefish generally remain below the guideline.

Mercury is released to the atmosphere from both natural and anthropogenic sources (Rydberg et al., 2010; AMAP, 2011; Kumari et al., 2015). Currently, anthropogenic sources dominate emissions and further release is expected to increase due to climate change (Lindqvist & Rodhe, 1985; AMAP, 2011; UNEP, 2019). Arctic and subarctic landscapes are a sink for atmospheric Hg because colder temperatures favor deposition (AMAP, 2005; Obrist et al., 2017). Inorganic and elemental forms of Hg can be methylated to methylmercury (MeHg), the toxic organic form, in catchments and in anoxic sediments (Branfireun et al., 1999; AMAP, 2011; Kumari et al., 2015). MeHg released from soil and sediment into the water column can enter aquatic food webs via sorption by primary producers, and then biomagnify with each trophic transfer (Lindqvist & Rodhe, 1985; Kumari et al., 2015). The bioavailability of Hg (i.e., concentrations and speciation) in the abiotic environment—that is, in water and sediment—is the first major driver of variability in [Hg] in aquatic food webs (AMAP, 2011; UNEP, 2019). Sorption onto primary producers in the water column is the only significant input of Hg to the pelagic food web; there is negligible uptake from the water column to organisms at higher trophic levels (AMAP, 2011; Kidd & Batchelar, 2012).

Mercury concentrations increase by several orders of magnitude (10,000-100,000X) between the water column and primary producers, but the magnitude of bioconcentration depends in part on the abundance of primary producers. Algal [Hg] decreases as an inverse function of algal biomass; this is known as the bloom dilution effect (Pickhardt et al., 2002; Berglund, 2003; Hill & Larsen, 2005; Walters, 2015). Once in aquatic food webs, Hg undergoes trophic biomagnification (Atwell et al., 1998; Pickhardt et al., 2002; Kidd & Batchelar, 2012). Rates of trophic biomagnification vary depending on a number of factors, including latitude and lake physicochemistry (e.g., DOC, total phosphorus, Hg deposition rate; Lavoie et al., 2013). Concentrations of Hg increase with trophic position (Hall et al., 1997; Atwell et al., 1998; Campbell et al., 2005) and other variables that affect bioaccumulation, such as growth rates. Metabolic rates and growth efficiency can affect concentrations of bioaccumulating

contaminants in animals, especially those at higher trophic levels. Previous researchers have shown that fish that grow more quickly and efficiently (i.e., gain more mass per unit food/contaminant intake) have lower [Hg] (Karimi et al., 2007; Ward et al., 2010).

The objective of this study was to compare [Hg] in three food fishes—Lake Whitefish, Northern Pike, and Walleye—between two subarctic lakes, Tathlina Lake and Kakisa Lake. Preliminary research has shown that [Hg] in Lake Whitefish and Walleye, important subsistence and commercial fish species for the KTFN, are higher in Tathlina Lake than in downstream Kakisa Lake, and Walleye [Hg] can exceed Health Canada's guideline for commercial sale (0.5 ppm wet weight; Low et al., 2017). Using data collected between 2014 and 2018, [Hg] in fish were compared directly between lakes and related to indicators of growth (age-at-size) and trophic ecology (stable isotope ratios of carbon and nitrogen). Also compared between lakes were rates of biomagnification and availability of MeHg in abiotic ecosystem compartments and at the bottom of each lake's food web. It was predicted that: 1) [MeHg] and %MeHg (of total Hg, THg) would be higher in water and sediment from Tathlina Lake than in Kakisa Lake; 2) primary productivity would be higher in Kakisa Lake than in Tathlina Lake; 3) fish growth rates would be higher in Kakisa Lake than in Tathlina Lake; and, 4) these differences would affect Hg biomagnification and ultimately [Hg] in top predator fishes.

#### 2.3 Materials & Methods

#### 2.3.1 Study area

Tathlina Lake (60.555086°N, 117.531469°W; Figure 2.1) is a wide, shallow lake with a surface area of 573 km<sup>2</sup> and an average depth of 1.5 m (maximum depth 2.7 m; Kennedy, 1962; DFO, 2010; Stewart et al., 2016). The Kakisa River flows into Tathlina Lake and continues downstream to Kakisa Lake (60.928438°N, 117.716127°W; Figure 2.1) before emptying into the

Mackenzie River at Beaver Lake (Figure 2.1). Kakisa Lake has a surface area of 331 km<sup>2</sup> and an average depth of 3.8 m (maximum 7 m; Roberge et al., 1986; Stewart & Low, 2000). Both Kakisa and Tathlina lakes are relied upon by the Ka'a'gee Tu First Nation (KTFN; denoted "Kakisa" in Figure 2.1) for commercial and subsistence fishing. Lady Evelyn Falls lies downstream of Kakisa Lake along the Kakisa River and, standing at 14.6 m tall, prevents fish movement to Kakisa and Tathlina Lakes from the Mackenzie River (Kennedy, 1962). Shortly upstream of Kakisa Lake lies a series of intense rapids that are believed to prevent fish movement between the lakes (Roberge et al., 1988; Stewart & Low, 2000). The Kakisa River watershed spans 14,900 km<sup>2</sup> (Roberge et al., 1988).

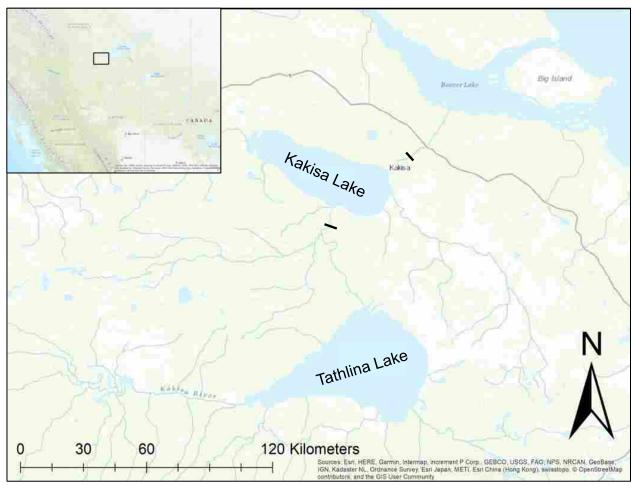


Figure 2.1: Locations of Tathlina and Kakisa Lakes and Ka'a'gee Tu First Nation (denoted "Kakisa;" Tom Pretty, 2019). Black lines indicate rapids and falls that are believed to prevent fish movement into and out of Kakisa Lake.

# 2.3.2 Field sampling

In each lake, samples of fish, sediment, water, and benthic invertebrates were collected for analyses of stable isotope ratios, THg concentration ([THg]), and [MeHg]. Water was also analyzed for a suite of standard chemical parameters. Tathlina Lake was sampled in the summers of 2014 and 2018 as part of ongoing monitoring efforts, including efforts undertaken by Indigenous guardians. Kakisa Lake was sampled in the summers of 2015, 2017, and 2018. Surface sediments were collected at the deepest point of each lake with a 6" x 6" x 6". Wildco Ekman dredge. The top 1-2 cm of sediment in the filled dredge was removed with a Teflon scraper, and placed into a 4 oz. WhirlPak bag before being frozen at -20°C in the field.

Water was collected for ultra-trace Hg analysis using EPA protocol 1669 (US EPA, 1996). The clean-hands dirty-hands technique was used to collect 250 mL unfiltered water samples. Filtered water samples (250 mL) were also collected using the clean-hands dirty-hands technique; water for filtered samples was pumped through acid-washed Teflon tubing and through a muffled QMA quartz filter ( $2.2 \mu m \times 47 mm$ ) using a peristaltic pump. Both filtered and unfiltered water were preserved with 1.25 mL of trace metal grade HCl to acidify each sample to 0.5% by volume.

Surface water samples for chlorophyll *a* analysis were collected in brown Nalgene bottles and then filtered through ethanol-rinsed glass fiber filters (1.6  $\mu$ m x 47 mm; typically 250 mL) on the same day. Chlorophyll *a* filters were frozen at -20°C in the field after processing.

Littoral and profundal macroinvertebrates were collected by kick net and Ekman dredge, respectively. Three locations per lake were selected to represent the diversity of available littoral and profundal habitats; within habitats (littoral vs. profundal), samples were combined due to biomass constraints. Macroinvertebrates were visually sorted to Order in the field before being frozen at -20°C in Whirlpak bags.

Forage fishes were captured with baited minnow traps deployed along the shoreline of each lake. After capture, fork lengths (mm) and wet weight (g) were recorded, and the fish were frozen whole in Whirlpaks at -20°C.

Gill nets were used to collect Walleye, Northern Pike, and Lake Whitefish. In each lake, KTFN fishers identified optimal netting locations. When sampling began in 2014 with a target of

10-15 individuals per lake, one to two multi-mesh gill nets (2 cm to 14 cm stretched mesh; 50 m) were set overnight (average 14 h). After qualitative assessment of catches in the first netting efforts, fishing locations and net meshes and lengths were adapted to minimize by catch. Because quantitative abundance and catch per unit effort were not objectives of this study, fishing methods were not standardized among years or lakes. Instead, methods have been adapted as necessary to: 1) maximize the captured size ranges of the target species; 2) manage catch volume such that all individuals could be processed in the field on the day of collection; and 3) minimize the risk of stranded nets and fish spoilage. In general, nets varied in length from 15 m to 50 m, with a mesh size between 2 mm and 14 mm stretched mesh. Nets were set for 3 to 14 h. Upon capture, fork length (mm) and whole weight (g) were recorded for each fish. Muscle tissue and aging structures were then immediately dissected. Collected aging structures were opercles in Walleye (Scott & Crossman, 1979), cleithra in Northern Pike (Faust et al., 2013), scales in Lake Whitefish, and otoliths (all species). Otoliths and scales were placed in Rite in the Rain envelopes and dried in the field; cleithra and opercles were placed in Whirlpak bags and frozen in the field at -20°C. Muscle tissue was sampled anterior to the dorsal fin and above the lateral line, and placed into Whirlpak bags before being frozen at -20°C in the field.

Frozen sediment, invertebrate, and fish tissues were shipped to the Swanson Lab at the University of Waterloo (UW) for further processing. Water samples destined for Hg analyses were shipped to the Analytical Services Laboratory at the Biotron Experimental Climate Change Research Centre at Western University, London, ON. Frozen filters (for chlorophyll *a* analysis) and water samples destined for general chemical analyses were shipped to the University of Alberta Biogeochemical Analytical Service Laboratory.

### 2.3.3 Laboratory processing

Frozen sediment, invertebrate, and fish tissues were freeze-dried (-55 °C, 0.01 mBar, 48 h; Labconco Freezone 2.5 Liter Freeze Dry System; Labconco, Kansas City, MO; Lewis, 2018) before being homogenized with scissors, which were rinsed with Milli-Q water and 90% ethanol between samples. Samples were stored in acid-washed 20 mL glass scintillation vials before they were sent for stable isotope analysis (SIA) and Hg analysis (see "Laboratory analyses" below). Benthic macroinvertebrates were sorted to Family or probable genus using a Leica dissecting microscope (10-60x) and dichotomous keys (Merritt et al., 1978; Thorp & Rogers, 1991). To ensure that there was sufficient mass for Hg and SIA analyses, congeneric invertebrates were combined in scintillation vials before freeze-drying and homogenization. Fish opercles and cleithra were manually cleaned of soft tissue using warm water and forceps before being sent to AAE Tech Services Inc. for analysis.

#### 2.3.4 Laboratory analyses

Freeze-dried and homogenized samples were sent to the Biotron Experimental Climate Change Research Centre at Western University (Biotron) for analyses of THg and MeHg. MeHg concentrations in sediment, water, and invertebrates were analyzed by cold vapor atomic fluorescence spectrophotometry (CVAFS) on a Tekran 2700 (Tekran Instruments Corporation, Toronto, ON) using a modified version of EPA 1630 (TM.0812). DORM-4 fish protein (National Research Council, Ottawa, ON) was used as a certified reference material. THg concentrations in water were analyzed with a Tekran 2600, while sediment and fish THg were analyzed on a DMA-80 Dynamic Mercury Analyzer (Milestone Srl, Italy) using modified versions of EPA 7473 (TM.0813) and EPA 1631 (TM.0811). DORM-4 fish protein was again used as a certified reference material. Because the majority of fish THg is in the form of MeHg (Bloom, 1992), and THg is simpler to measure, only THg analyses were performed on fish. Blanks and certified reference materials were analyzed at the beginning of every run and between every 10 samples.

After invertebrate and fish samples were freeze-dried and homogenized, 0.30-0.35 mg of those tissues was weighed into 3.5 mm tin capsules and sent to the UW Environmental Isotope Lab (EIL) to be analyzed for  $\delta^{13}$ C and  $\delta^{15}$ N stable isotope ratios. Analyses were performed on a 4010 Elemental Analyzer (Costech Instruments, Italy) with a Delta Plus XL (Thermo-Finnigan, Germany) continuous flow isotope ratio mass spectrometer (CFIRMS). For quality control, EIL uses international reference materials (IAEA-N1 & N2, IAEA-CH3 & CH6, USGS-40 & 41) and in-house reference materials (EIL-72, EIL-3, JSEC-01), as well as a reference material chosen to suit the particular analysis (NIST-1557b – bovine liver for fish and invertebrates). Twenty percent of all sample runs consisted of standard material. Results were reported with error within 0.3‰ and 0.2‰ for  $\delta^{15}$ N and  $\delta^{13}$ C, respectively.

Fish aging structures were sent to AAE Tech Services Inc. (La Salle, MB) for age estimations. Lake Whitefish and Walleye otoliths were aged using the crack and burn method (Christensen, 1964). For Northern Pike, cleithra were aged whole and otoliths were aged by sectioning. Ages were estimated by a primary aging technician and QAQC checks were made independently on 50% of the samples by a second aging technician.

At the University of Alberta Biogeochemical Analytical Service Laboratory, total nitrogen and phosphorus were analyzed by flow injection analysis on a Lachat QuickChem QC8500 FIA Automated Ion Analyzer (Hach, Loveland, CO), using modified versions of EPA method 353.2 and APHA method 4500-P-G, respectively. Alkalinity and pH were analyzed by titration on a Mantech PC-Titration Plus System (Mantech Inc., Guelph, ON) using modified versions of APHA method 4500-H+ B and 2320 B, respectively. Conductivity was analyzed on

the same machine using a conductivity probe. Chlorophyll *a* was analyzed on a Shimadzu RF-1501 Spectrofluorophotometer (Shimadzu, Columbia, MD) according to a modified version of Welschmeyer (1994).

### 2.3.5 Data analysis

Data from certain samples were excluded from analysis. Fish collected in winter months were excluded because seasonality is known to have an effect on [Hg] in individuals (Lemly, 1996; Harris & Bodaly, 1998; Zhang et al., 2012). Haemophagic leeches were excluded because their trophic niche does not fit the assumptions of biomagnification analyses. For further details and justifications, see Appendix 1.

Species-specific fish [Hg] were compared between Tathlina Lake and Kakisa Lake with an analysis of covariance (ANCOVA), using lake as the fixed factor, log<sub>10</sub>(fork length) as the covariate, and an interaction term (lake\*log<sub>10</sub>(fork length)). Data are presented as least-squares means estimated at a standardized fork length of 450 mm for Walleye and Lake Whitefish and 650 mm for Northern Pike; these sizes were chosen to minimize extrapolation. Pairwise comparisons of least-squares means were achieved with a Tukey's test.

Clams from the Family Sphaeriidae were collected in both lakes, and used to correct for between-lake differences in baseline  $\delta^{15}N$ . Average  $\delta^{15}N$  for sphaerids in Kakisa Lake was 4.31‰, and thus  $\delta^{15}N_{adj}$  in fish from Kakisa Lake was calculated as: fish  $\delta^{15}N - 4.31$ . Average  $\delta^{15}N$  in sphaerids from Tathlina Lake was 3.76‰, and thus Tathlina Lake fish  $\delta^{15}N_{adj}$  was calculated as: fish  $\delta^{15}N - 3.76$ . Species-specific  $\delta^{15}N_{adj}$  and  $\delta^{13}C$  ratios for Walleye, Northern Pike, and Lake Whitefish were then compared between lakes with an ANCOVA, where lake and  $\log_{10}$ (fork length) were the fixed factor and covariate, respectively. When interaction terms were

significant, they were retained in the model. Least-squares means at the above-stated standardized sizes were calculated and compared between lakes with a Tukey's test. As fork length was not significantly related to  $\delta^{13}$ C in Lake Whitefish (p=0.454), arithmetic means were compared between lakes with a Student's t-test.

Fish growth rates were compared between lakes using both Von Bertalanffy and age-atsize growth models. Testing of the model residuals indicated that age-at-size was the more robust approach. Age-at-size was calculated for each species/lake combination using the following ANCOVA model:

### Log<sub>10</sub>(age) ~ Log<sub>10</sub>(fork length), Lake, Log<sub>10</sub>(fork length)\*Lake

Values reported are the least-squares means estimates at the standardized fork lengths described above; these least-squares means were compared between lakes with a post-hoc Tukey's test. For results of Von Bertalanffy growth models, see Appendix 2.

Using both benthic invertebrate and fish data, rates of Hg biomagnification through the food web of each lake were estimated and compared to each other using an ANCOVA, where the responding variable  $\log_{10}([Hg])$  (total Hg for fish, MeHg for invertebrates) was modeled as a function of the covariate  $\delta^{15}N$  and lake (fixed factor). For all invertebrate taxa, [MeHg] and  $\delta^{15}N$  were arithmetic means of samples within the same Family and functional feeding group; where samples were confamilial but of different functional feeding groups, they were treated as separate groups with unique mean [Hg] and  $\delta^{15}N$ . For each fish species, size-standardized, least-squares means of  $\delta^{15}N$  were used.

When assumptions of normality of residuals and homogeneity of variance appeared to be violated, nonparametric tests were performed (Quade's ANCOVA, Welch's T test). Nonparametric results did not change interpretations, thus the original parametric results were used. Alpha was set to 0.05 for all significance testing. The general linear models described above, including comparisons of age-at-size, [Hg],  $\delta^{15}N_{adj}$ , and  $\delta^{13}C$  were performed using the open-access RStudio statistical software version 1.1.423 and the lsmeans and emmeans packages. The biomagnification model and all figures relating to general linear models were created with SPSS Statistics version 24 (IBM Corp., Armonk, NY) and SigmaPlot version 12.0 (Systat Software, Inc., San Jose, CA).

### 2.4 Results & Discussion

#### 2.4.1 Comparisons of fish [Hg] between lakes

Size-standardized [Hg] in Lake Whitefish and Walleye were significantly higher in Tathlina Lake than in Kakisa Lake (Tukey's test, p<0.0001; Figure 2.2, Table S2.1), but there was no significant difference in size-standardized [Hg] in Northern Pike between the two lakes (Tukey's test, p=0.7109; Figure 2.2, Table S2.1). ANCOVA model interaction terms (Lake\*Log<sub>10</sub>(fork length)) were significant for Lake Whitefish (p=0.002) and Northern Pike (p=0.004), but not for Walleye (p=0.500), indicating that the rate of increase of [Hg] with fork length differed significantly between lakes for Lake Whitefish and Northern Pike, but that the rate of increase of [Hg] with fork length for Walleye was similar between the two lakes. The between-lake difference in Walleye [Hg] was 3.6 x the between-lake difference in Lake Whitefish [Hg] (Figure 2.2).

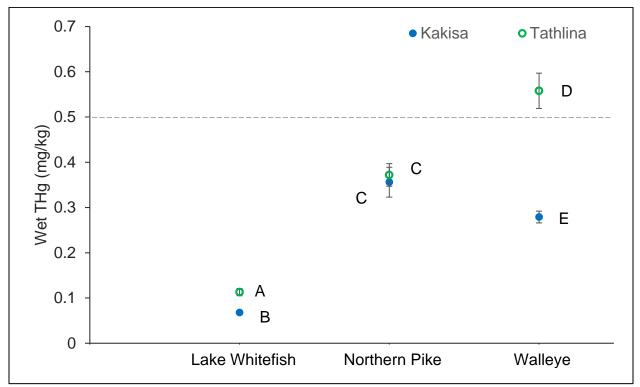


Figure 2.2: Size-standardized [Hg] ( $\pm$ SE) of food fishes in Tathlina and Kakisa Lakes. The dotted line indicates Health Canada's guideline for commercial sale (0.5 ppm wet weight). Values are least-squares means estimated at standardized fork lengths of 450 mm (Walleye and Lake Whitefish) and 650 mm (Northern Pike). Letters indicate significant pairwise differences (Tukey's test, Walleye & Lake Whitefish p<0.0001, Northern Pike p=0.7109).

At standardized sizes of 650 mm and 450 mm for Northern Pike and Lake Whitefish respectively, [Hg] in both lakes were below Health Canada's (2010) commercial sale guideline of 0.5 ppm THg (wet weight; Figure 2.2). The number of individual Northern Pike and Walleye that exceeded the Health Canada guideline are shown in Table 2.1; no Lake Whitefish exceeded the guideline.

	Tatl	ılina Lake	Kakisa Lake		
	Walleye (n=21)	Northern Pike (n=42)	Walleye (n=71)	Northern Pike (n=43)	
Number exceedances	13 (62%)	13 (30%)	2 (3%)	6 (14%)	

Table 2.1: Number of fish exceeding Health Canada [THg] guideline of 0.5 ppm (wet weight) for commercial sale of fish. %=percent of each species/lake combination in exceedance. No Lake Whitefish exceeded the guideline.

It was surprising that size-standardized [Hg] differed significantly between lakes for Lake Whitefish and Walleye, but not for Northern Pike. Given the lack of differences in [Hg] between Kakisa and Tathlina lakes for Northern Pike, and the fact that KTFN relies less on Northern Pike than on Walleye and Lake Whitefish to support commercial and subsistence fisheries (T. Chicot, KTFN, personal communication; G. Low, Dehcho First Nations, personal communication), the remainder of the analyses focused on explaining why [Hg] in Walleye and Lake Whitefish were higher in Tathlina Lake than in Kakisa Lake.

#### 2.4.2 Analyses of sediment and water

While sediment in Tathlina Lake had lower [THg] than sediment in Kakisa Lake, [MeHg] and %MeHg of THg were both higher in Tathlina Lake than Kakisa Lake (Table 2.2), indicating that net methylation rates are higher in Tathlina Lake. Although absolute differences in [MeHg] and %MeHg in sediment were small, higher values in Tathlina Lake mean that more mercury is available for uptake into sediment-dwelling organisms and for diffusion into overlaying waters, and this can result in greater accumulation in organisms. A similar pattern was seen in water: [MeHg] and %MeHg in filtered and unfiltered water samples were higher in Tathlina Lake than in Kakisa Lake (Table 2.2). Total mercury concentrations in unfiltered water were higher in Kakisa Lake, indicating that most of the THg in water from Kakisa Lake is bound to particulate matter.

	K	akisa Lak	ke	Tathlina Lake				
	Unfiltered	Filtered	Sediment	Unfiltered	Filtered	Sediment		
	Water	Water	(ng/g)	Water	Water	(ng/g)		
	(ng/L)	(ng/L)		(ng/L)	(ng/L)			
THg	1.694	0.736	98.432	1.003	1.106	84.528		
MeHg	0.038	0.031	0.228	0.094	0.088	0.335		
%MeHg	3.883	4.662	0.233	10.737	9.611	0.456		

Table 2.2: Mean [Hg] in water and sediment in each lake. %MeHg of THg.

Higher %MeHg in Tathlina Lake sediments and waters suggest a higher rate of net methylation in Tathlina Lake than Kakisa Lake, which could reflect differences in in-lake bacterial community composition (i.e., relative abundance of SRBs and other methylators), photodemethylation rates, and/or redox conditions in sediment, but further research is required (e.g., Gilmour & Henry, 1991; Kidd & Batchelar, 2012; Paranjape & Hall, 2017). Theoretically, a turbid lake such as Tathlina (Table 2.3) would undergo a lower rate of photodemethylation than a clearer one such as Kakisa Lake (Lehnherr & St. Louis, 2009), and this would contribute to the observed difference between net methylation rates. Annually recurring hypoxia in the winter, as reported by Stewart et al. (2016) would also contribute to anoxia in sediments, which is favorable for methylation. Assuming that filtered water samples represent availability of Hg that can be absorbed by or adsorbed to phytoplankton, Table 2.2 indicates that bioavailability of Hg is higher in Tathlina Lake than Kakisa Lake (Hill & Larsen, 2005).

As hypothesized, [chlorophyll *a*] was higher in Kakisa Lake than Tathlina Lake (Table 2.4). This higher productivity may be at least partially influenced by phosphorus availability, as total phosphorus concentrations were five times higher in Kakisa Lake than Tathlina Lake (Table 2.4). Furthermore, primary production may be limited in Tathlina Lake as a result of low light availability. As a shallow lake (average depth 1.5 m), it is subject to considerable wind mixing.

Stewart et al. (2016) indicated that algal production in Tathlina Lake has increased since 1967, citing [chlorophyll *a*] in sediment. Climate warming may be driving this increase and could cause continued increases pending changes in other limiting factors such as light availability, which may be affected by changes in mixing and wind-driven turbidity.

Based on [chlorophyll *a*], Kakisa and Tathlina Lakes can be designated as mesotrophic (2.5-8  $\mu$ g/L chlorophyll *a*) and oligotrophic (<2.5  $\mu$ g/L chlorophyll *a*), respectively; total phosphorus measurements dictate designations of eutrophy (>35 TP) and mesotrophy (10-35  $\mu$ g/L TP), respectively (Dodds, 2002). Regardless, it is clear that Kakisa Lake is more productive than Tathlina Lake. These findings are consistent with Redfield ratio calculations, which indicate that Tathlina Lake is likely phosphorus-limited (N:P<sub>Tathlina</sub> = 55:1), whereas Kakisa Lake has an excess of phosphorus relative to nitrogen (N:P<sub>Kakisa</sub> = 9:1; Dodds, 2002).

The relative concentrations of chlorophyll *a* indicate that Kakisa Lake has more algal biomass per L of water, and therefore more algal biomass per mol MeHg, than Tathlina Lake (Table 2.4; Pickhardt et al., 2002; Berglund, 2003; Hill & Larsen, 2005; Walters, 2015). As such, algae should have higher [MeHg] in Tathlina Lake than in Kakisa Lake, which is supported by the intercept values of the trophic biomagnification slopes. Results of regressions of log<sub>10</sub>([Hg]) vs.  $\delta^{15}$ N show that the Y-intercept, which can be interpreted as an estimate of [Hg] at the base of the food web (Lavoie et al., 2013), was significantly higher (ANCOVA, p<0.0005) in Tathlina Lake than in Kakisa Lake (Table S2.2). This is consistent with the bloom dilution hypothesis, although testing the hypothesis directly on algal samples was outside the scope of this work. Considered in relation to Walleye [Hg] (Figure 2.2; Table S2.1), these findings are consistent with previous researchers who have shown a negative correlation between lake chlorophyll *a* concentrations and Walleye [Hg] in lakes in the Dehcho region (Low et al., 2017).

Table 2.3: 2018 field water chemistry parameters. A caveat to note is that Tathlina Lake's Secchi depth was taken in the western basin where inflow keeps turbidity low. Tathlina Lake tends to be highly turbid in the main basin; Secchi depth in Tathlina Lake in the main basin in 2019 was 0.2 m (no measurements available for main basin in 2018).

	Secchi depth (m)	DO (mg/L)	pН	TDS (mg/L)	Conductivity (µS/cm)	Temp. ( <sup>O</sup> C)
Kakisa	0.4	11.0	7.53		235.1	10.4
Tathlina	1.6 (bottom)	11.4	7.86	183.95	283	8.6

Table 2.4: 2018 analytical water chemistry parameters. Chlorophyll a values are mean (±SE) concentrations from all sampling seasons.

	TP	TN	pН	Alkalinity	Conductivity		Chlorophyll $a$
	$(\mu g/L P)$	$(\mu g/L N)$	_	(mg/L CaCO <sub>3</sub> )	(µS/cm)	(mg/L HCO <sub>3</sub> )	$(\mu g/L)$
Kakisa	60	523	8.2	105.8	240	129.1	$4.16\pm0.48$
Tathlina	12	657	8.2	144.5	301	176.3	$0.42\pm0.55$

### 2.4.3 Biotic analyses: stable isotope ratios and fish growth rates

There was a significant and positive relationship between  $\delta^{15}N_{adj}$  and  $Log_{10}$ (fork length) for Walleye in both lakes (linear regression,  $p_{Tathlina}=0.001$ ,  $R^2_{Tathlina}=0.465$ ,  $df_{Tathlina}=20$ ,  $p_{Kakisa}<0.0005$ ,  $R^2_{Kakisa}=0.288$ ,  $df_{Kakisa}=70$ ) and Lake Whitefish in Kakisa Lake (linear regression, p=0.002,  $R^2=0.211$ , df=42), but not for Lake Whitefish in Tathlina Lake (linear regression, p=0.553,  $R^2=0.006$ , df=63). Because of the significant differences observed, comparisons of  $\delta^{15}N_{adj}$  between lakes were made at a standardized size. While Walleye  $\delta^{15}N_{adj}$  differed significantly between lakes (Table 2.5), this did not help explain the observed differences in [Hg]. Contrary to initial predictions, Walleye had significantly higher  $\delta^{15}N_{adj}$  in Kakisa Lake than in Tathlina Lake, and there was no significant difference in Lake Whitefish  $\delta^{15}N_{adj}$  between lakes (Table 2.5). In general,  $\delta^{15}N_{adj}$  ratios increase ~3-4 ‰ with each trophic transfer (DeNiro & Epstein, 1981), so Walleye in the two lakes are feeding at a similar trophic level. The observed difference (0.5 ‰) in Walleye  $\delta^{15}N_{adj}$  between lakes likely has little ecological relevance and it therefore does not appear that Lake Whitefish and Walleye in Tathlina Lake have higher [Hg] because they occupy a higher trophic position.

Table 2.3. 5	IZC-Stanuar		Vadj OI IC	Jou IIs	n popula	lons, me	luuing 1	ukey spe	st-not test.
Species	Lake	δ <sup>15</sup> Nadj	SE	df	Lower	Upper	R <sup>2</sup>	Tu	key's
	Lake	(‰)	SE	ui	CL	CL	K	t	р
Lake	Kakisa	5.532	0.111	104	5.313	5.751	0.211	0.704	0.4832
Whitefish	Tathlina	5.453	0.108	104	5.239	5.667	0.006	0.704	0.4652
Walleye	Kakisa	8.086	0.099	89	7.888	8.284	0.288	3.141	0.0023*
	Tathlina	7.548	0.150	89	7.250	7.845	0.465	5.141	0.0023

Table 2.5: Size-standardized  $\delta^{15}N_{adj}$  of food fish populations, including Tukey's post-hoc test.

Differences between lakes in fish [Hg] could not be explained by rates of biomagnification, as these were statistically indistinguishable between Tathlina and Kakisa Lakes. As reported above, [Hg] was significantly and positively related to  $\delta^{15}$ N when analyzed across all taxa collected (ANCOVA, p<0.0005; Figures 2.3, 2.4; Tables S2.2, S2.3). The interaction term Lake\* $\delta^{15}$ N was not statistically significant (p=0.427), indicating that rates of biomagnification were similar between lakes and did not explain observed between-lake differences in Walleye and Lake Whitefish [Hg]. The slopes are consistent with Lavoie et al.'s (2013) estimates of biomagnification slopes for THg in freshwater systems. Biomagnification through the fish community alone (Figure 2.4; Table S2.3) also did not explain observed difference in fish [Hg] between lakes (ANCOVA, interaction term p=0.065).

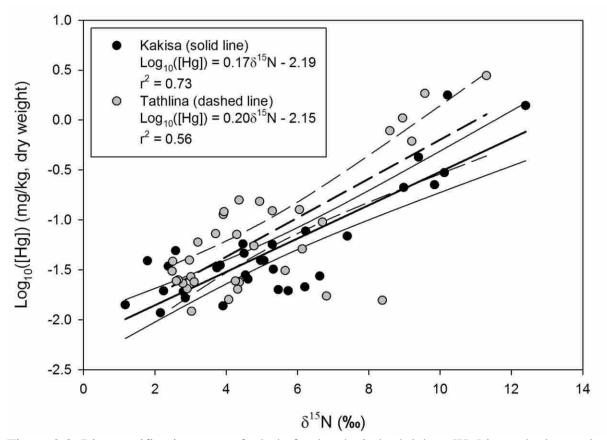


Figure 2.3: Biomagnification rates of whole food webs in both lakes. [Hg] is on the log scale and measured in mg/kg MeHg (dry mass) in invertebrates and mg/kg THg (dry mass) in fish. Thin lines indicate 95% confidence intervals.

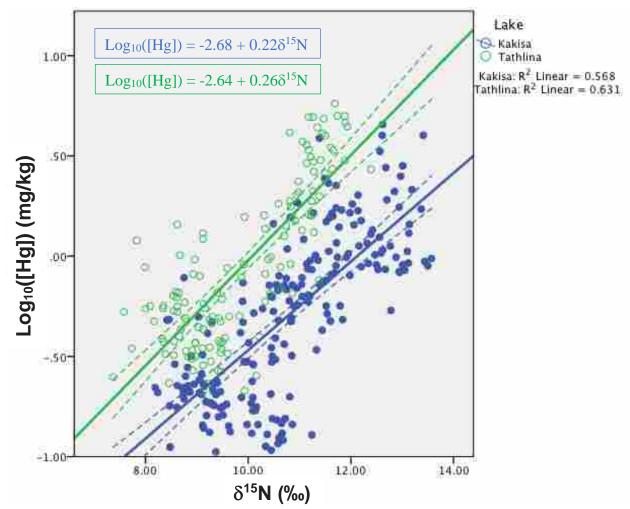


Figure 2.4: Biomagnification rates of fish in both lakes. [Hg] is measured on a log scale in mg/kg THg (dry mass). Dotted lines indicate 95% confidence intervals.

Figure 2.5 depicts a stable isotope biplot, including all sampled taxa. Data points are averaged by Family and functional feeding group in invertebrates, and size-standardized by species in fish. Delta<sup>13</sup>C ratios were more negative in Tathlina Lake than in Kakisa Lake, especially for organisms occupying lower trophic positions. Fishes also had more negative  $\delta^{13}$ C ratios in Tathlina Lake compared to Kakisa Lake (Table 2.6). These data suggest that Tathlina Lake's organic carbon source is more pelagic, while Kakisa Lake has more littoral/terrigenous input (Fry, 2006; Eloranta, 2013). Relative importance of pelagic vs. benthic carbon can have important implications for Hg bioaccumulation, as pelagic food webs tend to have higher [Hg] than benthic food webs (Power et al., 2002; St. Louis et al., 2011; Keva et al., 2017).

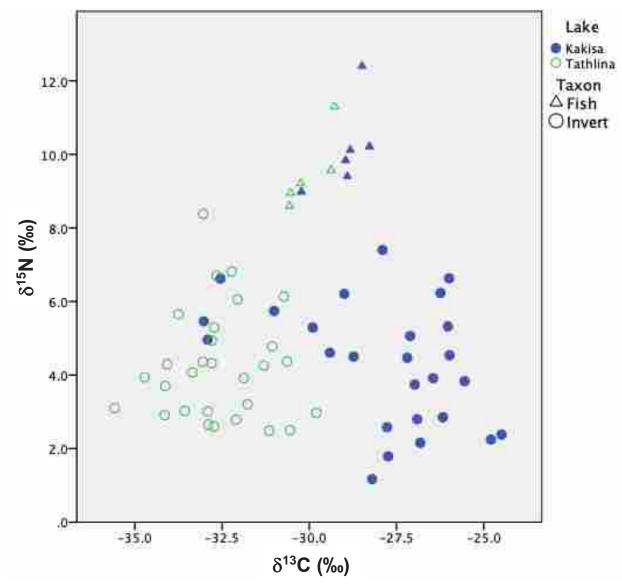


Figure 2.5: Stable isotope biplot of all sampled taxa.

To investigate whether differences in  $\delta^{13}$ C could help explain between-lake differences in [Hg] in Lake Whitefish and Walleye, Log<sub>10</sub>([Hg]) was regressed against  $\delta^{13}$ C for each species. Walleye and Lake Whitefish  $\delta^{13}$ C were significantly and negatively related to Log<sub>10</sub>([Hg])

(linear regression, p<0.0005;  $R^2_{Walleye}=0.179$ ,  $R^2_{Lake Whitefish}=0.142$ ; Table 2.6; Figures 2.6, 2.7), indicating that Walleye and Lake Whitefish that relied on more pelagic carbon sources had higher [Hg] than those relying on more littoral carbon sources (Power et al., 2002; St. Louis et al., 2011; Keva et al., 2017).

Table 2.6:  $\delta^{13}$ C of food fish populations. Walleye values are least-squares means estimates at the standardized fork lengths; Lake Whitefish values are population means. For both species,  $\delta^{13}$ C was significantly less negative in Kakisa Lake than in Tathlina Lake.

Species	Lake	δ <sup>13</sup> C	SE	SE df	Lower	Upper	R <sup>2</sup>	T	ukey's	
	Lake	(‰)	SE	ai	CL	CL	K-	t	р	
Lake	Kakisa	-28.965	0.156	105	-29.274	-28.655	-	6385	< 0.0001*	
Whitefish	Tathlina	-30.254	0.128	105	-30.507	-30.000	-	0303	<0.0001	
Wallowa	Kakisa	-28.667	0.073	88	-28.811	-28.523	0.182	3.502	0.0007*	
Walleye	Tathlina	-29.122	0.108	88	-29.336	-28.908	0.480	5.302	0.0007*	

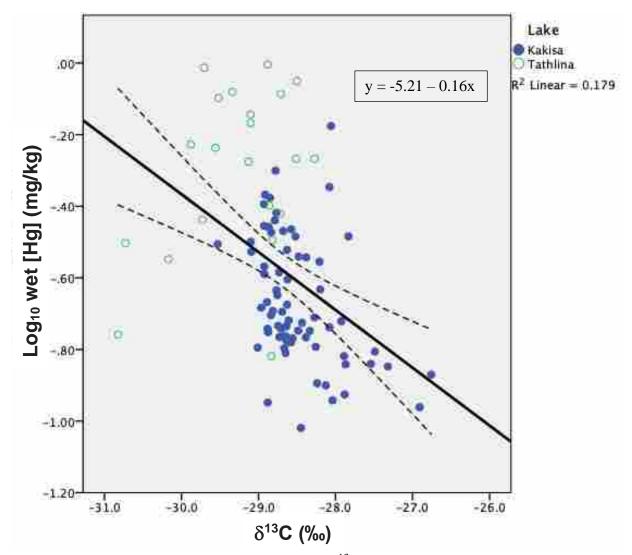


Figure 2.6: Relationship between  $log_{10}([Hg])$  and  $\delta^{13}C$  in Walleye in Tathlina and Kakisa Lakes. Dotted lines indicate 95% confidence intervals.

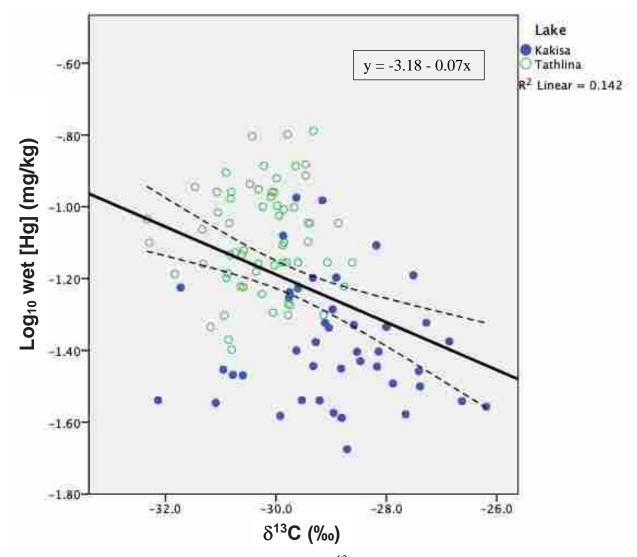


Figure 2.7: Relationship between  $\log_{10}([Hg])$  and  $\delta^{13}C$  in Lake Whitefish in Tathlina and Kakisa Lakes. Dotted lines indicate 95% confidence interval.

Montgomery et al. (2000) demonstrated a negative correlation between [MeHg] and  $\delta^{13}$ C ratios in fine particulate matter (i.e., more negative pelagic  $\delta^{13}$ C signal is correlated to higher [MeHg]), implying that carbon sourcing may play an important role in determining MeHg availability to the food web; similarly, Power et al. (2002) demonstrated a link between more negative  $\delta^{13}$ C ratios and higher fish [Hg]. The findings of Montgomery et al. (2000) and Power et al. (2002) are consistent with those presented here, where Tathlina Lake is associated with

more negative  $\delta^{13}$ C ratios and higher [MeHg] in the food web (Table 2.6; Figures 2.6, 2.7; see also Power et al., 2002; St. Louis et al., 2011; Keva et al., 2017).

In addition to differences in source of carbon between lakes, differences in growth rate may help explain why Walleye in Tathlina Lake have higher [Hg] than in Kakisa Lake. Walleye grow significantly more slowly in Tathlina Lake than in Kakisa Lake (Tukey's test, p<0.01; Table 2.7). No difference in growth rates was found between Lake Whitefish populations, although this was likely due, at least in part, to low power in the model ( $\pi$  =0.328; Table 2.7). Lake Whitefish and Walleye both may be subject to system-level drivers such as MeHg availability and bloom dilution, but the larger between-lake difference in Walleye [Hg] may be additionally driven by growth rates. Higher [Hg] in Lake Whitefish in Tathlina Lake cannot be explained by slower growth rates but may be explained by the lake-level factors discussed earlier, including abiotic MeHg availability and bloom dilution. Growth models typically demand larger sample sizes than those available for this work, and additional data are necessary to make a stronger conclusion about between-lake variability in growth rates and associated impacts on fish [Hg].

Spacing	Laka	Age	SE	df	Lower	Upper	R <sup>2</sup>	Tukey's	
Species	Lake	( <b>yr</b> )	SE	aı	CL	Upper CL	K-	t	р
Lake	Kakisa	12.499	1.778	52	9.396	16.628	0.682	-0.773	0 443
Whitefish	Tathlina	14.429	1.723	52	11.354	18.337	0.715	-0.775	0.445
Wallowa	Kakisa	10.397	0.398	65	9.631	11.224	0.744	-3.036	0.0034*
Walleye	Tathlina	14.605	1.53665	65	11.838	18.017	0.882	-3.030	0.0054*

Table 2.7: Age-at-size estimates of food fish populations, including results of a Tukey's post-hoc test.

Growth efficiency can be affected by a number of biotic factors, including nutrient absorption efficiency, prey nutritional quality, and inter- or intra-specific competition, or abiotic factors such as durations of seasonal ice-over or anoxia (Verta, 1990; Karimi et al., 2007; Ward et al., 2010; Stewart et al., 2016). Tathlina Lake's winter anoxia and long ice-over season (Stewart et al., 2016) may be causing more stress on its fish populations and thereby reducing growth rates.

While high Hg exposure can cause developmental impairment in humans, there is little evidence of growth impairment in fish as a result of Hg exposure. Male Walleye have been demonstrated to experience a small but significant reduction in growth after six months on a consistent diet containing 1 mg/kg MeHg; Walleye from lower exposure treatments and female Walleye from the same and lower exposure treatments showed no difference in growth (Friedmann et al., 1996). Because none of the biota sampled in this work reached comparable [Hg], growth impairment due to Hg exposure is assumed not to be an important factor in these analyses.

### **2.5 Summary of Main Findings**

While all of the Hg drivers explored in this work have been well presented and supported in existing literature, there has been considerably less work in assessing many drivers simultaneously in the same lake.

- At a standardized size, [Hg] in Walleye and Lake Whitefish were significantly higher in Tathlina Lake than in Kakisa Lake. There was no significant difference between lakes in size-standardized [Hg] in Northern Pike.
- Methylmercury is more concentrated in the sediment and water of Tathlina Lake compared to Kakisa Lake. Net methylation rate, as indicated by %MeHg of THg, also appears to be higher in Tathlina Lake than in Kakisa Lake.

- 3. Primary production, as indicated by water chlorophyll *a* concentrations, is higher in Kakisa Lake than in Tathlina Lake. This may contribute to a bloom dilution effect that effectively lowers [Hg] at the base of the food chain in Kakisa Lake. While many previous research efforts have demonstrated this mechanism under experimental conditions (Pickhardt et al., 2002; Hill & Larsen, 2005), this work provides some support for demonstrating the principle *in situ*.
- 4. Food web-wide trophic biomagnification slopes are statistically indistinguishable between the two lakes. The slopes are consistent with the results of previous studies (Lavoie et al., 2013). Tathlina Lake consistently has higher mercury concentrations than Kakisa Lake at any given trophic position, including the slopes' intercepts, indicating that Tathlina Lake has higher [Hg] at the base of the food web (Lavoie et al., 2013).
- Differences in fish trophic position between lakes did not explain differences in [Hg];
   Walleye and Lake Whitefish had similar trophic positions in each lake.
- 6. Growth rates in Lake Whitefish were similar between lakes, whereas Walleye grew more slowly in Tathlina Lake than in Kakisa Lake. Therefore, lower [Hg] in Walleye in Kakisa Lake may partially reflect faster fish growth rates and therefore a growth dilution effect. These findings are consistent with many previous studies (Verta, 1990; Karimi et al., 2007; Ward et al., 2010).
- 7. [Hg] in Northern Pike did not differ significantly between the two lakes, and the reasons for this deserve further study.
- 8. Subsistence fishing continues to occur in both Kakisa Lake and Tathlina Lake; current commercial fishing efforts are limited to Kakisa Lake. Larger Walleye and Northern Pike are at highest risk of excessive Hg contamination, while Lake Whitefish have

consistently low [Hg]. While it is outside the scope of this work to propose consumption

advisories, these general trends will be important for assessing exposure risk and

generating community-specific consumption guidelines.

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### **2.7 Supplementary Material**

Table S2.1: Size-standardized [THg] of food fish populations, including Tukey's post-hoc test.

Species	Lake	[THg]	SE	df	Lower	Upper	Tu	ıkey's	
species	Lake	(mg/kg) SE		ul	CL	CL	t	р	
Lake	Kakisa	0.068	0.004	155	0.061	0.076	-7.413	<0.0001*	
Whitefish	Tathlina	0.113	0.008	155	0.099	0.129	-7.415	<0.0001	
Northern	Kakisa	0.356	0.033	81	0.296	0.428	0 272	0.7100	
Pike	Tathlina	0.372	0.025	81	0.325	0.425	-0.372	0.7109	
Wallowo	Kakisa	0.279	0.013	89	0.255	0.306	0 715	<0.0001*	
Walleye	Tathlina	0.558	0.039	89	0.486	0.640	-0./13	<0.0001	
	Tathlina Kakisa	0.372         0.025           0.279         0.013		81 89	0.325 0.255	0.425	-0.372 -8.715	0.7109 <0.0001*	

Table S2.2: Model output of biomagnification in whole food webs (ANCOVA, R<sup>2</sup>=0.634).

Source	Type III SS	df	F	Sig.
<b>Corrected Model</b>	14.036	2	53.688	< 0.0005
Intercept	59.501	1	455.179	< 0.0005
Lake	0.552	1	4.226	0.044
δ <sup>15</sup> N	13.798	1	105.557	< 0.0005
Error	8.105	62		
Total	119.776	65		
<b>Corrected Total</b>	22.141	64		

Source	<b>Type III SS</b>	df	F	Sig.
<b>Corrected Model</b>	37.160	3	189.380	< 0.0005
Intercept	34.763	1	531.502	< 0.0005
Lake	0.001	1	0.018	0.893
$\delta^{15}N$	30.169	1	461.261	0.000
Lake*δ <sup>15</sup> N	0.225	1	3.434	0.065
Error	22.303	341		
Total	73.473	345		
<b>Corrected Total</b>	59.463	344		

Table S2.3: Model output of biomagnification in food fish (ANCOVA,  $R^2=0.625$ ).

Table S2.4: Size-standardized  $\delta^{13}$ C of Northern Pike.  $\delta^{13}$ C was significantly less negative in Kakisa Lake than in Tathlina Lake.

Laka	δ <sup>13</sup> C	SE	df	Lower	Upper	<b>R</b> <sup>2</sup>	Tu	key's
Lake	(‰)	SE	ui	CL	CL	K	t	р
Kakisa	-27.913	0.082	82	-28.077	-27.749	0.418	13.142	<0.0001*
Tathlina	-29.286	0.072	82	-29.428	-29.144	0.494	15.142	<0.0001*

The model of Northern Pike Log<sub>10</sub>([Hg]) as a function of  $\delta^{13}$ C was not significant (linear regression, p=0.202,  $R^2=0.02$ ). General linear models show a positive relationship between  $\delta^{15}$ N<sub>adi</sub> and Log<sub>10</sub>(fork length) for Northern Pike in both lakes (linear regression, p<sub>Tathlina</sub><0.0005,  $R^{2}_{Tathlina}=0.668$ ,  $p_{Kakisa}<0.0005$ ,  $R^{2}_{Kakisa}=0.469$ ); that is, trophic position increased significantly with fish size, and thus comparisons between lakes were made at a standardized size (650 mm). While the difference between  $\delta^{15}N_{adj}$  was significant (Table S2.5), there is likely not an ecologically relevant difference in trophic level, which is consistent with size-standardized [Hg] results (Figure 2.2).

 $\delta^{15}N_{adj}$ Lower Upper Tukey's **R**<sup>2</sup> Lake SE df CL CL (‰) t р Kakisa 7.561 0.107 7.349 7.774 82 0.469 0.0120\* 2.570 Tathlina 7.213 0.093 82 7.029 7.398 0.668

Table S2.5:  $\delta^{15}N_{adj}$  of Northern Pike, size-standardized to 650 mm fork length.

Northern Pike grow significantly more slowly in Tathlina Lake than in Kakisa Lake (Tukey's test, p<0.01; Table S2.6). While a slower growth likely does contribute to higher [Hg] in Tathlina Lake Walleye, slower growth in Northern Pike in Tathlina Lake evidently does not result in higher [Hg], though species-specific factors such as digestive efficiency or ontogenetic feeding behavior are likely to confound a direct comparison between Walleye and Northern Pike.

Tukey's Age Lower Upper **R**<sup>2</sup> SE df Lake (yr) CL CL t р 5.027 Kakisa 5.817 0.421 44 6.732 0.872 -2.762 0.0083\* Tathlina 7.409 0.364 44 6.710 8.181 0.825

Table S2.6: Age-at-size of Northern Pike, estimated at 650 mm fork length.

	-	Jaka		Fork Length	A	Wet	[THg] mg/kg	[THg] ppm	5420	54 F NI	C:N		
ID 10194	Species NRPK	Lake Tathlina	Year 2014	(mm) 455	Age	Weight (g) 585	(dry wt.)	(wet wt.)	δ13C	δ15Ν	Ratio	sex	maturity
							1.990	0.40	-29.56	11.39	3.18776		
10195	NRPK	Tathlina	2014	563		360	0.595	0.12	-29.69	10.33	3.21332		
10196	NRPK	Tathlina	2014	366		380	0.652	0.13	-29.46	10.86	3.15529		
10197	NRPK	Tathlina	2014	252		105	0.881	0.18	-30.94	7.99	3.15901		
10198	NRPK	Tathlina	2014	745		3245	1.610	0.32	-28.89	11.18	3.07897		
10199	LKWH	Tathlina	2014	364		747	0.473	0.09	-29.38	8.17	3.30592		
10200	NRPK	Tathlina	2014	550		1055	1.550	0.31	-29.47	10.86	3.13995		
10201	LKWH	Tathlina	2014	284		335	0.233	0.05	-29.14	8.46	3.10391		
10202	LKWH	Tathlina	2014	324		542	0.321	0.06	-29.59	8.55	3.45680		
10203	NRPK	Tathlina	2014	899		6075	4.640	0.93	-28.48	11.89	3.25569		
10204	NRPK	Tathlina	2014	872		5430	2.600	0.59	-28.94	11.50	3.07946		
10205	NRPK	Tathlina	2014	630		1695	1.260	0.25	-29.03	10.70	3.04801		
10206	NRPK	Tathlina	2014	835		4750	4.380	0.88	-28.66	11.35	3.10562		
10207	NRPK	Tathlina	2014	895		5965	4.940	0.99	-28.79	11.18	3.08879		
10208	NRPK	Tathlina	2014	680		1900	2.080	0.42	-28.72	10.98	3.02739		
10209	NRPK	Tathlina	2014	625		1820	1.370	0.27	-29.69	10.33	3.04355		
10210	WALL	Tathlina	2014	310		200	2.910	0.58	-29.56	11.14	3.21027		
10211	NRPK	Tathlina	2014	412		480	1.500	0.30	-29.52	10.92	3.03974		
10212	NRPK	Tathlina	2014	315		190	0.659	0.13	-30.20	9.40	3.09696		
10213	NRPK	Tathlina	2014	393		475	0.909	0.18	-29.79	9.92	3.07549		
10214	NRPK	Tathlina	2014	384		380	0.692	0.10	-29.78	10.64	3.01444		
10215	NRPK	Tathlina	2014	475		765	1.600	0.14	-29.81	10.31	2.94067		
10216	LKWH	Tathlina	2014	392		964							
10210	LKWH	Tathlina	2014	365		810	0.388	0.08	-29.42	8.32	3.35142		
10217	LKWH	Tathlina	2014	390		1025	0.527	0.11	-31.07	7.58	3.45241		
10210		ratinid	2014	590		1025	0.551	0.11	-30.03	8.34	3.17199		

Table S2.7: Fish data used for this work. NRPK=Northern Pike; LKWH=Lake Whitefish; WALL=Walleye; WHSC=White Sucker; LNSC=Longnose Sucker; CISC=Cisco.

ID	Species	Lake	Year	Fork Length (mm)	Age	Wet Weight (g)	[THg] mg/kg (dry wt.)	[THg] ppm (wet wt.)	δ13C	δ15N	C : N Ratio	sex	maturity
10219	LKWH	Tathlina	2014	360		745	0.315	0.06	-30.64	8.83	3.45504		
10220	LKWH	Tathlina	2014	309		440	0.213	0.04	-30.80	9.12	3.22359		
10221	LKWH	Tathlina	2014	340		650	0.371	0.07	-29.88	8.97	3.16584		
10222	LKWH	Tathlina	2014	365		655	0.351	0.07	-29.81	9.25	3.32911		
10223	LKWH	Tathlina	2014	335		585	0.432	0.09	-28.87	8.69	3.23630		
10224	LKWH	Tathlina	2014	375		832	0.374	0.07	-29.60	9.50	3.23140		
10225	LKWH	Tathlina	2014	353		720	0.635	0.13	-29.64	8.54	3.21703		
10226	LKWH	Tathlina	2014	374		847	0.491	0.10	-30.24	8.78	3.44602		
10227	LKWH	Tathlina	2014	382		739	0.471	0.09	-30.85	8.79	3.84503		
10228	LKWH	Tathlina	2014	362		825	0.442	0.09	-29.42	9.04	3.64356		
10229	LKWH	Tathlina	2014	325		598	0.269	0.05	-29.78	8.92	3.24112		
10230	LKWH	Tathlina	2014	326		571	0.308	0.06	-28.76	8.90	3.25065		
10231	LKWH	Tathlina	2014	400		858	0.588	0.12	-29.99	8.76	3.25858		
10232	LKWH	Tathlina	2014	355		714	0.339	0.07	-29.07	8.91	3.44557		
10233	WALL	Tathlina	2014	420		915	1.600	0.32	-28.82	11.23	3.18324		
10234	WALL	Tathlina	2014	536		1591	4.950	0.99	-28.88	11.26	3.15495		
10235	WALL	Tathlina	2014	445		975	3.380	0.68	-29.10	11.18	3.11179		
10236	WALL	Tathlina	2014	523		1380	2.710	0.54	-28.27	12.39	3.11910		
10237	WALL	Tathlina	2014	476		1079	1.880	0.38	-28.72	11.24	3.13690		
10238	WALL	Tathlina	2014	430		748	4.440	0.89	-28.50	11.93	3.08776		
10239	WALL	Tathlina	2014	535		1550	4.090	0.82	-28.71	11.45	3.19083		
10240	WALL	Tathlina	2014	478		1021	4.140	0.83	-29.34	10.81	3.13656		
10241	WALL	Tathlina	2014	490		1255	2.640	0.53	-29.13	11.28	3.10849		
10242	WALL	Tathlina	2014	409		755	2.020	0.40	-28.86	10.94	3.13159		
10243	WALL	Tathlina	2014	520		1575	2.720	0.54	-28.51	11.64	3.08607		
10244	WHSC	Tathlina	2014	475		1945	1.220	0.24	-31.12	9.12	3.49905		
10245	WHSC	Tathlina	2014	512		2380	1.200	0.24	-32.01	7.83	4.05976		

ID	Species	Lake	Year	Fork Length (mm)	Age	Wet Weight (g)	[THg] mg/kg (dry wt.)	[THg] ppm (wet wt.)	δ13C	δ15N	C : N Ratio	sex	maturity
10246	WHSC	Tathlina	2014	440		1360	0.548	0.11	-31.10	8.00	3.23505		
10247	WHSC	Tathlina	2014	460		1565	1.440	0.29	-29.07	8.62	3.18959		
10248	WHSC	Tathlina	2014	492		1875	1.010	0.20	-29.57	8.67	3.08780		
10249	LKWH	Tathlina	2014	440		1363	0.663	0.13	-30.22	8.59	3.17619		
10250	NRPK	Tathlina	2014	275		155	0.500	0.10	-29.07	9.63	3.02169		
10251	LKWH	Tathlina	2014	493		2130	0.564	0.11	-30.81	8.09	4.13636		
10252	LNSC	Tathlina	2014	423		1000	0.963	0.19	-31.44	8.96	3.25105		
10253	LKWH	Tathlina	2014	330		523	0.336	0.07	-28.62	8.79	3.04318		
10433	LKWH	Kakisa	2015	359		675	0.211	0.04	-26.86	9.46	3.45444		
10434	LKWH	Kakisa	2015	401		910	0.186	0.04	-28.47	9.26	4.02975		
10435	LKWH	Kakisa	2015	457		1575	0.415	0.08	-29.87	9.85	4.44690		
10436	LKWH	Kakisa	2015	414		1160	0.322	0.06	-27.51	9.11	3.89969		
10437	CISC	Kakisa	2015	139		31	0.226	0.05	-30.62	8.62	3.52178		
10438	CISC	Kakisa	2015	151		39	0.225	0.05	-30.12	9.14	3.42069		
10439	CISC	Kakisa	2015	152		40	0.208	0.04	-30.04	9.05	3.33641		
10440	CISC	Kakisa	2015	140		36	0.231	0.05	-30.18	9.13	3.41399		
10441	CISC	Kakisa	2015	138		30	0.226	0.05	-30.52	8.83	3.51393		
10442	CISC	Kakisa	2015	150		38	0.205	0.04	-30.36	9.38	3.55978		
10443	CISC	Kakisa	2015	147		36	0.209	0.04	-30.18	8.24	3.35372		
10444	WHSC	Kakisa	2015	505		1905	0.483	0.10	-28.05	8.46	3.48411		
10445	LNSC	Kakisa	2015	495		1810	0.471	0.09	-29.36	9.29	3.39831		
10446	LNSC	Kakisa	2015	450		1255	0.480	0.10	-28.86	8.42	3.50660		
10447	NRPK	Kakisa	2015	452		640	0.482	0.10	-28.17	11.84	3.30849		
10448	NRPK	Kakisa	2015	541		1160	1.450	0.29	-28.52	10.49	3.36337		
10449	NRPK	Kakisa	2015	506		950	0.630	0.13	-28.05	11.20	3.29650		
10450	NRPK	Kakisa	2015	490		865	0.599	0.12	-28.12	10.80	3.28176		
10451	NRPK	Kakisa	2015	519		990	0.633	0.13	-28.15	10.44	3.33752		

ID	Species	Lake	Year	Fork Length (mm)	Age	Wet Weight (g)	[THg] mg/kg (dry wt.)	[THg] ppm (wet wt.)	δ13C	δ15N	C : N Ratio	sex	maturity
10452	CISC	Kakisa	2015	142		33	0.194	0.04	-30.27	8.75	3.63025		
10453	LKWH	Kakisa	2015	420		1080	0.170	0.03	-30.78	10.14	4.39815		
10454	LKWH	Kakisa	2015	407		910	0.142	0.03	-31.09	9.62	4.63743		
10455	LNSC	Kakisa	2015	471		1615	0.781	0.16	-29.00	8.75	3.51818		
10456	WHSC	Kakisa	2015	486		1665	0.737	0.15	-28.81	9.90	3.64555		
10457	WHSC	Kakisa	2015	484		1725	0.499	0.10	-28.49	9.03	3.44770		
10458	WHSC	Kakisa	2015	490		1680	0.468	0.09	-28.84	9.90	3.61586		
10459	WHSC	Kakisa	2015	448		1420	0.185	0.04	-27.78	9.26	3.40406		
10460	WALL	Kakisa	2015	305		286	0.710	0.14	-27.32	10.56	3.30954		
10461	WALL	Kakisa	2015	328		365	0.547	0.11	-26.91	10.28	3.30986		
10462	WALL	Kakisa	2015	361		470	0.759	0.15	-27.89	11.52	3.28062		
10463	WALL	Kakisa	2015	478		1190	3.333	0.67	-28.06	12.57	3.32232		
10464	WALL	Kakisa	2015	485		1200	1.434	0.29	-28.38	12.06	3.33338		
10465	NRPK	Kakisa	2015	552		1360	0.626	0.13	-26.34	10.47	3.33432		
10466	NRPK	Kakisa	2015	728		2465	4.532	0.91	-27.90	12.62	3.24607		
10467	CISC	Kakisa	2015	146		36	0.215	0.04	-30.15	8.73	3.33083		
10468	NRPK	Kakisa	2015	564		1155	1.562	0.31	-28.42	10.82	3.31472		
10469	LKWH	Kakisa	2015	399		940	0.198	0.04	-28.14	9.87	3.96096		
10470	LKWH	Kakisa	2015	401		990	0.317	0.06	-29.33	10.47	3.50507		
10471	CISC	Kakisa	2015	139		33	0.211	0.04	-30.08	8.86	3.36005		
10472	CISC	Kakisa	2015	152		41	0.187	0.04	-30.11	9.18	3.48732		
10473	CISC	Kakisa	2015	141		33	0.205	0.04	-29.93	9.09	3.26124		
10474	NRPK	Kakisa	2015	513		865	1.845	0.37	-28.04	10.96	3.26502		
10475	NRPK	Kakisa	2015	486		870	1.213	0.24	-28.26	11.08	3.30560		
10476	CISC	Kakisa	2015	148		40	0.203	0.04	-30.69	8.73	3.59976		
10477	LKWH	Kakisa	2015	384		910	0.131	0.03	-29.92	10.07	4.08672		
10478	LKWH	Kakisa	2015	383		820	0.106	0.02	-28.71	9.37	4.19455		

ID	Species	Lake	Year	Fork Length (mm)	Age	Wet Weight (g)	[THg] mg/kg (dry wt.)	[THg] ppm (wet wt.)	δ13C	δ15N	C : N Ratio	sex	maturity
10479	LKWH	Kakisa	2015	379		835	0.158	0.03	-27.39	9.42	3.52972		
10480	LKWH	Kakisa	2015	410		1055	0.234	0.05	-28.59	9.38	3.73096		
10481	LKWH	Kakisa	2015	366		710	0.129	0.03	-28.81	10.37	3.66054		
10482	WHSC	Kakisa	2015	508		2135	0.595	0.12	-28.26	9.89	3.36029		
10483	WHSC	Kakisa	2015	492		1775	0.719	0.14	-29.92	10.55	3.73908		
10484	NRPK	Kakisa	2015	205		63	0.178	0.04	-29.31	9.50	3.27080		
10485	NRPK	Kakisa	2015	227		90	0.160	0.03	-28.82	9.19	3.22894		
10486	NRPK	Kakisa	2015	706		2270	2.436	0.49	-27.56	11.49	3.21123		
10487	WALL	Kakisa	2015	356		510	0.563	0.11	-28.88	10.89	3.25629		
10488	WALL	Kakisa	2015	439		955	1.639	0.33	-28.52	11.63	3.26050		
10489	WALL	Kakisa	2015	448		1040	2.252	0.45	-28.08	11.71	3.24315		
10490	CISC	Kakisa	2015	140		30	0.216	0.04	-30.36	8.84	3.47775		
10491	LNSC	Kakisa	2015	168		62	0.112	0.02	-26.65	8.48	3.37507		
10492	LKWH	Kakisa	2015	401		1000	0.144	0.03	-29.21	10.19	3.79529		
10493	LKWH	Kakisa	2015	411		1120	0.145	0.03	-32.14	9.98	8.22583		
10494	LKWH	Kakisa	2015	369		780	0.139	0.03	-26.19	9.13	3.84213		
10495	NRPK	Kakisa	2015	526		1080	0.714	0.14	-28.06	10.70	3.28722		
10496	NRPK	Kakisa	2015	348		310	0.182	0.04	-28.68	9.30	3.23613		
10497	WALL	Kakisa	2015	444		970	1.698	0.34	-28.68	11.82	3.20959		
10498	WALL	Kakisa	2015	480		1045	1.394	0.28	-28.21	11.77	3.21666		
10499	WALL	Kakisa	2015	372		635	0.720	0.14	-27.87	11.39	3.23913		
10500	LNSC	Kakisa	2015	390		855	0.252	0.05	-29.31	8.78	3.61674		
10501	LNSC	Kakisa	2015	448		1330	0.288	0.06	-29.92	10.10	3.44574		
10503	LNSC	Kakisa	2015	477		1445	0.412	0.08	-29.12	9.21	3.47077		
10504	LKWH	Kakisa	2015	310		415	0.132	0.03	-27.65	9.25	3.34608		
10505	LKWH	Kakisa	2015	305		400	0.144	0.03	-26.63	9.11	3.35859		
10506	LKWH	Kakisa	2015	410		1050	0.390	0.08	-28.18	10.53	3.51701		

ID	Species	Lake	Year	Fork Length (mm)	Age	Wet Weight (g)	[THg] mg/kg (dry wt.)	[THg] ppm (wet wt.)	δ13C	δ15N	C : N Ratio	sex	maturity
10507	LKWH	Kakisa	2015	395		870	0.174	0.03	-27.41	9.47	3.74599		
10508	LKWH	Kakisa	2015	426		1355	0.177	0.04	-28.82	10.16	4.58474		
10509	LKWH	Kakisa	2015	375		880	0.145	0.03	-29.53	10.86	4.58977		
10510	LKWH	Kakisa	2015	381		885	0.133	0.03	-28.96	9.65	3.57615		
10511	LKWH	Kakisa	2015	425		1360	0.231	0.05	-28.00	9.33	4.11675		
10512	LKWH	Kakisa	2015	424		1065	0.197	0.04	-28.53	10.09	5.51183		
10513	LKWH	Kakisa	2015	408		1035	0.170	0.03	-30.60	10.09	3.85484		
10514	LKWH	Kakisa	2015	398		990	0.199	0.04	-29.63	10.81	3.80439		
10515	CISC	Kakisa	2015	147		34	0.183	0.04	-29.85	9.58	3.22684		
10516	CISC	Kakisa	2015	142		33	0.242	0.05	-30.32	9.05	3.39747		
10517	CISC	Kakisa	2015	151		39	0.218	0.04	-30.25	9.31	3.39732		
10518	CISC	Kakisa	2015	142		35	0.205	0.04	-30.00	9.36	3.20516		
10519	LKWH	Kakisa	2015	150		36	0.161	0.03	-27.88	9.51	3.19497		
10520	CISC	Kakisa	2015	135		32	0.222	0.04	-30.33	8.74	3.35431		
10521	NRPK	Kakisa	2015	554		1045	0.717	0.14	-28.23	10.86	3.17869		
10522	NRPK	Kakisa	2015	625		1455	2.763	0.55	-28.41	12.68	3.16756		
10523	NRPK	Kakisa	2015	689		2305	2.819	0.56	-28.14	13.01	3.18062		
10725	WALL	Kakisa	2017	409	13	805	1.560	0.31	-29.53	11.59	3.18655	Μ	Μ
10726	WALL	Kakisa	2017	435	11	860	1.503	0.30	-28.63	12.20	3.20987	F	Μ
10727	WALL	Kakisa	2017	445	8	910	0.987	0.20	-28.84	12.88	3.19864	F	Μ
10728	WALL	Kakisa	2017	420	8	720	1.015	0.20	-28.81	12.47	3.20485	F	Μ
10729	WALL	Kakisa	2017	425	13	890	2.143	0.43	-28.91	13.19	3.22661	Μ	Μ
10730	WALL	Kakisa	2017	405	10	720	1.742	0.35	-28.87	11.71	3.16191	Μ	IM
10731	WALL	Kakisa	2017	425	9	780	1.287	0.26	-28.92	12.39	3.15500	Μ	Μ
10732	WALL	Kakisa	2017	441	12	900	1.755	0.35	-28.93	12.94	3.20251	Μ	Μ
10733	WALL	Kakisa	2017	405	8	720	1.057	0.21	-28.63	13.10	3.19782	Μ	Μ
10734	WALL	Kakisa	2017	420	9	730	1.351	0.27	-28.93	12.42	3.17276	Μ	Μ

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10735	WALL	Kakisa	2017	450	11	800	1.486	0.30	-29.09	11.52	3.16692	F	Μ
10736	WALL	Kakisa	2017	416	6	695	0.905	0.18	-28.88	12.50	3.19392	М	М
10737	WALL	Kakisa	2017	365	7	475	0.905	0.18	-28.71	13.01	3.18045	F	IM
10738	WALL	Kakisa	2017	420	5	820	0.864	0.17	-28.65	11.16	3.20027	F	IM
10739	WALL	Kakisa	2017	420	9	760	1.074	0.21	-28.89	12.62	3.22047	М	М
10740	WALL	Kakisa	2017	406	7	760	0.858	0.17	-28.73	12.53	3.16973	Μ	М
10741	NRPK	Kakisa	2017	300	1	180	0.364	0.07	-28.69	10.70	3.21141	F	IM
10742	WHSC	Kakisa	2017	375	3		0.113	0.02	-28.75	10.34	3.21460	F	М
10743	WHSC	Kakisa	2017	460	4	1520	0.152	0.03	-29.01	10.56	3.21865	F	М
10744	WHSC	Kakisa	2017	470	12	1500	0.682	0.14	-29.69	10.74	3.23269	Μ	М
10745	WHSC	Kakisa	2017	445	10	1400	0.349	0.07	-28.60	10.45	3.27507	М	М
10746	WHSC	Kakisa	2017	420	5	110	0.147	0.03	-28.77	10.65	3.14951	F	М
10747	WHSC	Kakisa	2017	418	5	1120	0.205	0.04	-29.16	10.44	3.25029	М	М
10748	WHSC	Kakisa	2017	355	3	730	0.108	0.02	-29.13	10.45	3.23685	М	IM
10749	WHSC	Kakisa	2017	350	3	690	0.124	0.02	-29.30	10.60	3.16739	М	IM
10750	WHSC	Kakisa	2017	360	3	720	0.121	0.02	-28.69	10.68	3.18972	U	IM
10751	WHSC	Kakisa	2017	365	3	720	0.116	0.02	-29.03	10.58	3.19162	U	IM
10752	LKWH	Kakisa	2017	440	21		0.521	0.10	-29.16	10.11	3.27655	F	М
10753	LNSC	Kakisa	2017	445	17		0.475	0.09	-29.05	11.60	3.39925	F	М
10754	CISC	Kakisa	2017										
10755	CISC	Kakisa	2017										
10756	CISC	Kakisa	2017										
10757	CISC	Kakisa	2017										
10758	WALL	Kakisa	2017	432	14	800	1.818	0.36	-28.79	12.03	3.13886	Μ	М
10759	WALL	Kakisa	2017	278	4	230	0.571	0.11	-28.04	10.96	3.16345	М	IM
10760	WALL	Kakisa	2017	335	8	405	0.956	0.19	-28.61	11.36	3.11689	М	IM
10761	WALL	Kakisa	2017	392	8	640	0.857	0.17	-28.39	11.24	3.12145	Μ	М

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10762	WALL	Kakisa	2017	255	3	170	0.798	0.16	-28.67	10.60	3.11884	М	IM
10763	WALL	Kakisa	2017	332	4	370	0.674	0.13	-26.76	10.58	3.12759	М	IM
10764	WALL	Kakisa	2017	355	7	460	1.010	0.20	-28.69	11.31	3.14362	F	IM
10765	WALL	Kakisa	2017	432	13	925	1.908	0.38	-28.77	12.70	3.18335	М	М
10766	WALL	Kakisa	2017	416	12	750	1.716	0.34	-28.57	13.37	3.15130	М	М
10767	WALL	Kakisa	2017	432	10	860	1.166	0.23	-28.20	11.72	3.16524	F	М
10768	WALL	Kakisa	2017	424	7	870	0.895	0.18	-28.48	11.71	3.16056	F	М
10769	WALL	Kakisa	2017	411	5	790	0.806	0.16	-28.26	11.62	3.15843	F	М
10770	WALL	Kakisa	2017	440	9	925	1.124	0.22	-28.76	12.77	3.12738	F	М
10771	WALL	Kakisa	2017	428	9	775	1.244	0.25	-28.62	12.56	3.11453	М	М
10772	WALL	Kakisa	2017	445	13	1010	2.503	0.50	-28.78	12.97	3.16794	М	М
10773	WALL	Kakisa	2017	416	10	825	1.037	0.21	-28.96	12.59	3.13124	F	М
10774	WALL	Kakisa	2017	440	10	830	1.584	0.32	-29.10	11.65	3.12182	F	М
10775	WALL	Kakisa	2017	411	9	780	0.886	0.18	-28.88	12.20	3.15162	М	М
10776	WALL	Kakisa	2017	400	9	785	0.830	0.17	-28.58	12.95	3.15174	М	М
10777	WALL	Kakisa	2017	394	7	640	0.852	0.17	-28.56	13.05	3.13852	М	М
10778	WALL	Kakisa	2017	365	7	635	0.974	0.19	-28.27	13.57	3.17938	F	М
10779	WALL	Kakisa	2017	430	8	960	0.924	0.18	-28.74	13.12	3.17530	F	М
10780	WALL	Kakisa	2017	410	8	760	1.440	0.29	-28.48	13.14	3.14125	М	М
10781	WALL	Kakisa	2017	410	12	815	2.101	0.42	-28.85	12.85	3.13786	М	М
10782	WALL	Kakisa	2017	415	9	875	0.920	0.18	-28.64	12.34	3.19907	F	М
10783	WALL	Kakisa	2017	495	12	1205	2.017	0.40	-28.93	12.14	3.10359	F	М
10784	WALL	Kakisa	2017	442	10	890	1.159	0.23	-28.77	12.32	3.13007	F	М
10785	WALL	Kakisa	2017	435	10	845	1.301	0.26	-28.73	13.13	3.12026	F	М
10786	WALL	Kakisa	2017	342	7	445	0.941	0.19	-28.44	13.44	3.14729	М	М
10787	WALL	Kakisa	2017	456	9	910	1.683	0.34	-28.84	12.14	3.18255	F	М
10788	WALL	Kakisa	2017	409	9	790	0.859	0.17	-28.65	12.67	3.13429	F	М

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10789	WALL	Kakisa	2017	390	7	690	0.893	0.18	-28.33	13.50	3.14934	F	Μ
10790	WALL	Kakisa	2017	374	7	560	0.838	0.17	-28.64	12.94	3.16149	F	М
10791	WALL	Kakisa	2017	405	13	680	1.640	0.33	-27.83	11.27	3.13204	F	IM
10792	NRPK	Kakisa	2017	755	7	2825	4.006	0.80	-27.55	13.41	3.18345	F	М
10793	NRPK	Kakisa	2017	885	10	5915	4.001	0.80	-27.11	12.61	3.18577	F	Μ
10794	NRPK	Kakisa	2017	515	3	770	2.318	0.46	-28.18	11.56	3.14571	М	Μ
10795	NRPK	Kakisa	2017	255	1	140	0.341	0.07	-28.49	11.33	3.13968	М	IM
10796	NRPK	Kakisa	2017	570	6	1265	1.057	0.21	-28.03	11.57	3.16212	М	Μ
10797	NRPK	Kakisa	2017	540	5	910	1.644	0.33	-28.13	11.98	3.16752	F	Μ
10798	NRPK	Kakisa	2017	435	2	530	0.743	0.15	-28.00	11.47	3.17904	М	Μ
10799	NRPK	Kakisa	2017	535	4	860	1.027	0.21	-28.16	11.95	3.16939	F	Μ
10800	NRPK	Kakisa	2017	490	3	840	0.565	0.11	-28.82	11.79	3.20815	F	Μ
10801	NRPK	Kakisa	2017	545	6	1025	3.876	0.78	-28.39	11.39	3.18979	F	М
10802	NRPK	Kakisa	2017	495	4	880	0.825	0.16	-27.88	11.68	3.18260	М	Μ
10803	NRPK	Kakisa	2017	430	2	565	0.427	0.09	-28.00	11.44	3.18672	F	IM
10804	NRPK	Kakisa	2017	220	1	85	0.297	0.06	-29.41	9.98	3.16722	М	IM
10805	LKWH	Kakisa	2017	411	20	825	0.530	0.11	-29.63	10.49	3.37381	М	IM
10806	LKWH	Kakisa	2017	420	12	1110	0.298	0.06	-31.73	9.92	6.10666	F	М
10807	LKWH	Kakisa	2017	380	4	800	0.180	0.04	-29.32	9.34	3.59842	М	IM
10808	LKWH	Kakisa	2017	330	4	575	0.289	0.06	-29.75	8.57	3.31939	F	IM
10809	LKWH	Kakisa	2017	406	7	1050	0.238	0.05	-27.27	9.39	3.42841	М	М
10810	LKWH	Kakisa	2017	325	4	515	0.179	0.04	-28.17	8.47	3.39522	F	IM
10811	LKWH	Kakisa	2017	398	7	1115	0.238	0.05	-29.11	10.08	3.40302	F	М
10812	LKWH	Kakisa	2017	335	7	665	0.259	0.05	-28.97	9.09	3.38563	F	М
10813	LKWH	Kakisa	2017	300	3	390	0.279	0.06	-29.76	8.82	3.22448	М	IM
10814	LKWH	Kakisa	2017	235	3	170	0.210	0.04	-29.28	8.86	3.19345	U	IM
10815	WALL	Kakisa	2017	400	7	640	0.951	0.19	-27.92	11.21	3.12911	М	IM

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10816	WALL	Kakisa	2017	280	3	235	0.638	0.13	-28.24	11.19	3.17943	М	IM
10817	WALL	Kakisa	2017	300	4	265	0.781	0.16	-27.49	11.05	3.12334	М	IM
10818	WALL	Kakisa	2017	225	3	105	0.594	0.12	-27.88	11.16	3.11724	F	IM
10819	WALL	Kakisa	2017	185	2	65	0.478	0.10	-28.45	10.59	3.22158	F	IM
10820	LKWH	Kakisa	2017	220	3	135	0.230	0.05	-29.04	8.95	3.25550	U	IM
10821	WHSC	Kakisa	2017	475	15	1680	0.716	0.14	-28.59	10.41	3.35053	М	М
10822	LKWH	Kakisa	2017	420	12	950	0.318	0.06	-28.91	8.89	3.50142	F	М
10823	WHSC	Kakisa	2017	462	9	1485	0.370	0.07	-28.71	10.01	3.33566	М	М
10824	WHSC	Kakisa	2017	435	9	1335	0.319	0.06	-29.57	10.85	3.38500	М	М
10825	WHSC	Kakisa	2017	480	9	1590	0.306	0.06	-29.18	10.53	3.28764	F	М
10826	WHSC	Kakisa	2017	435	4	1265	0.155	0.03	-29.10	10.72	3.38494	F	М
10827	WHSC	Kakisa	2017	405	5	1120	0.131	0.03	-29.27	10.66	3.26852	F	М
10828	WHSC	Kakisa	2017	403	5	1020	0.155	0.03	-29.07	10.47	3.21145		
10829	WHSC	Kakisa	2017	440	5	1290	0.185	0.04	-28.71	10.06	3.36717	F	М
10830	WHSC	Kakisa	2017	411	5	1105	0.128	0.03	-29.38	11.24	3.25716	F	М
10831	LKWH	Kakisa	2017	420	11	1120	0.296	0.06	-29.60	10.46	3.27492	М	М
10832	WHSC	Kakisa	2017	446	5	1130	0.221	0.04	-26.38	8.19	3.26269	F	М
10833	WALL	Kakisa	2017	372	6	605	0.802	0.16	-29.01	11.98	3.19148	М	М
10834	NRPK	Kakisa	2017	522	4	920	0.989	0.20	-28.17	11.74	3.16354	М	IM
10835	NRPK	Kakisa	2017	475	3	740	0.693	0.14	-27.60	10.95	3.19850	М	М
10836	NRPK	Kakisa	2017	550	6	1140	1.087	0.22	-27.83	10.76	3.14237	F	М
10837	WALL	Kakisa	2017	360	6	465	0.914	0.18	-28.08	11.14	3.14991	М	IM
10838	NRPK	Kakisa	2017	512	3	700	1.124	0.22	-28.60	12.16	3.17873	F	IM
10839	WALL	Kakisa	2017	330	5	335	0.722	0.14	-27.54	11.04	3.13461	М	IM
10840	WALL	Kakisa	2017	340	6	455	0.775	0.15	-28.65	10.74	3.21224	М	IM
10841	NRPK	Kakisa	2017	503	3	885	0.535	0.11	-28.79	12.78	3.16663	F	IM
10842	NRPK	Kakisa	2017	385	2	400	0.473	0.09	-29.23	10.84	3.18028	М	IM

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10843	NRPK	Kakisa	2017	415	2	455	0.516	0.10	-28.64	11.54	3.19920	F	IM
10844	NRPK	Kakisa	2017	440	2	700	0.494	0.10	-29.51	10.84	3.19896	U	U
10845	WALL	Kakisa	2017	300	4	290	0.629	0.13	-28.13	11.05	3.18535	F	IM
10846	NRPK	Kakisa	2017	445	3	635	0.564	0.11	-27.90	11.52	3.20464	М	IM
10847	NRPK	Kakisa	2017	470	4	685	0.987	0.20	-28.54	11.91	3.15600	F	IM
10848	NRPK	Kakisa	2017	325	1	275	0.475	0.09	-29.05	10.83	3.24602	М	IM
10849	LKWH	Kakisa	2017	265	3	260	0.176	0.04	-30.95	8.61	3.58246		
15296	LKWH	Tathlina	2018	407	12	1075	0.559	0.11	-30.31	8.62	3.13075	F	Μ
15297	LKWH	Tathlina	2018	367	7	640	0.473	0.09	-29.95	9.46	3.13997	Μ	М
15298	LKWH	Tathlina	2018	325	3	540	0.317	0.06	-30.90	8.97	3.09937	F	Μ
15299	LKWH	Tathlina	2018	332	4	540	0.326	0.07	-30.87	9.60	3.34591	М	Μ
15300	LKWH	Tathlina	2018	339	4	625	0.266	0.05	-29.74	9.47	3.15841	Μ	М
15301	LKWH	Tathlina	2018	409	13	965	0.503	0.10	-29.98	9.83	3.25475	М	Μ
15302	LKWH	Tathlina	2018	410	10	915	0.498	0.10	-29.67	9.30	3.24005	М	Μ
15303	LKWH	Tathlina	2018	311	3	430	0.325	0.06	-31.83	8.39	3.08418	F	М
15304	LKWH	Tathlina	2018	322	4	440	0.391	0.08	-29.88	9.55	3.14442	Μ	М
15305	LKWH	Tathlina	2018	350	4	595	0.374	0.07	-30.73	8.35	3.15400	Μ	М
15306	NRPK	Tathlina	2018	953	11	8240	1.877	0.38	-29.56	10.81	3.14240	F	М
15307	NRPK	Tathlina	2018	646	7	1980	0.914	0.18	-29.60	10.82	3.11952	F	М
15308	NRPK	Tathlina	2018	653	5	2175	1.320	0.26	-29.35	11.28	3.09542	F	М
15309	NRPK	Tathlina	2018	639	5	1895	0.883	0.18	-29.24	10.63	3.10956	F	М
15310	NRPK	Tathlina	2018	863	8	5570	1.589	0.32	-29.41	11.23	3.12371	F	М
15311	NRPK	Tathlina	2018	794	12	4725	2.109	0.42	-29.26	11.26	3.13109	F	М
15312	NRPK	Tathlina	2018	815	8	4125	3.478	0.70	-28.68	11.61	3.09745	F	М
15313	LKWH	Tathlina	2018	373	8	750	0.610	0.12	-29.46	9.49	3.18760	Μ	М
15314	LKWH	Tathlina	2018	355	9	710	0.432	0.09	-31.34	9.12	3.30896	М	М
15315	LKWH	Tathlina	2018	313	3	455	0.378	0.08	-30.58	9.43	3.16566	Μ	Μ

ID	Species	Lake	Year	Fork Length (mm)	Age	Wet Weight (g)	[THg] mg/kg (dry wt.)	[THg] ppm (wet wt.)	δ13C	δ15N	C : N Ratio	sex	maturity
15316	LKWH	Tathlina	2018	453	17	1460	0.813	0.16	-29.32	8.93	3.36711	F	М
15317	LKWH	Tathlina	2018	388	12	885	0.579	0.12	-30.47	9.13	3.20928	М	М
15318	LKWH	Tathlina	2018	355	5	620	0.398	0.08	-29.85	10.21	3.17031	М	Μ
15319	LKWH	Tathlina	2018	336	4	565	0.213	0.04	-30.86	9.93	3.48036	М	Μ
15320	LKWH	Tathlina	2018	255	2	240	0.347	0.07	-31.32	7.73	3.11631	М	IM
15321	LKWH	Tathlina	2018	382	7	865	0.550	0.11	-30.08	9.16	3.11477	F	М
15322	NRPK	Tathlina	2018	931	11	6990	3.018	0.60	-29.05	11.41	3.14939	F	М
15323	NRPK	Tathlina	2018	859	10	5555	3.380	0.68	-28.57	11.66	3.13934	F	М
15324	NRPK	Tathlina	2018	812	10	5135	3.006	0.60	-28.79	11.88	3.16956	F	М
15325	NRPK	Tathlina	2018	985	16	9080	5.007	1.00	-29.00	11.74	3.13245	F	М
15326	LKWH	Tathlina	2018	113		16							
15327	LKWH	Tathlina	2018	106		12							
15328	LKWH	Tathlina	2018	398	6	1005	0.527	0.11	-30.82	8.62	3.06751	F	М
15329	LKWH	Tathlina	2018	430	15	995	0.656	0.13	-29.46	8.65	3.18306	Μ	М
15330	LKWH	Tathlina	2018	394	11	840	0.785	0.16	-30.43	9.87	3.33800	Μ	М
15331	WALL	Tathlina	2018	338	6	420	0.759	0.15	-28.83	11.38	3.10010	F	IM
15332	LKWH	Tathlina	2018	340	4	630	0.483	0.10	-31.05	9.24	3.62548	F	М
15333	LKWH	Tathlina	2018	341	3	555	0.367	0.07	-30.83	9.46	3.25858	Μ	М
15334	LKWH	Tathlina	2018	342	4	650	0.286	0.06	-30.26	9.12	3.16907	Μ	М
15335	LKWH	Tathlina	2018	353	3	730	0.347	0.07	-30.32	9.11	3.16846	F	М
15336	LKWH	Tathlina	2018	328	4	485	0.269	0.05	-29.78	9.41	3.21968	Μ	М
15337	LKWH	Tathlina	2018	357	5	570	0.533	0.11	-30.09	8.55	3.11260	F	М
15338	LKWH	Tathlina	2018	340	4	615	0.231	0.05	-31.19	9.23	3.91252	Μ	М
15339	LKWH	Tathlina	2018	360	4	780	0.368	0.07	-30.61	8.97	3.30441	F	М
15340	LKWH	Tathlina	2018	290	4	400	0.330	0.07	-30.36	9.46	3.19260	F	М
15341	NRPK	Tathlina	2018	356	3	310	0.650	0.13	-30.90	10.42	3.19825	Μ	
15342	NRPK	Tathlina	2018	1042	14	9390	3.711	0.74	-29.55	11.06	3.20759	F	Μ

ID	Species	Lake	Year	Fork Length (mm)	Age	Wet Weight (g)	[THg] mg/kg (dry wt.)	[THg] ppm (wet wt.)	δ13C	δ15N	C : N Ratio	sex	maturity
15343	NRPK	Tathlina	2018	723	11	3255	1.770	0.35	-29.54	10.83	3.15431	F	M
15344	NRPK	Tathlina	2018	474	7	705	1.349	0.27	-28.26	11.05	3.33824	F	М
15345	NRPK	Tathlina	2018	580	11	1315	2.362	0.47	-29.59	10.98	3.26896	М	М
15346	NRPK	Tathlina	2018	429	4	520	0.815	0.16	-30.19	10.33	3.34202	М	М
15347	NRPK	Tathlina	2018	363	4	320	0.965	0.19	-30.19	10.39	3.38486	F	М
15348	NRPK	Tathlina	2018	473	5	740	0.990	0.20	-29.61	10.44	3.18232	М	М
15349	LKWH	Tathlina	2018	307	3	390	0.254	0.05	-30.06	10.16	3.40003	М	IM
15350	LKWH	Tathlina	2018	404	9	1105	0.462	0.09	-32.33	9.29	4.18351	F	Μ
15351	LKWH	Tathlina	2018	392	18	845	0.622	0.12	-30.91	8.61	3.83748	Μ	М
15352	LKWH	Tathlina	2018	389	11	925	0.795	0.16	-29.79	9.06	3.23492	F	М
15353	LKWH	Tathlina	2018	376	7	725	0.344	0.07	-30.03	9.48	3.34349	F	Μ
15354	LKWH	Tathlina	2018	319	2	480	0.299	0.06	-30.59	9.27	3.28222	F	М
15355	LKWH	Tathlina	2018	285	2	355	0.249	0.05	-30.93	7.37	3.52306	М	IM
15356	LNSC	Tathlina	2018	526	24	1800	1.302	0.26	-29.64	9.12	3.60626	F	М
15357	NRPK	Tathlina	2018	939	10	6970	3.125	0.62	-28.84	11.22	3.34808	F	М
15358	NRPK	Tathlina	2018	979	15	7095	5.765	1.15	-28.93	11.69	3.12901	F	М
15359	NRPK	Tathlina	2018	936	13	7245	4.994	1.00	-28.53	11.88	3.16904	F	М
15360	NRPK	Tathlina	2018	262	2	110	0.377	0.08	-30.83	8.56	3.10029	Μ	IM
15361	WALL	Tathlina	2018	335	4	390	1.569	0.31	-30.73	9.93	3.14940	F	IM
15362	WALL	Tathlina	2018	437	20	875	4.852	0.97	-29.70	11.47	3.32844	Μ	М
15363	WALL	Tathlina	2018	420	10	565	3.592	0.72	-29.10	11.14	3.08491	F	IM
15364	WALL	Tathlina	2018	276	2	235	0.871	0.17	-30.82	9.59	3.20338	F	IM
15365	LKWH	Tathlina	2018	271	4	320	0.567	0.11	-31.46	8.35	3.16316	Μ	IM
15366	LKWH	Tathlina	2018	379	9	795	0.491	0.10	-29.86	9.43	3.42781	Μ	М
15367	LKWH	Tathlina	2018	260	3	255	0.397	0.08	-32.30	8.66	3.31489	М	IM
15368	WALL	Tathlina	2018	388	6	605	1.825	0.36	-29.73	10.59	3.30235	М	М
15369	WALL	Tathlina	2018	512	18	1420	3.995	0.80	-29.52	11.50	3.13995	F	М

				Fork Length		Wet	[THg] mg/kg	[THg] ppm			C : N		
ID	Species	Lake	Year	(mm)	Age	Weight (g)	(dry wt.)	(wet wt.)	δ13C	δ15N	Ratio	sex	maturity
15370	WALL	Tathlina	2018	413	14	740	2.963	0.59	-29.88	11.27	3.33915	М	М
15371	WALL	Tathlina	2018	339	5	400	1.416	0.28	-30.17	10.74	3.37046	М	М
15372	LKWH	Tathlina	2018	102		10							

### **CHAPTER 3: GENERAL CONCLUSIONS**

### **3.1 Significance**

Whereas many publications in the fields of SIA, Hg ecotoxicology, or fisheries tend to focus primarily single topics such as modeling food webs, characterizing exposure risk, or stock viability assessments, this thesis expands the literature by combining these various disciplines to assemble a holistic, ecosystem-level understanding of Hg dynamics in natural boreal lakes. The benefit of this work is not so much the use of the analytical tools (e.g., SIA, growth rate analysis, etc.) of each compartment of Hg's journey through a lake, but rather the synthesis and assembly of each compartment into the context of the others in an ecological framework. The findings of this project can help fishers and community members make safer, more informed choices in terms of which species to target and eat.

Results of this project will inform a PhD dissertation in a collaborating lab (Dr. Brian Laird, UW Health Sciences), who also works with the KTFN and other Indigenous communities in the Dehcho region in response to concerns about fish [Hg]. That dissertation uses probabilistic exposure modeling to estimate Hg dosage based on lake, fish species, human consumption patterns, and the age and sex of the human consumers. Those findings will be balanced with probabilistic exposure models of selenium and fatty acids in order to create a more holistic prosand-cons approach to forming fish consumption advisories. Bioaccessibility of Hg, selenium, and fatty acids will be tested in order to bridge between fish muscle profiles (external dose) and human internal doses as measured from hair, urine, and blood samples of community members. Consumption recommendations from that research may come in the form of age-dependent suggestions for frequency and species of fish consumption, as determined by the risk profiles of the lakes used by each community (S. Packull-McCormick, pers. comm., 2019).

The data and results of this project may also be important for industrial applications in the future. Within the Kakisa Lake watershed, the Cameron Hills (Figure 3.1; note the proximity of the drilling sites to Kakisa and Tathlina Lakes) is the development site for quickly progressing oil and gas infrastructure, with land use licenses in effect despite legal disputes in 2005 between the KTFN and the Responsible Ministers (comprised of Environment Canada, Indian and Northern Affairs Canada, Fisheries and Oceans Canada, and the Government of the Northwest Territories Department of Environmental and Natural Resources). These licenses grant permission for well drilling, campsites, airstrips, quarries, and other related infrastructure (Cli-Michaud, 2018). The Mackenzie Valley Environmental Impact Review Board (as cited in Halwas, 2009) stated that there is potential for considerable detrimental effects from the fossil fuel extraction efforts on the environment in the form of large-scale contamination of waterways and erosion. The potential for construction- or maintenance-related environmental contamination and the risk of pipeline leaks are cause for concern for the local wildlife and the KTFN, as the runoff would be within the Kakisa Lake watershed and therefore impact water and habitat quality in and around the KTFN. Data from this project will be of great value in environmental impact assessments in the event of a contamination event. Baseline data will also be highly useful for monitoring the impacts of effluent or runoff from the fossil fuel extraction facilities. Indirect ecosystem disruption like erosion, waterway diversion, or changes in faunal behavior or abundance, though impossible to predict with specificity, have the potential to influence the movement and accumulation of Hg in the watershed and otherwise impact the wild foods upon which the KTFN relies.

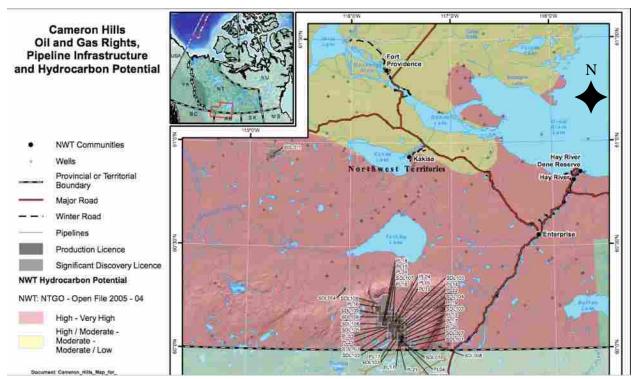


Figure 3.1: A map of the study lakes including potential incoming local oil and gas infrastructure, which represents a risk of potential hydrocarbon contamination and land disruption (Adapted from ITI, 2018, contacted for permission for republication).

## **3.2 Future Directions**

### 3.2.1 Future study on bottom-up Hg drivers

Carbon sourcing may affect Hg in lake systems. Bravo et al. (2017) determined that lake sediments dominant in allochthonous carbon (i.e., less negative signal) have higher [MeHg] and lower methylation rates ( $k_{meth}$ ) than sediments dominated by autochthonous carbon (i.e., less negative  $\delta^{13}$ C is related to increase [MeHg] and decreased  $k_{meth}$ . This is partially consistent with the findings of this work. Here, Kakisa Lake, which is less negative in biotic  $\delta^{13}$ C (Figures 2.5, 2.6, 2.7; Table 2.6), is associated with lower sediment [MeHg] but also lower  $k_{meth}$  (Table 2.2). Future efforts may benefit from  $\delta^{13}$ C analysis of sediments to determine differences in relative importance of terrigenous organic matter on Hg speciation.

#### 3.2.2 Management methods for high fish [Hg]

Certain tactics have been successful in reducing fish [Hg]. Verta (1990) used intensive fishing as a means to reduce Northern Pike [Hg]; when the population was severely reduced, the growth rate of the remaining population drastically increased due to a reduction in competitive pressure. This mechanism does not require a decline in Hg loading to the system or to the species; rather, it takes advantage of the growth dilution mechanism to dilute the consistent Hg load to the individual. Similar efforts in Dehcho's Sanguez Lake have returned promising preliminary results (Dixon et al., 2020).

Another known strategy is nutrient addition—when phosphorus is added to a lake, primary productivity increases, resulting in more energy and overall abundance of biomass in the system, reducing the effects of inter- and intraspecific competition and allowing growth dilution to reduce fish [Hg] (Essington & Houser, 2003; Mailman et al., 2006). However, inducing eutrophication may exacerbate Tathlina Lake's anoxia described by Stewart et al. (2016), and is likely impractical to implement, given the lake's large surface area and remote setting.

#### **3.3 Further Speculations**

While this work has focused on explaining why [Hg] differs between the two Lake Whitefish and Walleye populations, it has largely ignored the issue of why there is not a difference in [Hg] between the Northern Pike populations. Because Northern Pike are opportunistic ambush predators, their diets are more flexible, and therefore may be more representative of the relative availability of all potential prey items (Harvey, 2009). Lake Whitefish and Walleye have more consistent dietary preferences (Coad et al., 1995; Hartman, 2009); [Hg] of Lake Whitefish and Walleye's preferred diet items are likely higher in Tathlina Lake than Kakisa Lake due to bloom dilution and/or MeHg availability. The flexibility of

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Northern Pike diet may be a confounding factor affecting final [Hg] of individuals of that species. This may partially explain why it was impossible to distinguish between sizestandardized [Hg] in Northern Pike between lakes. Furthermore, water and sediment samples have all been taken from pelagic areas, representing the dominant habitat for Walleye and Lake Whitefish. Sampling from littoral areas would provide results that are more applicable to Northern Pike, which prefer littoral habitats (Chapman & Mackay, 1984). While benthic macroinvertebrates were collected from both nearshore and offshore locations in this study, targeted investigation of differences in benthic macroinvertebrate [MeHg] between habitat types have not yet been completed.

Habitat-specific differences will likely continue to be a confounding factor in speciesspecific [Hg] in the future. Lake-level factors will also likely affect the [Hg] of both lakes' fish assemblages moving forward. Climate change modeling has suggested that fish growth rates may increase with global climate change (Hill & Magnuson, 1990). Climate warming leads to longer ice-free periods in the subarctic, causing a longer growing season (Rolls et al., 2017). Warming also causes increased primary production as a result of increased nutrient availability and solar energy, which would be a bottom-up driver of total energy in the food web (Rolls et al., 2017). Incidentally, this may also lead to an increase in algal biomass, which may drive further bloom dilution. Increased ambient temperatures will also cause direct impacts on fish growth by increasing metabolic rate and therefore growth efficiency (Hill & Magnuson, 1990). Furthermore, the higher algal biomass in Kakisa Lake (Table 2.4) may be an indicator of higher total energy in that food web; more available energy may partially explain faster growth in the Kakisa Walleye population (Table 2.7). Because Tathlina Lake is shallower and therefore less capable of thermal buffering from climate warming, we might expect to see a higher increase in fish growth rates in this lake, but still some increase in fish growth rates in Kakisa Lake as climate warming continues. This could push growth dilution in both lakes, with a more significant impact on Tathlina Lake fish than Kakisa Lake fish, but more detailed bioenergetics and climate change modeling would be necessary to generate firmer predictions.

Tathlina Lake is consistently shallow, whereas Kakisa Lake has more variability in its depth profile, and therefore potentially more types of invertebrate habitat. Hypoxic winter conditions (Stewart et al., 2016) may also favor lower diversity, or at least lower invertebrate biomass. The habitat diversity hypothesis posits that habitat diversity of an area is the most important metric for determining biodiversity of a site, more so than the species-area relationship (Williams, 1964 in Boecklen, 1986). Given the variability in depth and therefore temperature and light availability of Kakisa Lake, and presuming a higher degree of homogeneity of habitat in Tathlina Lake, the invertebrate community may well be more diverse in Kakisa Lake than in Tathlina Lake. As such, competition would drive total invertebrate biomass down, whereas Kakisa Lake's invertebrate community would be spread out over more niche space and therefore be capable of sustaining the larger biomass. Lower abundance of invertebrates in Tathlina Lake would have a detrimental impact on fish growth. Hypoxia in the winter (Stewart et al., 2016) may also affect more sensitive taxa and further drive down Tathlina Lake's invertebrate diversity.

#### **3.4 Statement on the Integrative Nature of the Project**

This MSc thesis draws on tools from across the spectrum of biological investigation. The concept of bloom dilution is built on principles of large-scale ecosystem function and cycling of

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nutrients and contaminants while demanding knowledge and use of biogeochemical analysis. Stable isotope analysis is based in foundational principles of physical chemistry, but can be used in a trophic ecology context, creating a meeting point between atomic behavior, biogeochemistry, and ecology. Modeling the food web requires sampling across the range of aquatic taxa. The results will help inform other research framed around human health.

This project is an extension of the Dehcho Aboriginal Aquatic Resources and Oceans Management (AAROM) project and, as such, builds on previous research in mercury in these and other Dehcho lakes. Swanson Lab (University of Waterloo) projects, including this one, investigate the biological and ecological path of mercury in the Dehcho, Branfireun Lab (Western University) projects investigate abiotic and bacterial drivers of mercury methylation in the Dehcho, and Laird Lab (University of Waterloo) projects investigate the human health aspects specific to fish mercury consumption in the Dehcho. Additionally, this project furthers previous research in the MacLatchy Lab which includes assessment of the reproductive health of the Tathlina and Kakisa Lake Walleye and Lake Whitefish populations using physiological, organismal, and population-level endpoints. The present thesis work focuses on the abiotic and ecological mechanisms that connect mercury with ecology and life history. These transdisciplinary collaborations support each other in assembling an integrated picture of mercury in the environment, as well as fish health and mercury sources for people who eat fish from these lakes; they are united in a goal to address food security for northern and especially Indigenous communities.

### **3.5 Final Thoughts**

I experienced firsthand the high grocery prices in Yellowknife and Hay River, NT. Accessing commercial foods of decent nutritional value while maintaining financial stability

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seems a great challenge. I was grateful in those moments for our grants and their capacities for feeding us while traveling. I could not see a feasible way of living in the Dehcho region without relying on wild-caught foods for subsistence. It is easy to conduct a literature review about food security and safety in the north. To experience the reality of paying high grocery bills in Hay River (and slightly less high prices in Yellowknife) was a significant motivator both during field efforts and while analyzing and writing in Waterloo.

Support from the KTFN has been invaluable in this project and the ongoing monitoring. The experience was greatly enriched by personal relationships built with Chief Lloyd Chicot, Tarek Chicot, and Dawson Landry, whose welcoming attitudes of teamwork and camaraderie made sampling easier and more enjoyable. Just as motivating as the grocery prices was the time I spent in camps with KTFN community members, whose respect for and connection with the land and water was clear. My perspective, a result of an environmentally conscious and stewardshipfocused upbringing, fit well into the framework of community-driven research aimed at cooperative land stewardship and Indigenous empowerment; finding that common ground with KTFN community members had great personal value for me. These are the memories and motivations which have driven this product and will continue to propel my career.

An important caveat to note in interpreting the results of this work is that one should not place excessive emphasis on the role of any one mercury-driving mechanism detailed here. Mercury research is known to be complicated because of the large number of factors that can affect its final concentration in fish. I am confident in the results presented here because I have shown that there are many facets to Hg in these lakes. I would not presume any one driver (e.g., abiotic compartments, bloom dilution, or growth dilution) to be a "fix-all" component. As is often the case in ecology, there is no one simple answer. Mercury naturally occurs; no food item is without at least a small concentration of it. It is outside of my expertise to suggest a consumption guideline, but our collaborators are soon to distribute region- and community-specific risk assessments. In the meantime, while it is true that fish can sometimes have higher mercury concentrations than terrestrial animals, total avoidance of fish may reduce overall nutrition and removes an avenue of connection with the lakes.

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#### APPENDICES

#### **Appendix 1: Data Exclusions**

The following two subsections (A1.1, A1.2) detail justifications for excluding certain data from analyses in this thesis. Biological and/or statistical considerations are provided to justify these exclusions.

#### A1.1: Exclusion of fish collected in winter months

While most data and samples were collected in the summer season (August or September), some were collected in March. Winter conditions (e.g., lower temperature, shorter photoperiod) cue fish to reduce metabolism and therefore food intake, which can cause a decrease in condition factor (Lemly, 1996). Given the low rate of Hg excretion and this seasonal weight loss in fish, there is reason to believe that [Hg]<sub>fish</sub> could be higher in winter than in summer. Zhang et al. (2012) showed that some fish do indeed exhibit a seasonal change in [Hg], although they attributed the difference to dietary shifts and/or a bottom-up cascade of increasing bioavailable Hg. Harris & Bodaly (1998) found a significant interaction between temperature/metabolism and [Hg], but this was a minor effect (explaining 8% of [Hg] variation); that study compared two lakes during the summer only, and states a need for further study on the specific relationship between fish weight and [Hg].

Before any further analysis, I tested for seasonal differences in fish Hg. I performed an independent samples T-test on condition factors ( $CF = (W/(L^3))*100,000$ ) between summer and winter for all six species/lake combinations to test the hypothesis that there is a statistically significant difference in condition between seasons (Table A1).

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Species	Lake	n	t	df
Walleye	Tathlina	0.034*	-2.396	11.837
v	Kakisa	0.719	-0.365	18.704
Northern Pike	Tathlina	0.008*	-2.935	21.094
	Kakisa	0.548	-0.612	20.052
Lake Whitefish	Tathlina	0.066	1.865	77
	Kakisa	< 0.001*	-5.242	77.656

Table A1: Comparison of condition factors between summer- and winter-collected samples (independent samples T-test).

I then performed analyses of covariance (Log<sub>10</sub>(Hg)~Log<sub>10</sub>(FL), Season,

 $Log_{10}(FL)$ \*Season) for all six species/lake combinations and generated least squares mean [Hg] for each season. The interaction term was removed if it was found to be insignificant by the model (p > 0.05; Table A2).

Species	Lake	Season	[Hg]	SE	df	Lower	Upper	T	ukey		
						CL	CL	р	t	df	
Lake	Tathlina	S	0.123	0.010	73	0.104	0.145	0.492	0.707	72	
Whitefish		W	0.114	0.011	73	0.094	0.139	0.482	0.707	73	
	Kakisa	S	0.045	0.004	78	0.038	0.053	<0.0001*	6 156	70	
		W	0.106	0.011	78	0.086	0.130	<0.0001*	-6.456	78	
Northern	Tathlina	S	0.372	0.022	48	0.330	0.419	0.259	1.141	48	
Pike		W	0.315	0.042	48	0.242	0.411	0.239	1.141	40	
	Kakisa	S	0.362	0.034	56	0.300	0.437	0.459	-0.745	56	
		W	0.402	0.044	56	0.322	0.501	0.439	-0.743	56	
Walleye	Tathlina	S	0.566	0.051	26	0.471	0.680	0.228	-1.233	26	
		W	0.771	0.181	26	0.476	1.250	0.228	-1.233	20	
	Kakisa	S	0.276	0.012	82	0.253	0.302	0.002*	2 1 5 9	82	
		W	0.366	0.028	82	0.314	0.427	0.002*	0.002* -3.158		

Table A2: Comparison of size-standardized [Hg] between summer- and winter-collected samples, including Tukey's post-hoc test. S = Summer, W = Winter.

Because two of the six species/lake combinations show a significant difference in size-

standardized [Hg] between seasons, I chose to exclude winter data from further analyses.

I chose to include winter data for growth rate modeling. Both model types I performed (age-at-size and Von Bertalanffy) are derived from age at capture and length at capture, neither of which are subject to seasonal variation. These models are intended to output population-level parameters, so there is no concern for comparing growth in individuals to [Hg] or other variables in individuals.

These data may be useful if more winter sampling occurs. Future analyses may include seasonality in analytical approaches, but the current winter dataset is not yet robust enough to warrant its use.

#### A1.2: Exclusion of haemophagic leeches

During analysis and graphical output of biomagnification trends, some outliers were identified as leeches of the family Piscicolidae. As haemophages ("blood eaters"), Piscicolidae and Glossiphoniidae do not fit cleanly into the assumptions necessary for trophic biomagnification and accumulation, as they feed on single tissue types, rather than consuming whole prey. As consumers of blood only, they consume a mercury-depleted tissue relative to the whole fish (Kidd & Batchelar, 2012), but their  $\delta^{15}$ N values would be closer to those typical of a piscivore. These families held three data points which were removed before final analyses were performed.

### **Appendix 2: Growth Model Comparisons**

To determine if growth dilution is a factor in fish [Hg], I modeled growth rates for all six species-lake combinations using both age-at-size models and Von Bertalanffy growth models. Both modeling approaches typically demand sample sizes much larger than the current dataset afforded, so I output residuals for all models (3 spp. x 2 lakes x 2 models) and tested them for normality in order to determine which model would be more reliable. Age-at-size estimates were found to be more reliable are therefore presented in chapter 2.

## A2.1: Von Bertalanffy growth models of the three food fish species

The Von Bertalanffy growth model is employed to account for the logarithmic nature of fish growth. This model can be used with either length or weight, which have a sigmoid relationship to age that approaches a horizontal asymptote. The length-based variation of the Von Bertalanffy growth model is expressed by the following equation:

$$l_t = L_{\infty} \left( 1 - e^{-k(t-t_0)} \right)$$

Where:

 $l_t$  = length at age t; i.e., length at capture  $L_{\infty}$  = asymptotic length; i.e., length at which growth stops k = a measure of whole-organism catabolic rate; interpreted as the growth constant t = age t; i.e., age at capture  $t_0$  = theoretical age at which length is 0

 $l_t$  and t are observed values, and statistical software is used to estimate the remaining parameters. The growth constant k provides a simple means of comparing growth rates among populations (Beverton & Holt, 1957).

Species	Lake	Parameter	Estimate	SE	Lower CI	Upper CI
	Kakisa	L∞	428.272	5.360	417.501	439.042
		k	0.428	0.064	0.299	0.557
Lake		t <sub>0</sub>	0.741	0.409	-0.081	1.562
Whitefish	Tathlina	L∞	442.190	20.437	401.141	483.239
		k	0.142	0.045	0.051	0.232
		t <sub>0</sub>	-5.743	2.161	-10.084	-1.402
	Kakisa	L∞	845.656	106.244	630.576	1060.737
		k	0.142	0.053	0.034	0.249
Northern		t <sub>0</sub>	-2.408	1.044	-4.522	-0.295
Pike	Tathlina	L∞	972.257	143.706	678.771	1265.744
		k	0.172	0.094	-0.020	0.363
		$t_0$	0.022	1.505	-3.053	3.096
	Kakisa	L∞	473.584	13.713	446.248	500.920
		k	0.227	0.037	0.154	0.300
Walleye		t <sub>0</sub>	-0.521	0.580	-1.676	0.635
waneye	Tathlina	L∞	459.177	21.007	414.401	503.953
		k	0.164	0.057	0.043	0.285
		t <sub>0</sub>	-3.570	2.116	-8.079	0.939

Table A3: Estimates of Von Bertalanffy growth model parameters.

Table A4: Comparison of Von Bertalanffy's k between fish populations (Two-tailed T-test).

ι	Р
3.667	0.0004*
0.292	0.7715
0.781	0.4367
	0.292

Von Bertalanffy growth rate (k) estimates of Lake Whitefish are significantly different between both lakes, showing that the Tathlina Lake population grows more slowly than the Kakisa Lake population. However, the Tathlina Lake population reaches a higher asymptotic length, implying that Tathlina Lake's Lake Whitefish live longer than those in Kakisa Lake, which would further contribute to bioaccumulation of mercury. Tathlina Lake Walleye reach a lower asymptotic fork length and generally grow more slowly. However, the 95% confidence intervals for the estimated k overlap with one another, so it is difficult to interpret with confidence.

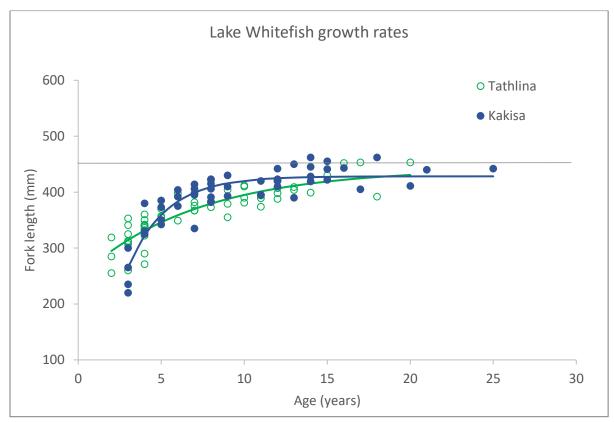


Figure A1: Von Bertalanffy growth model of Lake Whitefish in both lakes. Reference line represents the standardized fork length of 450 mm.

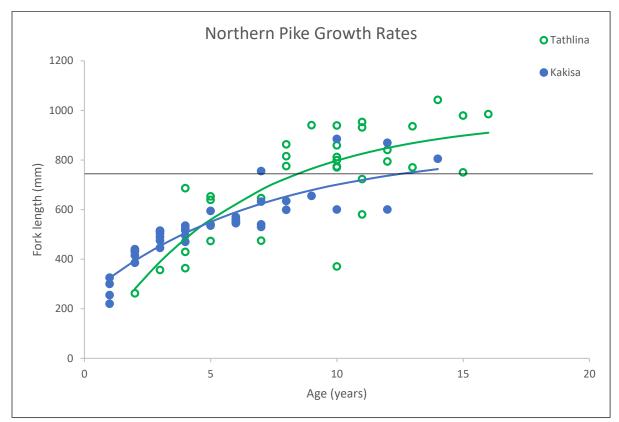


Figure A2: Von Bertalanffy growth model of Northern Pike in both lakes. Reference line indicates the standardized fork length of 650 mm.

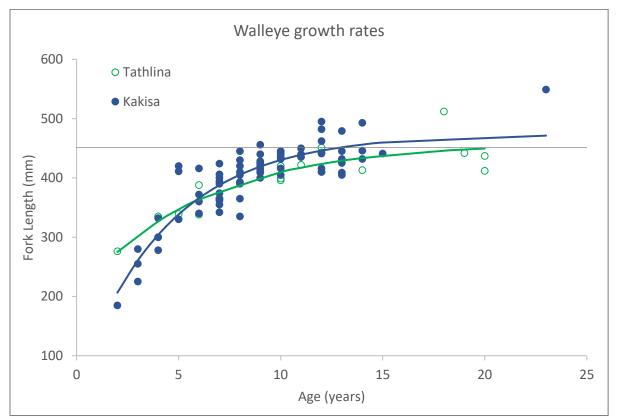


Figure A3: Von Bertalanffy growth model of Walleye in both lakes. Reference line indicates the standardized fork length of 450 mm.

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