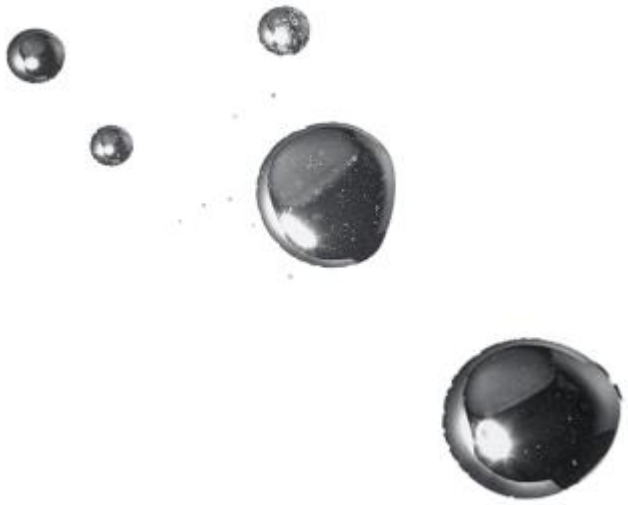
# PATTERNS OF MERCURYACCUMULATION IN NORTHERN ONTARIO WALLEYE AND RELATIONSHIP WITH WATERSHED CHARACTERISTICS

by

Sarah Francis



Source: Lourie 2003

FACULTY OF NATURAL RESOURCES MANAGEMENT

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# PATTERNS OF MERCURYACCUMULATION IN NORTHERN ONTARIO WALLEYE AND RELATIONSHIP WITH WATERSHED CHARACTERISTICS

by

Sarah Francis

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# ABSTRACT

Francis, S.E. 2018. Patterns of mercury accumulation in northern Ontario walleye and relationship with watershed characteristics. 60pp.

Keywords: aquatic ecosystem, coniferous forest, dissolved organic carbon, forest operations, harvest disturbance, mercury, mercury accumulation, methylmercury, methylation, mobilization, nitrogen, northern Ontario, pH, terrestrial ecosystem, walleye (*Sander vitreus*), watershed

Mercury is a well known toxic contaminate that poses health risks to humans and wildlife. In Ontario 85% of consumption restrictions of fish from inland lakes is due to mercury contamination. The purpose of this study was to investigate the relationship between total mercury concentration in walleye and lake chemistry, watershed characteristics, and forest harvesting. Data from the Ministry of Natural Resources and Forestry Broad Scale Fisheries Monitoring Program, Land Information Ontario, and Ontario Land Cover 2000 were utilized for analysis in this study. Linear regressions showed that lake surface area (9.8%), total land area (6.1%), total forest area (4.1%), coniferous forest area (5.3%), dissolved organic carbon (8.9%), pH (16.4%), nitrate and nitrite (8.1%), and dissolved inorganic carbon (13.8%) had a significant relationship with total mercury. Further, multivariate regressions showed that lake surface area and coniferous forest area (13.3%), dissolved organic carbon and pH (23%), and dissolved organic carbon and nitrate and nitrite (12.6%) had a significant relationship with a higher R-square value. This study supports that lake chemistry variables and landscape variables including dissolved organic carbon, pH, nitrate and nitrite, dissolved inorganic carbon, coniferous forest area, total forest area, total land area, and total lake surface area played a larger role on the impacts on total mercury concentration in high predatory fish than harvest disturbance and wetlands within the watershed which showed no significant relationship with mercury concentrations.

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# INTRODUCTION

## LITERATURE REVIEW

### Global Mercury Contamination

Mercury is a natural element found throughout Earth and has unique properties which affect mercury's chemical processes. Natural emissions of mercury include volcanic eruptions, forest wildfire, hot springs, and weathering of minerals within the natural environment (Lourie 2003). Since the industrial revolution approximately 200 years ago, anthropogenic mercury emissions have consistently exceeded natural emissions (United Nations Environment Program 2013). This additional mercury within the natural environment entering the mercury cycle is a result of anthropogenic activities. Anthropogenic sources of mercury may include coal burning electrical generation, mining activities, cement production, incinerating waste, and oil refining (United Nations Environment Program 2013). Of all anthropogenic sources in Canada, coal burning electrical generation contributes 27% and metal smelting processes of copper and zinc contributes another 25% of atmospheric mercury emissions (Lourie 2003). In 2016, the MOECC reported mercury emissions from coal burning electrical generation and metal smelting processes contributed a total of 40% down from the 2003 value, reported by Lourie (2003), at a combined value of 52%.

In 2013, the United Nations Environmental Program reported natural geological mercury emissions in the natural environment are responsible for ten percent of the global mercury, approximately thirty percent of annual mercury emissions coming from current anthropogenic sources and approximately sixty

percent from re-emissions of built up stored mercury that was previously released into the environment. The original source of the re-emitted mercury emissions cannot be conclusively determined however, they are considered a result of anthropogenic emissions because natural emissions have remained smaller than anthropogenic activities for two centuries (UNEP 2013).

Sources of mercury are stored on Earth in soil and the ocean (Jagtap and Maher 2015). The ongoing build up and storage of mercury emissions are a current widespread and future challenge because re-emission stores will continue to globally pollute the world (Lourie 2003), even though there has been a reduction of environmental mercury emissions. In the 1970's and 80's, new regulations were passed in Canada and the United States of America to reduce emissions with successful results reflecting Canada reduced emissions by more than 90% from 80 to 6 tonnes of mercury between 1970's – 2010 and the United States reduced mercury emissions as well (Gandhi et al. 2014). As a consequence, direct emissions have been significantly reduced in North America although sources of mercury re-emission continue. For example, in old mines located in Nova Scotia will continue to emit mercury for 60-70 years following the mine closures (MOECC 2016). Between 1990 and 2010, atmospheric mercury emissions declined by 85% in Canada. With the reduction of mercury emissions in Canada and the United States of America since 1970's, reductions in fish mercury concentration levels were anticipated to also decrease however, lower mercury concentrations in fish were not realized. Mercury trends in walleye (*Sander vitreus*), from Ontario lakes representing one-

third of the world's freshwater, were analyzed. Gandhi et al. (2014) reported fish mercury concentrations were increasing in more than half of the lakes sampled in northern Ontario. The results from Gandhi et al. (2014) were attributed to possible factors of climate change and global emissions. Globally, mercury emissions remained stable between 1990's to 2000's but have been recently increasing in Asia due to coal burning electrical generation which account for approximately 50% of the total global emissions (MOECC 2016). The growth of industrialization in Asia accounts for 48% of the global total mercury emissions (UNEP 2013).

Mercury in the natural environment circulates through waterways and since mercury is volatile, it can evaporate to enter the atmosphere (Lourie 2003). Both wet and dry atmospheric depositions release mercury into the environment from natural and anthropogenic sources (Jagtap and Maher 2015). Water is contaminated with anthropogenic mercury sources by wet deposition through storm sewers and sewage plant discharges directly into the aquatic ecosystem or through precipitation such as rain that is contaminated with atmospheric mercury (Lourie 2004). Dust particles within the atmosphere can be contaminated with mercury and are deposited by dry deposition on to surfaces such as lakes, rivers, foliage, and the forest floor (Lourie 2003). According to Lourie (2003), atmospheric mercury emissions may land on the Earth's surface within a 50 Kilometre radius from the anthropogenic sources such as a coal burning plant and is called local deposition. Mercury emissions from a coal burning plant may be deposited within several hundred kilometres downwind

from the source and is called regional deposition. Airborne mercury can also enter the global atmospheric pool which circles the globe for at least one year within major weather systems of the Earth. This mercury can be transported for thousands of kilometres before deposition (Lourie 2003). Further, because of mercury's extended residence time of up to 1.7 years, it remains in the atmosphere allowing it to circulate globally (Gonzalez-Raymat et al. 2017).

Mercury is also released by re-emission from a global distillation phenomenon which transfers mercury emissions from equatorial, subtropical and temperate regions to the polar arctic regions globally (Lourie 2003). Canada's arctic has mercury deposition from this global pool and current regional emission deposition from Asia and northern Europe (Lourie 2003). Ecosystems in remote locations geographically far from industrial emissions, can suffer from elevated mercury levels due to long-distance mercury dispersal (Fitzgerald et al. 1998).

Lourie reported (2003), the quantitative value of atmospheric mercury level globally had tripled since industrialization as a result of anthropogenic activities. Some of these sources of mercury included emissions from coal burning, incinerating waste, smelting, mining, and discarding products made with mercury. Some of the sources of liquid mercury included sewage treatment plant waste and industrial plant wastes directly entering waterways from anthropogenic activities. These and additional sources of mercury have resulted from industrialized human activities that enter and contribute to higher levels of mercury in the mercury cycle (Lourie 2003).

Impacts of Mercury on Human Health

Mercury is classified as a neurotoxin (Celo et al. 2006). Methylmercury is the most dangerous form of mercury because it is the most abundant and toxic form of mercury (Celo et al. 2006). All forms of mercury are harmful to human health and species. For example, mercury poisoning caused severe illness and death in Minamata Japan, in 1956 (Mackereth 2017) after people consumed contaminated fish in Minamata Bay. One of many forms of mercury called methylmercury was dumped into Minamata Bay from a chemical plastic manufacturing plant (Lourie 2003). Mercury poisoning in Minamata Japan was one of the first documented cases impacting human health (Lourie 2003, MOECC 2015). Inhalation, absorption and or ingestion of mercury can cause severe health effects to humans and other species. High exposure levels to mercury can cause deleterious impacts to human health including damage to the central nervous system, brain damage, kidney damage, lung damage, birth defects, and or death (Lourie 2003; Gonzalez-Raymat et al. 2017). Methylmercury can enter the bloodstream and organs (Lourie 2003; Ullrich et al. 2001). The brain is highly susceptible to methylmercury toxicity and poisoning consequently causing the entire nervous system to be compromised (Lourie 2003). People affected with chronic low-grade mercury exposure experience many adverse symptoms that may include fatigue, anxiety, depression, weight loss, difficulty concentrating, and memory loss (Bernhoft 2012). These subjective symptoms of mercury toxicity may also be indicative of a wide variety

of other medical conditions, leading to challenges in diagnosis of mercury poisoning.

Mercury Cycle

Mercury cycles through the atmosphere, water and soil. Elemental mercury can be deposited from the atmosphere into aquatic ecosystems through deposition. Mercury travels to the water where it is evaporated into the air, travels globally through wind, and re-deposits itself only to resume the cycle again. The numerous, complex interactions within the mercury cycle involve methylation, demethylation, and biotic processes. The mercury cycle is complex and mercury cycling does not follow one single pathway (Grigal 2002; Lourie 2003; Gonzalez-Raymat et al. 2017).

Elemental mercury (Hg0) is soluble in water and can be sorbed onto dissolved organic matter or suspended particulate matter (Gonzalez-Raymat et al. 2017). Mercury can enter into the aquatic food web when organisms ingest organic matter with sorbed mercury. Once elemental mercury is sorbed, it can then undergo oxidation reactions to form mercuric ($Hg^{2+}$). Mercuric increases the availability for methylation processes to occur. Mercuric can also be converted back to elemental mercury and re-enter the atmosphere. Elemental mercury can be methylated by abiotic or biotic pathways. Methyliodide is believed to contribute to the abiotic methylation process. Methyliodide is produced from cyanobacteria, seaweed, algae, and fungi within the ecosystem. The biotic pathway of methylation is thought to be due to primarily sulphate-reducing anaerobic bacteria (Gonzalez-Raymat et al. 2017). Methylation within a

watershed causes inorganic mercury to transform through natural microbial processes into the toxic form of methylmercury which is transported in water bodies (United Nations Environment Program 2013).

Mercury Contamination in Food Webs

Methylmercury bioaccumulates in aquatic ecosystems by binding with organisms. In bioaccumulation, contaminants are taken up by the organism faster than they can eliminate them, causing contaminants to accumulate in the body (Lourie 2003). Bioaccumulation of mercury in aquatic ecosystems is driven by chemical and biological processes that only require low initial quantities of mercury to have significant impacts (Fitzgerald et al. 1998). Due, to biomagnification, increasingly larger amounts of mercury accumulate with increasing trophic levels. Consequently, top predators have the highest methylmercury concentrations in their body tissue (Weiner et al. 2006). The aquatic ecosystem is the main source of mercury exposure for humans and wildlife since methylmercury is stored in the muscle tissue of fish (MOECC 2015). It is estimated that predatory fish consumed by humans have one hundred thousand to one million times more methylmercury in their bodies compared to the surrounding lake water (Lourie 2003).

In Ontario's 2015-2016 Guide to Eating Ontario Fish, it was noted that 85% of eating restrictions of fish from inland lakes is due to mercury contamination (MOECC 2015). With mercury poisoning posing a serious health concern, one course of action to combat mercury toxicity is to decrease the consumption of contaminated food sources. Government officials from a variety

of ministries have worked collaboratively to inform the public and to provide guidelines on the recommended portions and frequency of fish consumption. These recommendations are published by the Ministry of Natural Resources and Forestry based on their field research and analysis of the Broad Scale Fish Monitoring Program (Sandstorm 2013). Therefore, limits to consumption of fish are recommended to reduce mercury exposure through ingestion.

Children are given special consideration within the guidelines of the recommended amount of fish consumption because their cellular repair system is not fully developed (Lourie 2013). Medical research suggests that there is no safe level of methylmercury in a child's body due to the potential poisonous effects on development and growth (Lourie 2003). However, this suggestion for the total elimination of fish from children's diet has not resulted in a recommendation for a complete ban on children eating lake fish in Ontario (Lourie 2003) possibly because there are benefits of having fish in the diet (MOECC 2015). In addition, evidence to eliminate fish from children's diet would require specific and current research in support of the claim.

Pregnant women are also given special consideration within the guidelines of the recommended amount that may be consumed. Pregnant women should not consume high levels of contaminated fish due to the link between birth defects, neurological damage, learning disabilities, and negative effects on the gestational growth of the developing fetuses as a result of increased levels of mercury toxicity from contaminated fish consumption (Lourie 2003). The toxicity of mercury in a fetus causes serious and extended health

issues after birth. Modification to the pregnant woman's diet is enough to avoid the negative health consequences of their unborn baby (MOECC 2015).

Aquatic Ecosystems

There are many factors that influence mercury contamination in aquatic ecosystems. Lake characteristics and water chemistry elements are two factors that can influence methylation and demethylation processes (Garcia and Carignan 2000; Ullrich et al. 2001; Grigal 2002; Weiner et al. 2006; Drott 2007; Gonzalez-Raymat et al. 2017). The physical lake size is a potential factor contributing to mercury concentrations (Grigal 2002). Lake size has inversely related to the concentration of mercury present in the ecosystem. Therefore, smaller lakes have been found to have higher levels of total mercury concentrations present (Grigal 2002). However, this relationship between lake size and methylmercury was weak in the study of Grigal (2002), and possible explanations for this could be due to higher water temperature in smaller lakes and a less effective transport of mercury in smaller watersheds.

Measures of water chemistry including dissolved organic carbon, anaerobic bacteria, pH, redox conditions, and sulphide impact and contribute to oxidation and methylation (Ullrich et al. 2001; Gonzalez-Raymat et al. 2017). It has been found that increased concentrations of methylmercury in water, sediment and fish are positively correlated to increased levels of dissolved organic carbon. High concentrations of dissolved organic carbon influence mercury levels in watersheds (Grigal 2002). Mercury concentrations and dissolved organic carbon have a positive relationship explained by wetlands

having sources of organic matter and inorganic Hg(II) from atmospheric deposition onto wetlands causing methylation. The methylmercury that is produced binds with dissolved organic carbon and is transported in the hydrological watershed of lakes (Weiner et al. 2006). Acidification of lakes has been of concern because low pH lake water levels increase accumulation of methylmercury. Lakes with lower pH levels tend to have higher methylmercury levels in fish (Garcia and Carignan 2000; Weiner et al. 2006). There is greater methylation of inorganic mercury Hg (||) by microbial activity of bacteria at a lower pH (Weiner et al. 2006). Lower pH may impact the greater methylation process (Weiner et al. 2006). Further, additional sulfate levels increase methylation through sulfate-reducing bacteria (Ullrich et al. 2001; Drott 2007). Garcia and Carignan (2000) found the positive correlation between mercury levels of fish and sulphate concentration may be related to acidic pH levels and the presence of sulphate reducing bacteria. Weiner et al. (2006) reported sulfate stimulated the methylation process of inorganic Hg(II) by sulfate reduction bacteria.

Lucotte et al. (2016) and Garcia and Carignan (2000) reported contradictory results involving elements of water chemistry in fresh water lake methylation and mercury concentrations in fish. Lucotte et al. 2016 found the levels of lake water acidity were not a significant factor in methylation and methylmercury concentration levels in predatory fish were not high. Higher methylation was not supported with lower water pH levels either (Lucotte et al. 2016). In contrast, Garcia and Carignan (2000) reported a correlation between mercury concentrations in predatory fish is related to lake water pH. It is

considered an important predictor. High mercury concentrations in predatory fish were also found in lake water with a pH of 6.1 (Garcia and Carignan 2000).

Wetland Influences on Mercury

Wetlands are a site of mercury methylation and output (Grigal 2002). Interlake differences of methylmercury concentration in fish were found to be related to wetland occurrence in the surrounding watershed (St. Louis et al. 1994). Wetlands are associated with the transport of mercury to aquatic ecosystems because of their high dissolved organic carbon, water residence time, and hydrological pathway connections within watersheds. Braaten and de Wit (2018) identified hydrological pathways as playing a significant role in determining stream water chemistry and water chemistry factors impacting methylation. Wetland area is also linked with mercury methylation since their anoxic conditions promote the activity of sulphate-reducing bacteria (Braaten and de Wit 2016).

Peatlands include bogs, fens and swamps (Talbot et al. 2017). Peatlands have high organic matter production and low rates of decomposition. Due to the high organic matter content, peatlands can store atmospheric pollutants including mercury. It is evident that mercury and methylation hotspots are present within wetlands. Near the upland peatland interface where there is high quantity of runoff, hydrological mixing of upland and lowland waters, high sulfate concentration, acidic pH and high dissolved oxygen content hotspots are found (Mitchell et al. 2008). Higher mercury concentrations are found near the top layers of peatlands (Talbot et al. 2017). Bogs are the best indicator of mercury

deposition because they are not influenced by groundwater. Therefore, bogs only reflect atmospheric inputs. Climate change, increased decomposition rates, and degradation of peatlands could all potentially release large pools of mercury back into the atmosphere (Talbot et al. 2017).

Terrestrial Ecosystems

A natural source of mercury in the terrestrial environment comes from minerals in the earth such as cinnabar (United Nations Environment Program 2013). Yet, the main source of mercury in remote terrestrial environments is atmospheric deposition (Fitzgerald et al. 1998; Gonzalez-Raymat et al. 2017). Mercury can be deposited onto foliage, soil particles, and organic matter through dry deposition, precipitation, throughfall, and litterfall (Fitzgerald et al. 1998; Graydon et al. 2008) resulting in approximately half of the atmospheric mercury deposition from the atmosphere stored on the surface soil and foliage of the forest (Lourie 2003). Graydon et al. (2008) found that precipitation in open spaces has significantly lower mercury concentration than the soil surface beneath a forest canopy.

The forest is a sink for mercury (Gonzalez-Raymat et al. 2017). Soil stores of mercury are unpredictable in terms of long term storage stability because the mercury can either remain in place, be mobilized by water, or be re-emitted atmospherically (Rajani and Maher 2015). Organic matter, temperature, moisture, soil porosity, and soil surface area all impact and contribute to oxidation and methylation (Graydon et al. 2008). Runoff from the forest to aquatic ecosystems facilitates the mobilization and transfer of mercury in soil

and litter into the watershed (Gonzalez-Raymat et al. 2017). In addition to deposition from the atmosphere, elemental mercury can also be deposited onto soil by litterfall. Foliage that has deposited mercury on the surface results in mercury accumulation on the soil surface through litterfall (Braaten and de Wit 2016). Litterfall is the main pathway for mercury to enter the terrestrial environment from foliage because atmospheric mercury is deposited onto leaves and plant uptake of mercury is limited (Grigal 2002). Litterfall is the main mercury load contributor to soil in deciduous forests and throughfall is the main mercury load soil contributor in coniferous forests (Demers et al. 2007).

The unique characteristics of different forest vegetation types impact total mercury concentration and methylmercury loading potential. Leaves with greater surface area or leaf area index tend to adhere greater amounts of mercury (Graydon et al. 2008; Drenner et al. 2013). For example, Braaten and de Wit (2016) found that atmospheric deposition of mercury was highest in dense spruce forest and lowest in treeless peatland. Similarly, Drenner et al. 2013 found a positive linear relationship between coniferous forest composition and mercury contamination of nearby lakes. Total fish mercury concentrations can differ by more than 400% between watershed with different conifer densities, with higher conifer cover being a significant explanatory variable of fish mercury levels (Eagles-Smith et al. 2012). Graydon et al. (2008) found higher mercury accumulation rates under conifer canopies than deciduous forest canopies. The differences in mercury accumulation may be explained by the annual shedding of deciduous tree leaves not having the ability to store mercury over a longer time compared to coniferous tree leaves. Deciduous leaves annually have a

decreased mercury load within the foliage annually (Laacouri et al. 2013; Richardson and Friedland 2015). Blackwell et al. (2014) confirm deciduous litterfall and coniferous throughfall are the main mercury contributors to forest soil with coniferous forest deposition having a greater impact.

The mercury load to forest soil is lower from deciduous forest litterfall than coniferous forest throughfall (Graydon et al. 2008). This greater concentration of mercury load in the coniferous forest results in a mercury transfer potential from both litterfall and throughfall into the soil and possibly entering into entry of the aquatic ecosystem (Graydon et al. 2008). This leads to soils having greater mercury concentrations which support the forest stores and serving as a sink for mercury. However, Blackwell et al. (2014) discovered soil mercury pools were not significantly different between the coniferous and deciduous forests. This suggests that mercury contamination in the soil may not be the net sink of atmospheric mercury but that a mercury loss mechanism within the coniferous stand contributes significant to the mobilization of mercury into the aquatic watershed ecosystem which could support Blackwell et al. (2014) findings that coniferous and deciduous forest leaf structure do have significantly different amounts of deposited mercury on their leaves and an unknown mechanism facilitates mercury transfer out of the soil. Further research to clarify if the forest soil is or is not the net sink of atmospheric mercury is necessary (Blackwell et al. 2014). There may be a mechanism contributing mercury forest foliage being transported to a water body within the watershed by pathways after deposition. This release of mercury from forest soil could be

affected by variations in climatic conditions from increased precipitation and rising temperatures resultant of climate change (Blackwell et al. 2014).

Disturbance Impacts on Mercury

Forest harvesting can impact mercury methylation and mobilization in a number of ways (Eklöf 2016). Harvesting can result in changes to soil conditions and forest composition. If harvesting occurs within a watershed, the chemical changes to the soil nutrients can be transferred to aquatic systems (Kreutzweiser et al. 2008).

Logging in watersheds with a large number and area of wetlands may lead to increased nutrient export to aquatic ecosystems. Dissolved organic carbon may increase in soil following harvest, supporting increased methylation. Garcia and Carignan (2000) and Eklöf et al. (2016) found that organic carbon increased after logging, which they concluded could contribute to increased methylation and mobilization of mercury.

Additional terrestrial debris and materials on the forest floor after harvest contribute to the methylation process through increased soil organic matter (Garcia and Carignan 2000). The removal of trees during logging disrupts the hydrological cycle by reducing transpiration. This can potentially cause decreased output of elemental mercury to the atmosphere and higher groundwater levels which can potentially lead to increased mobilization of mercury from the watershed to aquatic ecosystems (Eklöf et al. 2016). Forest operations may cause soil compaction, which lowers water infiltration and

increases waterlogged conditions and runoff that supports methylation and mobilization respectively (Garcia and Carignan 2000; Eklöf et al. 2016).

Harvesting can potentially impact the potential for methylation by increasing soil temperature, carbon from logging residuals, i water tables, and organic matter. Mobilization of mercury can be caused by increasing hydrological connectivity between methylmercury hotspots and aquatic ecosystems (Sørensen et al. 2009). However, Sørensen et al. (2009) did not find a significant increase of mercury in the aquatic ecosystem after harvesting. Bishop et al. (2009) state that forest operations mobilize mercury to aquatic ecosystems, although they are not the only source of mercury in watersheds. Forestry is estimated to contribute approximately 10-25% of mercury in lakes through increased mercury runoff after forest operations contains (Bishop et al. 2009).

Forest disturbances from logging or thinning raise the groundwater table closer to the surface of the soil within the horizons (Bishop et al. 2009). This facilitates mixing of water with dissolved organic carbon, forest debris, and microbial content creating components ideal for the methylation processes within the watershed.

Stump harvest and ordinary site preparation did not result in a significant increase in mercury levels in Sweden (Eklöf et al. 2013). However, another separate mercury level study conducted found contradicting results revealing relatively high levels of total mercury and methylmercury following site preparation and stump harvest (Eklöf et al. 2013). The data analyses in the second study, revealed that organic carbon has the greatest correlation to

mercury suggesting that organic carbon, hydrology and temperature influence mercury (Eklöf et. al 2013). This finding suggests that the initial harvesting rather than stump harvest removal or site preparation affect the results. (Eklöf et al. 2013).

Industrial logging was not linked to increased methylation or methylmercury in large freshwater lakes in a Quebec study (Lucotte et al. 2016). Also, methylmercury levels in predatory fish were not significantly impacted by logging (Lucotte et al. 2016) and remained at acceptable levels below the mercury fish level findings of Garcia and Carignan (2000).

As a result of fire disturbance, an average pH of 6.5 in lake water within the immediate geographic surroundings was documented (Garcia and Carignan 2000). The range of acidity is variable however, methylmercury concentrations in predatory fish were consistently predicted (Garcia and Carignan 2000). Fire disturbance, which alters lake water pH, contributes to the methylation process. Garcia and Carignan (2005) studied how changes in nutrient levels after forest disturbance could be related to changes in mercury concentrations in lakes. They found increased phosphorus and nitrogen after fire was correlated with increased mercury concentrations in fish (Garcia and Carignan 2005). Kreutzweiser et al. (2008) also found increased nitrogen and phosphorus levels in freshwater lakes after forest disturbance. Kreutzweiser et al. (2008) found both nitrogen and phosphorus have soil mineralization potential after logging practices. Nitrogen cycling is strong between trees to litter layers and soil surfaces with nitrogen pools entering the watershed contributing to nitrogen uptake rates and microbial activity. After logging, phosphorus is exported from

the soil and phosphorus loads increase within surface waters and hydrological pathways of the watershed. Phosphorus loading can increase lake eutrophication, may increase cyanobacterial increasing organic material for deposition of mercury in the watershed (Krutzweiser et al. (2008). These shifts of nitrogen and phosphorus from the forest soil into the watershed are factors which may contribute to wet mercury deposition and the methylation process in the watershed increasing levels of methylmercury.

Since Soil mercury levels within the coniferous forest are relatively high, forest operations in coniferous forest can potentially contribute to greater release of mercury into the environment than operations in deciduous stands (Graydon et al. 2008). Forest operations must consider the potential risks of methylation and mobilization from harvesting and site preparation (Mackereth 2017).

## INTRODUCTION

Mercury is classified as a neurotoxin poisoning humans and wildlife by infiltrating organisms through the absorption of various forms of mercury (Celo et al. 2006). Methylmercury is the most abundant and poisonous form of mercury toxicity and contaminates food webs (Celo et al. 2006). In Ontario 85% of eating restrictions of fish from inland lakes is due to mercury contamination (MOECC 2015). Further challenges facing mercury toxicity and poisoning are bioaccumulation and biomagnification in food webs (Lourie 2003). Consequently, top predators have the highest concentrations in their body tissue (Lourie 2003; Weiner et al. 2006). In northern Ontario, walleye *(Sander vitreus)* and northern pike *(Esox lucius)* are top predatory fish with the highest amounts

of methylmercury in fresh water Ontario lakes (MOECC 2015) which have been studied extensively in methylmercury research (Gandhi et al. 2014). Humans are primarily exposed to mercury poisoning by eating fish contaminated with mercury (MOECC 2015). The effects of mercury poisoning in humans is on a continuum of symptoms ranging from neurological deficits, respiratory complications, reproductive side effects, cardiovascular complications, immune system compromise, hematological complications, fetal abnormalities with birth defects in newborn children, negatively affected growth and development of children to death (Ullrich et al. 2001; Lourie 2003; Gonzalez-Raymat et al. 2017).

Mercury cycles through the atmosphere, water and soil (United Nations Environment Program 2013). Elemental mercury can be released to the atmosphere and subsequently enter aquatic ecosystems through deposition (Gonzalez-Raymat et al. 2017). Methylation is a process occurring within a watershed and allows for inorganic mercury to be converted through natural microbial processes into the toxic form of methylmercury (UNEP 2013). Both watershed characteristics and water chemistry influence methylation and demethylation processes. Small lake size, high dissolved organic carbon, anaerobic bacteria, pH, redox conditions, and sulphide all impact and contribute to oxidation and methylation processes in aquatic ecosystems (Ullrich et al. 2001; Gonzalez-Raymat et al. 2017).

Wetlands are generally sites of methylation and output of mercury (Grigal 2002). Wetlands are associated with output of mercury to surrounding aquatic ecosystems because they generally have high dissolved organic carbon, long

water residence time, and high hydrological connectivity, and their anaerobic conditions favour the activity of sulphate-reducing bacteria associated with methylation of inorganic mercury (Grigal 2002).

Mercury can be deposited on to foliage, soil particles, and organic matter through dry deposition, precipitation, throughfall, and litterfall (Fitzgerald et al. 1998; Graydon et al. 2008; Gonzalez-Raymat et al. 2017). Therefore, forests can act as a sink for mercury (Rajani and Maher 2015). Harvesting causes landscape disturbance associated with changing soil conditions and forest composition. Chemical changes to the soil nutrients after harvesting, such as increased dissolved organic carbon or increased nitrogen, can result in downstream effects to aquatic systems (Kreutzweiser et al. 2008). The removal of trees disrupts the hydrological cycle which can potentially lead to increased mobilization of mercury within a watershed to aquatic ecosystems (Garcia and Carignan 2000; Eklöf et al. 2016). Harvesting can also potentially impact the potential for methylation by increasing soil temperature, increasing carbon from decomposing logging residuals, increasing organic matter (Sørensen et al. 2009), increasing nitrogen and increasing phosphorous (Kreutzweiser et al. 2008).

The risks to environmental contamination from mercury are concerning as mercury is a known toxin. Additional research is required to understand the impact and mitigation required to protect the environment and species health from Methylmercury contamination. Forest operations impact ecosystems and understanding these impacts within the dynamic nature of the environment supports a foundation of knowledge to ensure mitigation measures are

effectively implemented. Investigation into the contributing factors of mercury accumulation, methylation, and mobilization with respect to forest harvesting and supporting environmental conditions contributes to a greater understanding of the mercury cycle. The objective of this study is to investigate the relationship between total mercury concentration in walleye muscle and watershed characteristics including harvest disturbance and lake chemistry. Since coniferous forest cover (Graydon et al. 2008; Drenner et al. 2013; Laacour et al. 2013; Richardson and Friedland 2015; Braaten and de Wit 2016), wetlands (St. Louis et al. 1994; Grigal 2002; Mitchell et al. 2008; Braaten and de Wit 2016; Talbot et al. 2017), small lakes (Grigal 2002), dissolved organic carbon, sulphate, acidic pH (Garcia and Carignan 2000; Ullrich et al. 2001; Grigal 2002; Weiner et al. 2006; Drott 2007; Gonzalez-Raymat et al. 2017), nitrogen, and phosphorous (Garcia and Carignan 2005) have been previously shown to be associated with higher levels of accumulation, mobilization and methylation of mercury, then I predict mercury concentration of walleye to be highest in lakes possessing these characteristics.

# METHODS

## STUDY AREA

The region for this study is in northwestern Ontario which is within the boreal forest. For the purpose of this study, only data from fisheries management zones 1, 2, 3, 4, 5, and 6 (Figure 1) was utilized. These zones occupy similar latitude and climate conditions. Only medium sized lakes from 100 to 6750 hectares were studied to eliminate potential confounding effects of lake size on fish mercury levels. Data from lakes were collected and organized according to lake surface area and the subset of data for this study came from bins 2 (100 to 500 ha), 3 (500 to 1 500 ha), and 4 (1 500 to 5000 ha) from the mentioned fisheries management zones.

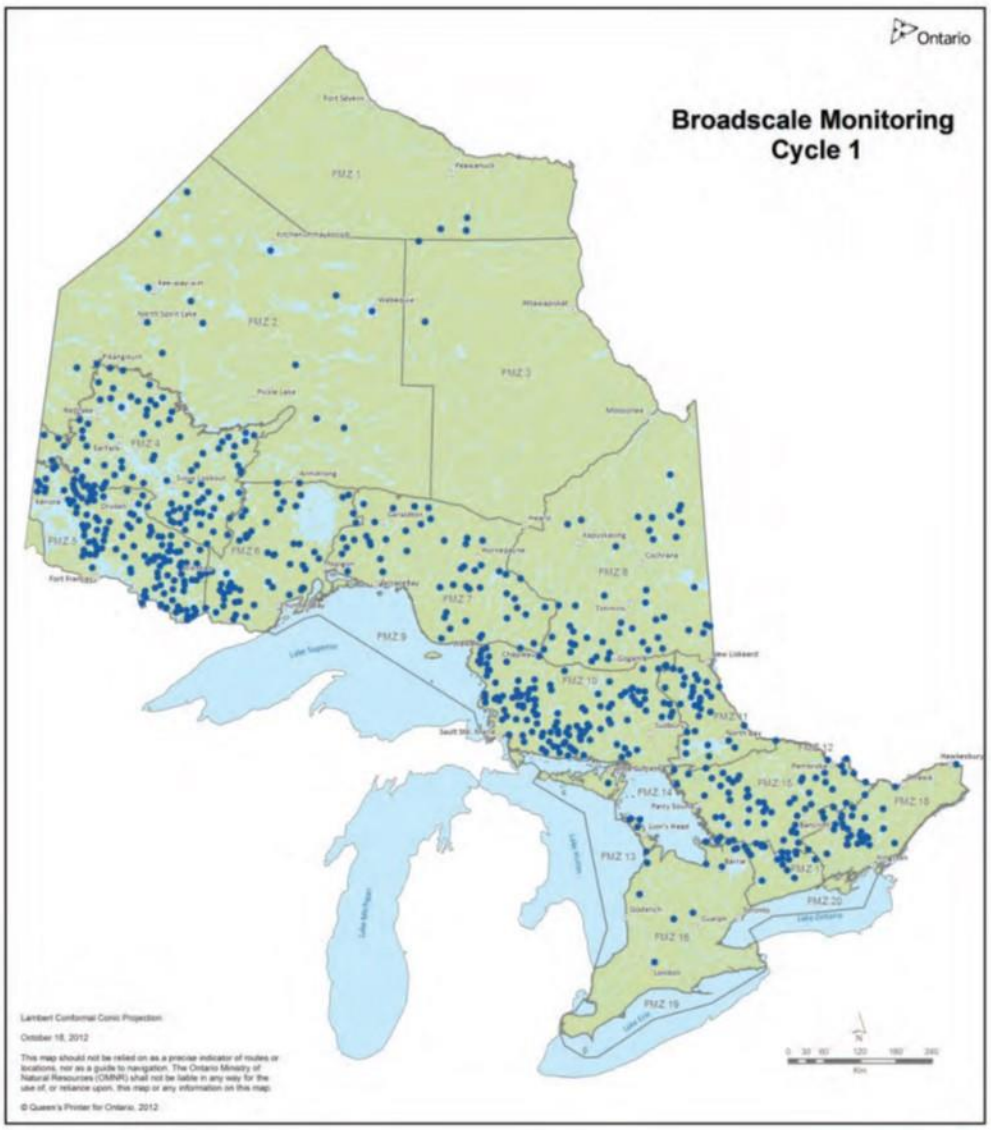


Figure 1. Lakes sampled within fisheries management zones in Ontario's broadscale monitoring cycle 1 (OMNR 2012).

Only mercury concentrations from walleye were considered to reduce data analysis to an amount tractable for the purposes of this study. Walleye are top predators and thus tend to contain high levels of mercury in their tissues and are of concern for human consumption, and thus were considered an

appropriate choice for this study (Weiner et al. 2006). All lakes within the study contain data from walleye.

## FISH MERCURY SAMPLING

Prior to 2004, recreational fisheries were monitored through individual lake management practices. The need for improved monitoring of Ontario's lakes led to the Minister of Natural Resources to initiative the Ecological Framework for Fisheries Management (EFFM) in 2004. This created an active management landscape approach within the Broad Scale Fisheries Monitoring program including regulatory streamlining, public involvement, and a standardized broad-scale fish community monitoring program.

From 2008 to 2017 inclusive, the Ontario Ministry of Natural Resources and Forestry sampled lakes as part of the Broad Scale Fisheries Monitoring Program. The data collection within the sample selection is derived from standard protocols, policies, and procedures which ensure transparency of data collection and the reliability of the data (Sandstrom et al. 2013). Random stratified data collection was implemented to ensure no bias in fish or location sampling was introduced. Ontario was divided into twenty fishery management zones. Every five years, samples of lakes were surveyed in each fishery management zone through random selection which included netting location on lakes set by depth strata (Sandstrom et al. 2013).

To facilitate sampling, netting begins when the water surface temperature reaches 18 degrees Celsius or warmer which typically happens in late May (Sandstrom et al. 2013). Once the temperature drops below 18 degrees Celsius,

data collection stops. Sampling during maximum summer water temperatures over a four to six week period is the ideal time of data collection (Sandstrom et al. 2013).

As part of the Broad Scale Fish Monitoring Program in Ontario, contaminant analysis of fish was conducted by staff from the Ontario Ministry of Natural Resources and Forestry and the Ministry of the Environment and Climate Change. The data collected on the fish, used for this study, was collected during 2008 to 2012 inclusive. Large mesh gill nets were used to collect walleye and northern pike. The nets were left overnight typically set between 13:00 to 17:00 hours and lifted between 08:00 to 11:00 hours the following day. Large nets needed to be immersed for a minimum of sixteen hours and a maximum of twenty-two hours with small nets being immersed for a minimum of twelve hours and a maximum of twenty-two hours (Sandstrom et al. 2013).

Information about each fish was recorded, including length, weight, age assessment, and sampling location. A fish sample from the dorsal muscle tissue was taken for analysis of total mercury. Sample size no smaller than 100 grams was required but tissue samples of 200-300 grams were more ideal since larger samples provide for a more representative and accurate analyses. The sample of muscle tissue was frozen prior to transporting it to the lab in Etobicoke, Ontario for contaminant analysis. The Ministry of Environment and Climate Change, Sport Fish Contaminant Monitoring Program in the Environmental Monitoring and Reporting Branch determined the mercury content in each fish sample (Sandstrom et al. 2013). In top predator fish, approximately 95% of total

mercury concentration measurements account for methylmercury (Celo et al. 2006).

For the purpose of this study, only data from walleye between 35.5 to 58.4 cm in length was used. This range was selected because it is the average length range of walleye. The number of walleye fish included in the study was 2 735. For each lake, an average mercury concentration was calculated on only the walleye in that size range.

The Broad Scale Fisheries Monitoring Program collaborated with the Ontario Ministry of Natural Resources and Forestry and the Ministry of the Environment and Climate Change to collect lake and fish samples. These samples were subsequently analyzed by the Ministry of the Environment and Climate Change to develop databases of mercury contaminant concentrations in fish across Ontario.

LAKE CHEMISTRY DATA

Lake chemistry used in this study was collected by MNRF between 2008 and 2010 as part of the broad-scale fisheries monitoring program. For this study, data from 200 northwestern Ontario lakes was included. Data collection for each of the sample lakes included lake depth, water transparency, and clarity. Lake depth was estimated by bathymetry surveys if the depth was not already known. Water transparency and clarity was measured at the deepest location of the lake by using a Secchi disc and reflected the trophic status of the water body. Lakes with multiple distinct basins required Secchi readings in each (Sandstrom et al. 2013). Lake chemistry data used in this study was also collected by MNRF as

part of the BSFMP, and included dissolved organic carbon (DOC), pH, nitrate and nitrite, dissolved inorganic carbon (DIC), total Kjeldahl nitrogen, sulphate, and phosphorus (Table 1). Temperature and dissolved oxygen were recorded using a digital YSI oxygen/temperature metre at the deepest location of the lake. Standardized depths of 0.5 m, which is considered the surface, were followed by measurements at 1.0 m intervals to 16 m, then at 2.0 m intervals to a depth of 35 m as well as 1 m off the bottom or at the maximum cable length (Sandstrom et al. 2013). Water samples for DOC, pH, nitrate and nitrite, DIC, total Kjeldahl nitrogen, sulphate, and phosphorus were taken shortly after the ice melted from the top of the lake. These samples were tested by the Ministry of the Environment and Climate Change for chemical analysis (Sandstrom et al. 2013).

Table 1. Lake chemistry variables used in this study.

| Variable | Units |
|---|---|
| Dissolved Organic Carbon | mg/L |
| Nitrate and Nitrite | µg/L |
| Dissolved Inorganic Carbon | mg/L |
| Potassium | mg/L |
| Total Kjeldahl Nitrogen | µg/L |
| Sulphate | mg/L |
| Phosphorus | µg/L |

## SPATIAL DATA ACQUISITION

Information on the geographic boundaries of individual lake watersheds was obtained from MNRF based on results of hydrological mapping analysis. Information on the geographic location and size of wetlands was obtained from the provincial wetlands layer using Land Information Ontario (MNRF 2017), supplemented with information on the occurrence of open bog, treed bog, open

fen, and treed fen from forest resource inventories, where available. Data layers representing forest composition coniferous forest, deciduous forest, sparse forest and mixed forest types was obtained based on GIS analysis of provincial forest inventory data. Each of the 13 Northwest Region standard forest units from the Boreal Landscape Guide (OMNR 2014) represented in the FRI was reclassified into one of the seven provincial forest types within The Forest Resources of Ontario (MNRF 2011) to represent broad forest cover types. Forest disturbance data from harvesting was extracted from the Ontario Land Cover 2000. The percentage of each provincial forest type and forest disturbance in each lake shed was calculated using ArcGIS 10.5 software. The variables for analysis included lake surface area, total land area, total forest cover area, conifer forest area, sparse forest area, deciduous forest area, mixed forest area, open fen area, treed fen area, open bog area, treed bog area, total wetland area, and the area disturbed by harvesting.

## STATISTICAL ANALYSIS

All statistical calculations were conducted using IBM SPSS statistics 25 and or Excel 2016. For each lake, the landscape variables including lake surface area, total land area, total forest area, conifer forest area, sparse forest area, deciduous forest area, mixed forest area, open fen area, treed fen area, open bog area, treed bog area, total wetland area, and harvested forest area were divided by the total catchment area to produce a proportional value to represent the amount of area for further analysis.

Outlier analysis using boxplots was used to ensure there were no outliers in the dataset and that each variable was in a normal distribution for further statistical analysis. The following variables were log transformed coniferous forest area, deciduous forest area, open fen area, treed fen area, open bog area, treed bog area, total wetland area, total lake surface area, harvested forest area, phosphorous, pH, nitrate and nitrite, and dissolved inorganic carbon.

Linear regressions were analyzed for each variable separately using the dependant variable of total mercury. All regressions were tested at an alpha level of 0.05 with 95% confidence. For each variable that had a significant relationship with total mercury, a scatter graph was generated to display the linear relationship.

A correlation test was conducted on the landscape variables and the lake chemistry variables together to calculate which variables were correlated to each other. Variables that were correlated were not used together in multivariate regression tests. Several multivariate regressions were conducted with a dependant variable of total mercury. Multivariate regressions were tested at an alpha level of 0.05 with 95% confidence.

# RESULTS

Lake size ranged from 100 to 6,760 hectares (ha). The catchment area for each lake ranged from 318 to 5,080 330 ha. The amount of area harvested with the catchment area ranged from 0 to 280,934 ha. Walleye total mercury ranged from 0.16 to 2.04 ppm. The average total mercury concentration in the walleye was 0.67 ppm in the 200 lakes sampled. Each study lake is listed with its corresponding average mercury concentration and surface area in APPENDIX I.

Lake surface area, total land area, total forest area, coniferous forest area, dissolved organic carbon, pH, nitrate and nitrite, and dissolved inorganic carbon all had a significance value lower than 0.05 indicating there was a significant relationship between those variables and total mercury (Table 2).

Table 2. Summary of linear regression results.

| Variable | Significance | R Square Value | Relationship |
|---|---|---|---|
| Lake Surface Area | 0.000 | 0.098 | Negative |
| Total Land Area | 0.000 | 0.061 | Positive |
| Total Forest Area | 0.004 | 0.041 | Positive |
| Conifer Forest | 0.001 | 0.053 | Positive |
| Sparse Forest | 0.189 | 0.009 | |
| Deciduous Forest | 0.663 | 0.001 | |
| Mixed Forest | 0.395 | 0.004 | |
| Open Fen | 0.763 | 0.001 | |
| Treed Fen | 0.963 | 0.000 | |
| Open Bog | 0.363 | 0.006 | |
| Treed Bog | 0.438 | 0.003 | |
| Total Wetland | 0.128 | 0.012 | |
| Harvested Forest | 0.616 | 0.001 | |
| Dissolved Organic Carbon | 0.000 | 0.089 | Positive |
| pH | 0.000 | 0.164 | Negative |
| Nitrate and Nitrite | 0.000 | 0.081 | Positive |
| Dissolved Inorganic Carbon | 0.000 | 0.138 | Negative |
| Potassium | 0.957 | 0.000 | |
| Total Kjeldahl Nitrogen | 0.704 | 0.001 | |
| Sulphate | 0.309 | 0.006 | |
| Phosphorous | 0.977 | 0.000 | |

The following variables had a significant relationship with total mercury concentration include coniferous forest area (Figure 2), total forest area (Figure 3), total land area (Figure 4), total lake surface area (Figure 5), dissolved organic carbon (Figure 6), pH (Figure 7), nitrate and nitrite (Figure 8), and dissolved inorganic carbon (Figure 9).

There is a significantly positive relationship between coniferous forest area and mercury concentration in fish (Figure 2). The results show that 5.3 % of

mercury concentration in fish is explained by the amount of coniferous forest within the watershed (Table 2). If the proportion of coniferous forest area increases, mercury in fish increases.

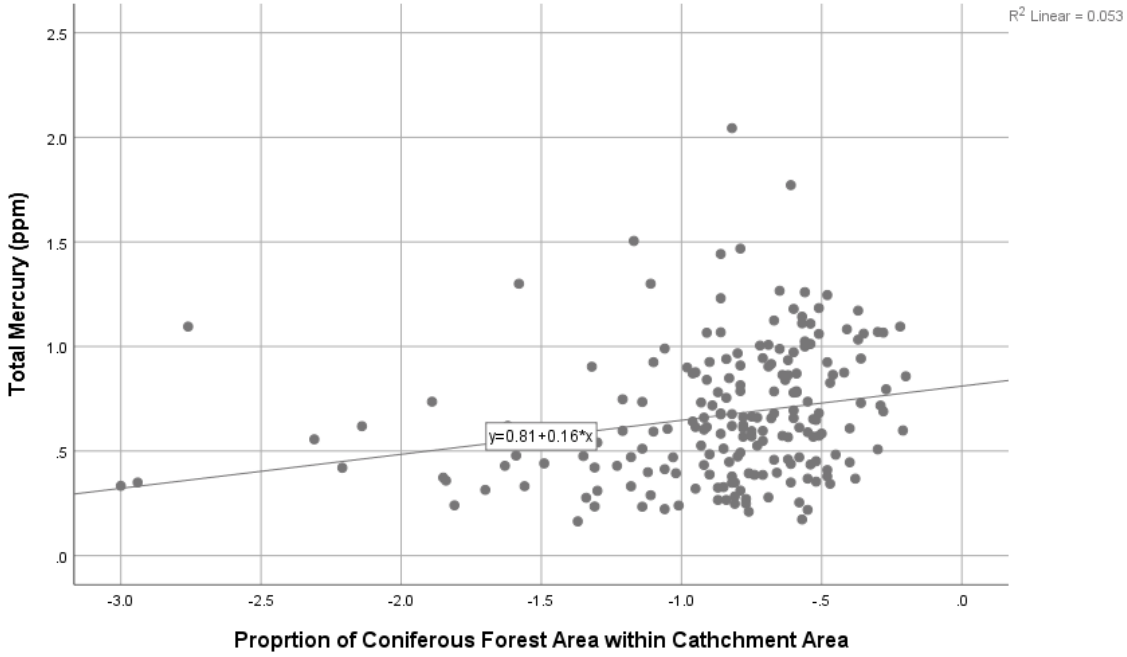


Figure 2. Scatter graph of the liner relationship between coniferous forest and total mercury.

Both the total forest area (Figure 3) and the total land cover (Figure 4) have a significantly positive relationship with total mercury concentration. The results show that 6.1% of mercury concentration is explained by the total land area within the watershed and that 4.1% of mercury concentration is explained by the total forest area within the watershed (Table 2).

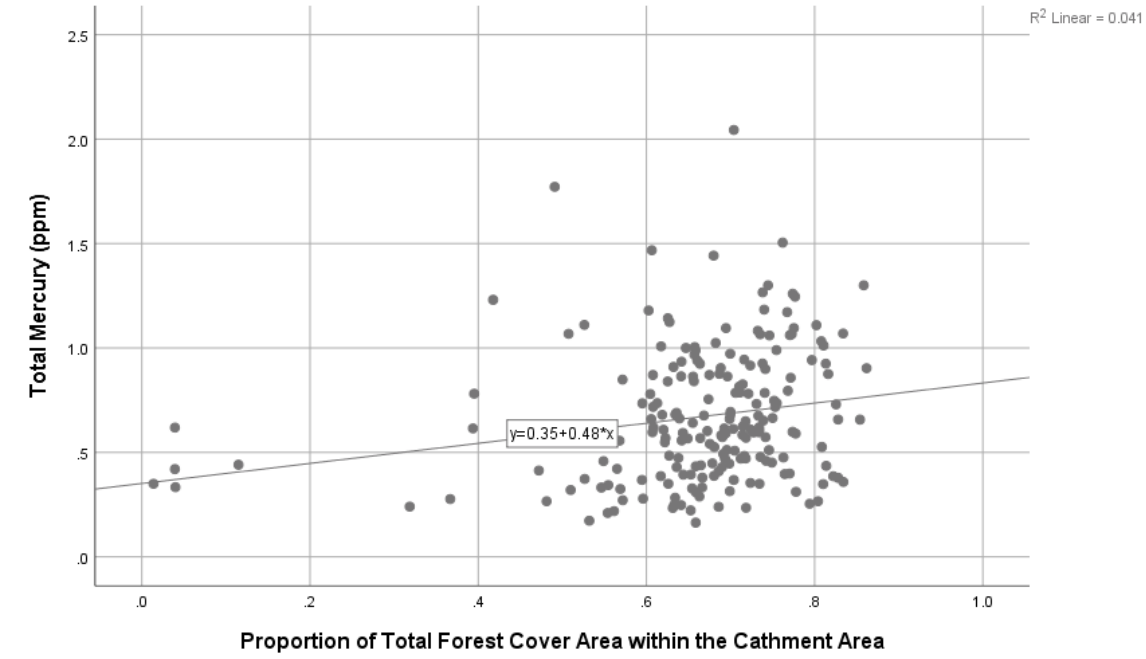


Figure 3. Scatter graph of the liner relationship between total forest cover and total mercury.

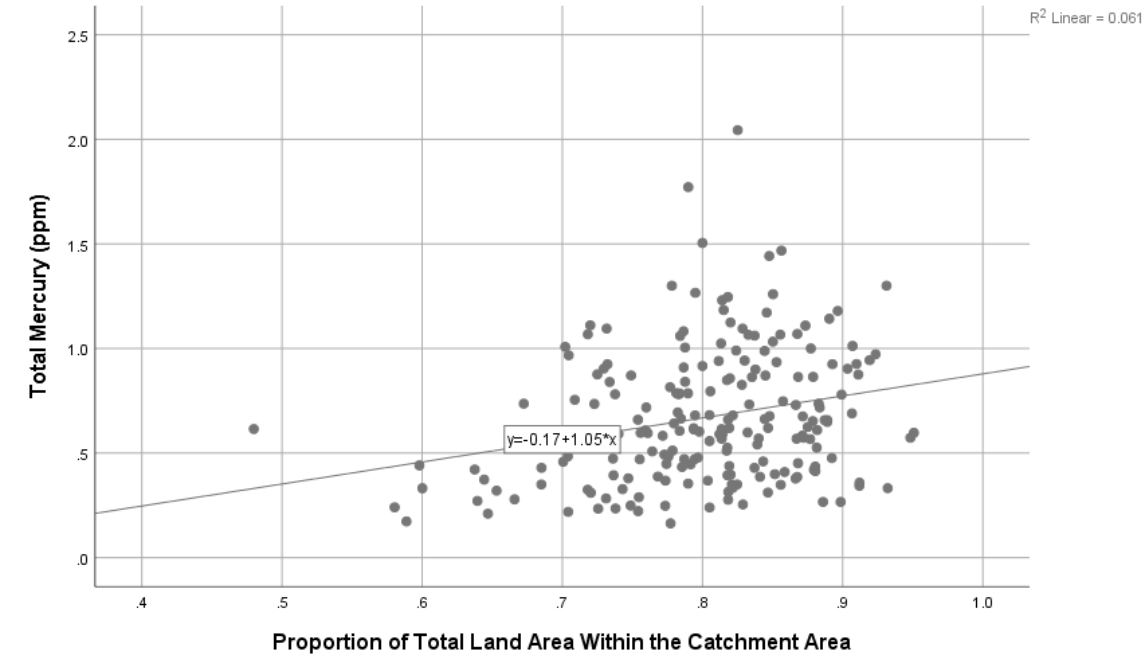


Figure 4. Scatter graph of the liner relationship between total land area and total mercury.

The lake surface area has a significant negative relationship explaining 9.8% of mercury concentration in lake fish (Figure 5 and Table 2).

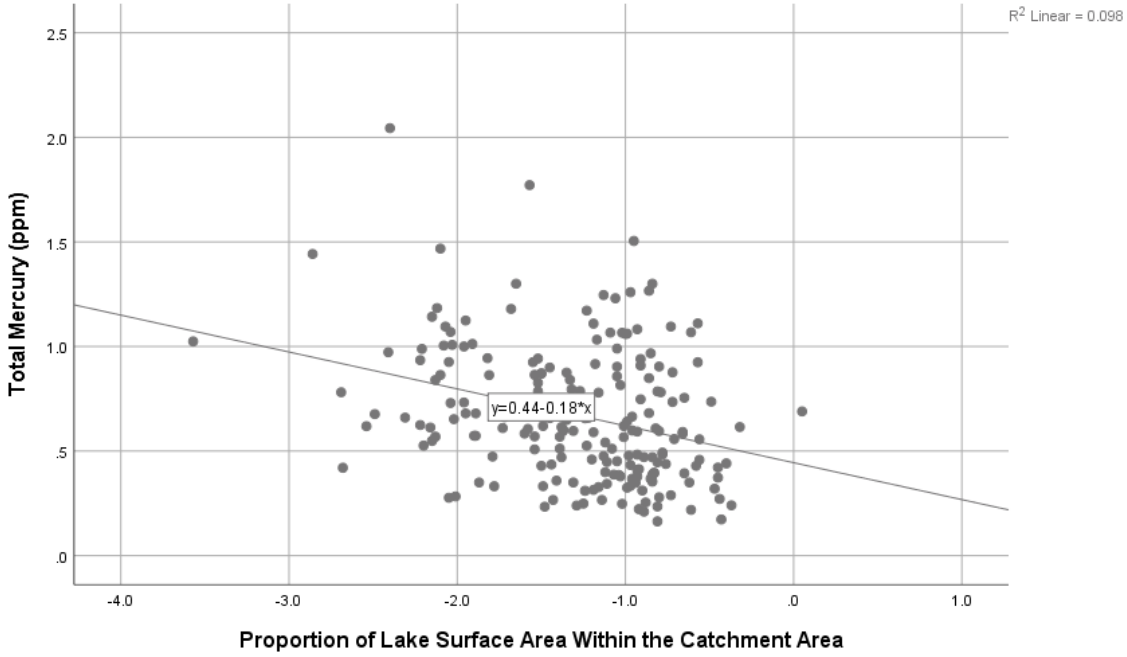


Figure 5. Scatter graph of the liner relationship between lake surface area and total mercury.

In this study, dissolved organic carbon has a significantly positive relationship with mercury concentration (Figure 6). Dissolved organic carbon has an R-square value of 0.089 which explains 8.9% of mercury concentration in fish (Table 2).

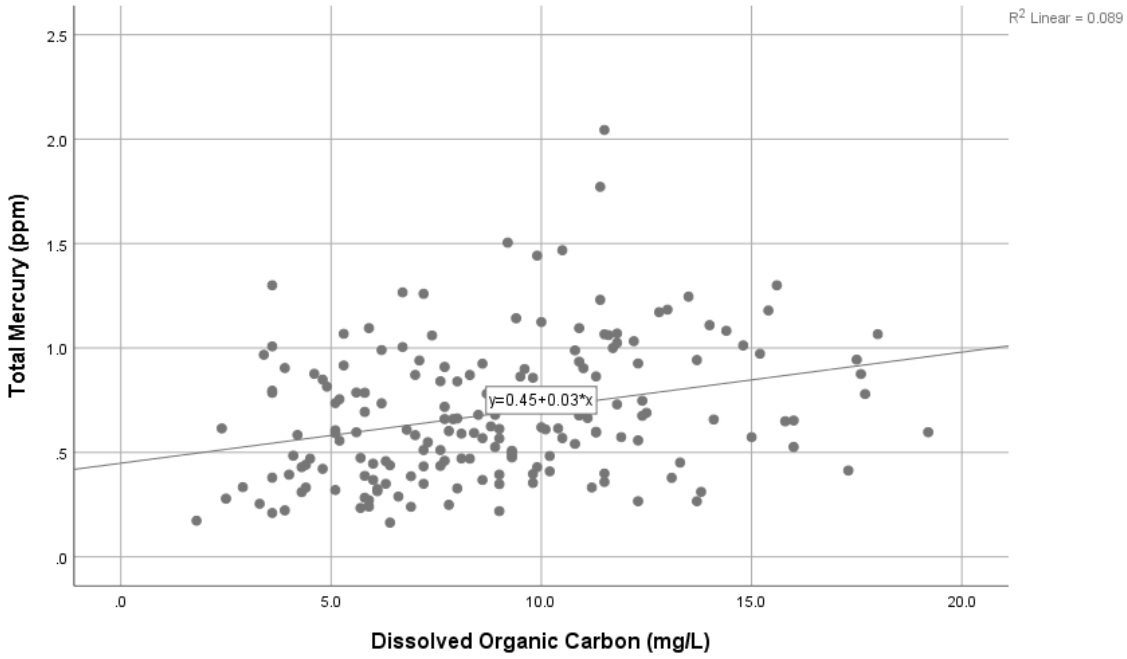


Figure 6. Scatter graph of the liner relationship between dissolved organic carbon and total mercury.

Low pH values are significantly positively associated with mercury concentration (Figure 7). Low pH explains 16.4% of mercury concentration in fish. The R-Square value was highest for this test when conducting the linear regressions

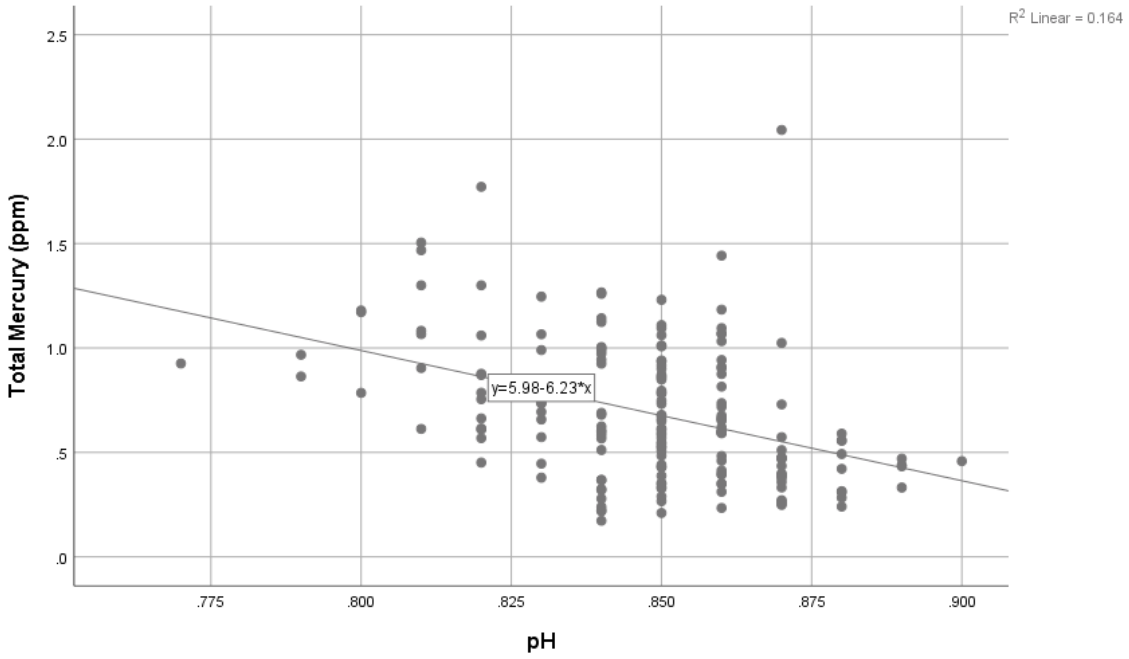


Figure 7. Scatter graph of the liner relationship between pH and total mercury.

In this study, nitrate and nitrite has a significantly positive relationship with mercury concentration (Figure 8). Nitrate and nitrite has an R-square value of 0.081 which explains 8.1% of mercury concentration in fish (Table 2).

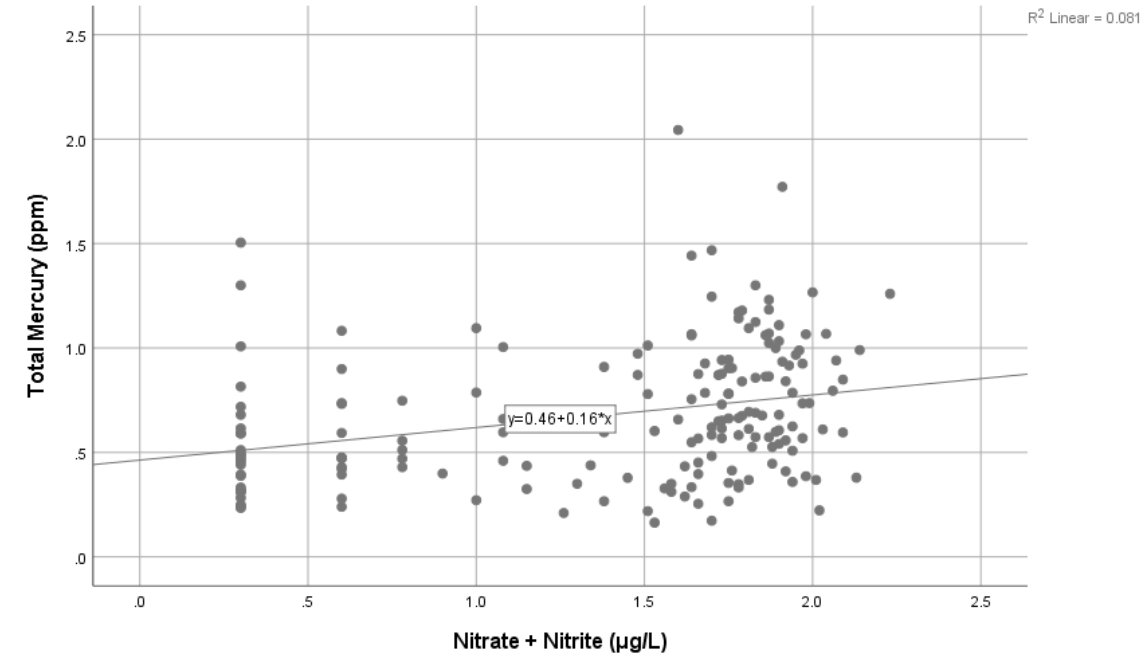


Figure 8. Scatter graph of the liner relationship between nitrate and nitrite and total mercury.

Dissolved inorganic carbon also has a significant negative relationship with mercury concentration (Figure 9). Dissolved inorganic carbon explains 13.8% of mercury concentration (Table 2).

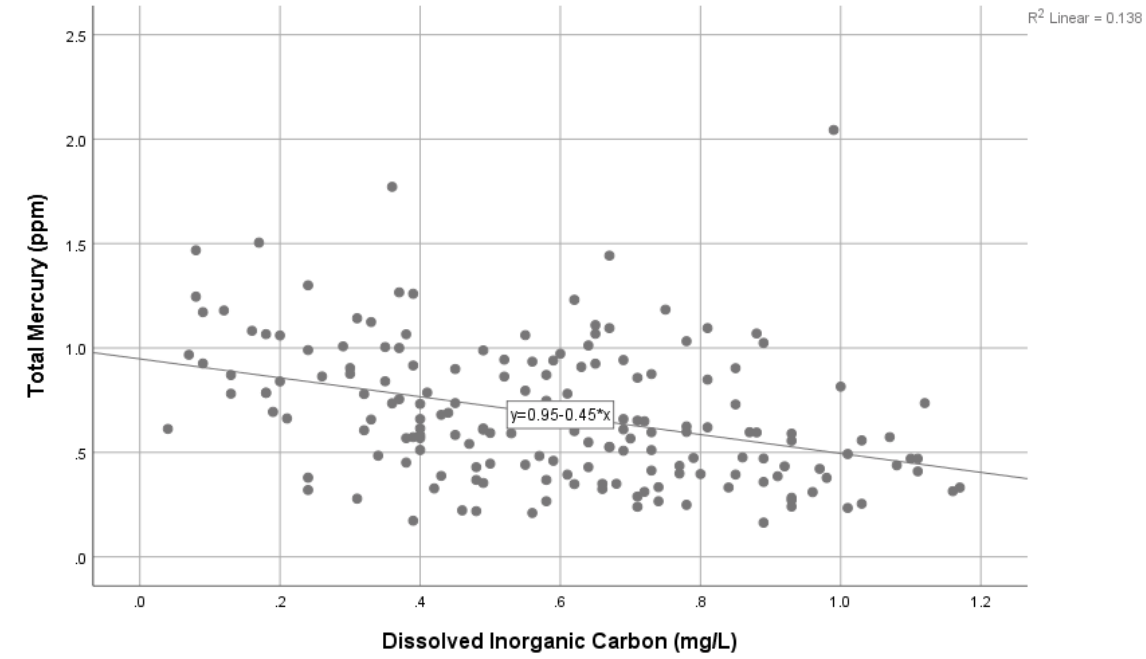


Figure 9. Scatter graph of the liner relationship between dissolved inorganic carbon and total mercury.

In summary, total land area (Figure 4), total forest area (Figure 3), coniferous forest area (Figure 2), dissolved organic carbon (Figure 6), and nitrate and nitrite (Figure 8) were all positively related to total mercury concentration. Lake surface area (Figure 5), pH (Figure 7), and dissolved inorganic carbon (Figure 9) were negatively related to total mercury concentration.

Three multivariate regressions produced a significance value lower than 0.05 indicating there was a significant relationship between the variables and total mercury concentrations (Table 3). Variables included in the significant multivariate regressions included lake surface area coniferous forest area, dissolved organic carbon, pH, nitrate, and nitrate. The multivariate regression

including lake surface area and coniferous forest cover produced a significant relationship explaining 13.3% of total mercury concentration in fish (Table 3). These variables combined explain a greater proportion of the relationship and interaction that impacts mercury levels in fish.

Both the nitrate and nitrite and dissolved inorganic carbon have a significant relationship with total mercury concentration. Their R-Square value was 0.126 from the multivariate regression which explains 12.6% of mercury concentration (Table 3). These variables combined explain a greater proportion of the relationship and interaction that impacts mercury levels in fish.

In this study, dissolved organic carbon and pH explained the greatest variation in fish mercury concentrations with an R-square value of 0.230 (Table 3). This value shows that 23% of mercury concentration in fish is explained by dissolved organic carbon and pH.

Table 3. Multivariate regression results.

| Variable | Significance | R Square Value |
|---|---|---|
| Lake Surface Area and Conifer Forest Area | 0.000 | 0.133 |
| Nitrate, Nitrite, and Dissolved Organic Carbon | 0.000 | 0.126 |
| Dissolved Organic Carbon and pH | 0.000 | 0.230 |

# DISCUSSION

The mean total mercury concentration in the walleye is 0.67 ppm in the 200 lakes sampled. This value exceeds what is deemed acceptable for human consumption, as mercury concentrations below 0.47 ppm are recommended under consumption guidelines (MOECC 2015). Mercury is a neurotoxin and the eating contaminated fish causes significant health complications (Bernhoft 2012). By limiting fish consumption to recommended guidelines, toxity thresholds will not be reached and prevent the manifestation of symptoms of mercury poisoning (MOECC 2015). Continuation of the broad scale fish monitoring program is recommended to proactively protect the public from methylmercury poisoning attributed to ingestion of contaminated fish.

This study found a positive relationship between coniferous forest area and fish mercury, similar to what has been reported in previous studies (Graydon et al. 2008; Drenner et al. 2013; Laacour et al. 2013; Richardson and Friedland 2015; Braaten and de Wit 2016). The relationship between coniferous forest area and fish mercury can be explained by the fact that coniferous trees have a greater leaf area index leading to greater atmospheric deposition of mercury contributing to contamination in lakes where coniferous forests are located within the watershed (Graydon et al. 2008; Drenner et al. 2013). Coniferous foliage also has higher mercury accumulation rates which contribute to the impacts of litterfall and throughfall mercury loads which facilitate a pathway of mercury into lakes supported by Braaten and de Wit (2016) finding dense spruce forest having the highest atmospheric mercury deposition. The

significant relationship between coniferous forests as a predictor of mercury contamination in lakes confirmed by Drenner et al. (2013) and supports the findings of Laacour et al. (2013) and Richardson and Friedland (2015) who identify longer leaf survival as the potentially contributing factor in coniferous trees having higher mercury concentrations. The work of Graydon et al. (2008) reported coniferous forests contributed greater mercury load to the soil although Blackwell et al. (2014) concluded mercury pools were not significantly different between coniferous and deciduous forests. They suggested that a mercury loss mechanism existed to release mercury rather than supporting soil as a sink for mercury. Ullrich et al. (2001) reported sediment was both a sink and a source of mercury. The factors of each mechanism of soil sediment as a sink and a source, remain to be identified and the chemical process of each.

Forest managers need to consider the atmospheric mercury content stored in the forest canopy realizing that mitigation due to forest operations need to be incorporated into the forest management plan. Replanting species and riparian buffer zones need to consider methylation processes and the complexity of interactions around mercury.

Smaller lakes generally contained fish with higher concentrations of mercury, which agrees with a previous study by Grigal (2002) reporting an inverse relationship between lake size and mercury presence. This relationship may be explained by smaller lakes having a less effective transport of mercury to larger watersheds and warmer temperatures (Grigal 2002). However, Grigal (2002) states that this relationship is weak and not always evident.

The finding of a positive relationship between total forest area and total landcover and fish mercury could be attributed to forests and soil acting as a sink for mercury (Fitzgerald et al. 1998; Lourie 2003; Graydon et al. 2008; Gonzalez-Raymat et al. 2017). If mercury stores increase within the total forest area and total land cover of the watershed, the amount of mercury would also increase. Therefore, there is a greater amount of mercury present in watersheds with increased land and forest cover.

The relationship positive relationship between dissolved organic carbon and fish mercury found herein is supported by several other studies including Ullrich et al. (2001), Grigal (2002), and Gonzalez-Raymat et al. (2017). It is generally accepted that dissolved organic carbon has a strong influence on mercury concentration. However, it should be noted that the percent variation in fish mercury explained by dissolved organic carbon was not particularly high in this study (8.9%). The finding of a significant negative association between dissolved inorganic carbon and fish mercury in this study further confirms the relationship of dissolved carbon and mercury.

In this study, pH had the strongest relationship with fish mercury indicating that the lower the pH the higher mercury concentration in fish (16.4%). Several other studies have noted this relationship and support these findings supporting that acidification of lakes increase accumulation of methylmercury (Garcia and Carignan 2000; Ullrich et al. 2001; Weiner et al. 2006). Greater methylation of inorganic mercury Hg (||) occurs due to increased microbial activity of bacteria at a lower pH (Weiner et al. 2006). Further, low pH supports

additional sulfate levels increase methylation through sulfate-reducing bacteria (Ullrich et al. 2001; Drott 2007). Garcia and Carignan (2000) found the positive correlation between mercury levels of fish and sulphate concentration may be related to acidic pH levels and the presence of sulphate reducing bacteria. Weiner et al. (2006) reported sulfate stimulated the methylation process of inorganic Hg(II) by sulfate reduction bacteria. Weiner et al. (2006) researched lake chemistry and fish mercury concentrations of one year old yellow perch (*Perca flavescens*) because they are prey fish that fed on zooplankton and small benthic invertebrates. The yellow perch variation in mercury concentrations was then assumed to be a result of ecosystem processes instead of the effects of increased mercury concentrations from bioaccumulation and biomagnification seen in top predatory fish such as walleye higher in the food chain. Weiner et al. (2006) found three lakes with the lowest fish concentration of mercury had the highest pH.

The contrasting research results between Lucotte et al. (2016) and Garcia and Carignan (2000) regarding pH impacting mercury require further investigation and consideration needs to be given to any number of possible factors that could impact results. Lucotte et al. (2016) looked only at large lakes in Quebec while Garcia and Carignan (2000) examined the effects of logging only in small lakes. Thus, the results of these studies must be considered in light of the potential confounding factor of lake size, which previous studies have shown to be correlated with fish mercury levels (Grigal 2002).

In this study, dissolved organic carbon and pH explained the greatest percent variation in fish mercury concentrations (23%). Ullrich et al. (2001) noted the presence of high dissolved organic carbon is often correlated to the presence of low pH, suggesting a complex interaction of these two factors in influencing fish mercury (Ullrich et al. 2001).

Higher fish mercury levels associated with high levels of nitrogen, as found in this study, have been previously reported by (Garcia and Carignan 2005). Kreutzweiser et al. (2008) also found increased nitrogen and phosphorus levels in freshwater lakes after forest disturbance. Kreutzweiser et al. (2008) found both nitrogen and phosphorus have soil mineralization potential after logging practices. Nitrogen cycling is strong between trees to litter layers and soil surfaces with nitrogen pools entering the watershed contributing to nitrogen uptake rates and microbial activity. After logging, phosphorus is exported from the soil and phosphorus loads increase within surface waters and hydrological pathways of the watershed. Phosphorus loading can increase lake eutrophication, may increase cyanobacterial increasing organic material for deposition of mercury in the watershed (Krutzweiser et al. (2008). Garcia and Carignan (2005) used a nitrogen isotope to identify the trophic position of a variety of fish species in 38 Ontario lakes to research the impacts of forest disturbance on mercury concentrations in the variety of fish studied. Some examples of fish collected for the research included northern pike (*Exox lucius*), walleye, yellow perch, white sucker (*Catostomus commersoni*), whitefish (*Coregonus clupeaformis*), and burot (*Lota lota*).They reported among-group

variations in mercury concentrations of fish is not a result of differenced in biomagnifications power but mercury is higher in fish from logged lakes from higher bioavailability of mercury. Variations in the amount of nitrogen found in lake water with total nitrogen concentrations higher in fire impacted lakes suggests that fire impacts and interferes with the terrestrial nitrogen cycle. The results of Garcia and Carignan (2005) found piscivorous species with high trophic levels in the food chain also have the highest mercury concentrations although nitrogen levels in lakes is another factor contributing to elevated mercury concentrations in fish. Overall, Garcia and Carignan (2005) showed higher levels of mercury concentrations in their study lakes, the average mercury concentration for standardized fish length of walleye in disturbed lakes was higher than 2.0 ppm while in this study the highest mercury concentration is 2.04 ppm and the average is 0.67 ppm.

In this study, wetland ecosystems did not show any significant relationship with mercury concentrations in lakes. This result does not support research findings in other studies showing that wetlands are sources of mercury hotspots (Mitchell et al. 2008) or sources of methylation and mercury output (Grigal 2002). Even though wetlands are reported to have suitable conditions for methylation (Grigal 2002) this study did not reflect that. This result could be due to wetlands in the study area not having a large enough area within watersheds to contribute significantly to fish mercury levels. Further, the analysis of wetland area may not have truly reflected their presence due to the broad scale analysis of this study.

Harvest disturbance also showed no significant impact on mercury concentrations in walleye lake fish. The lack of significant results of harvest disturbance on mercury concentrations in fish could be a reflection that either the proposed potential that harvesting could cause increased mercury mobilization and availability for methylation (Garcia and Carignan 2000; Graydon et al. 2008; Bishop et al. 2009; Sørensen et al. 2009; Braaten and de Wit 2016; Eklöf et al. 2016) does not in fact exist in nature or the mitigation measures forestry operations prescribe to minimize soil disturbances are effective. For example, buffer zones around water features are implemented to minimize erosion and runoff into watersheds and seasonal restrictions such as harvesting in the winter are implemented to reduce soil disturbance (OMNR 2014).

The increased mercury contamination in Ontario's inland lakes may be explained by atmospheric deposition, since mercury is known to have the ability to travel long distances and be deposited into ecosystems in remote locations far from the source of emission (Fitzgerald et al. 1998; Gonzalez-Raymat et al. 2017). To understand what causes atmospheric deposition to occur in certain locations more global studies and comparisons are required. This would also confirm if mercury emissions are the most likely cause of contamination in lakes.

To further understand the factors contributing to mercury contamination in Ontario's inland lakes additional research is needed for further analysis and accuracy. There are still several gaps in knowledge of the mercury cycle including the chemistry of mercury and mercury contamination in the natural

environment (Ullrich et al. 2011). Conclusive research findings continue to report unpredictable results based on previous knowledge as noted in the research by Burns and Riva-Murray (2018). Burns and Riva-Murray (2018) did not find any landscape metrics related to fish mercury concentrations regionally however, did report supporting results for mercury fish concentration levels and chemistry metrics analyzed in the water body. Landscape metrics have previously supported a significant relationship with mercury concentrations in fish. Currently there is no single or set of variables to associate with mercury concentrations environmentally variable. Understanding of the mercury cycle as a whole may be required as multiple factors have an impact of present research findings. The environment is dynamic and anthropogenic influences change over time. The initiatives through legislation to decrease mercury emissions in Canada were effective although a corresponding decrease in mercury concentration in freshwater inland lakes were not immediately realized (Lourie 2003) indicating that mercury processes are complex and mitigating factors do not respond immediately. Further, methylation is not a cause and effect relationship with one specific variable rather a synthesis of the collective body of research whereby one action does not define the outcome (Ullrich et al. 2001).

# CONCLUSION

This study supports that lake chemistry variables and landscape variables including dissolved organic carbon, pH, nitrate and nitrite, dissolved inorganic carbon, coniferous forest area, total forest area, total land area, and total lake surface area played a larger role on the impacts on total mercury concentration in high predatory fish than harvest disturbance and wetlands within the watershed which showed no significant relationship with mercury concentrations. However, the continued monitoring of mercury concentrations in the environment is essential in diverting the neurotoxic consequences on human and species health. Further research is recommended to gain a greater understanding of the environmental factors that contribute to mercury accumulation, methylation and mobilization.

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# APPENDIX I

Table A1. Study lakes.

| Lake | FMZ | Lake Surface Area (ha) | Mean Total Mercury (ppm) |
|---|---|---|---|
| Addie Lake | 6 | 1170 | 0.44 |
| Agnes Lake | 5 | 30118 | 0.90 |
| Amik Lake | 4 | 11616 | 1.11 |
| Andy Lake | 5 | 1627 | 0.47 |
| Arc Lake | 4 | 5963 | 1.10 |
| Arethusa Lake | 4 | 7679 | 0.72 |
| Arrow Lake | 6 | 32344 | 0.33 |
| Arrowroot Lake | 6 | 1771 | 0.53 |
| Athelstane Lake | 6 | 17649 | 0.29 |
| Bad Vermilion Lake | 5 | 17885 | 0.56 |
| Batwing Lake | 6 | 6146 | 0.35 |
| Bawden Lake | 4 | 3762 | 0.94 |
| Beaverhouse Lake | 5 | 19566 | 0.61 |
| Bedivere Lake | 6 | 22256 | 1.07 |
| Bell Lake | 4 | 41618 | 0.35 |
| Bending Lake | 5 | 11471 | 0.84 |
| Berens Lake | 4 | 26617 | 0.97 |
| Bertaud Lake | 4 | 4126 | 1.23 |
| Big Sandy Lake | 4 | 38081 | 0.43 |
| Birmingham Lake | 4 | 2675 | 0.41 |
| Black Lake | 5 | 3317 | 0.66 |
| Black Sturgeon Lakes | 5 | 30707 | 0.60 |
| Blair Lake | 4 | 1261 | 1.25 |
| Blindfold Lake | 5 | 5044 | 0.68 |
| Blueberry Lake | 5 | 1253 | 0.75 |
| Bluffy Lake | 4 | 24870 | 0.65 |
| Blunder Lake | 6 | 1256 | 0.57 |
| Bukemiga Lake | 6 | 7950 | 0.86 |
| Burchell Lake | 6 | 10449 | 0.85 |
| Burditt Lake | 5 | 14483 | 0.23 |
| Burt Lake | 5 | 7369 | 0.97 |
| Bury Lake | 4 | 4738 | 1.07 |
| Butler Lake | 5 | 8469 | 0.48 |
| Canyon Lake | 4 | 16979 | 0.87 |
| Captain Tom Lake | 5 | 1885 | 1.30 |

| Lake | FMZ | Lake Surface Area (ha) | Mean Total Mercury (ppm) |
|---|---|---|---|
| Carling Lake | 4 | 15556 | 1.03 |
| Caviar Lake | 5 | 31703 | 0.47 |
| Cedar Tree Lake | 5 | 5320 | 0.33 |
| Circle Lake | 6 | 3862 | 0.36 |
| Cirrus Lake | 5 | 22158 | 0.99 |
| Clay Lake | 4 | 27554 | 2.04 |
| Coli Lake | 4 | 21142 | 0.22 |
| Confusion Lake | 4 | 14630 | 0.94 |
| Conifer Lake | 4 | 11379 | 1.07 |
| Crayfish Lake | 6 | 5406 | 0.84 |
| Crooked Pine Lake | 5 | 16729 | 0.51 |
| Crowrock Lake | 5 | 18130 | 0.79 |
| Crystal Lake | 5 | 6239 | 0.62 |
| Crystal Lake (2) | 4 | 1158 | 1.01 |
| Cygnet Lake | 5 | 13179 | 0.43 |
| Delaney Lake | 4 | 12781 | 0.21 |
| Dibble Lake | 5 | 10773 | 1.01 |
| Dinorwic Lake | 5 | 49811 | 0.57 |
| Dogpaw Lake | 5 | 19669 | 0.28 |
| Dogtooth Lake | 5 | 26211 | 0.39 |
| Dovetail Lake | 5 | 4991 | 0.61 |
| Edward Lake | 5 | 5343 | 0.25 |
| Elbow Lake | 6 | 3166 | 0.57 |
| Eltrut Lake | 5 | 22698 | 1.00 |
| Empire Lake | 6 | 6814 | 0.37 |
| Expanse Lake | 4 | 8638 | 0.73 |
| Eye Lake | 4 | 1304 | 0.56 |
| Factor Lake | 5 | 6400 | 0.43 |
| Finlayson Lake | 5 | 14592 | 0.68 |
| Fitchie Lake | 4 | 11477 | 0.60 |
| Frank Lake | 6 | 5123 | 0.38 |
| Frazer Lake | 6 | 19679 | 0.16 |
| Gibraltar Lake | 4 | 1093 | 0.66 |
| Godson Lake | 5 | 2595 | 0.40 |
| Gooch Lake | 4 | 1430 | 0.78 |
| Grace Lake | 4 | 4757 | 0.60 |
| Grew Lake | 6 | 2801 | 0.45 |

| Lake | FMZ | Lake Surface Area (ha) | Mean Total Mercury (ppm) |
|---|---|---|---|
| Grey Trout Lake | 5 | 13332 | 0.28 |
| Hailstone Lake | 4 | 5152 | 1.07 |
| Hammell Lake | 4 | 8314 | 1.11 |
| Harmon Lake | 6 | 29473 | 0.68 |
| Hartman Lake | 4 | 5176 | 0.24 |
| Hawk Lake | 5 | 8874 | 0.22 |
| Hawkeye Lake | 6 | 4308 | 0.92 |
| Heathcote Lake | 2 | 11143 | 0.83 |
| Heathwalt Lake | 4 | 8218 | 0.25 |
| Henderson Lake | 6 | 1529 | 0.37 |
| Holinshead Lake | 6 | 19575 | 0.69 |
| Holly Lake | 6 | 3026 | 0.33 |
| I291 Lake | 2 | 5490 | 0.92 |
| Indian Lake NW Zone 4 | 4 | 40001 | 0.60 |
| Irene Lake | 5 | 14281 | 0.38 |
| Jacob Lake | 6 | 1732 | 0.35 |
| Jean Lake | 5 | 13404 | 0.88 |
| Jeanette Lake | 4 | 15807 | 0.48 |
| Jolly Lake | 6 | 1009 | 0.62 |
| Jubilee Lake | 4 | 9775 | 1.06 |
| Kahshahpiwi Lake | 5 | 5376 | 0.79 |
| Kashabowie Lake | 6 | 21633 | 0.61 |
| Kay Lake | 5 | 3467 | 0.87 |
| Kearns Lake | 6 | 9316 | 0.53 |
| Kekekuab Lake | 6 | 5459 | 0.27 |
| Kirkness Lake | 4 | 21446 | 0.61 |
| Kukukus Lake | 4 | 41677 | 0.66 |
| Lac du Milieu | 6 | 1207 | 0.27 |
| Lawrence Lake | 5 | 18773 | 0.57 |
| Lingman Lake | 2 | 5071 | 0.28 |
| Little Metionga Lake | 6 | 7254 | 0.99 |
| Little Sandbar Lake | 4 | 2185 | 0.46 |
| Little Turtle Lake | 5 | 23057 | 0.66 |
| Loganberry Lake | 6 | 4269 | 0.57 |
| Long Lake (2) | 5 | 18269 | 0.31 |
| Longbow Lake | 5 | 6951 | 0.39 |
| Longlegged Lake | 4 | 67603 | 0.91 |

| Lake | FMZ | Lake Surface Area (ha) | Mean Total Mercury (ppm) |
|---|---|---|---|
| Loonhaunt Lake | 5 | 20954 | 0.32 |
| Maggotte Lake | 6 | 1153 | 0.93 |
| Malachi Lake | 5 | 10543 | 0.59 |
| Manion Lake | 5 | 11385 | 0.51 |
| Marchington Lake | 4 | 33693 | 1.18 |
| Marmion Lake | 5 | 39600 | 0.42 |
| Mattawa Lake | 4 | 17673 | 0.57 |
| McCrea Lake | 4 | 40139 | 0.51 |
| Mercutio Lake | 5 | 16688 | 0.90 |
| Metionga Lake | 6 | 19945 | 0.86 |
| Minchin Lake | 4 | 15149 | 0.86 |
| Mount Lake | 5 | 10167 | 0.92 |
| Mud Lake | 4 | 1280 | 0.44 |
| Muskrat Lake | 6 | 5030 | 0.33 |
| Nelson Lake | 6 | 6531 | 0.45 |
| Nora Lake | 5 | 16127 | 0.48 |
| North Lake | 6 | 10537 | 0.25 |
| Nym Lake | 5 | 17925 | 0.32 |
| Obonga Lake | 6 | 37303 | 0.61 |
| Onnie Lake | 4 | 1648 | 1.77 |
| Otatakan Lake | 4 | 15197 | 0.88 |
| Other Man Lake | 5 | 1754 | 0.47 |
| Pelicanpouch Lake | 5 | 11252 | 0.54 |
| Penassi Lake | 4 | 14461 | 0.66 |
| Perch Lake | 5 | 7607 | 1.44 |
| Perreault Lake | 4 | 33021 | 0.47 |
| Pettit Lake | 5 | 11130 | 0.79 |
| Pickerel Lake (2) | 5 | 6018 | 0.74 |
| Pine Lake | 1 | 2988 | 0.62 |
| Poohbah Lake | 5 | 15170 | 0.75 |
| Populus Lake | 5 | 6941 | 0.59 |
| Premier Lake | 4 | 1522 | 0.55 |
| Press Lake | 4 | 36458 | 0.93 |
| Rawn Reservoir | 5 | 2096 | 0.48 |
| Redhead Lake | 2 | 4171 | 0.86 |
| Richardson Lake | 4 | 1948 | 0.27 |
| Rock Lake | 5 | 6425 | 0.24 |

| Lake | FMZ | Lake Surface Area (ha) | Mean Total Mercury (ppm) |
|---|---|---|---|
| Rude Lake | 4 | 5039 | 0.78 |
| Rugby Lake | 5 | 10205 | 0.58 |
| Saganagons Lake | 5 | 24700 | 0.35 |
| Sandbeach Lake | 5 | 6666 | 1.26 |
| Sandford Lake | 5 | 29121 | 0.17 |
| Sandison Lake | 6 | 3104 | 1.47 |
| Sandstone Lake | 6 | 7299 | 0.39 |
| Sarah Lake | 5 | 9446 | 1.30 |
| Savanne Lake | 6 | 3650 | 0.39 |
| Savoy Lake | 4 | 5557 | 0.58 |
| Schistose Lake | 5 | 3383 | 0.31 |
| Scotch Lake | 5 | 12544 | 1.06 |
| Selwyn Lake | 4 | 10033 | 1.14 |
| Shabu Lake | 4 | 7091 | 1.08 |
| Shamattawa Lake (Winisk River) | 1 | 9837 | 0.42 |
| Shebandowan Lake, L. | 6 | 59717 | 0.24 |
| Silcox Lake | 4 | 8739 | 0.40 |
| Silver Lake | 4 | 1528 | 0.80 |
| Singapore Lake | 4 | 5920 | 0.78 |
| Smye Lake | 4 | 2836 | 1.17 |
| South Scot Lake | 5 | 3974 | 0.68 |
| Sowden Lake | 4 | 37391 | 1.00 |
| Sparkling Lake | 6 | 12218 | 0.66 |
| Spruce Lake | 4 | 1147 | 0.41 |
| Sunbow Lake | 6 | 5597 | 0.37 |
| Sup Lake | 4 | 6758 | 0.49 |
| Thaddeus Lake | 4 | 15664 | 0.46 |
| This Man Lake | 5 | 3183 | 0.82 |
| Thompson Lake | 5 | 9271 | 0.59 |
| Thunder Lake | 5 | 11183 | 0.44 |
| Tide Lake | 4 | 13738 | 1.02 |
| Titmarsh Lake | 6 | 9683 | 0.45 |
| Totogan Lake | 2 | 27095 | 0.34 |
| Towers Lake | 4 | 1020 | 0.90 |
| Trap Lake | 5 | 2605 | 0.62 |
| Turtle Lake | 5 | 12085 | 0.69 |
| Tutu Lake | 2 | 3287 | 0.68 |

| Lake | FMZ | Lake Surface Area (ha) | Mean Total Mercury (ppm) |
|---|---|---|---|
| Union Lake | 5 | 2785 | 0.73 |
| Upper Medicine Stone Lake | 4 | 10762 | 1.27 |
| Vista Lake | 5 | 5566 | 1.10 |
| Wabinosh Lake | 6 | 17270 | 0.62 |
| Wapesi Lake | 4 | 23509 | 0.65 |
| Wapikaimaski Lake | 6 | 32900 | 1.12 |
| Wasaw Lake | 5 | 1700 | 0.60 |
| Wavell Lake | 4 | 8058 | 1.18 |
| Wawang Lake | 4 | 19627 | 0.39 |
| Weese Lake | 2 | 12626 | 0.72 |
| Weikabinonaw Lake | 6 | 12360 | 0.74 |
| Whitefish Lake (2) | 6 | 28713 | 0.74 |
| Whitemud Lake | 4 | 12414 | 0.94 |
| Wild Berry Lake | 3 | 21566 | 0.35 |
| Wintering Lake | 4 | 16542 | 0.31 |
| Your Lake | 5 | 1617 | 1.50 |
| Zizania Lake | 4 | 5084 | 0.64 |