

**Global stressors, regional impacts: How will climate change influence future cisco
(*Coregonus artedii*) distributions in Ontario? Are sport fish mercury levels affected by
climate?**

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A thesis submitted to the Faculty of Graduate Studies in partial fulfillment of the requirements
for the degree of Masters of Science

Graduate program in Biology

York University
Toronto, Ontario

December 2015

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ABSTRACT

As global air temperatures rise and precipitation events fluctuate as a result of climate change, environmental conditions for many freshwater fish are expected to change. Fish are particularly sensitive to climate change as their distributions and contaminant loads are influenced by water temperatures. My study focuses on two main objectives: (1) how the distributions of cisco (*Coregonus artedii*) may be altered by future climate change and (2) the role of climate and industrial emissions on fish mercury trends in Ontario. Data were obtained from multiple government and open sources. Future cisco occurrence models demonstrated a decline of 7-47% by 2070. Trend analysis and models of mercury levels in sport fish revealed increasing rates (0.2-0.4 ug/g/decade) within recent years, particularly influenced by changes in local climate. This period of rapid environmental change demands further investigation, to better inform fisheries management decisions and consumption advisories at various spatial and temporal scales.

ACKNOWLEDGEMENTS

To those integral mentors of this Master's degree: Thank you firstly to my supervisor, Dr. Sapna Sharma. I remember first joining Sapna's newly formed lab in September 2013, feeling nervous yet excited to begin whatever was needed to earn a graduate degree. Little did I know how much growth and character-building would occur in just two years! Good mentorship is necessary to navigate steep learning curves. Blessedly, Sapna was there each step of the way. From phone calls to emails to being present in the lab, my supervisor always made sure we knew she was close if we needed. I have learned so much under Sapna's guidance, not only about the biology, but of who I am as a scientist and a person. Thank you a thousand times over, Sapna.

Thank you to Dr. Dawn Bazely, committee member and mentor-extraordinaire! Observing the Twitter Queen at work has helped me to grow into a scientist who is unafraid to express her opinions about current issues in a public platform. One of my biggest cheerleaders, this journey would not have been possible without the support and encouragement of my committee member. Thank you, Dawn.

Thank you also to the guidance and mentorship from Dr. Satyendra Bhavsar of the Ontario Ministry of the Environment and Climate Change. Satyendra, you are such a patient, supportive, brilliant scientist and teacher! Though travelling to the MOECC office was often painful from Scarborough, it was always worth it to receive your guidance and input.

To the Sharmites a.k.a. the poor people who I asked to sit through all my presentations and read though all my drafts: Thank you for not only doing it, but *happily* doing it. Thomas Van Zuiden (a.k.a. Thomcat), my friend and partner through the fires and flames of this degree. Thank you for being your funny, sarcastic, considerate self—I will miss the conversations we had driving to Dorset, ON, looking out for turkeys on the road. Let me know when you begin that blog. Samantha Stefanoff (a.k.a Samanchi), my friend, sounding board, and Co-chair for

OE3C 2015! Thank you for helping me get through the hard days and supporting me in the good days. You give awesome pep talks. To the other current (Katrina Gaibisels, Lianna Lopez and Bailey Hewitt) and former Sharmites (Guang Zhang and Sara Masood), who together have made this lab such a warm place to work: I will never forget the late nights, snack-sharing, fridge-cramming days. I am honoured to have worked with each of you—thank you! Special thanks to the lab neighbours from the Lortie lab (Amanda Liczner, Ally Ruttan, Alex Filazzola, Taylor Noble, Diego Sotomayor) and Stutchbury lab (Sean Chin, Heidi Van Vliet, Tehmeena Chaudhry) for all their support and feedback as well.

To the family and friends who also endured many rounds of presentations and writing: I eternally thank you for the constant support and love you showered on me through the highs and lows of this degree. I love you Mom, Dad, Cheryl and Kenrick!

And of course, anyone who knows me knows that I am a deeply spiritual and religious person. So I must give thanks, praise, and glory to my Lord Jesus Christ. For everything. Period.

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GENERAL INTRODUCTION

Over the two decades, anthropogenic-driven climate change has been rapidly altering our natural environments (IPCC, 2013). Mean global temperature has increased by 0.8°C since 1880, two-thirds of which occurred between 1979 to 2005 at a rate of 0.15-0.20°C per decade (IPCC, 2013; Walther *et al.*, 2002). In North America, the frequency or intensity of heavy precipitation events has also likely increased (IPCC, 2013). Global temperatures are predicted to further increase between 2.0-4.0 °C and frequency of global precipitation events are expected to rise by the end of this century (IPCC, 2013). These increases in temperature and changes in precipitation events have major implications for ecosystem functioning including: phenological deviations (Magnuson, 2002; Parmesan and Yohe, 2003; Burrows *et al.*, 2011; Sharma and Magnuson, 2014), extirpation of local species (Thomas *et al.*, 2004), species range shifts (Rahel, 2002; Parmesan, 2006; Schindler, 2001; Heino *et al.*, 2009), introduction of invasive species (Rahel, 2002; Sharma *et al.*, 2009), and even increased bioaccumulation rates of fish mercury (Hg) (Dijkstra *et al.*, 2013; Pack *et al.*, 2014).

Climate change has been facilitating the movement of native and invasive species into new habitats (Sharma *et al.*, 2009; Alofs *et al.*, 2014). Range shifts of past, current and future distributions of fish due to climate change have been examined by, amongst others Chu *et al.*, 2005; Sharma *et al.*, 2009; and Alofs *et al.*, 2014. Hickling *et al.* (2006) reported that the mean northern range limit of 15 freshwater fishes in the United Kingdom shifted north between 33 and 51 km over a 25 year period. Many Ontario fish species currently occur in lakes farther north than they did historically (Alofs *et al.*, 2014). In particular, the distribution of coldwater fish has been a key indicator of how increases in water temperature may impact aquatic ecosystems. Cisco (*Coregonus artedii*) is an important coldwater forage fish and is a vital prey resource to

top predators such as lake trout (*Salvelinus namaycush*), northern pike (*Esox lucius*) and walleye (*Sander vitreus*) (Jacobson *et al.*, 2010). Research by Sharma *et al.* (2011) and Fang *et al.* (2012) suggests that cisco populations will be vulnerable to changes in water temperature under climate change. Projections of cisco populations in Wisconsin and Minnesota have suggested that this species is at risk of extirpation within lakes vulnerable to both climate change and the invasive warmwater fish, rainbow smelt (*Osmerus mordax*) (Sharma *et al.*, 2011; Fang *et al.*, 2012). Shifting aquatic community structures due to climate change have several implications on fisheries both economically and ecologically.

The influence of climate change on mercury (Hg) contamination in aquatic ecosystems is not currently well understood. Hg is a toxic, global pollutant that can have adverse implications for human health (Morel *et al.*, 1998; Grimalt *et al.*, 2010; UNEP, 2013). The primary pathway of human exposure to Hg is through fish consumption (UNEP, 2013). Despite declines in anthropogenic Hg emissions locally, fish Hg levels have recently begun increasing in Ontario (Monson *et al.*, 2011; Tang *et al.*, 2013; Gandhi *et al.*, 2014), and this is correlated with temperature and precipitation. Temperature and precipitation have been linked to the transport, solubility and bioaccumulation of Hg in aquatic environments (Grimalt *et al.*, 2010; Berg *et al.*, 2013). With increased precipitation and rising temperatures, we may expect to observe higher Hg levels in the food web, and thus our fish (Bodaly *et al.*, 1993; Canário *et al.*, 2007; Stern *et al.*, 2012). In order to understand how a global stressor such as climate change may impact fish Hg levels, and thus human health, we first need greater understanding of the relationship between fish Hg and climate at various temporal and spatial scales.

This thesis has two main objectives: (1) To examine how the distributions of cisco (*Coregonus artedii*), an important coldwater forage fish, may be affected by climate change in

the future; (2) to investigate the influence of climate change and industrial emissions on fish Hg trends in native coolwater (walleye, *Sander vitreus*; northern pike, *Esox lucius*) and warmwater (smallmouth bass, *Micropterus dolomieu*; largemouth bass, *Micropterus salmoides*) sport fishes in Ontario. With increasing water temperatures and precipitation events, climate change is predicted to limit the availability of preferred thermal habitat for cisco and increase the concentrations of Hg available for uptake by freshwater fish (Ficke *et al.*, 2007). We hypothesize that (1) with warmer waters and reduced concentrations of dissolved oxygen, cisco may become extirpated from their current ranges (Ficke *et al.*, 2007; Fang *et al.*, 2012) and (2) fish Hg levels will rise in Ontario's inland lakes (Canário *et al.*, 2007; Stern *et al.*, 2012; Dijkstra *et al.*, 2013; Pack *et al.*, 2014). We hope these studies help further inform future fish management strategies and fish advisories of changes that may occur to wildlife that live in multiple stressor environments.

CITATIONS FOR CHAPTERS SUBMITTED FOR PUBLICATION

This thesis was written as a series of two manuscripts, in collaboration with co-authors from Dr. Sapna Sharma's lab that have been submitted to journals for consideration for publication. The following is the citation list, including co-authorship from this thesis:

Van Zuiden*, T.M., Chen*, M., Stefanoff, S., Lopez, L., & Sharma, S. 2015. Projected impacts of climate change on three freshwater fishes and potential novel competitive interactions. *Diversity and Distributions*. In Press. (Chapter 1).

Note 1: * signifies co-first authors of this manuscript.

Note 2: All walleye writing and analysis, smallmouth bass writing and analysis and smallmouth bass-walleye/walleye-cisco occurrence boxplots were completed by T.M. Van Zuiden, S. Stefanoff and L. Lopez respectively. These components of the manuscript were excluded for thesis purposes.

Chen, M., Lopez, L., Bhavsar, S., & Sharma, S. 2015. What's hot about mercury? Examining the influence of climate and emissions on fish mercury levels in Ontario. Will be submitted to *Science of the Total Environment*. (Chapter 2).

Note 1: L. Lopez was responsible for obtaining all climate indices used in this study and running some of the historical/recent Sen's slopes that examine fish Hg levels.

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Chapter 1

Panic at the cisco: Predicting the effects of climate change on cisco distributions in Ontario

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Keywords: climate change, cisco, coldwater thermal guild, range shifts, biotic interactions

Chapter 1 is part of a larger manuscript accepted by Diversity and Distributions for publication.

ABSTRACT

Aim: As global air temperatures continue to rise in response to climate change, environmental conditions for many freshwater fish species will change. Warming air temperatures may lead to warming lake temperatures, and subsequently, the availability of suitable thermal habitat space. Our objectives are to identify the response of a coldwater forage fish to climate change in Ontario. We focus on lakes in Ontario because it is a dynamic region that encapsulates the southern range extents of coldwater fish species.

Location: Ontario, Canada

Methods: Using lake morphology, water chemistry, climate and fish occurrence data for cisco (*Coregonus artedii*), we modelled the occurrence rates in 2050 and 2070 under 126 scenarios of climate change.

Results: Cisco occurrence rates were predicted to decline by 8-37% ($\bar{x} = 20\%$) by 2050 and 7-47% ($\bar{x} = 26\%$) by 2070.

Main conclusions: These results highlight a unique response to climate change: range contraction for coldwater fish species. Alterations in distributions of this ecologically important fish species may lead to shifts in fish community structure and novel species interactions in Ontario lakes, exacerbating the vulnerability of native coldwater fish to climate change.

INTRODUCTION

Climate change and biological invasions are two major threats to biodiversity (Sala *et al.*, 2000) and the interacting impacts of multiple environmental stressors may further increase vulnerability of ecosystems (Sala *et al.*, 2000; Rahel and Olden 2008). As climate warms, species have been observed to shift their range northwards, including the Australian *Banksia* flowers at rates of 5 km per decade (Fitzpatrick *et al.*, 2008), voles in Yosemite National Park at elevation rates of 50 m per decade (Mortiz *et al.*, 2008), and marine commercial fishes, including cod, halibut, sole and herring (Mueter & Litzow, 2008). As climate warms, invasive species have also moved northwards including deer (Lankester, 2010; Frelich *et al.*, 2012) and warmwater sunfishes (Alofs *et al.*, 2014). In the North American boreal forest, the most dominant herbivore, moose (Karns, 1997), are being replaced at their southern range extent by smaller deer due to the warmer climates in these regions and a brain worm that deer carry, which is lethal to moose (Lankester, 2010; Frelich *et al.*, 2012). In freshwater systems, as global air temperatures rise, lake water temperatures are expected to also increase (Livingstone & Lotter, 1998). Changing water temperatures can influence the distribution of fish across landscapes by altering their available thermal habitat space (Magnuson *et al.*, 1990; Adrian *et al.*, 2009) and may also eliminate barriers that have historically prevented warmwater invasive species from colonising these lakes (e.g. Vinebrooke *et al.*, 2004; Rahel & Olden, 2008). The combination of climate change and invading warmwater sunfishes has been associated with large declines of cyprinids (Jackson & Mandrak, 2002), salmonids (Vander Zanden *et al.*, 1999), and percids (Fayram *et al.*, 2005) in northern lakes.

Our research focuses on how future changes in climate may modify distributions and potential future interactions of fish that prefer cold water (~15°C) habitats (Magnuson *et al.*,

1990). Fish from this habitat guild are expected to respond differently to climate change than those found in other thermal guilds (e.g. Shuter *et al.*, 2002; Chu *et al.*, 2005). Within the past 30 years, sport fish in Ontario have shifted their range northwards by 12.5-17.5 kilometres per decade while baitfish have shifted southwards in response to a changing climate and species interactions (Alofs *et al.*, 2014). Previous research has predicted that coldwater fish will decline from their current ranges and potentially shift their distributions northward under future scenarios of climate change (Sharma *et al.*, 2011; Herb *et al.*, 2014). We aim to predict the impacts of climate change on the coldwater forage fish cisco (*Coregonus artedii*). Alterations in coldwater fish species distributions resulting from climate change may facilitate changes in food web dynamics and ecosystem function with novel biotic interactions, leading to greater competition and predation pressures (Vander Zanden *et al.*, 1999; Sharma *et al.*, 2009). Non-native species expansions, such as smallmouth bass in Ontario can also have devastating effects on native biota, trophic structure, and ecosystem processes (Vander Zanden *et al.*, 1999; Jackson & Mandrak, 2002; Sharma *et al.*, 2009). There are also potential economic consequences that may be associated with the expansion of non-native species into more northern waters, as Canada's recreational and commercial fisheries are worth several billion dollars (Shuter *et al.*, 1998; Post *et al.*, 2002; Dove-Thompson *et al.*, 2011).

Research objectives

The overall goal of this study is to identify the differential response of an ecologically important coldwater forage fish to climate change and how distributional changes may alter the potential for biotic interactions. We focus our study in the province of Ontario, an especially dynamic region that encapsulates the southern range extent of coldwater species. More

specifically, our first objective is to identify the important abiotic and climatic predictors of cisco occurrence in Ontario lakes.

Our second objective is to develop a predictive model to forecast future occurrences of cisco in the years 2050 and 2070. We projected the occurrence of cisco under all 126 IPCC climate change scenarios in order to identify the likelihood of expansion or extirpation of this species by incorporating uncertainties in air temperature and precipitation from each climate model. We hypothesize that cisco populations will become extirpated from their southern extents in Ontario.

METHODS

Background information on cisco

Cisco are a coldwater forage species (preferred water temperature: 8-17°C) that are vital to the growth and success of many top predators (e.g. lake trout) in Canada and the northern United States (Matuszek *et al.*, 1990; Scott & Crossman, 1998; Jacobson *et al.*, 2010). As air temperatures increase under climate change, cisco distributions are predicted to shift northerly and decline from many of their southern extents (Jacobson *et al.*, 2010; Sharma *et al.*, 2011; Fang *et al.*, 2012). Increased mean annual air temperatures can lead to longer summer stratification periods and lower dissolved oxygen concentrations in the hypolimnion (Adrian *et al.*, 2009). Cisco prefer cold temperatures of the hypolimnion and become stressed as oxygen levels in this layer are depleted; this forces them to move into warm waters that are unsuitable for growth, survival or reproduction (Aku *et al.*, 1997; Ficke *et al.*, 2007).

Data acquisition: survey and climate data

Historical data were obtained from the Ontario Ministry of Natural Resources (OMNR) Aquatic Habitat Inventory (AHI) for 9885 lakes between 1957 and 1986 (Dodge *et al.*, 1985). The survey collected data on lake geography (latitude and longitude), morphology (i.e. mean depth, surface area), chemistry (i.e. secchi depth, pH) and climate variables (i.e. growing degree days). Additional data were also obtained from the OMNR Broad Scale Monitoring (BSM) Program for 722 lakes between 2008 and 2012 (Sandstrom *et al.*, 2010). Similar variables for lake geography, morphology, and chemistry data were included in this contemporary dataset. Of the 722 lakes, 605 overlapped with the AHI dataset which were subsequently updated to reflect more recent data. With 117 new lakes added from the BSM program, we compiled a dataset of 10,001 Ontario lakes (Table S1). A dataset comprised of 9736 lakes was resolved from the

10,001 as 265 lakes contained incomplete data. Fish occurrence data on 134 fish species from the AHI and 100 species from the BSM were also provided. Surveys in the contemporary period were able to effectively sample for both large- and small-bodied fish with the use of a wider range of gillnet and trapnet mesh sizes, while historical data likely under-sampled smaller fish. Northern regions of Ontario and the Hudson Bay lowlands continue to be undersampled (Minns, 1986; Sandstrom *et al.*, 2010).

Historical climate data and future climate change scenarios were obtained from the Intergovernmental Panel on Climate Change (IPCC) through the Worldclim database (see www.worldclim.org). Historical climate data were represented as climate averages between 1950-2000. Variables included total monthly precipitation, and monthly mean, minimum, and maximum air temperatures (Hijmans *et al.*, 2005). Future climate scenarios for 2050 (average for 2041-2060) and 2070 (average for 2061-2080) were also obtained from the latest IPCC 5 report (IPCC, 2013). Projected air temperature and precipitation values from 19 general circulation models (GCMs) under four greenhouse gas scenarios (representative concentration pathway (RCP) 2.6, 4.5, 6.0 and 8.5) were extracted for 2050 and 2070. These air temperatures and precipitation values are interpolations of average monthly climate data between weather stations summarized on a 1 km² spatial resolution grid (Hijmans *et al.*, 2005). Seasonal climate variables were also calculated to include in the matrix i.e. summer (June, July, and August) average mean temperatures. Eleven of the 19 GCMs projected future climate under all four RCPs for 2050 and 2070, while the remaining GCMs predicted for only select scenarios. Each GCM is unique and calculates climate values based on various assumptions of atmosphere, ocean, sea-ice, and land components (Hijmans *et al.*, 2005; Stocker *et al.*, 2013; IPCC, 2013). The scenarios of future greenhouse gas concentrations (including RCP 2.6, 4.5, 6.0, and 8.5) represent a gradient where

RCP2.6 is the most conservative estimate of future greenhouse gas (GHG) emissions, projecting a decrease in overall emissions by 2100, while RCP 8.5 is the ‘business-as-usual’ scenario, which estimates continuous increases of GHG emissions through 2100 (van Vuuren & Riahi, 2011; Moss *et al.*, 2010; Rojeli *et al.*, 2012). A total of 126 climate change scenarios were used to project cisco occurrence.

Data analysis: cisco occurrence models

We developed logistic regression models for cisco occurrence in Ontario lakes. We divided our combined AHI-BSM dataset (n=9736) into two random and independent subsets: 80% of the dataset was retained for model training, 20% for model validation. Variables were assessed for normality using a Shapiro-Wilk test; surface area, maximum depth, mean depth and secchi depth data were log-transformed to meet the assumptions of normality. Multicollinearity was found to be low ($r < 0.7$) among environmental predictor variables used in each species distribution model. To develop each species distribution model, a forward selection procedure with a dual-criterion ($\alpha = 0.05$ and R^2_{adj}) was used to identify significant environmental predictor variables for cisco occurrence (Blanchet *et al.*, 2008).

We used Receiver Operating Characteristics (ROC) curves to identify thresholds (0 – 1) that maximize the sensitivity (percent of correctly predicted presences) and specificity (percent of correctly predicted absences) of each species distribution model. This procedure is recommended when species presences and absences are not equal within the data (Fielding & Bell, 1997; Sharma & Jackson, 2008). A Cohen’s Kappa statistic for each logistic model was also calculated to assess the model’s predictive power (Fielding & Bell, 1997). All analyses were performed in the R-language environment (R Development Core Team, 2012). Methodological framework spanning from data acquisition to analysis has also been provided (Fig. S2).

Cisco projections under climate change

We predicted cisco occurrences under 126 future climate scenarios for the years 2050 and 2070. We used all possible climate scenarios to incorporate the variability between GCMs and RCPs on fish projections. The probability of cisco occurrence was calculated for each lake by averaging the predicted species occurrence rates under each climate scenario for both 2050 and 2070. Ordinary kriging was performed using ArcGIS 10.1 to illustrate the probability of each fish occurrence across the landscape of Ontario in 2050 and 2070 (ESRI, 2011) under 126 scenarios of climate change. Ordinary kriging is a smoothing process that interpolates the probability of cisco occurrence across landscapes. The probability of occurrence for each pixel across Ontario's landscape was calculated by averaging the probability of occurrence of the nearest 50 lakes.

RESULTS

Cisco occurrence model

Models found that cisco were predicted to occur more frequently in larger, deeper lakes, in cooler regions of Ontario (Table 1). Model validation yielded a classification success rate of 80% was observed from this model (Table 2).

Cisco projections under future climate change scenarios

Cisco were present in 23% ($n_{\text{cisco}}= 2257$, $n_{\text{total}}= 9736$) of lakes. By 2050 and 2070, cisco populations are projected to become extirpated from their southern and east-central range (Fig. 1b, c). Increasing greenhouse gas emissions will further push cisco populations into northern regions of Ontario (Fig. S1). Warming air temperatures correspond to a decline of cisco occurrence ranging from 8 to 37% ($\bar{x} = 20\%$) by 2050, and a loss of up to 7 to 47% ($\bar{x} = 26\%$) by 2070 (Fig. 2).

DISCUSSION

This study highlights the unique response of a fish species from a coldwater thermal guild to climate change in an especially sensitive region where these fishes are near their southern extent. We forecasted how cisco, a coldwater forage fish, may undergo range contractions into the most northern regions of the province. We expect that fish distributions will change faster and at times, in unexpected directions than previously projected by older climate models (e.g. projections made using the IPCC 2001 data). For example, extreme scenario fish projections based on earlier climate models (e.g., Chu *et al.*, 2005; Sharma *et al.*, 2007) are now considered conservative estimates of fish distribution changes as greenhouse gas emissions continue to increase. Even in the past 30 years, increases in mean annual air temperatures have been linked to northerly range shifts of warm- and coolwater sport fish species and southern range contractions of many baitfish at rates much faster than expected (Alofs *et al.*, 2014). Such drastic changes in projections for ecologically important coldwater forage fish will have implications for species interactions and community assembly for lakes in the future.

Future cisco distribution and implications of climate change on biotic interactions

We predict that by 2070, cisco will undergo a southern range contraction and will become extirpated from 80-100% of lakes in southern and central Ontario. In recent studies it has been suggested that cisco are already becoming extirpated from their southern range as a result of climate change and the invasion of coldwater rainbow smelt (Sharma *et al.*, 2011; Fang *et al.*, 2012; Jiang *et al.*, 2012). On average, one fourth of cisco populations could be extirpated by 2070 and remaining cisco populations could be forced into northern Ontario lakes. Cisco populations can only persist if well-oxygenated, cold-water habitat is available as they prefer larger, deeper lakes in cooler geographic regions. Well-oxygenated, coldwater habitat provides

suitable habitat refugia for cisco to grow and reproduce (Rudstam & Magnuson, 1985; Jacobson *et al.*, 2010; Fang *et al.*, 2012). In the late summer, however, these cold, well-oxygenated habitats become threatened, as insufficient dissolved oxygen concentrations in the hypolimnion decreases cisco survival. Cisco are then forced by this temperature-oxygen squeeze (Coutant, 1985; Ficke *et al.*, 2007), where the epilimnion becomes too warm and the hypolimnion becomes hypoxic, into unsuitable regions in the water column, resulting in reduced growth and higher summer kill rates (Becker, 1983; Aku *et al.*, 1997). With climate change predicted to increase water temperatures and decrease hypolimnetic dissolved oxygen concentrations, late summer mortalities of cisco may be exacerbated, leading to devastating losses of cisco populations at the southern extents of their range (e.g., Jacobson *et al.*, 2010; Sharma *et al.*, 2011).

Climate change is expected to impact individual fish populations directly through changes in temperature and precipitation. Changes in food web dynamics and ecosystem function may also be facilitated by climate change as it creates novel biotic interactions, leading to greater competition and predation pressures (Vander Zanden *et al.*, 1999; Sharma *et al.*, 2007). Non-native species invasions, such as smallmouth bass in Ontario, can have devastating consequences on native biota, trophic structure, and ecosystem processes (Shuter *et al.*, 1980; Vander Zanden *et al.*, 1999; Sharma *et al.*, 2007). Smallmouth bass are voracious predators and have been found to decimate populations of small bodied fish (particularly minnows), resulting in homogenization of fish communities (Rahel, 2002; Sharma *et al.*, 2007). Smallmouth bass are able to outcompete native top predators, such as lake trout and walleye, for energetically-rewarding littoral prey fish (Vander Zanden *et al.*, 1999; 2004). As a result, bass decouple food web interactions between coldwater fish predators and their prey (Vander Zanden *et al.*, 1999; 2004; Tunney *et al.*, 2014). When this occurs, alternate prey items must be available to buffer the

effect of bass competition. If pelagic food sources are available (e.g. cisco) then coldwater predators will be able to persist (Vander Zanden *et al.*, 1999; Sharma *et al.*, 2009). If climate change eliminates this buffer, coldwater predators will be forced to feed primarily on zooplankton, leading to slower maturation rates and stunted growth (Vander Zanden *et al.*, 1999). In Ontario, loss of cisco populations due to climate change would leave invaded lakes with no alternative prey sources to buffer the novel pressures of bass competition, resulting in loss of important top predator fish. Ultimately, changes in the distribution and subsequent interaction of cisco and other native and non-native fish species may result in altering trophic structure, resilience to changes in environmental conditions, and homogenization of fish communities under future climate change (Rahel, 2002; Tunney *et al.*, 2014).

Conclusion

With climate change, as lakes warm and non-native fish invade new lakes, the persistence of native fish assemblages becomes threatened. The warming and colonisations can result in homogenised lake communities, which may decrease the profitability of certain fisheries (Rahel, 2002; Jackson, 2002). Smallmouth bass represent a strong competitive pressure to native top predators, in both cool- (e.g. walleye) and coldwater (e.g. lake trout) fish guilds. We project that the likelihood of invasion of smallmouth bass and extirpation of walleye and cisco are substantially reduced under conservative climate change scenarios and reduced greenhouse gas emissions (e.g., RCP 2.6). Curbing greenhouse gas emissions is urgently needed to prevent the extirpation of important coldwater fish species from southern lakes and limit the invasion of warmwater predators into northern lakes.

ACKNOWLEDGEMENTS

We would like to thank Nigel Lester for providing updated fish community data and Shekhar Biswas, Brian Leung, and two anonymous reviewers for reviewing an earlier version of this manuscript. Funding for this research was provided by an NSERC Discovery Grant and York University to SS.

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TABLES

Table 1: Coefficients of significant ($p < 0.01$) predictors for the logistic regression model for cisco populations. Mean conditions of environmental characteristics in lakes with fish present and absent.

	Selected Variables	Model Coefficients	Environmental characteristics with fish present	Environmental Characteristics with fish absent
Cisco	Surface area (ha)	1.62	1037.0	148.5
	Mean depth (m)	1.75	7.8	4.9
	Mean annual air temperature (°C)	-0.09	1.9	2.7

Table 2: Classification success, specificity, sensitivity and kappa statistic values of the predictive cisco occurrence model.

	Classification success (%)	Specificity (%)	Sensitivity (%)	Kappa statistic
Cisco	80	87	56	0.43

FIGURES

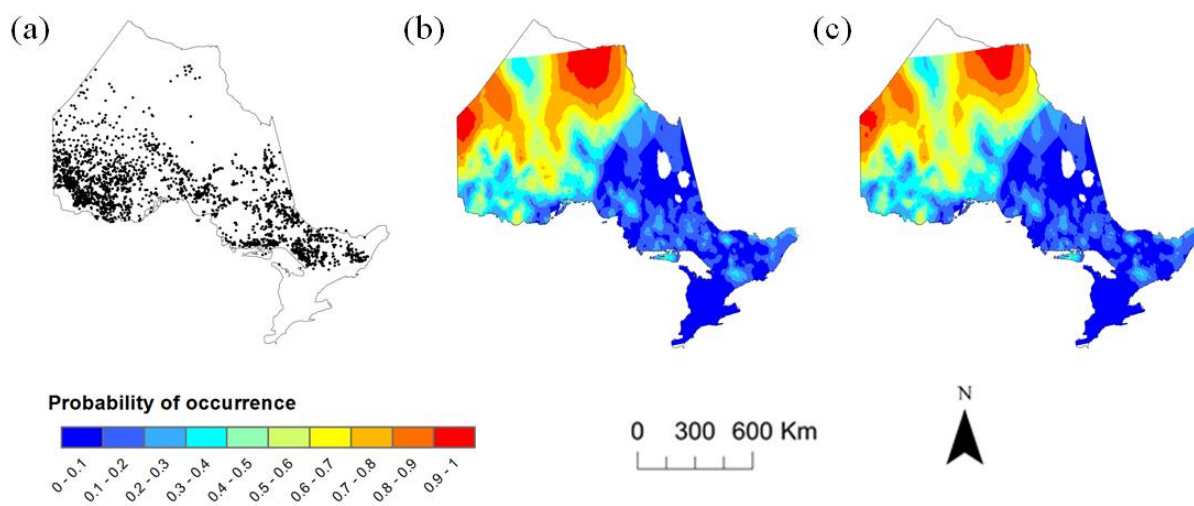


Figure 1: Distributions of cisco (a) historically, (b) in 2050, and (c) in 2070 under 126 scenarios of climate change.

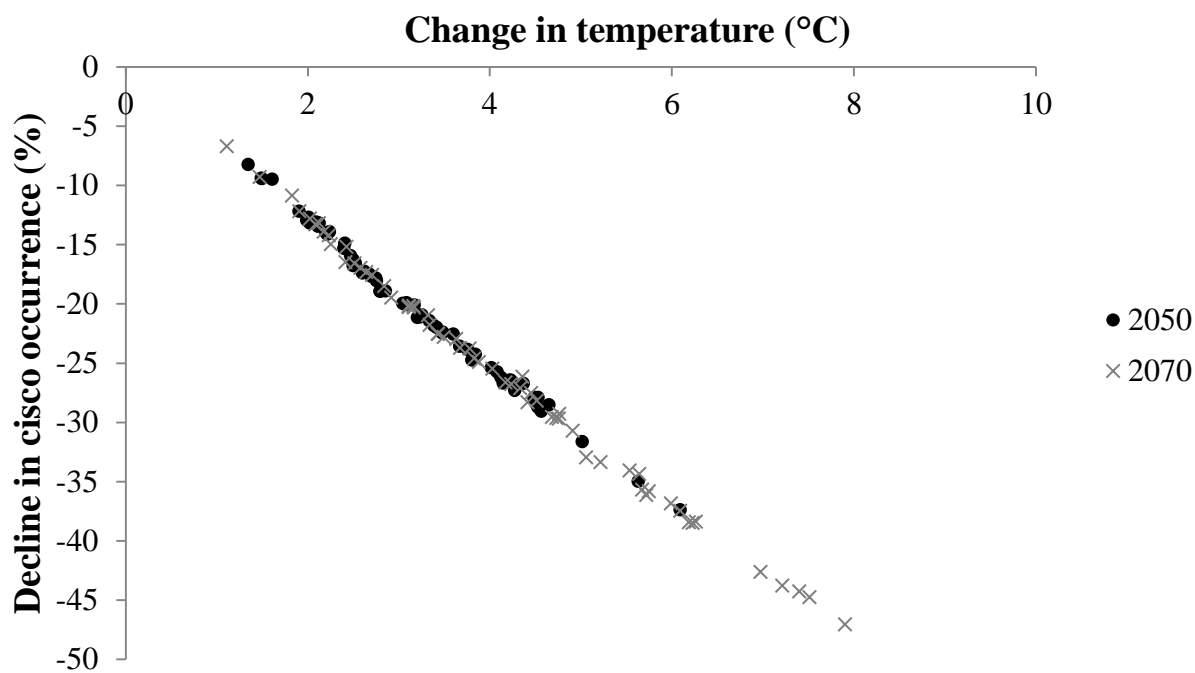


Figure 2: Percent change in cisco occurrence as temperature changes under 126 climate change scenarios in 2050 and 2070.

SUPPORTING INFORMATION

Table S1: Summary of geographic, environmental and climate variables of the lakes in our dataset.

Variable	Minimum	Maximum	Mean
Latitude	42.1	55.3	47.6
Longitude	-95.1	-74.5	-83.6
Surface area (ha)	0.1	79771.6	354.4
Perimeter (km)	0.1	938.3	14.0
Lake elevation (m)	42.0	875.0	338.9
Maximum depth (m)	0.2	213.5	17.0
Mean depth (m)	0.1	47.5	5.6
Secchi depth (m)	0.2	22	3.6
pH	4.0	10.0	7.4
Total dissolved solids (mg/L)	2.0	1294.0	59.1
Conductivity (uS/cm)	3.0	1243.1	86.8
Dissolved Oxygen (mg/L)	0.7	15.4	8.4
Mean July air temperature (°C)	14.1	22.4	17.8
Mean August air temperature (°C)	12.9	21.6	16.4
Mean annual air temperature (°C)	-4.2	9.5	2.5
July precipitation (mm)	55	106	82.2
Mean summer precipitation (mm)	66.7	97.3	85.0

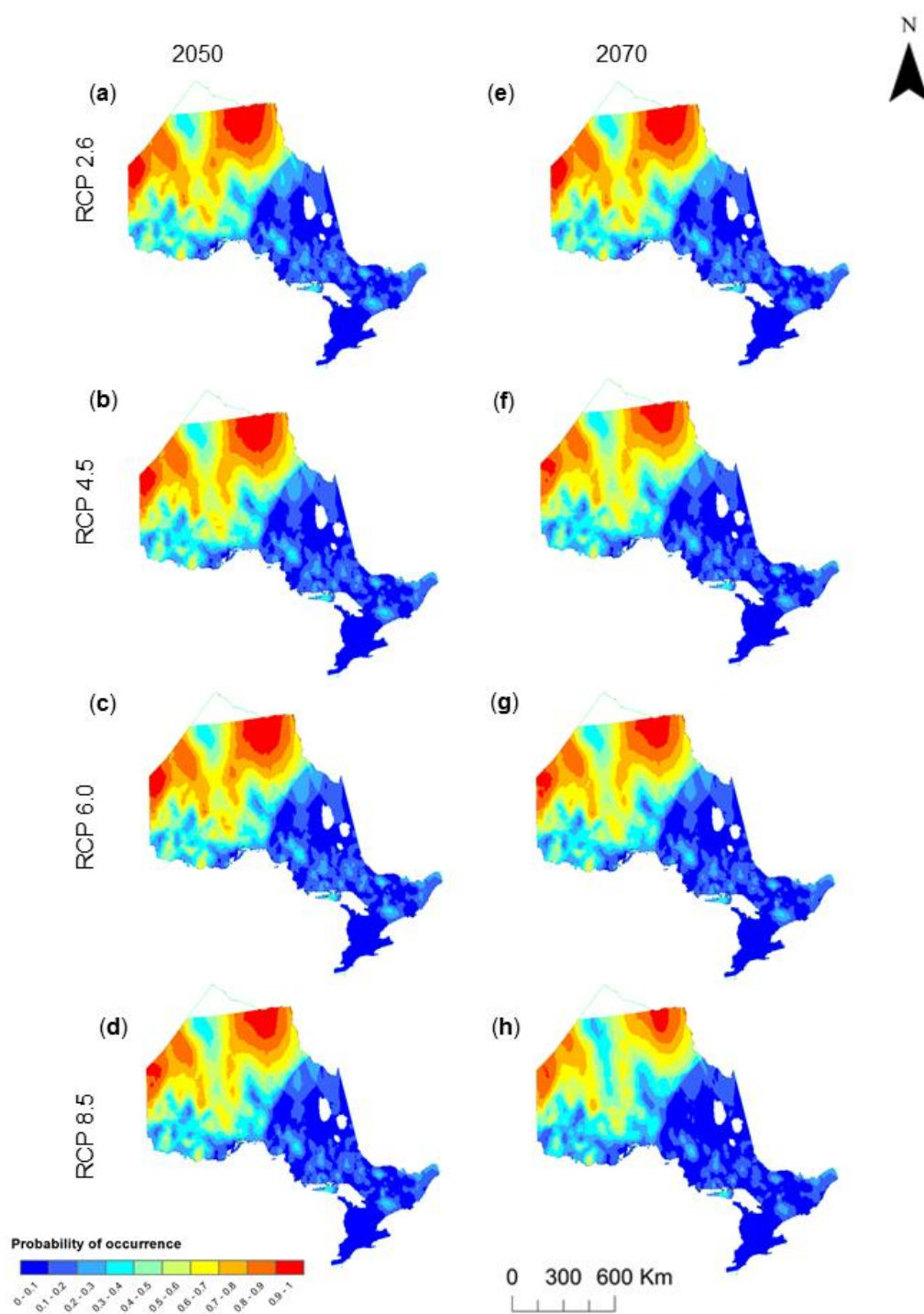


Figure S1: Probability of cisco occurrence for each greenhouse gas scenario in 2050 and 2070.

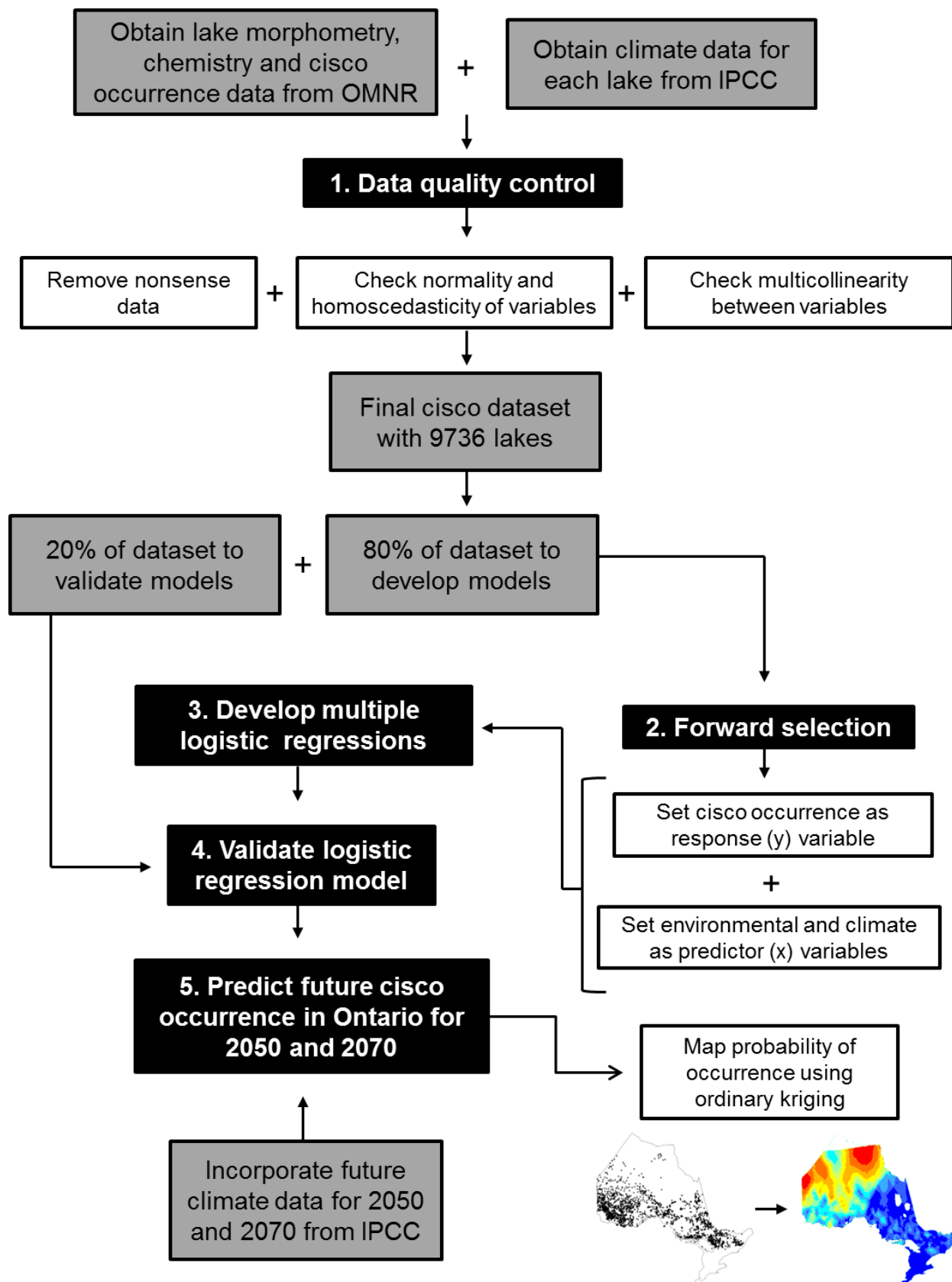


Figure S2: Methodological framework. Grey boxes indicate data or datasets; black boxes indicate data screening or analyses; and white boxes provide greater detail to the data analysis step above. Further details and description of steps can be found in the Methods section.

Chapter 2

What's hot about mercury? Examining the influence of climate and emissions on mercury levels in Ontario top predator fish

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Keywords: climate change, fish, mercury, industrial emissions, local, global, Ontario

Will be submitted to *Science of the Total Environment* for consideration of publication.

ABSTRACT

Mercury (Hg) levels in top predator fish have been increasing throughout Ontario in recent decades. With the onset of climate change, it is important to understand the role of climate in fish Hg patterns. The objective of this study was to examine the relationship between local and large-scale climate factors and Hg emissions with Hg trends in native coolwater (walleye and northern pike) and warmwater (smallmouth bass and largemouth bass) predatory sport fishes during historical (1970-1992) and recent (1993-2014) time periods. The results suggest that over 25% of secondary watersheds in Ontario shifted from historically declining to recently increasing fish Hg trends for all fishes studied. The recent Hg increases ranged between 0.0 to 0.20 $\mu\text{g/g/decade}$; however, the findings were significant only for walleye and northern pike. A complex interplay of local climate, global climate drivers, and Hg emissions influenced fish Hg levels. Although anthropogenic Hg emissions were correlated with fish Hg concentrations for mostly historical periods, local climate and global climate drivers were found to be important in recent years as temperatures and precipitation have more rapidly increased in Ontario. Distinguishing the effects of climate-related parameters and emissions are increasingly crucial to assess how changing multiple environmental stressors may impact health of wildlife and humans consuming fish.

INTRODUCTION

Mercury (Hg) is a toxic heavy metal that can bioaccumulate and biomagnify in the food web, and adversely impact humans through consumption of fish (Mergler *et al.*, 2007; Grimalt *et al.*, 2010). Fish Hg levels have been found to respond rapidly to changes in Hg concentrations within aquatic ecosystems (Harris *et al.*, 2007a), making them effective bio-indicators of Hg contamination in food webs. By the 1970s, fish Hg levels in North America were substantially elevated due to industrial emissions, particularly those from coal-fired power plants (Downs *et al.*, 1998). Though anthropogenic Hg emissions in North America have declined by approximately 75-90% between the 1970s and 2011 (Cain *et al.*, 2007; Risk Management Strategy for Mercury, 2010), recent studies have found that fish Hg levels are again increasing in the Province of Ontario, Canada (Bhavsar *et al.*, 2010; Monson *et al.*, 2011; Tang *et al.*, 2013; Gandhi *et al.*, 2014). This mismatch in the trends of Hg emissions and fish levels suggests that other factors are driving the Hg dynamics.

Fish Hg levels can be influenced directly by lake size (Bodaly *et al.*, 1993), lake acidity, hardness, DOC (Wren, 1989), food chain length (Cabana *et al.*, 1994; Pouilly *et al.*, 2013; Johnson *et al.*, 2015), trophic position (Coelho *et al.*, 2013), species, size, sex (Gewurtz *et al.*, 2011; Karimi *et al.*, 2013) as well as indirectly by forest fires, precipitation and temperature (Bodaly *et al.*, 1993; Coelho *et al.*, 2013; Dijkstra *et al.*, 2013; Pack *et al.*, 2014). The interactions of these factors can be complex, and become more complicated under climatic change (IPCC, 2013). The impacts of climate factors, such as increased temperatures, changes in precipitation, wind patterns or dust deposition, will change the distribution, mobility and uptake of Hg in freshwater ecosystems (Grimalt *et al.*, 2010; Berg *et al.*, 2013; Dijkstra *et al.*, 2013; Evans *et al.*, 2013).

Precipitation is one of the primary pathways through which elemental Hg enters lakes and watersheds, and influences the transport and distribution of Hg between systems (Outridge *et al.*, 2008; Grimalt *et al.*, 2010; Risch *et al.*, 2012). Temperature influences the rates of transformation between chemical Hg species and rates of transport between systems (Grimalt *et al.*, 2010; Pack *et al.*, 2014). With increased precipitation and rising temperatures, we can expect higher MeHg content in the food web (Bodaly *et al.*, 1993; Canário *et al.*, 2007; Stern *et al.*, 2012).

Fluctuations in air circulation patterns, or oscillations of large-scale climate drivers, can also influence local climate (Higgins *et al.*, 2002; Li *et al.*, 2013; Zhao *et al.*, 2013; Evans *et al.*, 2013), local weather patterns, and even support the transport of Hg from distant sources, especially those from Asian industrial locations (Selin *et al.*, 2007). For example, positive phases of El Niño Southern Oscillation (ENSO) and North Atlantic Oscillation (NAO) are associated with warmer winter temperatures (George *et al.*, 2000; Bonsal and Shabbar *et al.*, 2011) and below average winter precipitation in Ontario. Evans *et al.* (2013) found that the Pacific/North American Index (PNA) was important in explaining Hg levels in lake trout in Great Slave Lake, Northwest Territories, Canada. Deposition of atmospheric Hg has historically been the greatest contributor to Hg in fish (Pacyna *et al.*, 2006). Mercury is a global pollutant that can be transported long distances (Engstrom, 2007; Krabbenhoft and Sunderland, 2013). Deposition of Hg in Canada derived from trans-boundary sources now accounts for >95% of total Hg deposition (Risk Management Strategy for Mercury, 2010). Currently the role of Hg emissions in the current fish Hg narrative and climate change context is unclear.

We investigate the relative influence of climate, large-scale climate drivers, and atmospheric Hg emissions on fish Hg levels in native coolwater (walleye, *Sander vitreus*, WE;

northern pike, *Esox lucius*, NP) and warmwater (smallmouth bass, *Micropterus dolomieu*, SMB; largemouth bass, *Micropterus salmoides*, LMB) sport fishes in secondary watersheds of the Province of Ontario, Canada. Specifically, the objectives of this study are two-fold: (1) to examine how Hg levels in Ontario's top predator fish have changed between historical and recent time periods at a secondary watershed level; and (2) to investigate how local climate, large-scale climate and local/global Hg emissions are potentially driving fish Hg trends over time. To our knowledge, this is the one of few studies exploring the changes in fish Hg levels across a landscape and the interacting multiple environmental stressors, including climate change and atmospheric pollution. This study is aimed at providing a framework to critically evaluate observed fish Hg trends under multiple stressors, and is expected to improve our understanding of how intricate processes can impact fish Hg levels and thereby health of fish as well as wildlife and humans consuming fish.

METHODS

Data acquisition and screening

Fish Hg measurements were obtained from the Ontario Ministry of Environment and Climate Change (MOECC) Fish Contaminant Monitoring Program. This program was implemented to issue fish consumption advisories based on measured contaminants in Ontario fish. The program has collected Hg data since the 1970s from approximately 2,047 various lakes, reservoirs, rivers, creeks and streams. Fish samples were collected in partnership with the Ontario Ministry and Natural Resources and Forestry (MNRF) during late summer or early fall using a variety of methods, including gill netting, trap netting, electrofishing and angling. Total length, wet weight and sex (if possible) were recorded for each fish. Skinless boneless dorsal fillets were taken and stored at -20°C. Mercury analysis using MOECC protocols were performed, including acid digestion and cold vapor flameless atomic absorption spectroscopy as described by Bhavsar *et al.* (2010) and Neff *et al.* (2012). We considered top predator fish because they often exhibit higher Hg levels than lower trophic level fish due to biomagnification. Mercury data for four sport fish—WE, NP, SMB, and LMB—were screened for further analysis. For this study, Hg measurements for only natural inland lakes were retained, while river, creek and stream data were omitted. The final Hg dataset consisted of 36,639 WE measurements from 1,232 locations; 25,978 NP measurements from 1,313 locations; 11,879 SMB measurements from 652 locations; and 3,340 LMB measurements from 217 locations sampled between 1970 and 2014.

It is well-known that fish Hg concentrations increase with fish size (Gewurtz *et al.*, 2011). To reliably assess changes in fish Hg concentrations over time, Hg levels were first standardised at 3 lengths for each species, representing small, medium, and large size categories,

using power series regressions (Supplementary Table S1; Gandhi *et al.*, 2013). A total of 6159 power series regressions, one for every combination of fish species, location and year, were conducted (2033 WE, 1901 NP, 972 SMB and 300 LMB). Standard lengths representing Small, medium and large size were selected for each fish species based on previous literature (Scott and Crossman, 1998; Gewurtz *et al.*, 2011; Gandhi *et al.*, 2013). To avoid using over-extrapolated concentrations of Hg at the standard lengths, only sampling events within 15cm of the maximum and minimum lengths were considered. For example, to calculate the Hg concentration for a 40cm walleye at each location and year, only sampling events with the smallest walleye length being no greater than 55cm and the largest length no less than 25cm were retained (Table S1). The final standard length dataset consisted of 6069 Hg concentration data points for all species/length/location/year, though only medium sized fish (50cm for WE, 60cm for NP, and 30cm for SMB and LMB) were used for further analysis as this size provided the greatest abundance of observations per species.

Climate and emissions variables were obtained from multiple open access data sources (Table S2). Climate variables were split into three main categories: (1) local climate, (2) large-scale climate drivers, and (3) Hg emissions. Local climate variables included mean monthly precipitation (Precip) and average daily temperature (Temp) and were obtained from the Climatic Research Unit (CRU, see <http://www.cru.uea.ac.uk/>). A total of 9 global climate indices were also obtained: El Nino Southern Oscillation index (ENSO), Tropical/ Northern Hemisphere pattern (TNH), Polar/Eurasian pattern (PE), West Pacific pattern (WP), total Sunspot number (SunTOT), group Sunspot number (SunGN), North-Atlantic Oscillation index (NAO), Pacific Decadal Oscillation index (PDO) and North Pacific index (NP). Lastly, global and Canadian Hg emissions were acquired from Muntean *et al.* (2014), the European Monitoring and Evaluation

Programme (EMEP) and Environment Canada. We also included a lag of 1 year for all climate and emissions variables as it has been suggested that it takes within a year for bioaccumulation of Hg to reach top predator fish (Harris *et al.*, 2007).

The purpose of our study was to examine the relationships between climate and fish Hg levels at a landscape scale across all years between 1970-2013. As such, we also used a watershed approach and divided Ontario into its secondary watersheds. Data for the Ontario watersheds was obtained from the MNR metadata website (see www.ontario.ca/data).

Data analysis

Non-parametric Kruskal-Wallis (KW) tests were performed in order to assess if fish Hg concentrations were significantly different among lakes within the same secondary watershed. The KW tests were conducted among standard length fish Hg levels of the same species within a 5-year time period (e.g., lakes in secondary watershed A from 1970-1974). If fish Hg concentrations were not statistically different at significance $p=0.05$, they were merged to obtain watershed-specific standard length fish Hg concentrations to examine temporal and spatial trends. Only those cases with more than 2 observations for each fish species/size/time period were considered.

Temporal trend analysis

A nonparametric Sen's slope estimate was performed to assess fish Hg trends for the historical (1970-1992) and recent (1993-2014) time periods for each secondary watershed. These time periods were chosen because fish Hg trends for Ontario's inland lakes as well as Great Lakes region changed sometime between the late 1980s and early 1990s (Bhavsar *et al.*, 2010; Tang *et al.*, 2013; Gandhi *et al.*, 2014). Trends were also calculated for 1-year lag periods.

Sen's slopes are able to perform well with missing values and are suitable for data that are not normally distributed. Sen's slopes were calculated using the MAKESENS (Mann-Kendall test for trend and Sen's slope estimates) Microsoft Excel Template (Salmi *et al.*, 2002).

Correlation analysis with climate and emissions variables

Spearman's rank correlations were used to quantify the relationship between fish Hg levels and all climate and Hg emission variables for the historical, recent and 1-year lag time-periods for each species and watershed. To ensure that correlations between fish Hg and climate variables were not masked by trends in the local climate data, residuals of fish Hg and local climate variables (mean daily temperature and monthly precipitation) were used. All analyses were performed in the R-language environment (R Development Core Team, 2012) and mapped using ArcGIS software (ESRI, 2011). The data acquisition and analysis workflow framework is summarised in Fig. 1.

RESULTS

For each species and standard length, fish Hg levels within a secondary watershed and 5-year time period were not significantly different ($p>0.05$). As such, it was deemed appropriate to conduct fish Hg trend analysis at the secondary watershed level.

Historical and recent fish Hg trend analysis

Fish Hg levels for the two coolwater species (WE and NP) decreased during the historical period (Fig. 2a,c), but increased in the recent decades (Fig.2b, d). Between 1970 and 1992, 68% of the secondary watersheds experienced declines in fish Hg. More specifically, Hg levels in WE in the south-west Ontario decreased significantly ($p<0.05$) at the 0.20-0.70 $\mu\text{g/g/decade}$ (Fig. 2a). In contrast, for the recent time period (1993-2014), 57% of the secondary watersheds experienced increases in fish Hg. Hg levels in western Ontario WE increased at 0.20-0.40 $\mu\text{g/g/decade}$ (Fig. 2b). Between the time periods, 26% of the watersheds switched from decreasing to increasing Hg levels.

Similar to WE, 71% of the watersheds experienced declines in NP Hg with significant ($p<0.1$) declines in northern, western, and east-central Ontario at 0.10-0.70 $\mu\text{g/g/decade}$ (Fig. 2c). In the recent time period, 52% of the secondary watersheds showed increasing NP Hg, with the Hg levels in western Ontario NP increasing at 0.20-0.30 $\mu\text{g/g/decade}$. About 32% of the watersheds switched from historically decreasing to recently increasing NP Hg levels.

The warmwater predatory fishes (SMB and LMB) are currently restricted to southern and south-central Ontario (Fig. 2e,g) and have been expanding their range northwards during the time period of this study in response to warming temperatures (Alofs *et al.*, 2014). Historically, SMB Hg concentrations either remain unchanged or decreased (0.0-0.20 $\mu\text{g/g/decade}$), whereas

LMB Hg levels generally increased (0.0-0.10 $\mu\text{g/g/decade}$; Fig. 2g). Between 1993 and 2014, SMB Hg levels increased in 53% of the watersheds at 0.0-0.20 $\mu\text{g/g/decade}$ (Fig. 2f). Similarly, LMB Hg levels increased in 50% of the watersheds at 0.0-0.20 $\mu\text{g/g/decade}$ (Fig. 2h). Approximately 38% of the watersheds with SMB and 30% of the watersheds with LMB switched from decreasing to increasing Hg levels in the recent time period.

Correlations for 1-year lag fish mercury and climate/emissions variables

Spearman correlations of fish Hg and climate/emissions variables at a lag of 1-year suggested that not a single variable alone was driving the patterns in fish Hg levels, but rather a complex interplay between local climate, large-scale climate drivers, and emission patterns explained fish Hg levels (Fig. 3). Cross-correlations for medium WE Hg levels (Fig.3a) revealed that most local climate and large-scale climate indices have weak correlations ($r=\pm 0.1$) both historically and recently. Historical WE Hg levels were higher with positive correlations with El Niño Southern Oscillations (ENSO) ($r=0.3$), global ($r=-0.3$) and Canadian Hg emissions ($r=0.3$), although in recent decades these relationships becomes closer to zero. Recent WE Hg levels were correlated more pronouncedly with Sunspot count ($r=0.2$) and North Atlantic Oscillations (NAO) ($r=0.2$).

Correlations between fish Hg and climate/emission variables changed most drastically between periods for NP. The strongest correlations historically for NP Hg concentrations were with Tropical/Northern Hemisphere (TNH) patterns ($r=-0.4$), global ($r=-0.3$) and Canadian Hg emissions ($r=0.3$). Though global Hg emission remained correlated in recent periods, it was the local climate variables, average monthly precipitation ($r=0.3$) and average daily temperature ($r=0.2$), which came out as more strongly correlated.

Median correlations for SMB and LMB Hg levels epitomized the complex interplay between climate and emission on fish Hg trends. Historically, SMB Hg levels were correlated with local climate variables mean monthly precipitation ($r=0.2$) and average daily temperature ($r=0.2$), the climate indices ENSO ($r=0.3$) and TNH ($r=-0.25$), as well as global ($r=-0.4$) and Canadian Hg emissions ($r=0.45$). Correlations with recent SMB Hg levels maintained similar correlations between variables, with the addition of Sunspot count ($r=-0.2$). Historical LMB Hg levels had the strongest correlations with average monthly precipitation ($r=0.45$), mean daily temperature ($r=-0.5$), ENSO ($r=0.4$) and TNH ($r=0.2$). Recent correlations only included precipitation ($r=0.3$), TNH ($r=0.2$), Sunspot count ($r=0.2$). None of the Hg emissions had strong correlations with LMB Hg levels.

Though the median correlations presented were often not significant ($p>0.05$), secondary watersheds with greater fish Hg observations in the south-central regions of Ontario did contain significant correlations.

DISCUSSION

The results indicated that Hg levels in Ontario's sport fishes are increasing in the past two decades despite continued reductions in local and regional (North American) Hg emissions over the same time period. These results are consistent with other studies on this topic (Monson *et al.*, 2011; Tang *et al.*, 2013; Gandhi *et al.*, 2014). Gandhi *et al.* (2014) reported that the Ontario NP and WE Hg levels increased at 0.01-0.27 µg/g/decade between 1995 and 2012. For the boreal shield lakes in northern Ontario, Tang *et al.* (2013) found significant increases in rates of Hg bioaccumulation for WE and mean Hg concentration for NP between historical (1974-1981) and recent (2005-2010) time periods. Studies conducted for WE and LMB in Minnesota lakes demonstrated nonlinear Hg trends, with shifting upward Hg patterns in the early 1990s (Monson *et al.*, 2011). Though these studies often attribute the increase in fish Hg levels to multiple factors, such as global Hg emissions or climate change, the influence of climate on fish Hg levels is still unclear.

Historically, local climate variables were not strongly correlated with Hg levels in the coolwater predatory fishes (WE and NP), but more pronounced in the warmwater fishes (SMB and LMB). In contrast, local climate variables had generally stronger correlations to fish Hg in the recent time periods. Interestingly, the stronger correlations with temperature and precipitation in the recent periods coincide with climate change (i.e., increases in temperatures and changes in precipitation). This suggests that the local climate change related variables that historically did not influence coolwater fish Hg may now be relevant and may further exacerbate fish Hg trends in warmwater fish.

In recent years, mean monthly precipitation was found to be positively correlated with Hg levels in NP and LMB. Increased precipitation may facilitate increased Hg loading to aquatic

environments through (1) greater runoff from a watershed, and thus re-mobilization as a result of soil erosion, and (2) direct deposition (Risch *et al.*, 2012; Wiener *et al.*, 2012). Interestingly, we observed a negative correlation between temperature and Hg concentrations in all fish, particularly SMB. Though we expected a positive correlation (Bodaly *et al.*, 1993; Canário *et al.*, 2007; Stern *et al.*, 2012), this relationship may be attributed to possible changes in prey consumption with warmer waters. Freshwater fish subjected to warmer waters often have increased metabolism, and thus consume more resources (Pörtner *et al.*, 2007; Wang *et al.*, 2007). An increase in dietary intake may or may not lead to increased fish Hg levels (MacCrimmon *et al.*, 1983; Simoneau *et al.*, 2005; Karimi *et al.*, 2007). As Hg is bioaccumulative, fish size and age are positively correlated with Hg level (Gewurtz *et al.*, 2011). At a given age, faster growing fish have generally lower Hg concentrations due to *growth dilution* (Simoneau *et al.*, 2005; Karimi *et al.*, 2007).

Global climate drivers were correlated with fish Hg levels in all fish species throughout the study period across Ontario. These large-scale climate drivers are important in regulating local climate patterns (Bonsal and Shabbar, 2011). For example, coolwater WE and warmwater SMB and LMB Hg levels were positively correlated with ENSO Index. ENSO is known to influence the winter temperatures and total precipitation in Ontario, particularly the Great Lakes regions, with positive phases (El Niño) associated with cooler temperatures and higher precipitation (Shabbar and Khandekar, 1995; Mirza, 2004; Bonsal and Shabbar *et al.*, 2011; Yu *et al.*, 2015). Hg levels in WE were correlated with the NAO and Hg levels in SMB and LMB were correlated with the NP indices. The NAO has been found to influence temperatures over northeastern regions of the country (Bonsal and Shabbar, 2011), while the NP index can enhance precipitation and surface air temperature in coastal and south-central regions of Canada

(Linkin and Nigam, 2015). Sunspot count was also correlated to changes in recent fish Hg levels. Then sunspot cycle corresponds to solar magnetic activity which is often linked to air and sea surface temperatures in the Northern Hemisphere (Eddy, 1976; Reid, 1987; Friis-Christensen and Lassen, 1991). The mechanism that these large-scale climate drivers may impact fish Hg levels would be through the subsequent changes in local climate. Studies that have examined fish Hg trends against global climate drivers often find significant links between them (French *et al.*, 2006; Evans *et al.*, 2013). For example, French *et al.* (2006) determined that oscillations of total Hg concentrations in chinook salmon from the Bay of Quinte were associated with cooling La Niña trajectories and thus summer air temperatures. In our study, we show that global climate drivers, through their influence on local temperatures and precipitation, are linked to Hg levels in all four Ontario predator fishes considered in this study.

The recent slowdown or reversal of decreasing fish Hg trends may also be attributed to decreases in local Hg emissions and increases in global Hg emissions. Historically, Canadian Hg emissions were positively correlated with WE, NP and SMB Hg levels in Ontario. Over the last few decades, the U.S., Canadian and Ontario Hg emissions have declined due to stricter government regulations (Environment Canada, 2015). Though these changes are expected to prompt fish Hg declines, we find they are still increasing. For NP and SMB, global Hg emissions were correlated to fish Hg levels for both the historical as well as recent times. This suggests that contributions from global emissions may play a larger role in driving fish Hg trends than local emissions. It is likely that the decline in local emissions had an impact on the historical decreases of fish Hg trends, however, current rises of Hg levels are driven by the global emissions through transboundary transport (Monson, 2009; Monson *et al.*, 2011; Tang *et al.*, 2013; Gandhi *et al.*,

2014). Hg emissions, both local and global, are likely to remain significant determinants of fish Hg levels, among many other factors.

Climate change will neither act uniformly across the globe (Sala *et al.*, 2000), nor will its impacts be felt simply with increased temperatures and precipitation frequency. Similarly, Hg contamination will not be driven only by direct and indirect pathways related to climate and emissions. Other environmental stressors such as invasive species may cause an antagonistic or synergistic response to changes in Hg levels. However, many of the other stressors may be directly or indirectly related to the climate change. With increasing temperatures and precipitation events under the climate change, the threat of invasive species also rises (Parmesan and Yohe, 2003; Sharma *et al.*, 2007; Heino *et al.*, 2009). The introduction of a non-indigenous species to the environment has many consequences, ranging from new competition to predation to changes in the food chain length. Often when trophic structures in aquatic systems are altered, they impact the levels of Hg in top predators (Hrabik *et al.*, 1998; MacIssac, 1996; Vander Zanden and Rasmussen, 1996). The lengthening of food chains has been positively correlated with increases bioaccumulation of toxic contaminants in fish (Cabana *et al.*, 1994; Vander Zanden and Rasmussen, 1996).

Rennie *et al.* (2010) proposed that the establishment of an invasive invertebrate predator species *Bythotrephes* in Ontario inland lakes lengthen aquatic food chains and thus increased fish Hg concentrations. The majority of Ontario lakes that contain invasive fish species, such as rainbow smelt (*Osmerus mordax*) or SMB, generally have higher fish Hg levels than those without invasive fish species (Vander Zanden *et al.*, 2003). With a warming climate, warmwater invasive fish populations, such as SMB, may expand into neighbouring waterbodies, disrupting the native aquatic biota (Sharma *et al.*, 2007). SMB has been found to force top predators such as

lake trout into lower trophic positions (Vander Zanden *et al.*, 1999; Vander Zanden *et al.*, 2004). Species at lower trophic level are expected to have reduced Hg concentrations due to lower Hg biomagnification (Cabana *et al.*, 1994; Lavoie *et al.*, 2010). A recent study, however, suggests that this depends on the resources consumed. Johnson *et al.* (2015) developed simulations to assess how WE Hg concentrations would change under an altered diet. Their models suggested that WE consuming invertebrates may not only experience low growth but also contain high Hg levels, whereas WE that prey on fish may have higher growth efficiency and 85% lower Hg concentrations (Johnson *et al.*, 2015). When lower trophic position prey, such as invertebrates, contain higher Hg levels than prey fish, the implications of invasive species on native fish Hg dynamics becomes more complex.

Predicting how and why fish Hg levels have been changing in recent years requires further understanding of the underlying ecological processes and relative influence of environmental and climatic variables impacting Hg levels. Since Hg levels are driven by a complex interplay of multiple factors, with drivers that operate through synergistic and antagonistic feedback loops, it has been challenging to forecast changes in Hg particularly under a changing climate. In the Laurentian Great Lakes, for example, it has been found that fish Hg levels have been decreasing in Lakes Ontario and Huron but increasing in Lake Erie due to factors such as recycling of historical releases and changes in food web structures (Bhavsar *et al.*, 2010). For inland lakes, Hg levels in Ontario top predator fish have been increasing in recent years (Gandhi *et al.*, 2014), particularly in northern Ontario. Gandhi *et al.* (2014) discussed how factors, such as Hg emissions from Asian countries, climate change, invasive species, and acidity of a lake, could be contributing such fish Hg increases. With all these variables contributing to Hg levels, we need further understanding of (1) how Hg concentrations vary with Hg deposition,

methylation, and uptake by living organisms; (2) the relationship between methylation rates and climatic factors; and (3) the key processes related to cycling of Hg and global transport (UNEP 2013).

Conclusions

Recent decades have been marked with increasing fish Hg levels in some regions; however, identifying the factors that contribute the most to changes in fish Hg levels has been complex. Drivers of the recent fish Hg changes are different from historical periods, as temperature and precipitation are emerging as important factors influencing fish Hg trends for different species. Changes in local climate, large-scale climate drivers, and Hg emissions are not only important in predicting these trends, but also complicated and interlinked in producing direct and indirect impact on fish Hg levels. With rising temperatures, altered precipitation events, changing global climate indices and rising global Hg emissions, predicting how and why fish Hg will change in the next few decades will be both vital and challenging. Monitoring programs need to be designed carefully to distinguish the effects of various factors simultaneously influencing Hg concentrations in fish and wildlife. An improved understanding of how climate-related parameters impact the processes related to bioavailability and uptake of Hg by fish is necessary to make informed advisory and future preventative responses.

ACKNOWLEDGEMENTS

We thank MOECC for fish Hg data, Samantha Stefanoff, Thomas Van Zuiden, Katrina Gaibisels, Bailey Hewitt and Saudia Khan of York University for help in drafting the manuscript, and Sogee Spinner for support in R-code development.

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FIGURES

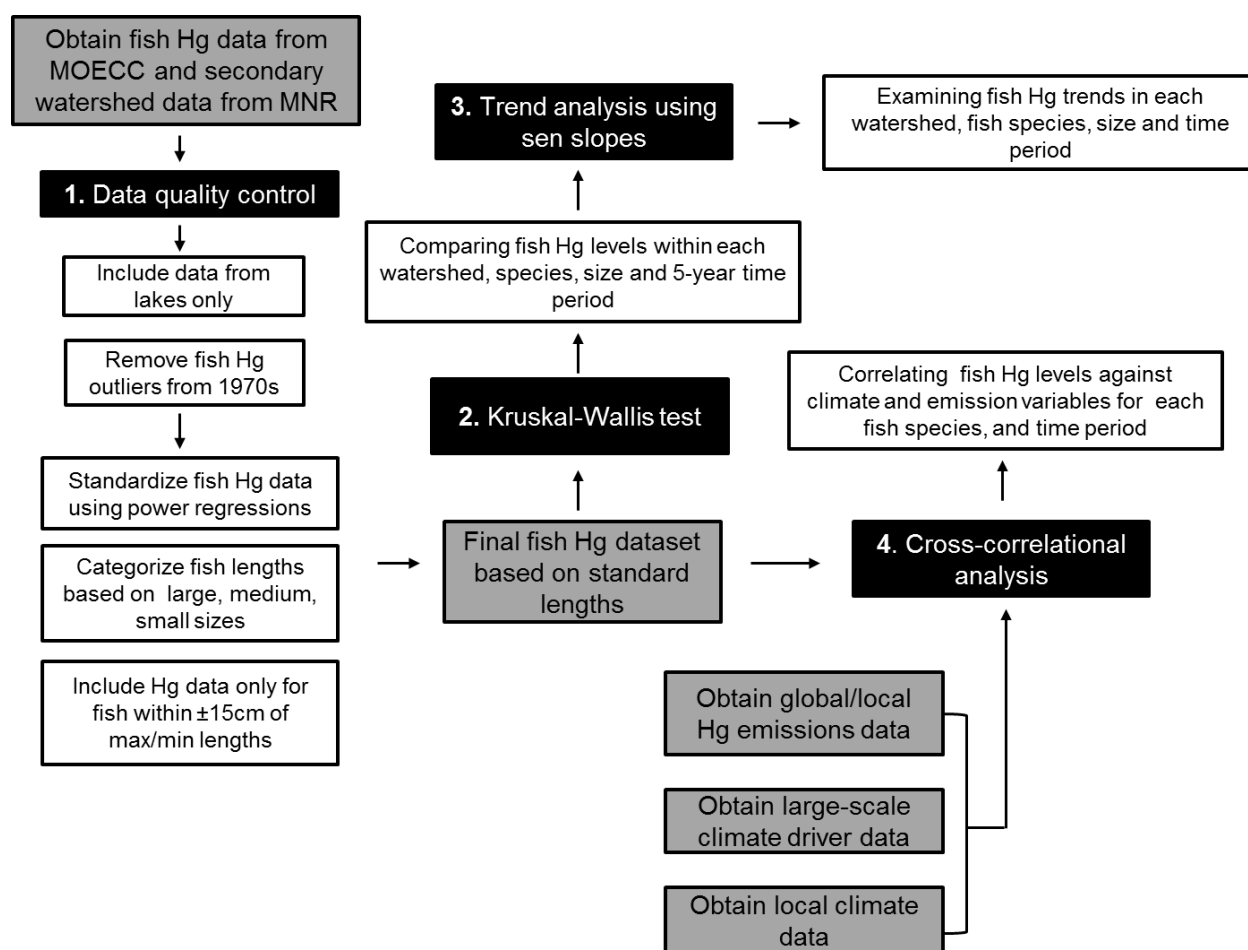


Figure 1: Method framework. Grey boxes indicate data or datasets; black boxes indicate data screening or analyses; and white boxes provide detail on the data analysis. Refer the Methods section for details.

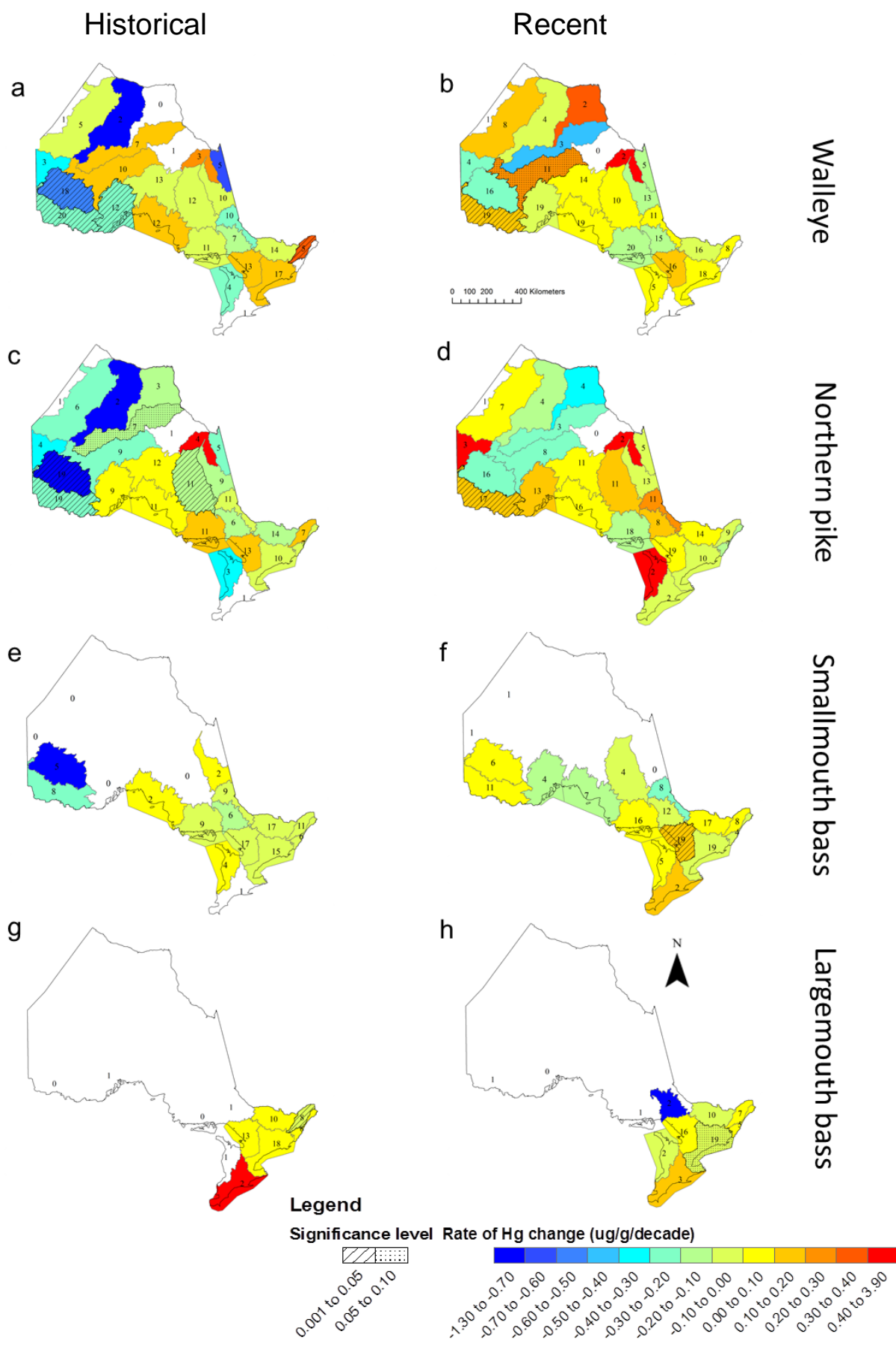


Figure 2: Fish Hg trends for walleye (a,b), northern pike (c,d), smallmouth bass (e,f), and largemouth bass (g,h) in each secondary watershed for the historical (1970-1992) and recent (1993-2014) periods estimated using Sen's Slope. The number for each watershed represents the number of lakes. Extreme values are binned into the upper and lower ends of the scale so the variation is not lost. White watersheds indicate insufficient data for the trend analysis.

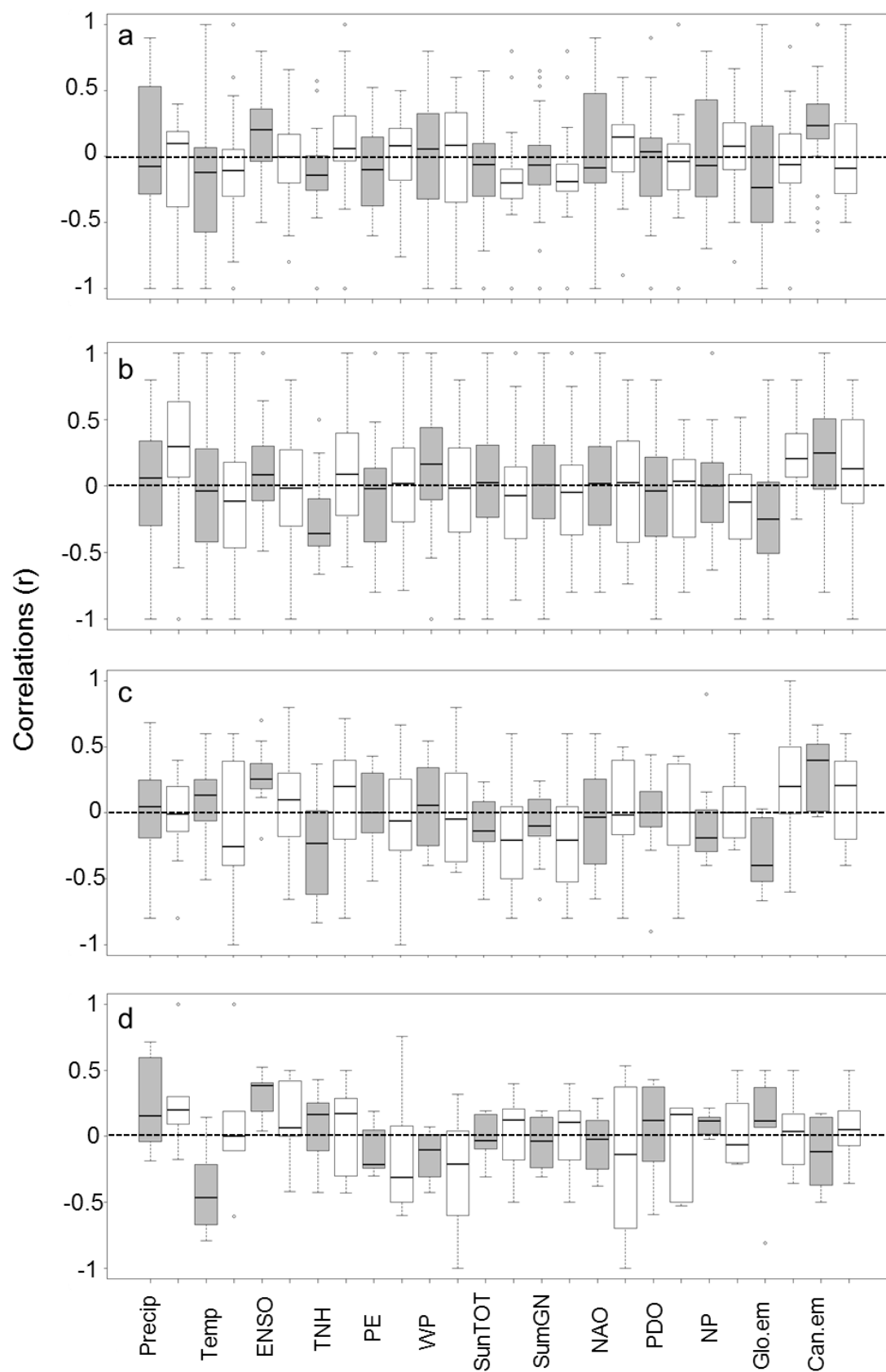


Figure 3: Boxplots of correlations between 1-year lag fish Hg levels and climate/emission variables for the historical (1970-1992; grey) and recent (1993-2014; white) periods for (a) walleye, (b) northern pike, (c) smallmouth bass and (d) largemouth bass. Precip: average monthly precipitation; Temp: mean daily temperature; ENSO: El Nino Southern Oscillation index; TNH: Tropical/ Northern Hemisphere pattern; PE: Polar/Eurasian pattern; WP: West Pacific pattern; SunTOT: total Sunspot number; SumGN: group Sunspot number; NAO: North Atlantic Oscillation index; PDO: Pacific Decadal Oscillation index; NP: North Pacific index; Glo.em: Global Hg Emissions; Can.em: Canadian Hg Emissions.

SUPPORTING INFORMATION

Table S1: Selected fish lengths to standardize Hg levels for walleye, northern pike, smallmouth bass and largemouth bass in Ontario. To avoid overly extrapolated Hg levels, only sites/years with Hg data for fish lengths between the minimum and maximum lengths were considered.

Fish species	Small (cm)	Medium (cm)	Large (cm)
Walleye	40	50	60
Min length (+15)	<55	<65	<75
Max length (-15)	>25	>35	>45
Northern pike	45	60	70
Min length (+15)	<60	<75	<85
Max length (-15)	>30	>45	>55
Smallmouth bass	20	30	40
Min length (+15)	<35	<45	<55
Max length (-15)	>5	>15	>25
Largemouth bass	20	30	40
Min length (+15)	<35	<45	<55
Max length (-15)	>5	>15	>25

Table S2: The record length, source and type of long-term climate indices data acquired for temporal analysis.

Climate variables	Record length	Source	Type of data
Average Monthly Precipitation (mm)	1901-2013	CRU	Annual
Average daily Temperature (°C)	1901-2013	CRU	Annual
El Nino Southern Oscillation Index (ENSO)	1876-2013	National Climate Centre, Australia	Monthly
North Atlantic Oscillation Index (NAO)	1865-2013	NOAA	Monthly/Annual
Pacific Decadal Oscillation Index (PDO)	1948-2013	NOAA	Monthly
Tropical Northern Hemisphere Oscillations (TNH)	1950-2015	NOAA	Average of DJF/Monthly
North Pacific Index (NP)	1899-2014	NOAA	Average of NDJFM/Monthly
Western Pacific Index (WP)	1950-2015	NOAA	Monthly
Polar/ Eurasia Oscillations (PE)	1950-2015	NOAA	Monthly
Total Sunspot Numbers	1700-2015	SILSO	Annual
Total Sunspot Group Number	1610-2015	SILSO	Annual
Global Hg Emissions (tonnes)	1970-2010	Muntean <i>et al.</i> , 2014	Annual
Canadian Hg Emissions (tonnes)	1970-2013	EMEP and Environment Canada	Annual

CRU = Climatic Research Unit

NOAA = National Oceanic and Atmospheric Administration

EMEP = European Monitoring and Evaluation Programme

SILSO = Sunspot Index and Long-term Solar Observations

GENERAL CONCLUSIONS

Global stressors, such as climate change and mercury contaminants, prompt regional responses in our freshwater fish populations. Climate change has been found to significantly impact the distribution of many fish species in Ontario and facilitate the introduction of non-native species into novel systems (Chu *et al.*, 2005; Sharma *et al.*, 2009; Alofs *et al.*, 2014). Together, native species are vulnerable to extirpation without mitigation of climate change or management strategies in place. In addition, climate change may also facilitate greater availability of Hg into our aquatic ecosystems. With warming temperatures and changes to precipitation, fish Hg levels may increase, and become hazardous for human consumption. The influences of climate- and emission-driven fish Hg trends are not well understood. Further understanding of how fish may respond under global stressors such as climate change is integral to maintain healthy aquatic ecosystems and human health.

Coldwater fish species are particularly vulnerable to the impacts of climate change. These predictions were supported in our study, as cisco ranges were found to contract by approximately 388 km from their southern extents in Ontario, resulting in a decline of cisco occurrence ranging from 8-37% (\bar{x} = 20%) by 2050, and up to a 7-47% (\bar{x} = 26%) decline by 2070. All climate change scenarios pointed to a decline in cisco populations in southern Ontario with minimal shifts northward, suggesting that water temperatures may be warming at a faster rate than these fish can respond. Along with the increased distribution of invasive species, such as rainbow smelt, these important forage prey fish may be further at risk to be extirpated in their native habitats (Jacobson *et al.*, 2010; Sharma *et al.*, 2011). Fisheries and conservation managers must examine and integrate these predictive models into their decisions by appropriately triaging

which lakes best protect cisco from extirpation and ultimately help to maintain the ecological health of Ontario lakes.

Despite the decreases in local Hg emissions, fish Hg levels in Ontario's top predator fish were found to be increasing in the past two decades (Tang *et al.*, 2013; Gandhi *et al.*, 2014). Though these were significant only for walleye and northern pike, these recent increases ranged between 0 to 0.20 ug/g/decade. As the non-native warmwater fish move northward, however, these Hg levels in all of our fish may change in response to a new competitor. For each fish species, we found that >25% of Ontario's secondary watersheds shifted from historically declining fish Hg trends to recent increasing patterns and $\geq 50\%$ of watersheds contained increasing fish Hg trends between 1993 and 2014. Complex combinations of local climate, global climate drivers, and Hg emissions influence fish Hg levels. Although industrial Hg emissions were correlated with fish Hg concentrations for both historical and recent periods, global climate drivers and local climate were found to be important more so in recent years as temperatures and precipitation has increased in Ontario. Differentiating the effects of climate and emissions on fish Hg levels is critical to assess how changing environmental stressors may impact the health of our ecosystems, wildlife and the human consumption of fish.

Inferences of multiple regression models are built upon correlations (Mac Nally, 2002). As we know, correlations do not prove there is a direct causal link between the variables being examined. The presence of significant ($p < 0.05$) correlations in models developed between the response variable (i.e. cisco occurrence or fish Hg concentrations) and predictors variables (i.e. abiotic and biotic variables), can however imply an unresolved causal structure (Shipley, 2002). Regression models must be used in combination with other forms of evidence to support the proposed casual structure. For example, Westoby and Wright (2006) suggest that correlations

between plant functional traits using scatterplots must be supplemented with cost-benefit modelling and field experiments. In this study, tests for multicollinearity between variables or use of a dual-criterion selection procedure were implemented for the purpose of building accurate, conservative models (Mac Nally, 2002; Blanchet *et al.* 2008). Researchers who have examined the direct links through micro- and mesocosm experimentation (Harris *et al.*, 2007; Dijkstra *et al.*, 2013; Pack *et al.*, 2014) and in statistical approaches, such as step-wise selection techniques or hierarchical partitioning (Mac Nally 2000; 2002), continue to support the interpretations of multiple regression models. When many lines of evidence reveal a consistent story, we can be more confident in an interpretation.

The response of aquatic species to changes in climate is complex. Global stressors such as climate change and mercury contamination can have severe implications on our natural environment, even threatening human health. Aquatic species today live in a multiple stressor environment, where freshwater fish populations are vulnerable to warming temperatures, forcing them to move or perish in the lakes they remain, and potentially causing the levels of Hg in our fish to rise. Consistent, wide-spread, and long-term monitoring programs are vital to understanding spatial and temporal trends in fish distributions and Hg trends under a changing climate.

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