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# What's hot about mercury? Examining the influence of climate on mercury levels in Ontario top predator fishes

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ABSTRACT

Mercury (Hg) levels in Ontario top predator fishes have been increasing in recent decades. These increases may be a result of many additive factors, including global climate change. Only recently has research been conducted on how climate change may impact Hg levels in freshwater fishes at large-scales. We examined the relationship between Hg trends and (1) local weather, (2) large-scale climate drivers, and (3) anthropogenic Hg emissions, in native cool water (walleye and northern pike) and warm water (smallmouth bass and largemouth bass) predatory fishes in Ontario, Canada, for historical (1970–1992) and recent (1993–2014) time periods. For each fish species studied, > 25% of Ontario's secondary watersheds shifted from historically declining to recently increasing fish Hg trends, and ≥ 50% of watersheds experienced increasing trends between 1993 and 2014. Recent fish Hg increased at up to 0.20 μg/g/decade; which were significant ($p < 0.05$) for walleye, northern pike and smallmouth bass. Multiple linear regressions revealed a complex interplay of local weather, large-scale climate drivers, and anthropogenic Hg emissions influencing fish Hg levels. Recent Hg levels for walleye and largemouth bass increased with changes in global climate drivers, while higher precipitation influenced smallmouth bass Hg levels the most. Walleye Hg levels increased during the positive phases of global climate drivers, reflecting the local influence of local temperatures and precipitation indirectly. Differentiating the effects of climate-related parameters and emissions is increasingly crucial to assess how changing multiple environmental stressors may impact health of wildlife and humans consuming fish.

## 1. 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). 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). Although anthropogenic Hg emissions in North America have declined by approximately 75–90% between the 1970s and 2011 (Cain et al., 2007; Environment Canada, 2015), fish Hg levels are again increasing in the Province of Ontario, Canada (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 may be driving Hg dynamics.

Fish Hg levels can be influenced directly by factors such as lake size (Bodaly et al., 1993), lake acidity, hardness, dissolved organic carbon (DOC) (Wren et al., 1991), 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), land use or land cover (Bank et al., 2006; Drenner et al., 2013), as well as indirectly by watershed disturbances such as forest fires and the invasion of non-native species in aquatic environments (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 changes (IPCC, 2013). The impacts of climate factors, such as increased temperatures, changes in precipitation, wind patterns or dust deposition, can change the distribution, mobility and uptake of Hg in freshwater ecosystems in both direct and indirect pathways (Grimalt et al., 2010; Dijkstra et al., 2013; Evans et al., 2013; Eagles-Smith et al., 2017).

Deposition of atmospheric Hg can be a major contributor to Hg in fish (Pacyna et al., 2006; UNEP, 2013) and the amount deposited on to aquatic ecosystems can be affected by climatic factors such as precipitation (Outridge et al., 2008; Grimalt et al., 2010; Risch et al., 2012). Mercury is a global pollutant that can be transported long distances (Engstrom, 2007; Krabbenhoft and Sunderland, 2013). This long distance transport is reflected in the total anthropogenic Hg deposited in Canada as > 95% is currently from trans-boundary sources (Risk

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Management Strategy for Mercury, 2010). Precipitation is one of the primary pathways through which reactive 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; ICMGP, 2017). Greater quantities of precipitation can elevate wet Hg deposition by incorporating water-soluble inorganic Hg ($Hg^{II}$) into rain or snow, and thereby increasing inputs watersheds and lakes (Lamborg et al., 2002), exposing fish and other aquatic organisms to this contaminant.

Temperature is another climatic factor that has been shown to influence fish Hg concentrations. Temperature can affect the amount of bioavailable methylmercury (MeHg) at the base of the food chain as it is highly correlated with epilimnetic temperatures and it can also influence the rates of atmospheric transport between systems (Adrian et al., 2009; Grimalt et al., 2010; Pack et al., 2014). Available MeHg can be directly affected by temperature in a variety of ways. For example, warmer temperatures have been shown to increase the conversion rate of $Hg^{II}$ to MeHg (Avramescu et al., 2011; Hecky et al., 1991; Bloom, 1992). In addition, warmer epilimnion temperatures have resulted in methylation rates exceeding demethylation by sulfur-reducing bacteria, one of the main converters of $Hg^{II}$, increasing the net MeHg available and exposure to aquatic organisms (Hecky et al., 1991). Elevated temperatures can also indirectly influence available MeHg in aquatic ecosystems by reducing dissolved oxygen (Jankowski et al., 2006). Mumley and Abu-Saba (2002) found that methylation of Hg quadrupled when dissolved oxygen concentration dropped below 6 mg/L. Lastly, global models have also projected that warmer temperatures may result in less oxidation, causing elemental mercury ($Hg^0$) levels to remain longer in the atmosphere, enhancing long-range transport of Hg (Wängberg et al., 2010). With rising temperatures due to climate change, we can expect higher methylmercury (MeHg) content in the food web (Bodaly et al., 1993; Canário et al., 2007; Stern et al., 2012).

The available Hg found in freshwater lakes can also be partially attributed to changes in global climate drivers. Fluctuations in air circulation patterns, or oscillations of large-scale climate drivers, have significant effects on three main factors: (1) local climate (Li et al., 2013; Zhao et al., 2013; Evans et al., 2013), (2) local weather patterns, and (3) the transport of Hg from distant sources to local lakes, especially those from Asian industrial locations (Selin et al., 2007). For example, negative phases of the El Niño Southern Oscillation (ENSO) are associated with warmer winter temperatures (George et al., 2000; Bonsal and Shabbar, 2011) and below average winter precipitation in Ontario (Bonsal and Shabbar, 2011). Changes in local air circulation, precipitation and temperature patterns attributed to large-scale climate drivers, can therefore indirectly affect fish Hg levels (Evans et al., 2013). Evans et al. (2013), for example, found that the Pacific/North American Index (PNA) was important in explaining Hg levels in lake trout in Great Slave Lake, Northwest Territories, Canada, indicating that trends may have been affected by patterns in air circulation.

Although most studies often attribute the increase in fish Hg levels to multiple factors, such as global Hg emissions or climate change (Tang et al., 2013; Gandhi et al., 2014; ICMGP, 2017), the proportional influence of climate on fish Hg levels is still unclear. We investigate the relative influence of local weather, large-scale climate drivers, and atmospheric Hg emissions on fish Hg levels in native cool water (walleye, *Sander viteus*, WE; northern pike, *Esox lucius*, NP) and non-native warm water (smallmouth bass, *Micropterus dolomieu,* SMB; largemouth bass, *Micropterus salmoides,* LMB) fishes in secondary watersheds of the Province of Ontario, Canada. Secondary watersheds are defined catchment basins of land, drained by a watercourse and its tributaries by the Ontario Ministry of Natural Resources. Secondary watersheds are subdivisions of the primary watersheds in Ontario and are either large river systems or groupings of small coastal streams. 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 weather, large-scale climate drivers, and local/global Hg emissions are potentially driving fish Hg trends over time. To our knowledge, this is one of few studies exploring the changes in fish Hg levels across a large landscape and interacting multiple environmental stressors, including climate change and atmospheric pollution. Since there is little climatic variation between lakes in the same region, we chose to examine fish Hg levels between watersheds in order to capture greater regional differences between watersheds. We predict that increases in fish Hg levels over time may more recently be attributed to changes in climate, specifically increases in precipitation events and temperature. This study is aimed to critically evaluate observed fish Hg trends under multiple stressors over broad spatio-temporal scales and improve our understanding of how intricate climatic processes can impact fish Hg levels and thereby health of fish, and the wildlife and humans consuming fish.

## 2. Materials and methods

### 2.1. Data acquisition and screening

Total 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 2047 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. Total mercury analysis using MOECC protocols were performed, including acid digestion and cold vapour 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 (Kamman et al., 2005). Mercury data for four sport fish—WE, NP, SMB, and LMB—were screened for further analysis as detailed in the following section. We chose to include several predator species as different species experience Hg dynamics in distinct ways. By including more than one species we were able to infer fish biology differences inherent in food webs and gain a better understanding of the temporal trends in predator fish Hg levels (Bhavsar et al., 2010). For this study, Hg measurements for only natural inland lakes (excluding the Great Lakes) were retained, while reservoir, river, creek, and stream data were omitted. The final Hg dataset before further screening consisted of 36,639 WE measurements from 1232 locations; 25,978 NP measurements from 1313 locations; 11,879 SMB measurements from 652 locations; and 3340 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 standardized at medium lengths for each species, using power series regressions (Supplementary Table A.1; Gandhi et al., 2014). A total of 6159 power series regressions, one for every combination of fish species, watershed, and year, were conducted (2033 WE, 1901 NP, 972 SMB and 300 LMB). Standard total lengths representing medium sizes were selected for each fish species based on previous literature (Scott and Crossman, 1998; Gewurtz et al., 2011; Gandhi et al., 2014). To avoid using over-extrapolated concentrations of Hg at the standard lengths, only fish sampled within 15 cm of the maximum and minimum medium lengths were considered. For example, to calculate the Hg concentration for a 40 cm WE at each location and year, only sampling events with the smallest WE length being no greater than 55 cm and the largest WE length no less than 25 cm were retained (Table S1). The final standard length