Environ. Sci. Technol. **2010,** *44,* 3273–3279

Changes in Mercury Levels in Great Lakes Fish Between 1970s and 2007

SATYENDRA P. BHAVSAR,*,† SARAH B. GEWURTZ,†,‡ DARYL J. MCGOLDRICK,§ MICHAEL J. KEIR,§ AND SEAN M. BACKUS§

Environmental Monitoring and Reporting Branch, Ontario Ministry of the Environment, 125 Resources Road, Toronto, Ontario M9P 3V6, Department of Ecology and Evolutionary Biology, University of Toronto, Toronto, Ontario M5S 3G5, and Water Quality Monitoring & Surveillance Division, Water Science and Technology Directorate, Environment Canada, 867 Lakeshore Road, P.O. Box 5050, Burlington, Ontario L7R 4A6

Received December 21, 2009. Revised manuscript received March 8, 2010. Accepted March 19, 2010.

A number of initiatives have curtailed anthropogenic mercury emissions in North America over the last two decades; however, various factors, including long-range transport of global emissions, may complicate the response of fish mercury levels to remedial actions. Since the Great Lakes of North America are together the largest surface freshwater body in the world and are under the influence of many complicating factors, trends of mercury in fish from the Great Lakes can reflect the overall impact of mercury management actions at local, regional, and perhaps global scales. Here we present a comprehensive view of mercury trends in Canadian Great Lakes fish using two large (total 5807 samples), different (fillet and whole fish), and long-term (1970s−2007) monitoring data sets. The spatial differences in lake trout and walleye mercury levels during this period have generally been within a factor of 2−3 with Lakes Erie and Superior having the lowest and highest concentrations, respectively. These spatial differences have diminished in the recent years (2000−2007). The concentrations have generally declined over the three decades (mid-1970s to 2007); however, in recent years, the concentration trends are flat in Lake Ontario walleye and appear to be increasing in Lake Erie walleye. There was a mismatch in the Lake Ontario lake trout and walleye temporal trends, which shows the importance of considering more than one fish species for proper spatial/temporal trend assessments.

Introduction

All freshwater fish in North America, and perhaps in the world, have at least trace levels of mercury in their tissues (*1*). Although mercury in fish could be a result of natural occurrence, a substantial portion of elevated fish mercury levels at many locations has been attributed to increased mercury emissions from anthropogenic activities (*2, 3*). This has resulted in mercury being a major contaminant causing restrictions on consumption of fish from most freshwater systems in North America (*4, 5*).

The Great Lakes of North America are together the world's largest surface freshwater body and are of significant economic importance to the U.S. and Canada. Both the federal and provincial/state U.S. and Canadian governments have acted to minimize the use and release of mercury by mandating its removal from many consumer items and through more stringent emission standards that can be achieved by implementation of improved emission control technologies (*6*). Since atmospheric deposition remains a major input process for mercury in many aquatic systems including the Great Lakes (*7, 8*), these emission reduction activities are expected to result in decreased fish mercury levels. Further, various actions taken to mitigate pollution problems at hot spots identified by the International Joint Commission (IJC) during the 1980s in the Great Lakes are also expected to improve fish mercury levels. However, many factors may complicate the expected outcome.

First, it is well-known that mercury is a global pollutant. Other countries such as China and Russia contribute a significant portion of today's worldwide mercury emissions (*9*) and their emissions may not be declining or may even be increasing (*10*). Second, the relationship between anthropogenic mercury emission and atmospheric concentration, which affect the rate of atmospheric deposition, can be confounded by contributions from natural emissions (*11*). Third, recycling of historically released mercury within the environment may dampen the rate of expected improvements (*12*). Finally, the Great Lakes span 7° 30′ of latitude and nearly 16° of longitude adjacent to areas of dense urban and industrial development, intensive agriculture, and forests. Due to the varied influences of these land uses and other anthropogenic impacts, such as the introduction of nonnative species, the Great Lakes are experiencing alterations in their food webs and chemistry which are also known to affect bioaccumulation of mercury in fish (e.g. refs *13*−*15*). For these reasons, mercury trends in the Great Lakes fish could serve as an indicator of the overall impact of mercury management actions at local and regional scales and, to a certain extent, on a global scale.

Different studies with varying size of data and regional coverage have reported diverse spatial and temporal trends of fish mercury levels for a number of sites in the Great Lakes (*16*−*20*) as well as the Great Lakes region (*21*−*23*). We provide a comprehensive view of mercury trends in Canadian Great Lakes fish using two large (total 5807 samples), and long-term (1970s−2007) provincial and federal monitoring data sets. The provincial (Ontario Ministry of the Environment; OMOE, Canada) data employed in this study are length-based skinless dorsal fillet measurements used for the fish consumption advisory purpose, while the federal (Environment Canada; EC) data are length- and age-based whole fish measurements used to assess overall environmental contamination and risk to fish and fish-consuming wildlife.

Materials and Methods

Sample Collection and Analysis. The OMOE, in collaboration with Ontario Ministry of Natural Resources and other agencies, collects samples of various fish species from each of the Canadian Great Lakes, namely Lakes Superior, Huron, Erie, and Ontario, generally on an annual or biennial basis since mid-1970s. The samples have been collected in varying numbers and sizes from several locations in each lake during

* Corresponding author phone: 416-327-5863; fax: 416-327-6519; e-mail: s.bhavsar@utoronto.ca or satyendra.bhavsar@ontario.ca.
† Ontario Ministry of the Environment.
‡ University of Toronto.
§ Environment Canada.

10.1021/es903874x
Published on Web 03/30/2010

late summer or early fall using gill nets and electro-fishing. After collection, fish are measured for length and weight, sexed, filleted (skin-off), and sent to OMOE laboratories in Toronto and stored at −20 °C until chemical analysis.

EC runs a long-term biomonitoring program in the Great Lakes which, unlike the OMOE program, measures contaminants in whole fish. Since 1977, individual samples of top predators such as walleye and lake trout, and composite samples of prey species such as alewife, smelt, and sculpin have been collected annually through deployment of trawls and gill nets at three to five offshore sites in each of Lakes Ontario, Erie, Huron, and Superior. Samples are transported to the lab where they are stored at −20 °C prior to processing. Before homogenization, samples are thawed at 5 °C, weighed, sexed, and a calcified structure such as scale or otolith is removed for aging. The fish are then cut up and run through a commercial meat grinder five times. Aliquots of whole fish homogenate are placed in acetone−hexane rinsed jars and stored at −20 °C until chemical analysis.

The OMOE samples were analyzed for mercury concentrations using the OMOE method HGBIO-E3057 (*24*), and EC samples were analyzed at the National Laboratory for Environmental Testing using NLET method 2801 (*25*). These methods are briefly described in the Supporting Information (SI).

Data Screening. We used lake trout (LT; *Salvelinus namaycush*) and walleye (WE; *Sander vitreus*) as indicator species to evaluate spatial and temporal trends because these fish are present throughout the Great Lakes (*26*), they are top predators and integrate exposure through the food web (*26*), and they have been used to assess the trends of a wide range of contaminants in the Great Lakes as part of Canadian and U.S. monitoring programs (e.g. ref *17, 20, 27−30*). Lake Erie is relatively shallow and has a low population of LT; therefore, we primarily relied on the WE measurements for this lake.

Mercury concentrations in fish increase with fish size (*31, 32*). The size of the OMOE and EC fish samples varied over a wide range on both an inter- and intra-annual basis. In order to minimize influence of fish size on the analyses, we selected mercury measurements for narrow fish size ranges of 55−65 and 45−55 cm for LT and WE, respectively. This method is consistent with the approach adopted previously for a PCB trend analysis (*28*), provides a good sample size for each year, and overcomes the requirements of the annual regression slopes of logarithmic concentration versus fish length being parallel (*33*). The OMOE Georgian Bay data were treated separately from the main basin of Lake Huron due to known differences in the limnology and contaminant levels between the two systems (*34, 35*). Data are reported on a wet weight basis and not on a lipid normalized basis because mercury primarily partitions into the protein, rather than the lipid phase, of muscle tissue (*36*).

In total, 3416 OMOE samples (1828 LT and 1588 WE) of the selected size ranges were analyzed for mercury over the years (1079, 91, 265, 34, and 359 LT, and 185, 227, 142, 737, and 297 WE from Lakes Superior, Georgian Bay, Huron, Erie, and Ontario, respectively). The OMOE samples were not necessarily collected from the same location in a lake every time. Although this may affect temporal trends at a smaller scale, it is believed that the long-term trends should not be influenced significantly, especially for medium to large sized top predatory fish, such as LT and WE, because of their relatively large home ranges (*26, 37*).

In contrast, the EC samples utilized in this study were collected from the same location every time. We used the most comprehensive data sets that exist for LT (Lakes Superior, Huron, and Ontario) and WE (Erie), which were from the following locations (number of samples in the length-based analysis): Thunder Bay—Pie Island (152), Marathon (51), and Whitefish Bay (131) in Lake Superior; North Channel (90) and Owen Sound (39) in Lake Huron; Pelee Island (205) in Lake Erie; and Niagara-on-the-lake (117), Port Credit (151), Port Hope—Cobourg (105), and eastern basin—Oswego (203) in Lake Ontario. Although forage fish measurements may not be a strong indicator of lake-wide changes in the Great Lakes, they can be used to corroborate the findings from predatory fish measurements as they provide a different perspective on contaminant trends based on local conditions and less influenced food web processes. As such, we also used 12−16 cm EC rainbow smelt (RS; *Osmerus mordax*) data for the same locations. The number of RS samples analyzed (in the same order of sites for Lakes Superior to Ontario as those for LT and WE locations except missing Marathon site in Lake Superior and additional Port Colborne in Lake Erie) were: 50, 39, 50, 55, 127 (Port Colborne, Lake Erie), 106, 107, 72, and 169, respectively. In total, 2019 EC samples (1244 LT and WE, and 775 RS) were considered in this length-based temporal trend analysis.

Mercury concentrations in fish typically increase with age; however, fish size (i.e., length) is obtained as a surrogate measure for the duration of contaminant exposure because it is easy and inexpensive to acquire. Since fish size can be influenced by many factors such as variation in growth rates due to food web changes/differences (*38*), use of a fixed size range over the years may not provide accurate temporal trends in contaminant levels due to possible differences in exposure time. For this reason, we also performed temporal trend analysis on 4 year old LT (WE for Erie) from the EC data set. Four years was the average age for the selected size ranges of LT and WE. This age-based subdata set had a total of 790 LT (Superior, Huron, and Ontario) and 91 WE (Erie) measurements of which 355 (45%) and 17 (19%), respectively, were not part of the length-based analyses because their sizes were outside the selected length ranges.

Statistical Procedures. For long-term temporal trend analysis, first mean OMOE LT and WE mercury measurements for each of the Canadian Great Lakes were compared for two time periods, 1980−1990 and 2000−2007, using General Linear Model Univariate analysis with Tukey's posthoc multiple comparison in SPSS (version 12.0.1, 2003; SPSS, Chicago, IL).

Second, a nonparametric Mann-Kendall (MK) test (*39*) was performed on the annual average values to assess monotonic temporal trends in the mercury levels. A nonparametric Sen's method (*39*) was then used to estimate magnitude of the trends. These methods are not sensitive to missing values and are suitable for the data that do not follow a normal distribution after transformation. The calculations were performed using an Excel (Microsoft Corp., Redmond, WA) template MAKESENS—Mann-Kendall test for trend and Sen's slope estimates (*40*).

Finally, linear regressions were performed on all individual data points to capture variability in annual sample sizes and measurements. Linear regression models rely on key assumptions of linearity of the relationship between dependent and independent variables and normal distribution of the error in data. In order to linearize relationships and stabilize variance, we also performed linear regression analysis on natural logarithmic-transformed data in addition to non-transformed data.

Results and Discussion

Spatial Differences. As shown in Figure 1 and SI Figure SI1, the spatial differences in LT and WE fillet mercury levels for the Canadian Great Lakes over the ∼30 years (mid-1970s−2007) have generally been within a factor of 2−3 with Lake Superior fish having the highest concentrations. Individual measurements for Lakes Ontario, Erie and Huron during the period ranged from (5th−95th percentile)

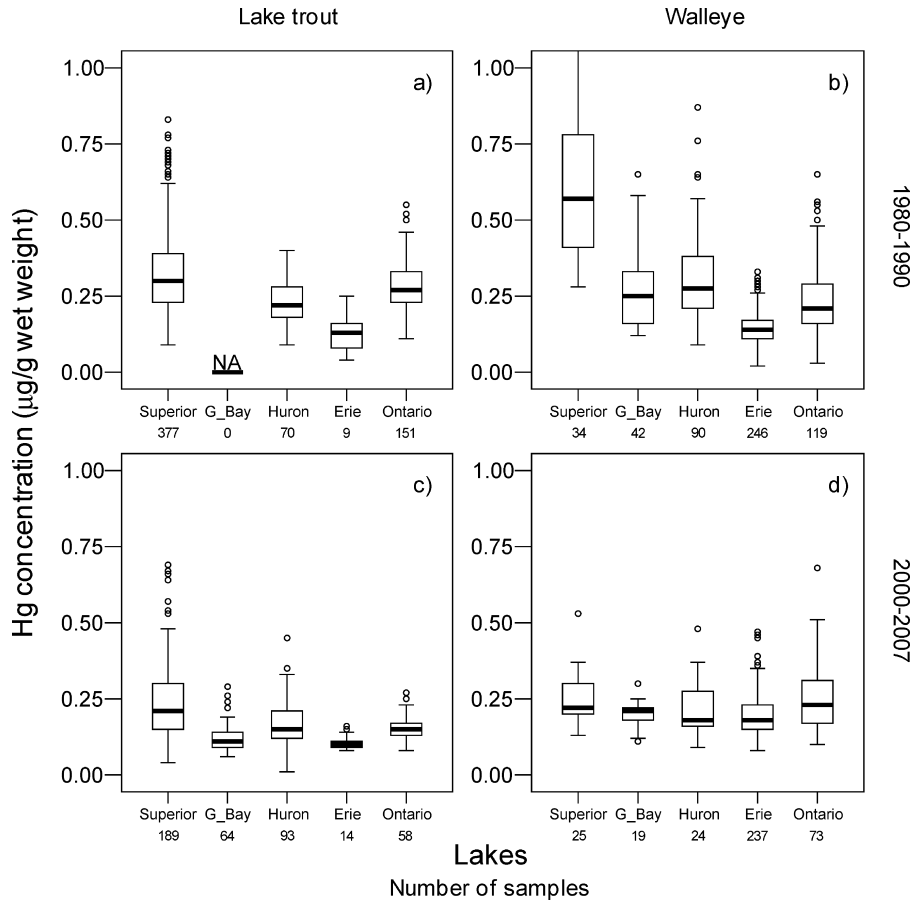

FIGURE 1. Measured Hg concentrations (μg/g wet weight) in OMOE skinless fillets. (a, c) 55−65 cm lake trout and (b, d) 45−55 cm walleye collected from Lakes Superior, Georgian Bay, Huron, Erie and Ontario during (a, b) 1980−1990 and (c, d) 2000−2007. Box plots: the line within the box indicates median, the box indicates 25th and 75th quartile values, the whiskers indicate the upper and lower values not classified as statistical outliers or extremes. The outliers (open circles) and extremes (not shown) were values more than 1.5 and 3 times 25th−75th interquartile range away from the closest end of the box, respectively. The numbers below lake names are for sample sizes (*N*). NA indicates not available.

0.07−0.38 μg/g for LT and 0.08−0.8 μg/g for WE; the corresponding ranges for Lake Superior were 0.13−0.88 and 0.15−0.8 μg/g, respectively (SI Figure S1). During the 1980s, LT and WE mercury levels were in the order of Erie < Huron ≈ Ontario < Superior ($P < 0.05$). These spatial differences have diminished in recent years (2000−2007) ($P > 0.05$; except for comparison between Superior and other LT).

MacEachen et al. (*41*) reported similar spatial patterns in skin-off muscle of sea lamprey and whole LT. Measurements from a study conducted by the Great Lakes Fish Monitoring Program (GLFMP) of the U.S. Environmental Protection Agency (*17*) are also in good agreement with the OMOE fillet data presented in this study, that is, lake-specific average values ranging from 0.11−0.23 μg/g and diminished spatial differences in recent years (more details in the SI). Reported Lake Michigan whole LT mercury concentrations (*19*) are similar to those for the Lakes Huron and Ontario LT.

The spatial pattern in fish mercury levels (i.e., lowest in Erie and highest in Superior) is in contrast to that for the sediments where the lowest levels of mercury are in Huron and Superior and the highest are in Ontario and western Erie (*42*). This mismatch needs further investigation. The pattern also contrasts with those observed for major contaminants causing restrictions on consuming fish (i.e., PCBs and dioxins/furans), where the concentrations are highest in Lake Ontario (*27, 28*). However, the spatial trend of fish mercury presented here is similar to that for toxaphene (*43, 44*), which is the only other contaminant currently causing restrictions on consuming Lake Superior fish (*5*). Similar to toxaphene, mercury is also considered mainly atmospherically derived in Superior (*42, 45*). However, local industrial activities such as mining, chlor-alkali production, and pulp and paper production may also be sources of mercury (*42*). High mercury concentrations in Lake Ontario as compared to Lake Erie have been attributed to local sources in the Niagara River, which is the major tributary of Lake Ontario (*16, 46*).

Long-Term Changes. A comparison of corresponding median mercury concentrations during the 1980s and 2000s for OMOE LT and WE fillets (Figure 1) showed significant declines in Superior LT and WE ($P < 0.001$), Huron WE ($P < 0.05$) and Ontario LT ($P < 0.001$). Georgian Bay WE, Huron LT, Erie LT and WE, and Ontario WE fillet mercury levels were not significantly different between the 1980s and 2000s ($P > 0.05$; Figure 1). The MK test performed on the individual annual averages for the entire period (i.e., 1970s−2000s) showed results similar to the decade-specific comparison for Superior, Erie, and Ontario but suggested significant ($P < 0.01$) declines in Georgian Bay WE and Huron LT (SI Figure SI2).

According to the MK test, the rates of long-term mercury declines in LT is 0.12, 0.04, and 0.08 μg/g ww per decade for Lakes Superior, Huron (main basin) and Ontario, respectively (SI Figure SI2). There was no significant change in G. Bay and Erie LT. The rates of long-term WE mercury declines for Superior, G. Bay, and Huron are 0.14, 0.12, and 0.08 μg/g ww per decade, respectively, with no significant changes in Erie and Ontario WE (SI Figure SI2). These long-term (1970s−2007) general declines (i.e., approximately 77, 47, and 57% for Superior, Huron, and Ontairo LT, respectively; 72, 68, and 61% for Superior, G. Bay, and Huron WE, respectively) match

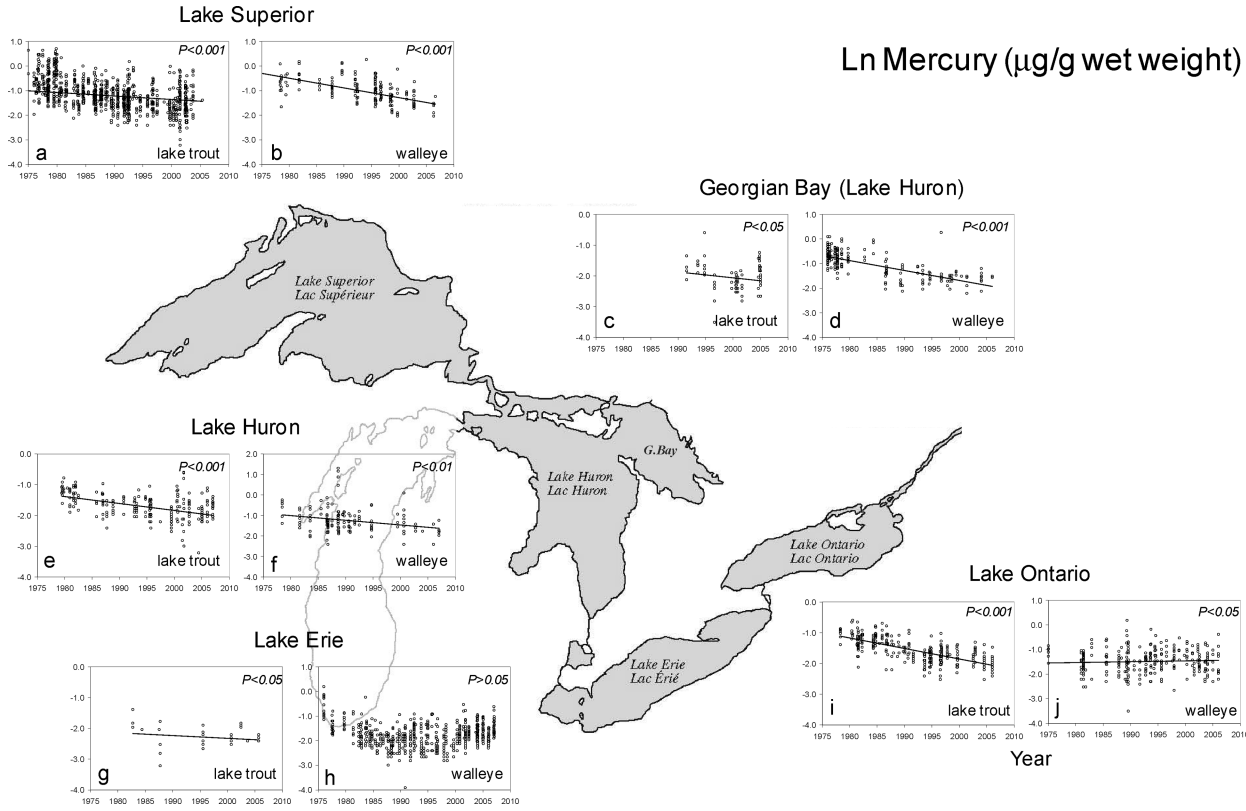

FIGURE 2. Temporal trends of mercury (natural log-transformed, μg/g ww) in OMOE skinless fillets. (a, c, e, g, i) 55−65 cm lake trout, and (b, d, f, h, j) 45−55 cm walleye collected from Lakes Superior (a-b), Georgian Bay (c,d), Huron (e,f), Erie (g,h), and Ontario (i,j) between 1973 and 2007. *P*-value is for statistical significance.

well with the declining mercury concentrations reported for the Ontario LT (*16*), Ontario salmon (*18*), and Great Lake sediments (∼25−80%) (*42, 47*).

Considering that a temporal trend analysis performed on decade-specific or annual mean values neglects variations in annual sample sizes over the period, next we performed linear regression analysis on natural log-transformed long-term (1970s−2000s) LT and WE mercury measurements (Figure 2). This analysis suggested significant ($P < 0.05$ to 0.001) declining trends in all cases except Lake Erie WE.

SI Figure SI3 shows regression analysis on length-based EC whole body LT and WE measurements. Long-term declining trends ($P < 0.001$) are evident for all locations considered except for LT from Owen Sound (Georgian Bay, Lake Huron) and Marathon (Lake Superior). These results correspond well with the trend analyses based on OMOE data except for Marathon in Superior and Pelee Island in Erie. A regression analysis performed on length-based EC whole RS measurements reflected the findings for LT and WE trends (except for North Channel in Huron); however, due to unavailability of RS data for Marathon (Superior), the increasing trends observed for LT and WE could not be confirmed (SI Figure SI4).

The trend analysis for age-based (4 year) EC whole LT and WE measurements generally reflected the length-based results and provided an explanation for the increasing LT and WE trend suggested for the EC Marathon (Superior) site by length-based analysis (SI Figure SI5). The 2006 Marathon measurements included in the 55−65 cm length-based analysis were for 5−6 years old LT, which were older than the corresponding 1980s−1990s data, where LT were on average 4 years old. Since the 2006 fish had more mercury exposure time compared to the 1980s−1990s fish, the increasing trend for Marathon suggested by the length-based analysis is probably an artifact of the different ages.

Both length- and age-based EC whole fish WE data showed a significant ($P < 0.001$) declining trend for Pelee Island in Erie (SI Figures SI3 and SI5), which is in contrast to the flat trend (no significant change, $P > 0.05$) shown by the OMOE data (Figures 2 and SI SI2). However, most of the EC measurements were collected during the 1970s−1990s with little post-1996 data. When we performed regression analysis on the OMOE 1970s−1996 Erie WE data (Figures 2 and SI1), a significant ($P < 0.001$) declining trend was observed (results not shown).

Recent Temporal Trends. The disagreement between the long-term (1970s−2007) flat and historically (1970s−1996) declining Erie WE mercury trends suggests that the concentrations declined in past and increased recently (1990s−2007). To examine this, next we performed temporal trend analysis on the post-1990 OMOE LT and WE data.

As shown in Figure 3, nonparametric MK tests ($P < 0.1$) and linear regressions on regular and natural log-transformed concentrations ($P < 0.001$) confirmed that the Lake Erie WE concentrations are indeed increasing in recent years at the rate of 0.04−0.05 μg/g ww per decade. Very limited Erie LT measurements also suggested increasing albeit insignificant trend ($P > 0.1$) (SI Figure SI6).

The tests also confirmed continued declining trends for Superior and Huron (WE: 0.18−0.2 and 0.05−0.07 μg/g ww per decade, respectively, Figure 3; LT: 0.02−0.03 μg/g ww per decade, SI Figure SI6) and insignificant ($P > 0.05$) declining trend for Georgian Bay (WE: 0.02−0.03 μg/g ww per decade, Figure 3; LT: 0.04 μg/g ww per decade, SI Figure SI6). Recent trends for Lake Ontario are similar to those of long-term: significant ($P < 0.001$) declining for LT (0.03 μg/g ww per decade, Figure SI6) but insignificant ($P > 0.1$) increasing trend for WE (0.01−0.02 μg/g ww per decade, Figure 3).

Similar to Lake Erie fish, Monson (*23*) found a downward trend before the mid-1990s and an upward trend thereafter in northern pike and WE from Minnesota lakes. Mixed declining, flat and increasing mercury trends between

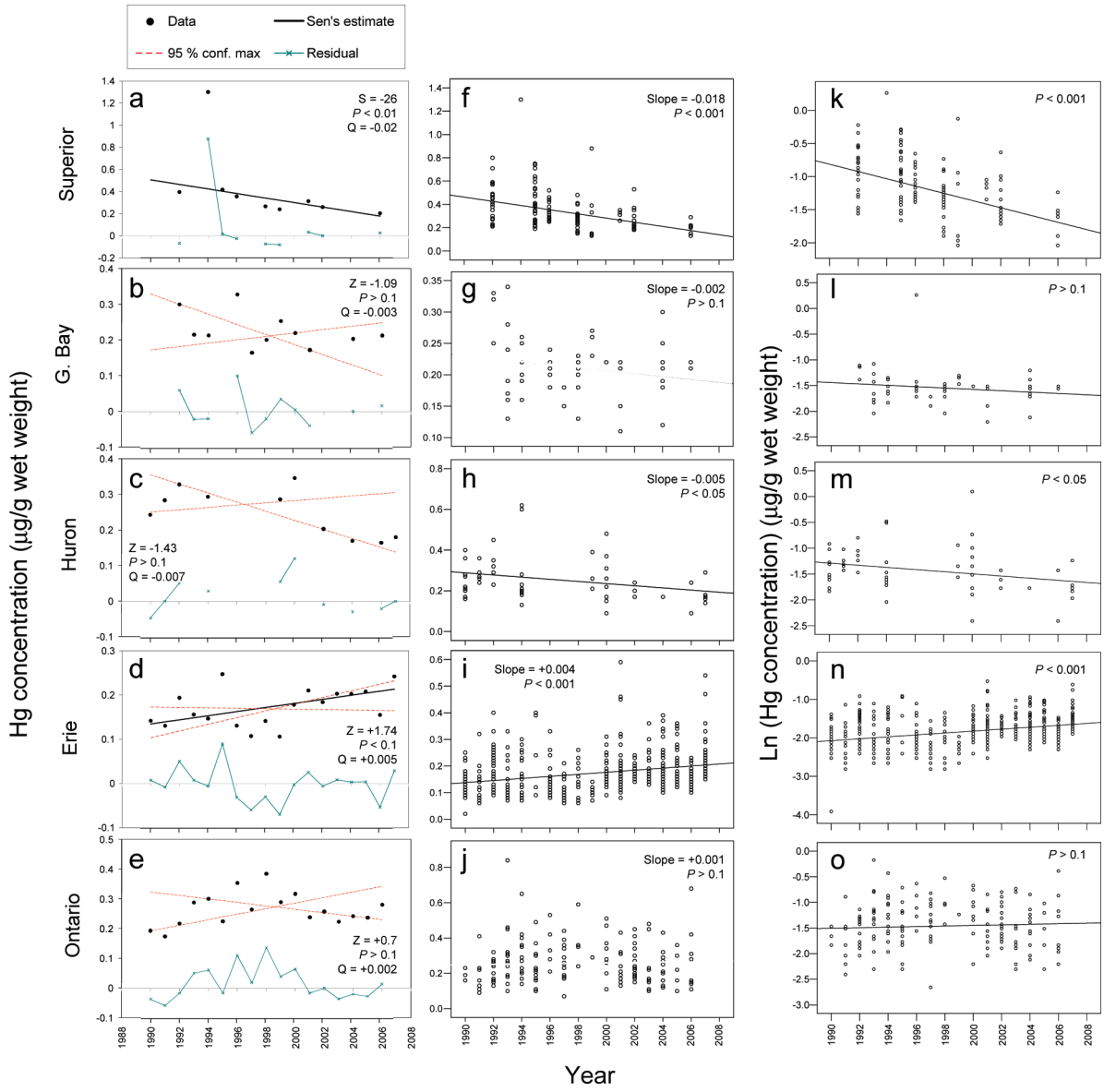

FIGURE 3. Temporal trends of mercury in OMOE skinless fillets of 45−55 cm walleye collected between 1990−2007. (a−e) Man-Kendall test, (f−j) linear regression on regular concentrations, and (k−o) linear regression on natural log-transformed concentrations. *P*-value is for statistical significance. Sen's estimate line is shown only for statistically significant ($P < 0.05$) upward/downward trend.

1982−2005 for Wisconsin WE have been reported (*22*). An average mercury decline of 14% in yellow perch from a group of Adirondack lakes (New York State) has been reported for the past 15 years (*21*).

Temporal data for U.S. air emissions indicate a significant decrease in emissions from municipal and medical waste incinerators between the early-1990s and ∼2001; however, emissions from coal-fired electricity generation and other sources were relatively constant (*48*). From 1990−2000, Canadian emissions decreased by ∼75%, largely due to process changes at metal smelting facilities (*48*). Model-estimated deposition to the Great Lakes decreased significantly between 1995−1996 and 1999−2001 with U.S. sources contributing much more to Great Lakes atmospheric mercury deposition than Canadian sources (*48*).

The time scale of the ecosystem response to changes in emissions and atmospheric deposition depends on various complicating factors and no reliable method is currently available to predict the response (*49*). Generally, transport of mercury through watersheds to aquatic system is slow with further delays of a few years for piscivorous fish to respond (*49*). As such, it is likely that the Great Lakes top predator fish have not yet fully responded to the recent reductions in mercury emissions in the Great Lakes basin and further declines may be observed in future.

In-Depth Erie Analysis. To further analyze the increasing mercury concentration in Erie WE, we checked if mean fish length differed among years, which might have influenced the temporal trends for the concentrations. Since some significant ($P < 0.05$) differences were observed, we narrowed down the selected WE size range from 45−55 to 48−52 cm, for which there was no significant ($P > 0.7$) difference in average lengths among years (results not shown). Even for this narrowed size range, there were significant ($P < 0.001$) differences in mercury levels among years with a positive regression coefficient (0.0036 µg/g/year; 95% confidence interval 0.0016−0.0056; $P < 0.001$; SI Figure SI7) suggesting an increasing concentration trend.

Since OMOE samples for a lake were not necessarily collected from the same locations every year, next we examined if pooling lake-wide data together had any influence on the Erie WE trend. For this analysis, the 45−55 cm

lake-wide WE data set was divided into four subsets (i.e., Blocks 1 through 4) according to the OMOE fish consumption advisory blocks (*5*). The coefficients for linear regression analyses performed on these Block 1−4 post-1990 OMOE subsets were positive (0.0037, 0.0043, 0.0023, and 0.0033 μg/g/year, respectively) with high statistical significance especially for Blocks 1 and 2 with a good sample sizes (*P*-values: <0.001, <0.0001, <0.1, and <0.05 for Blocks 1−4, respectively; SI Figure SI8).

Finally, we performed regression analysis on the block-specific OMOE Erie data with a narrowed size range of 48−52 cm. Although such constrains decreased the sample sizes in the analyses, all blocks still showed increasing trends with generally higher slopes and reasonable statistical significance (most $P < 0.1$) (SI Figures SI8 and SI9). Positive regression coefficients under all scenarios considered here strongly suggest that mercury levels in Lake Erie WE fillets are increasing at the approximate rate of 0.03−0.04 μg/g per decade. It has been reported that invasions of dreissenid mussels and round goby have modified Lake Erie's foodweb structure, which is causing mercury accumulation in smallmouth bass at the historical rate despite declines in the sediment mercury levels (*14*).

Use of the two large and different long-term fish contaminant monitoring data sets allowed us to present a comprehensive view of mercury trends in the Canadian Great Lakes. Varying effects of many factors have resulted in different spatial and temporal trends of fish mercury levels in the Great Lakes basin. The historical spatial differences of factors 2−3 between the lowest and highest concentrations in similar-sized Erie and Superior fish, respectively, have diminished in recent years. Concentrations in LT and WE from the upper Great Lakes (Superior and Huron), where mercury is occurring largely due to natural abundance and/or atmospheric input (*42*), are generally declining. In contrast, the lower Great Lakes (Erie and Ontario) are recently experiencing either flat or weak increasing trends for LT and/or WE. The majority of mercury in these lower lakes derive from point discharges, atmospheric deposition, and/or recycling of historical releases. In addition, changes in their food web structures and/or plankton abundance may also be contributing to sustained/increased mercury in the top predators. Differing long- and short-term trends of mercury in Lake Ontario WE and LT demonstrate the importance of considering more than one species when assessing temporal trends. The temporal trends from the length-based data were generally in agreement with those from the age-based measurements; however, addition of age-based analysis can be useful especially in cases where fish growth rates might have changed over the years.

Acknowledgments

We thank Andre Vaillancourt, Wolfgang Scheider, and all other OMOE staff who have contributed to the OMOE Sport Fish Contaminant Monitoring Program, Chantelle Gomes (OMOE) for assistance in statistical analysis, and Ram Prashad and Rusty Moody (OMOE) for sample analysis. We also acknowledge the contributions of Mandi Clark, Mary Malecki, the captain and crew of the CCGS *Kelso* and all other EC and DFO staff who have contributed to the EC/DFO Fish Contaminants Monitoring and Surveillance Program.

Supporting Information Available

Additional explanatory text and nine figures. This material is available free of charge via the Internet at http://pubs.acs.org.

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ES903874X