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# Fish Mercury Levels Appear to Be Increasing Lately: A Report from 40 Years of Monitoring in the Province of Ontario, Canada

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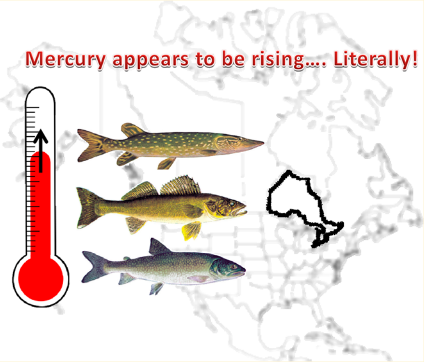
Supporting Information



**ABSTRACT:** Recent mercury levels and trends reported for North America suggest a mixed (positive/negative) outlook for the environmental mercury problem. Using one of the largest consistent monitoring data sets in the world, here we present long-term and recent mercury trends in Walleye, Northern Pike, and Lake Trout from the Province of Ontario, Canada, which contains about one-third of the world's fresh water and covers a wide geographical area (1.5 and 3 times larger than France and Germany, respectively). Overall, the results indicate that the fish mercury levels either declined (0.01−0.07 μg/g decade) or remained stable between the 1970s and 2012. The rates of mercury *decline* were substantially greater (mostly 0.05−0.31 μg/g decade) during the 1970s/80s possibly in response to reductions in mercury emissions. However, Walleye and Pike levels have generally *increased* (0.01−0.27 μg/g decade) in recent years (1995−2012), especially for northern Ontario (effect sizes for differences between the two periods ranged from 0.39 to 1.04). Proportions of Walleye and Pike locations showing a flat or increasing trend increased from 26−44% to 59−73% between the 1970s/80s and 1995−2012. Mercury emissions in North America have declined over the last few decades, and as such it is logical to expect recovery in fish mercury levels; however, other factors such as global emissions, climate change, invasive species, and local geochemistry are likely affecting the response time and magnitude.

## INTRODUCTION

Mercury is a neurotoxin that can also damage cardiovascular, immune, respiratory, gastrointestinal, and reproductive systems.[1] Elevated levels of mercury in fish has been a well-known environmental problem in North America and throughout the world.[2,3] Although mercury exists naturally, anthropogenic sources have contributed to an increase of mercury in the environment, particularly in top predatory fish.[4] Since biomagnification increases mercury levels in top predatory fish by about a million times compared to the surrounding water and fish can be a major part of human diet, fish consumption is a dominant pathway of mercury exposure for most humans.[1] As such, to protect human health, fish consumption advisories mainly due to elevated mercury have been issued for most freshwater systems in North America.[5,6]

To reduce mercury emissions to the North American environment, stricter regulations were developed during the 1970/80s by both the U.S. and Canada. Since it was recognized that air deposition was the major source of elevated mercury found at most places,[7] various actions were taken to reduce mercury emissions to air from, for example, switches in automobiles and from coal-fired electricity generation, which is the largest remaining anthropogenic source of mercury in Canada.[8] Further, the U.S. and Canada agreed on the Binational Toxics Strategy in 1997 to virtually eliminate persistent toxic substances including mercury from the Great Lakes basin.[9] In response to these actions, anthropogenic mercury emissions in Canada declined from approximately 80 to 6 tonnes (>90%) between the 1970s and 2010.[8] The use of mercury in the U.S. declined from 1500 to 2000 tons during the 1970/80s to <400 tons in the late 1990s (derived from Cain et al., 2008). Product-related U.S. mercury air emissions declined from >200 t in 1990 to about 30 tons in 2005 (derived from Cain et al., 2008), and total U.S. mercury emissions declined from about 246 tons in 1990 to 105 tons in 2005 and 61 tons in 2008.[11] Emissions for utility coal boilers still remain the largest (~50%) contributor of anthropogenic mercury emissions in the U.S.[11] In contrast, global atmospheric emissions during the 1990s and 2000s have been generally stable in the range of 1800−2000 tonnes per year. However, they appear to be increasing lately due to increasing emissions from Asia.[12]

Although atmospheric input of mercury is just one of the factors influencing fish mercury levels, it has been shown that changes in atmospheric input may be linearly correlated to mercury in aquatic biota in some cases.[13] As such, in response

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to reduced mercury emissions in North America, it could be expected that fish mercury levels declined, especially in recent years. This is particularly relevant to the Province of Ontario, Canada, where most (90%) of mercury emissions can be attributed to anthropogenic emissions.[14] However, some recent studies focusing on various parts of North America including the Great Lakes have shown conflicting and/or mixed temporal trends.[15−19]

In this study we used data from >200 000 consistent fish mercury measurements collected by Ontario Ministry of the Environment (OMOE), Ontario, Canada over the last 40 years (1970s to 2012) to examine long-term and recent trends. The province of Ontario covers a wide geographical area (approximately 1.5 and 3 times larger than France and Germany, respectively; spans approximately from 41.5° to 56.5° N and 73° to 95° W), and contains about one-third of the world's fresh water. As such, the trends derived from this data set may reflect changes and impacts on a large scale. Data analyses were conducted for overall Ontario as well as northern and southern Ontario separately to investigate if any regional differences in various affecting factors translated into diverging fish mercury trends.

## ■ METHODS

**Sample Collection and Analysis.** The OMOE, in partnership with the Ontario Ministry of Natural Resources and other agencies, has monitored mercury levels in fish from various locations (lakes/rivers/streams) in Ontario, including the Canadian waters of the Great Lakes, since the 1970s. Fish were collected using a variety of methods, such as gill netting, trap netting, electrofishing, and angling. Fish were measured for total length and weight, and also sexed in most cases. Most measurements are for skinless boneless dorsal fillets. The samples were ground and stored at −20 °C until mercury analysis using acid digestion and cold vapor flameless atomic absorption spectroscopy. The method has been previously described by Bhavsar et al.[18]

**Data Screening.** To perform a comprehensive trend analysis, consideration of more than one fish species is advisible.[18] Three species widely distributed in Ontario, namely, Walleye (WE, *Sander vitreus*), Northern Pike (NP, *Esox lucius*), and Lake Trout (LT, *Salvelinus namaycush*), were selected. Since these species are predatory, high mercury levels could be anticipated due to biomagnification. Further, all three species are among the popular Ontario game fishes. The initial data set included 47 797 WE, 30 819 NP, and 19 272 LT wet weight mercury measurements. We then excluded measurements collected from the Canadian waters of the Great Lakes and easily identifiable point-source impacted locations, as they may be experiencing trends that are not a reflection of other large scale changes. All river and creek locations were also not considered because samples may have been collected from different locations in a river/creek over time and, in general, rivers/creeks exhibit greater spatial variation in fish mercury than a lake. The final data set included 31 743 WE measurements from 1167 locations, 21 901 NP measurements from 1240 locations, and 13 539 LT measurements from 583 locations, collected between 1970 and 2012. The data was further screened to only include species/location/year combinations with a minimum of five measurements and a 10 cm size range to avoid inclusion of weak sampling events (i.e., difference between maximum and minimum fish lengths) (Supporting Information Figure S1).

**Standard Length Calculations.** For the species considered in this study, it has been shown that fish mercury concentrations increase with fish size.[20] To account for such an effect while evaluating concentration trends, power series regressions of fish mercury against size were constructed for each sampling event using the equation $Y = a\,X^b$ (where $Y$ is mercury concentration in $\mu$g/g wet weight, $X$ is fish length in centimeters, and $a,b$ are regression coefficients). The power series regressions were conducted by fitting linear regressions on logarithmically transformed values (i.e., $\log Y = \log a + b \log X$). We note that the exponential model (i.e., $Y = a{*}\exp(bX)$) is also applicable to the relationship between mercury concentration and fish length; however, we opted for the power function because it generally performs better than the exponential function.[20] This resulted in a total of 4234 power series regressions for every combination of species, location, and year (1748 WE, 1620 NP, 866 LT).

Based on the literature,[20] only sampling events resulting in positive relationships between fish length and mercury concentrations were considered. Using these regressions, mercury concentrations were calculated for three standard fish lengths (std-lengths) representing small, medium, and large sizes. Based on the literature[21] and measurements available in the data set, selected std-lengths were as follows: 40, 50, and 60 cm for WE; 45, 60, and 70 cm for NP; and 45, 60, and 70 cm for LT. To avoid large extrapolation of the power series regressions while calculating each std-length fish concentration, only sampling events with the smallest fish smaller than a std-length plus 15 cm and the largest fish larger than std-length minus 15 cm were considered. For example, as illustrated in Figure S1, to calculate the mercury concentration of a 50 cm WE for a particular year/location, the smallest WE measured for that sampling event should be smaller than 65 cm and the largest fish should be larger than 35 cm. Selection of the 15 cm buffer size was arbitrary and based on balancing maximization of locations and minimizing extrapolation.

In total, 11 264 std-length/species/location/year specific mercury concentrations were considered (small, medium, large: WE − 1614, 1616, 1616; NP − 1394, 1411, 1411; LT − 748, 727, 727; respectively). The number of locations varied with size categories for each species and time period considered, and have been presented in Table 1 and described in the Results section.

**Temporal Trend Analysis.** To compare historical (1970−1985) and recent (2000−2012) fish mercury levels, the mean of standardized mercury concentrations for all available time points within each of the two periods for each location/species/std-length was calculated. Detailed temporal trends were also analyzed, using four different time periods: (1) For the full scenario, which captures all available years (1970−2012), only those locations/species/std-lengths with at least one time point before 1985 and one time point after 2000 were considered. (2) For a historical trend scenario (1970s−1990), only those locations/species/std-lengths with at least one time point before 1982 and one time point between 1983 and 1990 (inclusive) were considered. (3) For the intermediate scenario (1985−2005), only those locations/species/std-lengths with at least one time point within 1985−1994 (inclusive) and one time point within 1996−2005 (inclusive) were considered. (4) For the recent scenario (1995−2012), only those locations/species/std-lengths with at least one time point within 1995−2002 (inclusive) and one time point within 2004−2012 (inclusive) were considered. The selection of time periods was based on studies indicating that trends of mercury and other contaminants in the Great Lakes region might have changed in the late 1980s or 1990s.[16−18,22−24]

Table 1. Summary of Spatial and Temporal Trends in Fish Mercury Levels for the Province of Ontario, Canada[a]

| | | Overall Ontario | | | | | | | | | Northern Ontario | | | | | | | | | Southern Ontario | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | Walleye | | | Northern Pike | | | Lake Trout | | | Walleye | | | Northern Pike | | | Lake Trout | | | Walleye | | | Northern Pike | | | Lake Trout | | |
| Std L (cm) → | | 40 | 50 | 60 | 45 | 60 | 70 | 45 | 60 | 70 | 40 | 50 | 60 | 45 | 60 | 70 | 45 | 60 | 70 | 40 | 50 | 60 | 45 | 60 | 70 | 45 | 60 | 70 |
| # of locations | 1970−2012 | 187 | 186 | 186 | 125 | 128 | 128 | 108 | 105 | 105 | 128 | 127 | 127 | 102 | 105 | 105 | 42 | 39 | 39 | 59 | 59 | 59 | 23 | 23 | 23 | 66 | 66 | 66 |
| | 1970−1990 | 48 | 48 | 48 | 40 | 42 | 42 | 23 | 22 | 22 | 35 | 35 | 35 | 28 | 29 | 29 | 12 | 10 | 10 | 13 | 13 | 13 | 12 | 13 | 13 | 11 | 12 | 12 |
| | 1985−2005 | 58 | 58 | 58 | 34 | 34 | 34 | 19 | 17 | 17 | 39 | 39 | 39 | 20 | 20 | 20 | 11 | 10 | 10 | 19 | 19 | 19 | 14 | 14 | 14 | 8 | 7 | 7 |
| | 1995−2012 | 85 | 85 | 85 | 58 | 62 | 62 | 20 | 19 | 19 | 53 | 53 | 53 | 38 | 40 | 40 | 11 | 11 | 11 | 32 | 32 | 32 | 20 | 22 | 22 | 9 | 8 | 8 |
| Δ[Hg] (μg/g ww/decade) | 1970−2012 | −0.03 | −0.04 | −0.05 | −0.01 | −0.01 | 0.00 | −0.02 | −0.04 | −0.06 | −0.03 | −0.03 | −0.04 | −0.01 | 0.00 | 0.01 | 0.00 | 0.00 | 0.00 | −0.03 | −0.05 | −0.07 | −0.03 | −0.03 | −0.03 | −0.03 | −0.07 | −0.10 |
| | 1970−1990 | −0.07 | −0.09 | −0.10 | −0.15 | −0.19 | −0.25 | 0.03 | −0.07 | −0.19 | −0.05 | −0.08 | −0.11 | −0.19 | −0.23 | −0.31 | 0.03 | −0.07 | −0.13 | −0.12 | −0.11 | −0.08 | −0.05 | −0.09 | −0.13 | 0.02 | −0.06 | −0.25 |
| | 1985−2005 | −0.05 | −0.09 | −0.13 | 0.03 | 0.03 | 0.01 | 0.00 | −0.02 | −0.02 | −0.06 | −0.10 | −0.16 | 0.03 | 0.01 | −0.04 | 0.01 | 0.03 | 0.06 | −0.04 | −0.05 | −0.07 | 0.03 | 0.06 | 0.08 | −0.01 | −0.08 | −0.14 |
| | 1995−2012 | 0.07 | 0.10 | 0.13 | 0.01 | 0.07 | 0.15 | 0.05 | 0.15 | 0.27 | 0.09 | 0.12 | 0.16 | 0.01 | 0.09 | 0.19 | 0.01 | 0.02 | 0.02 | 0.03 | 0.05 | 0.07 | 0.02 | 0.04 | 0.07 | 0.09 | 0.34 | 0.60 |
| Decline at (% of locations) | 1970−2012 | 61% | 60°% | 55% | 57% | 51% | 47% | 58% | 60% | 62% | 59% | 54% | 49% | 55% | 50% | 46% | 38% | 41% | 44% | 66% | 73% | 68% | 65% | 57% | 52% | 71% | 71% | 73% |
| | 1970−1990 | 65% | 58°% | 56% | 68% | 69% | 74% | 30% | 36% | 45% | 63% | 57% | 57% | 71% | 69% | 72% | 33% | 60% | 60% | 69% | 62% | 54% | 58% | 69% | 77% | 27% | 17% | 33% |
| | 1985−2005 | 62% | 71% | 67% | 35% | 41% | 53% | 47% | 47% | 47% | 67% | 74% | 72% | 35% | 55% | 70% | 45% | 40% | 30% | 53% | 63% | 58% | 36% | 21% | 29% | 50% | 57% | 71% |
| | 1995−2012 | 34% | 27% | 35% | 41% | 31% | 32% | 40% | 37% | 32% | 21% | 23% | 28% | 47% | 30% | 30% | 45% | 45% | 45% | 56% | 34% | 47% | 30% | 32% | 36% | 33% | 25% | 13% |
| Δ[Hg] for locations with -ve trend (μg/g ww/decade) | 1970−2012 | −0.07 | −0.10 | −0.17 | −0.05 | −0.07 | −0.10 | −0.05 | −0.11 | −0.16 | −0.07 | −0.11 | −0.18 | −0.05 | −0.07 | −0.10 | −0.04 | −0.08 | −0.11 | −0.08 | −0.09 | −0.15 | −0.06 | −0.09 | −0.11 | −0.06 | −0.12 | −0.17 |
| | 1970−1990 | −0.17 | −0.26 | −0.41 | −0.24 | −0.32 | −0.40 | −0.07 | −0.65 | −1.12 | −0.16 | −0.27 | −0.42 | −0.28 | −0.39 | −0.48 | −0.05 | −0.24 | −0.42 | −0.20 | −0.24 | −0.38 | −0.14 | −0.16 | −0.22 | −0.10 | −1.88 | −2.18 |
| | 1985−2005 | −0.14 | −0.19 | −0.33 | −0.06 | −0.10 | −0.13 | −0.04 | −0.10 | −0.15 | −0.15 | −0.21 | −0.34 | −0.07 | −0.11 | −0.16 | −0.01 | −0.03 | −0.07 | −0.10 | −0.14 | −0.29 | −0.05 | −0.06 | −0.05 | −0.08 | −0.17 | −0.20 |
| | 1995−2012 | −0.09 | −0.13 | −0.18 | −0.11 | −0.13 | −0.15 | −0.04 | −0.09 | −0.18 | −0.08 | −0.08 | −0.11 | −0.10 | −0.12 | −0.12 | −0.03 | −0.07 | −0.14 | −0.10 | −0.19 | −0.25 | −0.12 | −0.14 | −0.18 | −0.05 | −0.14 | −0.38 |
| Δ[Hg] for locations with +ve trend (μg/g ww/decade) | 1970−2012 | 0.04 | 0.06 | 0.10 | 0.04 | 0.06 | 0.10 | 0.03 | 0.05 | 0.09 | 0.03 | 0.06 | 0.10 | 0.04 | 0.06 | 0.10 | 0.03 | 0.05 | 0.09 | 0.05 | 0.08 | 0.12 | 0.03 | 0.04 | 0.06 | 0.03 | 0.05 | 0.09 |
| | 1970−1990 | 0.12 | 0.16 | 0.30 | 0.04 | 0.09 | 0.16 | 0.07 | 0.26 | 0.58 | 0.14 | 0.18 | 0.31 | 0.02 | 0.10 | 0.16 | 0.07 | 0.17 | 0.31 | 0.07 | 0.11 | 0.27 | 0.07 | 0.06 | 0.16 | 0.07 | 0.30 | 0.72 |
| | 1985−2005 | 0.09 | 0.17 | 0.28 | 0.08 | 0.12 | 0.17 | 0.04 | 0.06 | 0.09 | 0.12 | 0.21 | 0.31 | 0.09 | 0.15 | 0.25 | 0.03 | 0.07 | 0.11 | 0.03 | 0.11 | 0.24 | 0.08 | 0.09 | 0.13 | 0.05 | 0.04 | 0.02 |
| | 1995−2012 | 0.15 | 0.18 | 0.29 | 0.10 | 0.16 | 0.29 | 0.10 | 0.30 | 0.47 | 0.14 | 0.18 | 0.26 | 0.11 | 0.18 | 0.32 | 0.04 | 0.10 | 0.16 | 0.20 | 0.18 | 0.36 | 0.09 | 0.13 | 0.22 | 0.16 | 0.50 | 0.74 |
| Mixed Effect Model Results | Length (cm) → | 35−45 | 45−55 | 55−65 | 40−50 | 55−65 | 65−75 | 40−50 | 55−65 | 65−75 | 35−45 | 45−55 | 55−65 | 40−50 | 55−65 | 65−75 | 40−50 | 55−65 | 65−75 | 35−45 | 45−55 | 55−65 | 40−50 | 55−65 | 65−75 | 40−50 | 55−65 | 65−75 |
| # of locations | 1970−1990 | 641 | 623 | 362 | 599 | 645 | 485 | 238 | 224 | 178 | 537 | 516 | 273 | 520 | 569 | 420 | 122 | 124 | 84 | 104 | 107 | 89 | 79 | 76 | 65 | 116 | 100 | 94 |
| | 1995−2012 | 682 | 664 | 532 | 493 | 605 | 505 | 312 | 317 | 243 | 546 | 524 | 425 | 384 | 486 | 409 | 225 | 237 | 175 | 136 | 140 | 107 | 109 | 119 | 96 | 87 | 87 | 68 |
| Hg measurements | 1970−1990 | 6435 | 4387 | 1192 | 3072 | 3668 | 1800 | 1422 | 1135 | 674 | 5579 | 3566 | 836 | 2644 | 3322 | 1605 | 757 | 568 | 274 | 856 | 821 | 356 | 428 | 346 | 195 | 665 | 567 | 400 |
| | 1995−2012 | 5659 | 4160 | 1973 | 1864 | 2787 | 1603 | 1679 | 1577 | 737 | 4493 | 3142 | 1590 | 1387 | 2216 | 1292 | 1127 | 1128 | 528 | 1166 | 1018 | 383 | 477 | 571 | 311 | 552 | 552 | 209 |
| Δ[Hg] (μg/g ww/decade) | 1970−1990 | **-0.08** | **-0.07** | **-0.10** | **-0.15** | **-0.17** | **-0.18** | 0.00 | **0.09** | 0.11 | **-0.09** | **-0.10** | **-0.17** | **-0.16** | **-0.18** | **-0.19** | **-0.06** | −0.01 | **-0.32** | −0.04 | 0.00 | 0.01 | **-0.11** | **-0.10** | −0.10 | **0.07** | **0.15** | 0.19 |
| | 1995−2012 | **0.05** | **0.08** | **0.11** | **0.02** | **0.09** | **0.13** | **0.06** | **0.08** | 0.07 | **0.07** | **0.10** | **0.11** | **0.03** | **0.09** | **0.14** | 0.06 | **0.13** | **0.14** | −0.03 | 0.02 | 0.10 | 0.02 | **0.05** | 0.04 | 0.06 | 0.01 | 0.04 |
| Δ[Hg] (annual % change, APC) | 1970−1990 | **-1.23** | −0.29 | 0.26 | **-3.00** | **-2.21** | **-1.52** | 0.56 | 0.69 | 0.47 | **-1.67** | **-0.98** | **-0.98** | **-2.98** | **-2.13** | **-1.54** | −0.10 | 0.17 | **-3.83** | 0.45 | **1.33** | **2.02** | **-3.09** | **-2.25** | −1.64 | 1.31 | 1.08 | 1.21 |
| | 1995−2012 | **1.55** | **1.31** | **1.38** | 0.55 | **1.71** | **1.93** | 1.07 | 0.50 | 0.03 | **2.19** | **1.72** | **1.42** | 0.39 | **1.96** | **2.36** | 1.13 | **1.41** | 1.29 | −0.14 | 0.40 | 1.16 | 0.72 | 0.87 | 0.11 | 1.51 | 1.51 | −1.23 |

[a] The results presented here required a minimum of 2 time points for each time period considered. Please refer the Method section for details on the data screening. Δ[Hg] is for change in fish mercury concentrations. Mixed Effect Modeling (MEM) results are presented as change/decade and annual percentage (%) change (APC) in fish mercury levels for small, medium, and large sized fish. The statistically significant MEM change/decade and APC values are highlighted in bold fonts. [Please refer to the Supporting Information for detailed results of MEM.]

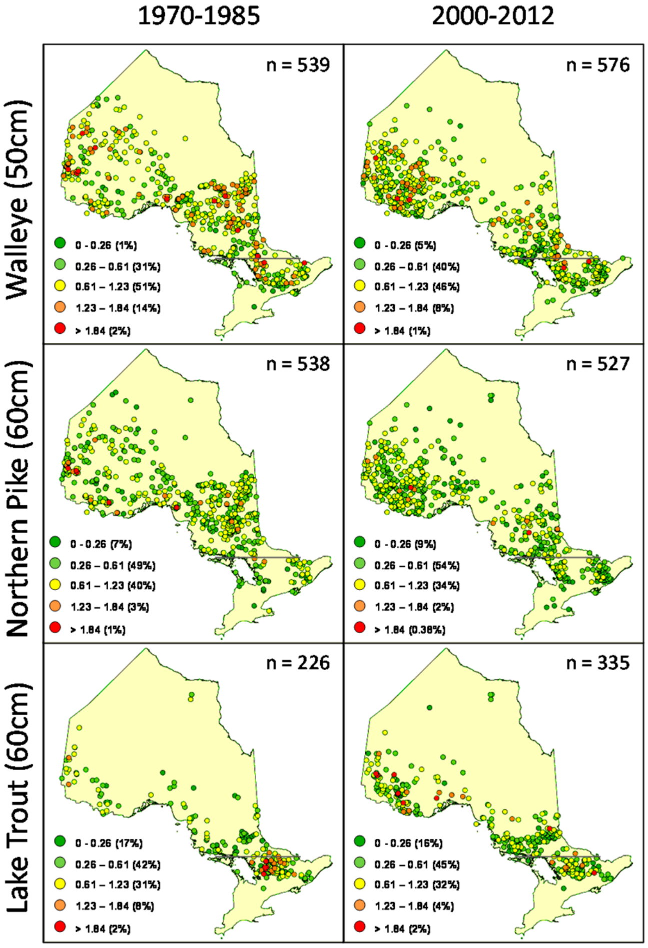

**Figure 1.** Mercury concentrations ($\mu$g/g wet weight) for two time periods (historical 1970−1985, recent 2000−2012) in skin-off fillets of medium sized Ontario Walleye, Northern Pike, and Lake Trout. The concentrations have been grouped into the various categories used by the Ontario Ministry of the Environment for the issuance of fish consumption advisories.[28] *n* represents number of locations. The horizontal line shows approximate demarcation between northern and southern Ontario described in this study.

For all four scenarios, linear regression was performed on mercury measurements for each location/species/std-length against year, and the slope of the regression was multiplied by 10 to calculate rate of change in fish mercury levels per decade. For all time periods and geographic regional scenarios (described below) as well as location/species/std-length, another set of linear regressions was also conducted by further constraining the requirement of available time points to a minimum of three (Table S1) compared to the previous requirement of a minimum of two (Table 1).

We conducted mixed effect linear modeling (MEM), using maximum likelihood estimation, to examine the temporal mercury trends.[16] The first analysis was species- and region-specific for logarithmically transformed mercury against fixed effect of sample year, and random effects of sample year, fish length, and sampling location. The results are presented as linear temporal trend, random year deviations from the linear trend, and LOWESS (locally weighted scatterplot) smoothing, which is a nonparametric regression method that combines multiple regression models in a *k*-nearest-neighbor-based meta-model.[16] The smoothing parameter value was set at 0.25 for LOWESS. The second analysis was conducted for both original and logarithmically transformed mercury against sample year as a fixed effect and sampling location as a random effect. In this analysis, fish length was accounted for by applying the model separately on small, medium, and large sized fish. The size classes were 10 cm in range around the species-specific, three standard lengths described above.

**Spatial Trend Analysis.** Separate analyses were conducted for overall Ontario, northern Ontario (>46°N), and southern Ontario (<46°N) to capture geographical differences. Most of northern Ontario is on a rocky plateau known as the Canadian Shield, and experiences extreme temperatures. Mining, forestry, and waterpower are the major industries in northern Ontario. The data screening resulted in a varying number of locations for each scenario ranging as high as 186−187 for the WE/overall Ontario/1970−2012 scenario (Table 1). In general, WE locations were equal or greater in number than those for NP and LT (Table 1).

**Statistical Comparison.** A statistical comparison of historical and recent fish mercury concentrations as well as rates of change was conducted using *t* tests on logarithmically transformed values. A constant was added to manage negative values for the rates. When the assumptions of normal distribution and/or equal variance in the observations tested using Shapiro and Bartlett tests, respectively, were not valid, the nonparametric Mann−Whitney U-test was conducted. Statistical significance was set at $p < 0.05$. The analyses were conducted using R v 3.0.1 (www.r-project.org).

Effect sizes, along with 95% confidence intervals, quantifying the difference between the rate of change in fish mercury concentration for two time periods (1970−1990 and 1995−2012) were also calculated using the Cohen's d method, pooled standard deviation, and Effect Size Calculator Excel template.[25] Effect size emphasizes the magnitude of the difference rather than confounding this with sample size and statistical significance, and can be a better indicator of importance of findings.[26] Effect sizes of 0.2, 0.5, and 0.8 are considered small, medium, and large, where a large difference corresponds to the difference between the heights of 13 and 18 year old girls.[25,27]

Fish consumption advisory benchmarks used to provide context of mercury levels are from the OMOE[28] and as follows: 0.26 $\mu$g/g to change an advisory from 8 to 4 meals/month for the sensitive population of children and women of child-bearing age; and 0.61, 1.23, and 1.84 $\mu$g/g to change an advisory from 8 to 4 meals/month, 4 to 2 meals/month, and 2 to 0 meals/month for the general population, respectively.

## ■ RESULTS

**Overall Ontario.** *Historical vs Recent.* Historically (1970−1985), 1%, 7%, and 17% of 226−539 *medium size* WE, NP, and LT locations, respectively, had fish mercury levels <0.26 $\mu$g/g (Figure 1). The corresponding values for the recent scenario (2000−2012) were generally better at 5%, 9%, and 16% of the 335−576 locations (Figure 1). The proportions of WE, NP, and LT locations with fish mercury levels <0.61 $\mu$g/g were 32%, 56%, 59% historically and 45%, 63%, 61% recently, respectively (Figure 1). Compared to the historical mercury levels, recent levels were significantly lower for WE and NP ($p < 0.001$) and unchanged for LT ($p = 0.92$) (Figure 2). The corresponding results for the small and large size fish were similar (Figure S2). These results indicate that overall, fish mercury levels declined or remained stable in Ontario between the 1970s and 2012.

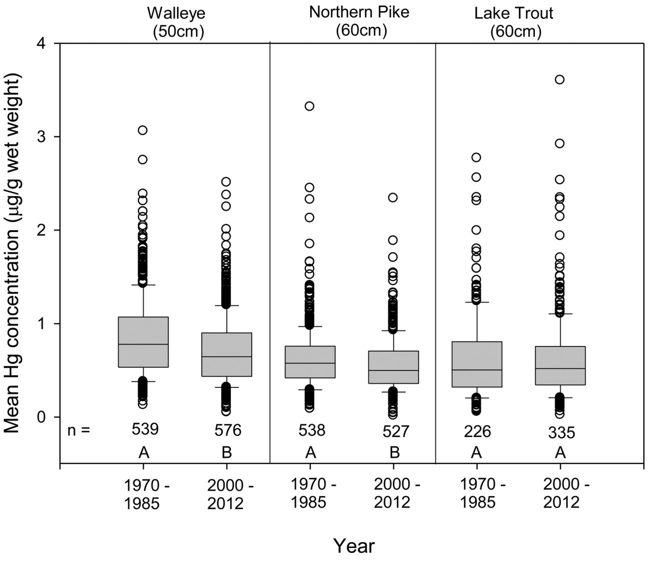

**Figure 2.** Mercury concentrations (µg/g wet weight) for two time periods (historical 1970−1985, recent 2000−2012) in skin-off fillets of medium sized Ontario Walleye, Northern Pike, and Lake Trout. $n$ represents the number of locations. For each species, identical letters belong to the same statistical group, where group A is significantly different ($p < 0.05$) from group B.

*Rates of Change.* For the 1970s−2012 period, mercury in Ontario declined at average rates of 0.03−0.05 µg/g/decade for WE, 0.002−0.01 µg/g/decade for NP, and 0.02−0.06 µg/g/decade for LT (Table 1, Figure 3). For each species, the declines in large fish were generally 2−3-fold greater than the corresponding small fish. A breakdown of the time period revealed that the rates of mercury decline were substantially greater during the 1970s/80s (average in µg/g/decade: WE, 0.07−0.10; NP, 0.15−0.25; LT, (0.03)−0.19; Table 1, Figure 3). For the intermediate time period of 1985−2005, average mercury concentrations were still declining for WE (0.05−0.13 µg/g/decade) and LT (0.001−0.02 µg/g/decade); however, the concentrations tended to increase for NP (0.01−0.03 µg/g/decade) (Table 1, Figure 3). In contrast, for the recent (1995−2012) time period, the average rates of change were positive, suggesting increases in fish mercury levels (at the rates of 0.07−0.13 µg/g/decade for WE, 0.01−0.15 µg/g/decade for NP, and 0.05−0.27 µg/g/decade for LT; Table 1, Figure 3). Medians of rates of change for 1995−2012 were also positive for all types/sizes of the fish considered (except close to zero for medium sized LT) (Figure 3). The statistical comparisons also indicate significant differences in the spreads of rates of mercury changes in WE and NP between 1970−1990 and 1995−2012 (Figure 3). Effect sizes of 0.59−0.76 for WE and 0.76−1.04 for NP for the differences in the rates of mercury change during the two periods highlight a substantial shift in trends of fish mercury levels (Figure 3). In contrast, effect sizes (0.19−0.34) and statistical significance tests for LT suggest a negligible shift in the trends (Figure 3).

*Proportion of Locations.* Fish mercury levels overall declined at 47−62% of the locations between the 1970s and 2012 (55−61% for WE, 47−57% for NP, 58−62% for LT; Table 1, Figure 3). Similar (WE), greater (NP), and lower (LT) proportions of locations experienced declines during the 1970s/80s (56−65% for WE, 68−74% for NP, 30−45% for LT; Table 1, Figure 3). The proportions of WE and NP locations showing declines in mercury levels have decreased over time (only 27−35% of WE and 31−41% of NP locations experienced a declining trend during the 1970s/80s; Table 1, Figure 3). The proportions of LT locations showing declines in mercury levels have generally remained stable with time (Table 1, Figure 3).

*Rates of Change for Specific Locations.* For the locations that showed declines, the average rates of declines dropped between 1970s/80s and 1995−2012 (from 0.17 to 0.41 to 0.09−0.18 µg/g/decade for WE, 0.24−0.40 to 0.11−0.15 µg/g/decade for NP, and 0.07−1.12 to 0.04−0.18 µg/g/decade for LT; Table 1). For WE and LT locations showing increasing trends, the rates of increases have not changed between the 1970s/80s and 1995−2012 (0.12−0.30 to 0.15−0.29 µg/g/decade for WE and 0.07−0.58 to 0.10−0.47 µg/g/decade for LT); however, proportions of WE locations showing increasing trends have increased (from 35−44% to 65−73%) (Table 1, Figure 3). In contrast, for NP, both the average rates of increases (for the locations showing increasing trends) and proportions of locations showing increasing trends have grown between the 1970s/80s and 1995−2012 (0.04−0.16 to 0.10−0.29 µg/g/decade, 26−32% to 59−69%; Table 1).

**Northern and Southern Ontario.** Mercury levels for northern Ontario generally declined (WE and NP; $p < 0.05$) or remained unchanged (LT; $p > 0.05$) between the 1970s and 2012 (Figure S2). In contrast, measurements for southern Ontario indicate mostly flat mercury concentrations over the past four decades (Figure S2). Although the LT regional pattern differed from those for WE and NP, this disagreement could be a result of the low number of LT locations meeting the data screening criteria and these limited number of locations showing a wide range of rates of mercury change (Table 1, Figure S3). We limit the following discussion to WE and NP.

Over the past four decades, about half (46−59%) of the WE and NP northern Ontario locations experienced declines in fish mercury (0.01−0.04 µg/g/decade; Table 1, Figure S3a). The corresponding observations for southern Ontario were better (52−73% and 0.03−0.07 µg/g/decade; Table 1, Figure S3b). WE and NP mercury levels *declined* substantially between the 1970s and 1990 (average rates: 0.05−0.11 and 0.19−0.31 µg/g/decade for northern Ontario, and 0.08−0.12 and 0.05−0.13 µg/g/decade for southern Ontario, respectively; Table 1). In recent years (1995−2012), WE and NP mercury levels are *increasing* in northern Ontario (at average rates of 0.09−0.16 and 0.01−0.19 µg/g/decade, respectively; Table 1, Figure S3a). In comparison, the corresponding increases for southern Ontario are moderate (0.03−0.07 and 0.02−0.07 µg/g/decade, respectively; Table 1, Figure S3b).

The proportions of northern Ontario locations showing declines in WE and NP mercury levels decreased from 57−72% during the 1970s/80s to 21−47% during 1995−2012 (Table 1, Figure S3a). In contrast, the corresponding drops were lower for southern Ontario (from 54−77% to 30−56%; Table 1, Figure S3b). The average rate of mercury decline for northern Ontario locations showed that the declining trends are much lower for the recent time period compared to those during the 1970s/80s (for WE and NP: Recent: 0.08−0.11 and 0.10−0.12 µg/g/decade, respectively; 1970s/80s: 0.16−0.42 and 0.28−0.48 µg/g/decade, respectively; Table 1).

For northern Ontario NP, the proportion of locations showing increasing trends increased between the 1970s/80s and 1995−2012. Additionally, the rates of increase for those locations also rose (average from 0.02−0.16 to 0.11−0.32 µg/g/decade). However, the corresponding WE rates were almost unchanged (0.14−0.31 and 0.14−0.26 µg/g/decade; Table 1). In contrast,

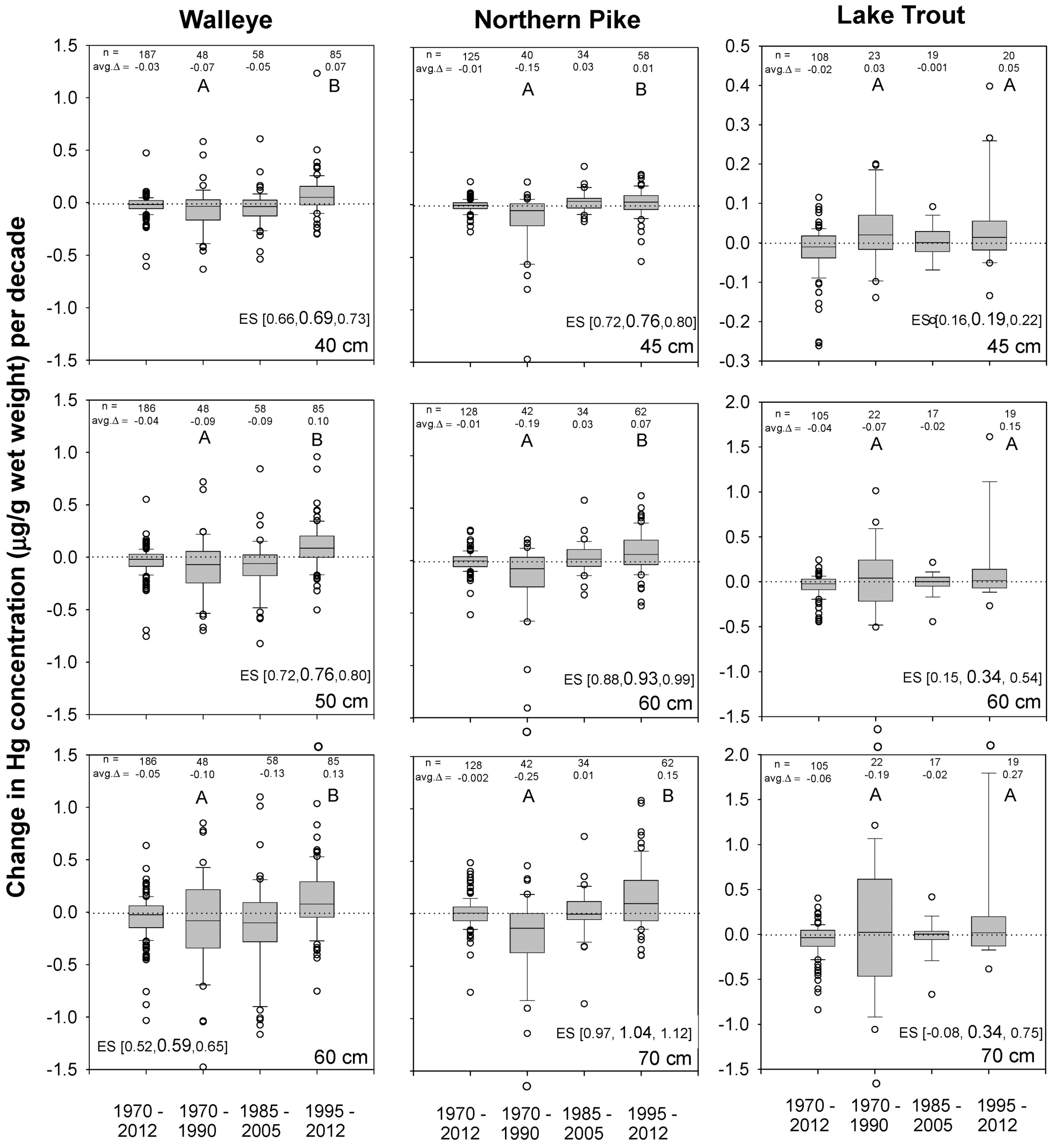

**Figure 3.** Box plots of rates of mercury change (*µ*g/g wet weight per decade) for different time periods in skin-off fillets of small, medium, and large sized Walleye, Northern Pike, and Lake Trout. The dotted line represents no change. Average values of mercury change are also presented. *n* represents the number of locations. For each species/std-length and the 1970–1990 and 1995–2012 time periods, ES (effect size) indicates the difference (with 95% confidence intervals), and identical letters belong to the same statistical group, where, for example, group A is significantly different ($p < 0.05$) from group B. Measurements plotted outside the chart area represent values outside the range of the *Y*-axis.

although the proportion of southern Ontario WE and NP locations with increasing mercury trends has increased over time (Figure S3b), the rates of mercury increase for those locations showed only a moderate increase (0.07–0.27 to 0.20–0.36 *µ*g/g/decade for WE and 0.07–0.16 to 0.09–0.22 *µ*g/g/decade for NP; Table 1). Effect sizes for the differences in the rates of change for northern Ontario WE and NP mercury concentrations between 1970s–1990 and 1995–2012 ranged from 0.70–0.84 and 0.83–1.15, respectively (Figure S3a), indicating a substantial shift toward increasing trends of fish mercury levels in recent years. The corresponding effect sizes for southern Ontario ranged from 0.39 to 0.66 for WE and 0.60–0.83 for NP, suggesting a relatively smaller change in fish mercury trends (Figure S3b).

Spatial distributions of rates of change in mercury levels of small, medium, and large size fish for various time periods show a clear change in fish mercury trends from *declining* during the 1970s/80s to *flat/increasing* during 1995–2012, especially for northern Ontario (Figures 4 and S4a,b). A more constrained data set with a requirement of a minimum of three time points generally produced similar results as presented above for the data set based on a minimum of two time points (Tables 1 and S1).

To further confirm the above findings, we also compared fish mercury levels for only those locations that met our data screening criteria for all four temporal trend scenarios. This required at least one time point in measurements for each of pre-1982, 1983–1990, 1985–1994, 1996–2005, 1995–2002, and 2004–2012 time periods for the same location (overlaps in the periods due to overlapping requirements in the individual scenarios). This more stringent requirement, however, reduced the available number of locations for the analysis (WE: 15, NP: 8–9, LT: 4; Figure S5). In this scenario, fish mercury levels for different time periods are compared for the same set of locations for each species/size. As shown in Figure S5, the results were similar to the findings from the above analysis and further strengthened the

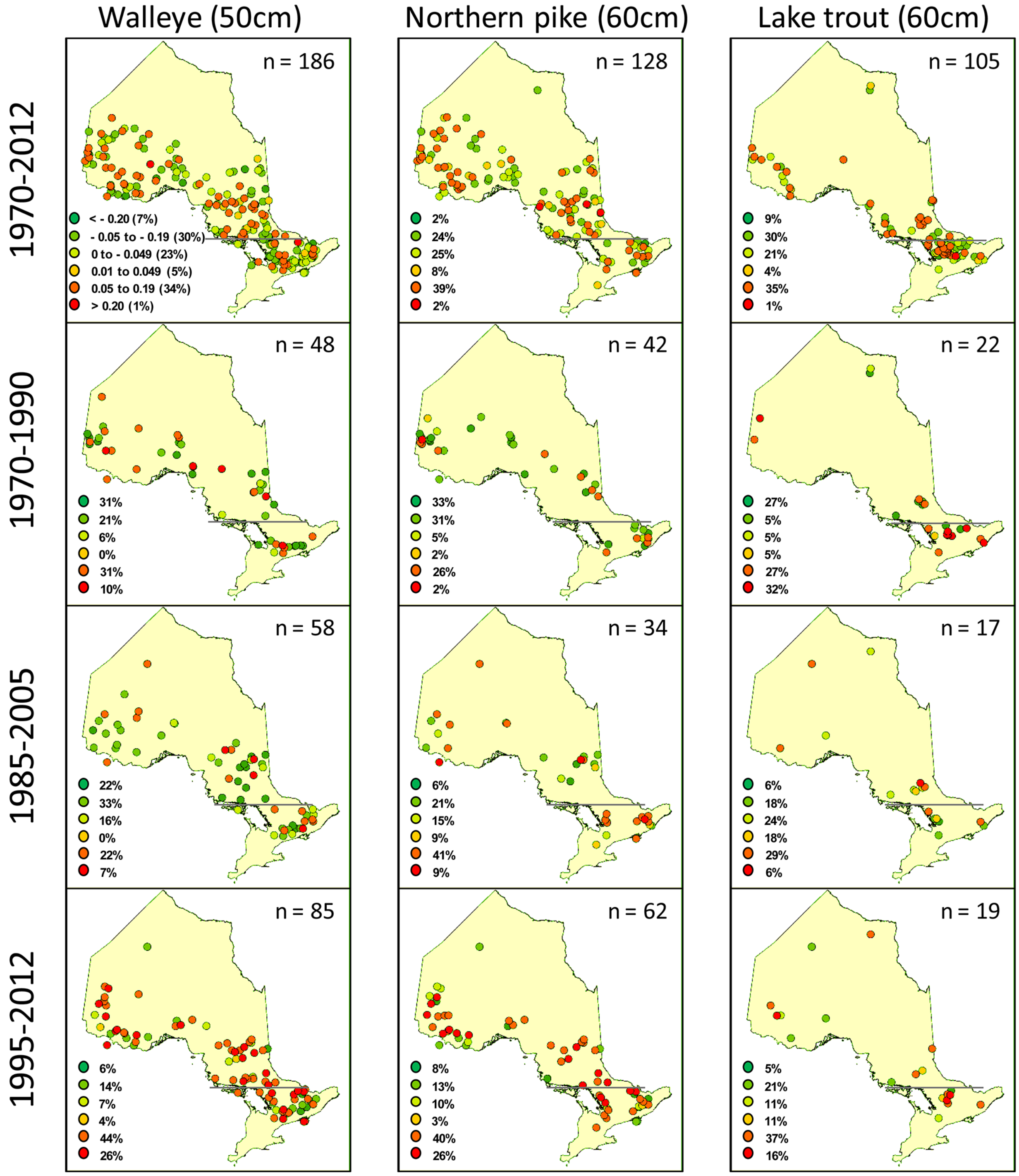

**Figure 4.** Spatial distribution of rates of mercury change ($\mu$g/g wet weight per decade) for different time periods in skin-off fillets of medium sized Ontario Walleye, Northern Pike, and Lake Trout. The rates have been grouped into various categories. Percentage of total locations within each category is also presented. *n* represents the number of locations for each time period and species. The horizontal line shows approximate demarcation between northern and southern Ontario described in this study.

conclusion that mercury levels in Ontario fish (especially in WE and NP from northern Ontario) appear to be increasing in recent years.

Overall species- and region-specific mercury trends from the mixed effect modeling are similar to those described above (Figure 5). Strong decline rates are evident for the 1970−1990 period for WE, NP (northern Ontario), and LT (southern Ontario) (Figure 5). Increasing trends are evident for the recent times for all scenarios except WE from southern Ontario (Figure 5). The rates of mercury change from the mixed effect modeling for the three size classes of each of three species for overall as well as northern and southern Ontario are also similar to those described above (Tables 1, S2).

## ■ DISCUSSION

This study showed that mercury levels in fish from the province of Ontario, Canada, declined at 47−62% of the locations sampled between the 1970s and 2012. The results are consistent with largely (89%) flat/downward trends from

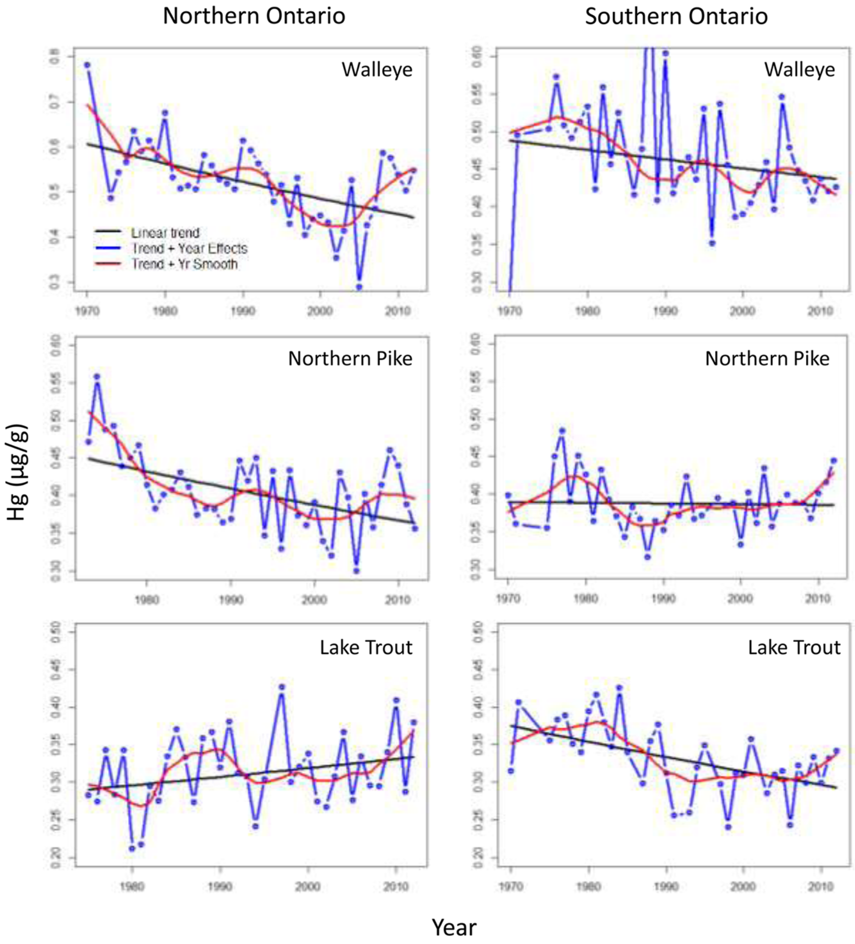

**Figure 5.** Mixed effect modeling results presented as linear temporal trend, random year deviations from the linear trend, and LOWESS smoothers for the trends for Walleye, Northern Pike, and Lake trout for northern and southern Ontario, separately.

1969 to 2005 for 90 U.S. locations, long-term walleye mercury declines in Wisconsin (77% of 420 lakes between 1982 and 2005), and an overall downward trend in five fish species from the Hudson River, NY (1970–2004) as well as yellow perch from New York lakes.[15,19,29,30] The results are also more or less consistent with the reported overall flat trend of mercury in seven fish species from 73 lakes in northern Ontario[31] and minor differences in findings can be attributed to marginal differences in the data analysis methods (e.g., differences in time period considered, 1974–1981 and 2005–2010 versus pre-1985 and 2000–2012).

A breakdown of the 1970s–2012 time period revealed that the declines in Ontario occurred at proportionally more locations and at higher rates during the 1970s/80s compared to the recent years. These results are consistent with reported declines in fish mercury at about 85% of 50 locations in the U.S. during the same time period and 64% Minnesota lakes between 1982 and 1995.[15,16] A decline in fish mercury levels during the 1970s/80s has also been reported for the Great Lakes and its basin.[16,18,23,32–35] These findings are in line with a substantial reduction in anthropogenic mercury emissions during the 1980s both in the USA (from >2000 tons in 1980 to about 700 tons in 1990) and worldwide.[4,36] In contrast, atmospheric mercury levels in northern hemisphere perhaps remained flat during the 1980s.[4] However, historical declines in fish mercury have been linked with stricter regulatory standards to curtail direct mercury discharges and declines in sediments during a similar time period.[15]

Our analysis indicates that trends in mercury levels in Ontario fish switched from declining to generally increasing during the late 1990s through 2012. Spatial analysis in this study identified the northern Ontario region with stronger increasing trends compared to southern Ontario, which has overall flat or weak/moderately increasing trends. Overall, these results are in line with generally flat fish mercury levels in recent years in U.S. lakes and rivers, an upward trend in some parts of the Great Lakes, and recent increases in fish mercury levels in 60% of Minnesota lakes.[15–19] Although the spatial (north–south) pattern is opposite that reported for Wisconsin lakes,[23] Wisconsin covers a much smaller area (169 639 versus 1 076 395 km$^2$) and has a smaller north–south spread (42° 37′ N to 47° 05′ N versus 41°41′ to 56°51′ N) than Ontario. During this same period, relatively constant atmospheric gaseous elemental mercury concentrations and wet mercury depositions at 76% of 49 Mercury Deposition Network sites in the eastern U.S. and Canada have been reported.[4,37,38]

Anthropogenic mercury emissions in Canada declined by about 90% between the 1970s and 2011: with a steady decline from 80 to 35 tonnes between 1970 and 1992, a steep decline to 13 tonnes between 1992 and 1994, and then a steady decline to 7 tonnes by 2003 and 4 tonnes by 2011.[39–41] Similarly, mercury emissions in Ontario are also declining with about 2 and 1 tonnes emitted in 2003 and 2011, respectively, representing about 25% of the anthropogenic mercury emissions in Canada.[39,40] However, trans-boundary flows of mercury are increasing and now account for >95% of mercury deposition in Canada.[8] It is estimated that the greatest contributions of mercury deposition in Canada are from sources in China (42%) and the U.S. (17%) (Environment Canada, unpublished data). Similarly, only 25−32% of mercury deposited over the continental U.S. originates from North American anthropogenic sources, while Asian anthropogenic emissions contribute 5−36% and natural emissions contribute 6−59%.[42] This is not surprising considering that 2/3 of the global anthropogenic mercury emissions in 2005 originated from Asian countries[43] and atmospheric circulation processes have a major influence on the fate of airborne mercury.[44] As such, Asian mercury emissions, which almost doubled from 700 to 1300 tonnes between 1990 and 2005,[45] may have played a role in recent fish mercury increases in Ontario.

Canada's weather has been warmer over the last 60 years with a greater impact in the high latitude regions.[46] Increasing temperatures have been reported for all parts of Ontario with a greater impact in the northwest region.[34] Increased temperatures may have raised fish mercury levels by remobilizing mercury in soil and sediments, accelerating the conversion of divalent mercury to more biologically available methylmercury, and uptake into fish.[47]

Changes in food web structures can affect mercury dynamics in an aquatic system, particularly fish mercury levels, due to bioaccumulation and biomagnification.[48] Non-native Bass species, Rainbow Smelt, and Spiny Water Flea (*Bythotrephes longimanus*) have been introduced to many Canadian Shield lakes,[49] which make up the majority of Ontario lakes considered in this study (Figure S6) and generally have higher fish mercury levels compared to other Ontario lakes.[50] Since Shield lakes are unproductive and support relatively few biotic species, Shield lakes have relatively simple, species-poor food webs that may be more vulnerable to perturbations.[49] As a result, even with relatively few invasive species, Shield lake ecosystems are experiencing substantial impacts,[49] and mercury in fish could be one of the influences. Invasive fish species can lead to increases in mercury concentrations in top piscivores, even though occasional studies have reported no such impact.[34,51−54] On the other hand, piscivorous Bass species can reduce littoral prey fish diversity, abundance, and community structure in north-temperate lakes, subsequently forcing LT to a lower trophic level.[49] Since dietary exposure (biomagnification) is a major route of mercury accumulation in fish,[51] a lower trophic level can result in reduced mercury in a top predator such as LT. This, in combination with potentially increasing mercury levels in northern Ontario lakes based on findings for WE and NP, could explain mixed (positive/negative) trends for LT in the recent times.

Mercury mobility and thereby its level in fish decreases with increasing lake pH because methylation of inorganic mercury declines due to lower abundance of sulfate-reducing bacteria.[55] The pH of many Shield lakes has been recovering (increasing) since the 1980s/90s due to decreased $SO_2$ emissions, most notably from metal smelters in Sudbury, Ontario.[56,57] This factor may have contributed to past declines in fish mercury; however, the significance of this factor may have weakened in recent times and/or been outweighed by other contrasting factors resulting in net increases in fish mercury in Ontario. For example, increasing pH can aid in invasion by Rainbow Smelt, which can persist in a variety of aquatic systems if minimal habitat and environmental conditions are available. This species can increase mercury levels in top predators, including LT and WE, by elevating their trophic levels even though increased growth rates may counter the effects.[49]

The average temperature in the North American Great Lakes basin, which covers a major portion of Ontario, could increase by about 4.5 °C by 2055 with greater impacts in northern Ontario.[58] By 2100, water temperatures in Canada may increase by 5−18 °C, with the greatest increase at northern latitudes.[59] Although many Ontario inland lakes have been invaded by some invasive species as discussed above, there is a potential for these species to expand further, especially with warming weather.[49] Many other non-native species such as rusty crayfish have the potential to invade numerous Shield lakes and could have serious impacts.[49] Introduction of more non-native species in inland locations of Ontario will be affected by climate change and other factors such as the establishment of invasive species in the Great Lakes, patterns of development, educational and outreach efforts, and legislations.[49]

Although it is logical to expect declines in fish mercury levels in response to reduced emissions in North America and lower atmospheric concentrations in recent years, other factors such as climate change,[58] invasive species,[35,60] local geochemistry, and possible increases in mercury emissions from other regions[43,61] may affect the response time and magnitude. This hypothesis is supported by differences observed in fish mercury trends for Lakes Erie and Ontario, where changes in food web structure due to invasive species, more prolonged anoxia, and possibly other local factors may have affected fish mercury trends.[18,62]

In summary, we evaluated long-term fish mercury levels and trends for the Province of Ontario, Canada. Although the mercury levels have generally declined or remained stable between the 1970s and 2012, this was mainly a result of declines which occurred during the 1970s/80s. In recent years, fish mercury levels appear to be increasing at more than half of the locations sampled, particularly in northern Ontario. It would be interesting to learn if these trends continue especially in light of the potential influence of complex factors.

## ■ ASSOCIATED CONTENT

### Ⓢ Supporting Information

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

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### Notes

The authors declare no competing financial interest.

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# Supporting Information

# Fish mercury levels appear to be increasing lately: a report from 40 years of monitoring in Province of Ontario, Canada

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**Table S1:** Summary of spatial and temporal trends in fish mercury levels for the Province of Ontario, Canada. The results presented here required minimum 3 time points for each time period considered. Please refer the Method section in the manuscript for details on the data screening.

| | | **Overall Ontario** | | | | | | | | | **Northern Ontario** | | | | | | | | | **Southern Ontario** | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | **Walleye** | | | **Northern Pike** | | | **Lake Trout** | | | **Walleye** | | | **Northern Pike** | | | **Lake Trout** | | | **Walleye** | | | **Northern Pike** | | | **Lake Trout** | | |
| | **Std L (cm) ->** | **40** | **50** | **60** | **45** | **60** | **70** | **45** | **60** | **70** | **40** | **50** | **60** | **45** | **60** | **70** | **45** | **60** | **70** | **40** | **50** | **60** | **45** | **60** | **70** | **45** | **60** | **70** |
| **Total number of locations** | **1970-2012** | 105 | 105 | 105 | 61 | 62 | 62 | 50 | 49 | 49 | 69 | 69 | 69 | 46 | 47 | 47 | 16 | 14 | 14 | 36 | 36 | 36 | 15 | 15 | 15 | 34 | 35 | 35 |
| | **1970-1990** | 22 | 22 | 22 | 13 | 13 | 13 | 6 | 4 | 4 | 14 | 14 | 14 | 11 | 10 | 10 | 4 | 2 | 2 | 8 | 8 | 8 | 2 | 3 | 3 | 2 | 2 | 2 |
| | **1985-2005** | 23 | 23 | 23 | 10 | 12 | 12 | 10 | 10 | 10 | 12 | 12 | 12 | 9 | 10 | 10 | 5 | 5 | 5 | 11 | 11 | 11 | 1 | 2 | 2 | 5 | 5 | 5 |
| | **1995-2012** | 21 | 21 | 21 | 14 | 15 | 15 | 6 | 6 | 6 | 10 | 10 | 10 | 10 | 10 | 10 | 3 | 3 | 3 | 11 | 11 | 11 | 4 | 5 | 5 | 3 | 3 | 3 |
| **Avg change/Decade (µg/g ww)** | **1970-2012** | -0.03 | -0.04 | -0.05 | -0.01 | -0.01 | -0.01 | -0.03 | -0.06 | -0.09 | -0.03 | -0.04 | -0.05 | -0.01 | -0.01 | -0.01 | 0.01 | 0.02 | 0.02 | -0.04 | -0.05 | -0.05 | -0.01 | 0.00 | 0.01 | -0.04 | -0.09 | -0.13 |
| | **1970-1990** | -0.08 | -0.13 | -0.17 | -0.30 | -0.31 | -0.42 | 0.05 | -0.15 | -0.28 | -0.08 | -0.16 | -0.26 | -0.36 | -0.40 | -0.53 | 0.07 | -0.31 | -0.55 | -0.08 | -0.07 | -0.02 | 0.01 | -0.02 | -0.06 | 0.02 | 0.01 | -0.01 |
| | **1985-2005** | -0.04 | -0.05 | -0.08 | 0.05 | 0.04 | 0.05 | -0.02 | -0.06 | -0.09 | -0.07 | -0.10 | -0.17 | 0.07 | 0.05 | 0.05 | 0.00 | 0.01 | 0.01 | -0.01 | 0.00 | 0.01 | -0.10 | -0.03 | 0.02 | -0.05 | -0.13 | -0.19 |
| | **1995-2012** | -0.02 | -0.01 | 0.01 | -0.02 | 0.06 | 0.15 | -0.01 | -0.02 | -0.03 | 0.04 | 0.04 | 0.03 | -0.04 | 0.07 | 0.21 | -0.01 | -0.05 | -0.09 | -0.06 | -0.05 | -0.01 | 0.04 | 0.04 | 0.03 | 0.00 | 0.01 | 0.02 |
| **Decline in (% of locations)** | **1970-2012** | 64% | 63% | 58% | 49% | 52% | 50% | 60% | 61% | 63% | 61% | 58% | 55% | 48% | 53% | 53% | 31% | 36% | 50% | 69% | 72% | 64% | 53% | 47% | 40% | 74% | 71% | 69% |
| | **1970-1990** | 73% | 64% | 64% | 77% | 69% | 77% | 17% | 50% | 75% | 79% | 71% | 71% | 91% | 70% | 80% | 25% | 100% | 100% | 63% | 50% | 50% | 0% | 67% | 67% | 0% | 0% | 50% |
| | **1985-2005** | 57% | 74% | 70% | 30% | 58% | 67% | 70% | 60% | 50% | 67% | 83% | 75% | 22% | 50% | 70% | 60% | 40% | 20% | 45% | 64% | 64% | 100% | 100% | 50% | 80% | 80% | 80% |
| | **1995-2012** | 57% | 48% | 52% | 57% | 33% | 33% | 67% | 50% | 33% | 30% | 50% | 60% | 80% | 40% | 40% | 67% | 67% | 67% | 82% | 45% | 45% | 0% | 20% | 20% | 67% | 33% | 0% |
| **Avg Decline/Decade at locations with -ve trend (µg/g ww)** | **1970-2012** | -0.08 | -0.10 | -0.16 | -0.05 | -0.07 | -0.11 | -0.07 | -0.14 | -0.20 | -0.07 | -0.11 | -0.17 | -0.05 | -0.08 | -0.12 | -0.04 | -0.07 | -0.09 | -0.09 | -0.10 | -0.16 | -0.05 | -0.06 | -0.06 | -0.07 | -0.15 | -0.23 |
| | **1970-1990** | -0.16 | -0.29 | -0.44 | -0.40 | -0.50 | -0.56 | -0.10 | -0.31 | -0.37 | -0.16 | -0.31 | -0.50 | -0.40 | -0.63 | -0.68 | -0.10 | -0.31 | -0.55 | -0.17 | -0.25 | -0.31 | NA | -0.04 | -0.10 | NA | NA | -0.02 |
| | **1985-2005** | -0.11 | -0.11 | -0.20 | -0.10 | -0.08 | -0.08 | -0.05 | -0.12 | -0.22 | -0.14 | -0.14 | -0.26 | -0.10 | -0.10 | -0.09 | -0.01 | -0.03 | -0.10 | -0.06 | -0.07 | -0.12 | -0.10 | -0.03 | -0.01 | -0.08 | -0.17 | -0.24 |
| | **1995-2012** | -0.09 | -0.14 | -0.19 | -0.06 | -0.08 | -0.11 | -0.02 | -0.06 | -0.15 | -0.05 | -0.06 | -0.09 | -0.06 | -0.09 | -0.11 | -0.03 | -0.08 | -0.15 | -0.10 | -0.22 | -0.31 | NA | -0.04 | -0.13 | -0.01 | -0.01 | NA |
| **Avg Increase/Decade at locations with +ve trend (µg/g ww)** | **1970-2012** | 0.05 | 0.06 | 0.10 | 0.04 | 0.06 | 0.09 | 0.03 | 0.07 | 0.11 | 0.04 | 0.05 | 0.09 | 0.04 | 0.07 | 0.10 | 0.03 | 0.07 | 0.13 | 0.07 | 0.09 | 0.13 | 0.04 | 0.05 | 0.06 | 0.04 | 0.06 | 0.10 |
| | **1970-1990** | 0.14 | 0.16 | 0.30 | 0.01 | 0.11 | 0.07 | 0.08 | 0.01 | 0.01 | 0.20 | 0.20 | 0.33 | 0.00 | 0.14 | 0.09 | 0.13 | NA | NA | 0.08 | 0.12 | 0.26 | 0.01 | 0.03 | 0.03 | 0.02 | 0.01 | 0.01 |
| | **1985-2005** | 0.05 | 0.12 | 0.18 | 0.12 | 0.20 | 0.30 | 0.04 | 0.03 | 0.03 | 0.07 | 0.10 | 0.09 | 0.12 | 0.20 | 0.38 | 0.02 | 0.03 | 0.04 | 0.03 | 0.13 | 0.25 | NA | NA | 0.05 | 0.07 | 0.04 | 0.01 |
| | **1995-2012** | 0.08 | 0.11 | 0.22 | 0.03 | 0.13 | 0.29 | 0.01 | 0.02 | 0.03 | 0.07 | 0.13 | 0.20 | 0.02 | 0.17 | 0.43 | 0.01 | 0.02 | 0.04 | 0.09 | 0.09 | 0.23 | 0.04 | 0.06 | 0.07 | 0.01 | 0.02 | 0.02 |

**Table S2:** Summary of Mixed Effect Modelling (MEM) results for small, medium and large sizes of Walleye (WE), Northern Pike (NP) and Lake Trout (LT) for the recent (1995-2012) and historical (1970-1990) time periods. The results are presented as annual percentage change (APC) (based on the use of logarithmically transformed mercury values in the models) and change in amount of mercury in unit amount of fish per decade (based on the use of regular mercury values in the models). The values are presented as averages along with ±2 standard error values (i.e., 95% confidence limit).

| Region | Species | Size | Time Period | Obs | Locations | Annual % Change (-2SE, Avg, +2SE) | Change/Decade (µg/g ww) (-2SE, Avg, +2SE) |
|---|---|---|---|---|---|---|---|
| Ontario | WE | Small | Historical | 6435 | 641 | -1.63, -1.23, -0.83 | -0.11, -0.08, -0.05 |
| | | | Recent | 5659 | 682 | 1.19, 1.55, 1.91 | 0.03, 0.05, 0.07 |
| | | Mediu | Historical | 4387 | 623 | -0.73, -0.29, 0.15 | -0.11, -0.07, -0.03 |
| | | | Recent | 4160 | 664 | 0.86, 1.31, 1.76 | 0.04, 0.08, 0.11 |
| | | Large | Historical | 1192 | 362 | -0.48, 0.26, 1.01 | -0.18, -0.10, -0.02 |
| | | | Recent | 1973 | 532 | 0.83, 1.38, 1.92 | 0.05, 0.11, 0.16 |
| | NP | Small | Historical | 3072 | 599 | -3.69, -3.00, -2.31 | -0.18, -0.15, -0.11 |
| | | | Recent | 1864 | 493 | -0.06, 0.55, 1.15 | 0.00, 0.02, 0.05 |
| | | Mediu | Historical | 3668 | 645 | -2.82, -2.21, -1.59 | -0.23, -0.17, -0.12 |
| | | | Recent | 2787 | 605 | 1.15, 1.71, 2.28 | 0.05, 0.09, 0.12 |
| | | Large | Historical | 1800 | 485 | -2.24, -1.52, -0.81 | -0.25, -0.18, -0.10 |
| | | | Recent | 1603 | 505 | 1.22, 1.93, 2.64 | 0.07, 0.13, 0.18 |
| | LT | Small | Historical | 1422 | 238 | -0.55, 0.56, 1.68 | -0.04, 0.00, 0.04 |
| | | | Recent | 1679 | 312 | -0.11, 1.07, 2.25 | 0.01, 0.06, 0.11 |
| | | Mediu | Historical | 1135 | 224 | -0.20, 0.69, 1.58 | 0.00, 0.09, 0.18 |
| | | | Recent | 1577 | 317 | -0.45, 0.50, 1.46 | 0.01, 0.08, 0.14 |
| | | Large | Historical | 674 | 178 | -0.82, 0.47, 1.75 | -0.07, 0.11, 0.28 |
| | | | Recent | 737 | 243 | -1.32, 0.03, 1.38 | -0.04, 0.07, 0.18 |
| North | WE | Small | Historical | 5579 | 537 | -2.12, -1.67, -1.23 | -0.12, -0.09, -0.06 |
| | | | Recent | 4493 | 546 | 1.76, 2.19, 2.61 | 0.05, 0.07, 0.10 |
| | | Mediu | Historical | 3566 | 516 | -1.49, -0.98, -0.47 | -0.14, -0.10, -0.05 |
| | | | Recent | 3142 | 524 | 1.18, 1.72, 2.27 | 0.06, 0.10, 0.14 |
| | | Large | Historical | 836 | 273 | -1.86, -0.98, -0.10 | -0.28, -0.17, -0.06 |
| | | | Recent | 1590 | 425 | 0.81, 1.42, 2.04 | 0.05, 0.11, 0.17 |

|  |  |  |  |  |  |  |  |
|---|---|---|---|---|---|---|---|
|  | NP | Small | Historical | 2644 | 520 | -3.76, -2.98, -2.21 | -0.19, -0.16, -0.12 |
|  |  |  | Recent | 1387 | 384 | -0.39, 0.39, 1.18 | 0.00, 0.03, 0.05 |
|  |  | Mediu | Historical | 3322 | 569 | -2.79, -2.13, -1.47 | -0.25, -0.18, -0.12 |
|  |  |  | Recent | 2216 | 486 | 1.27, 1.96, 2.65 | 0.05, 0.09, 0.13 |
|  |  | Large | Historical | 1605 | 420 | -2.30, -1.54, -0.79 | -0.28, -0.19, -0.11 |
|  |  |  | Recent | 1292 | 409 | 1.54, 2.36, 3.19 | 0.08, 0.14, 0.20 |
|  | LT | Small | Historical | 757 | 122 | -1.74, -0.10, 1.54 | -0.12, -0.06, -0.01 |
|  |  |  | Recent | 1127 | 225 | -0.35, 1.13, 2.62 | -0.01, 0.06, 0.13 |
|  |  | Mediu | Historical | 568 | 124 | -1.28, 0.17, 1.63 | -0.11, -0.01, 0.08 |
|  |  |  | Recent | 1128 | 237 | 0.20, 1.41, 2.62 | 0.04, 0.13, 0.22 |
|  |  | Large | Historical | 274 | 84 | -7.19, -3.83, -0.47 | -0.56, -0.32, -0.07 |
|  |  |  | Recent | 528 | 175 | -0.44, 1.29, 3.02 | 0.00, 0.14, 0.28 |
| South | WE | Small | Historical | 856 | 104 | -0.46, 0.45, 1.37 | -0.10, -0.04, 0.01 |
|  |  |  | Recent | 1166 | 136 | -0.84, -0.14, 0.56 | -0.07, -0.03, 0.00 |
|  |  | Mediu | Historical | 821 | 107 | 0.43, 1.33, 2.23 | -0.07, 0.00, 0.06 |
|  |  |  | Recent | 1018 | 140 | -0.41, 0.40, 1.21 | -0.03, 0.02, 0.08 |
|  |  | Large | Historical | 356 | 89 | 0.65, 2.02, 3.39 | -0.10, 0.01, 0.12 |
|  |  |  | Recent | 383 | 107 | -0.04, 1.16, 2.36 | -0.02, 0.10, 0.21 |
|  | NP | Small | Historical | 428 | 79 | -4.65, -3.09, -1.53 | -0.17, -0.11, -0.05 |
|  |  |  | Recent | 477 | 109 | -0.20, 0.72, 1.63 | -0.01, 0.02, 0.05 |
|  |  | Mediu | Historical | 346 | 76 | -3.94, -2.25, -0.56 | -0.17, -0.10, -0.02 |
|  |  |  | Recent | 571 | 119 | -0.11, 0.87, 1.85 | 0.00, 0.05, 0.10 |
|  |  | Large | Historical | 195 | 65 | -3.91, -1.64, 0.63 | -0.23, -0.10, 0.03 |
|  |  |  | Recent | 311 | 96 | -1.31, 0.11, 1.52 | -0.06, 0.04, 0.15 |
|  | LT | Small | Historical | 665 | 116 | -0.21, 1.31, 2.82 | 0.01, 0.07, 0.13 |
|  |  |  | Recent | 552 | 87 | -0.47, 1.51, 3.49 | -0.02, 0.06, 0.15 |
|  |  | Mediu | Historical | 567 | 100 | -0.05, 1.08, 2.20 | 0.01, 0.15, 0.28 |
|  |  |  | Recent | 552 | 87 | -0.47, 1.51, 3.49 | -0.07, 0.01, 0.09 |
|  |  | Large | Historical | 400 | 94 | -0.06, 1.21, 2.48 | -0.02, 0.19, 0.41 |
|  |  |  | Recent | 209 | 68 | -3.30, -1.23, 0.84 | -0.12, 0.04, 0.21 |

**Figure S1a.** Illustration of screening of sampling events for calculating 50cm std-length mercury concentration. To avoid large extrapolation of the power series regressions while calculating each std-length fish concentration, only sampling events with the smallest fish smaller than a std-length plus 15cm and the largest fish larger than std-length minus 15cm were considered. For example, as illustrated in this figure, to calculate 50cm WE mercury for a particular year/location, the smallest WE measured for that sampling event should be smaller than 65cm and the largest fish should be larger than 35cm.

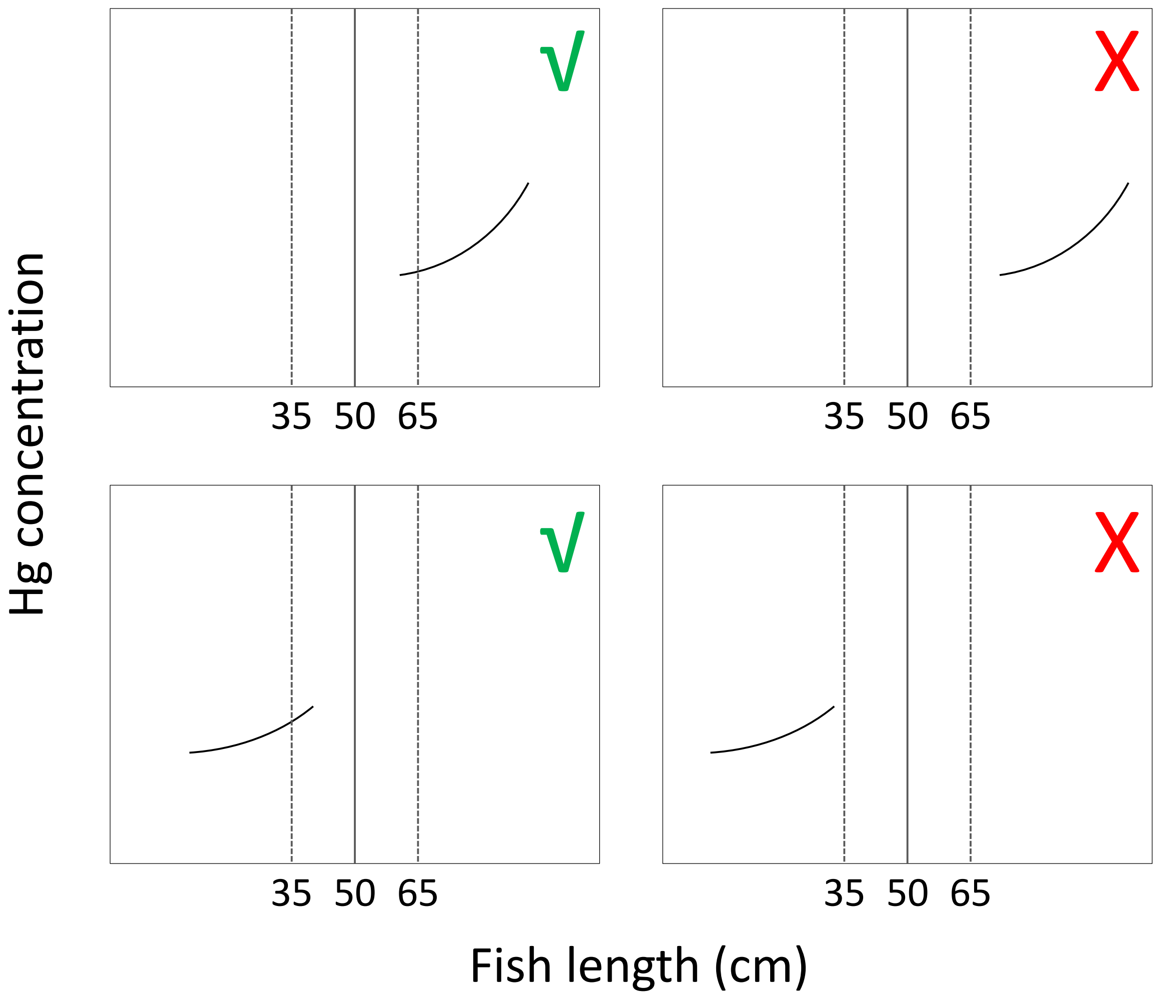

**Figure S1b.** Illustration of screening of sampling events to only include species/location/year combinations with (a) a minimum of five measurements and (b) a 10cm size range (i.e., difference between maximum and minimum fish lengths).

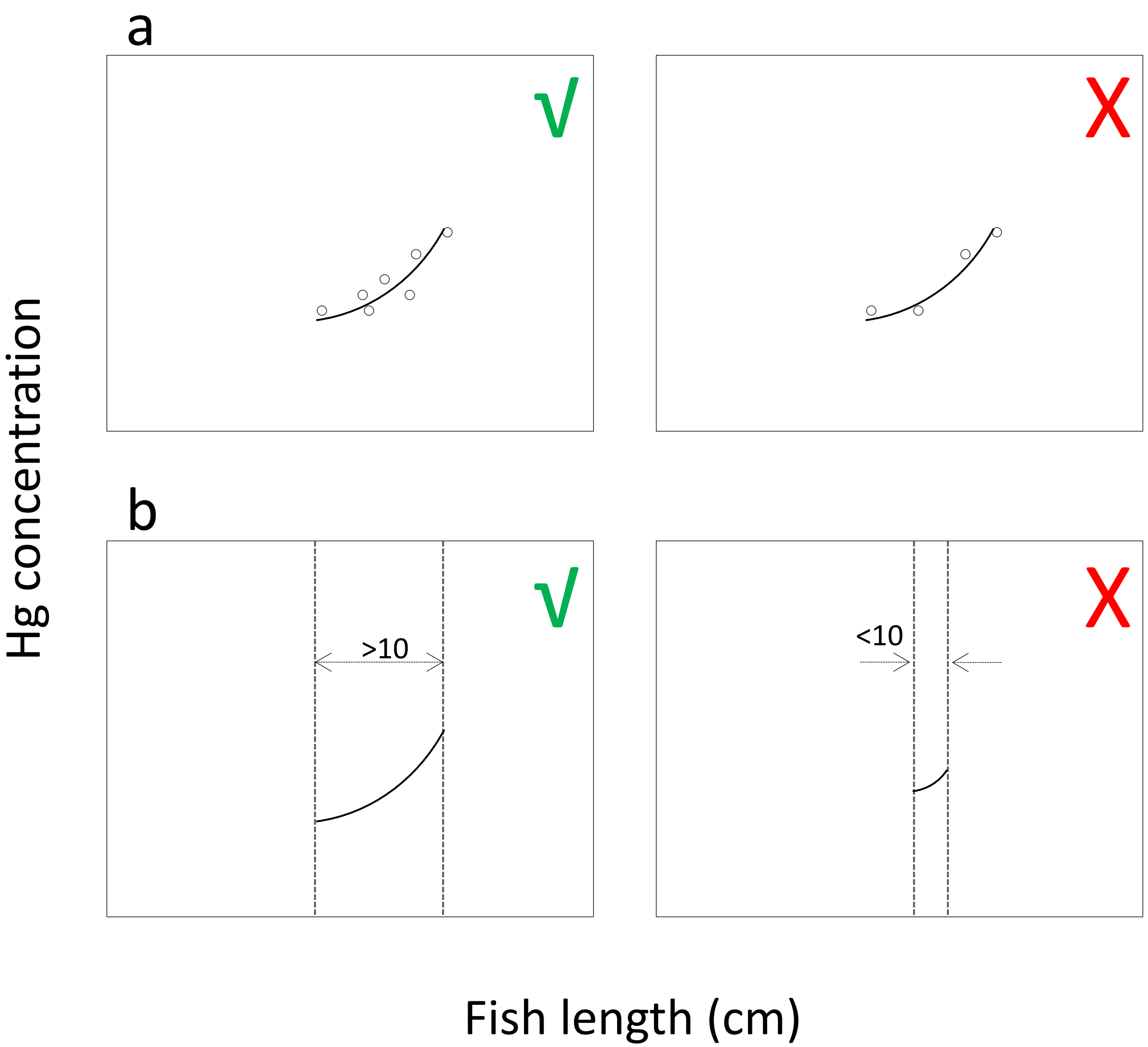

**Figure S2a:** Mercury concentrations (µg/g wet weight) for two time periods (historical 1970-1985, recent 2000-2012) in skin-off fillets of **small sized** overall Ontario, Northern Ontario and Southern Ontario walleye, northern pike and lake trout. *n* represents number of locations. For each species, identical letters for each region belong to the same statistical group, where group A is significantly different ($p<0.05$) from group B. Measurements plotted outside the chart area represent values outside the range of Y-Axis.

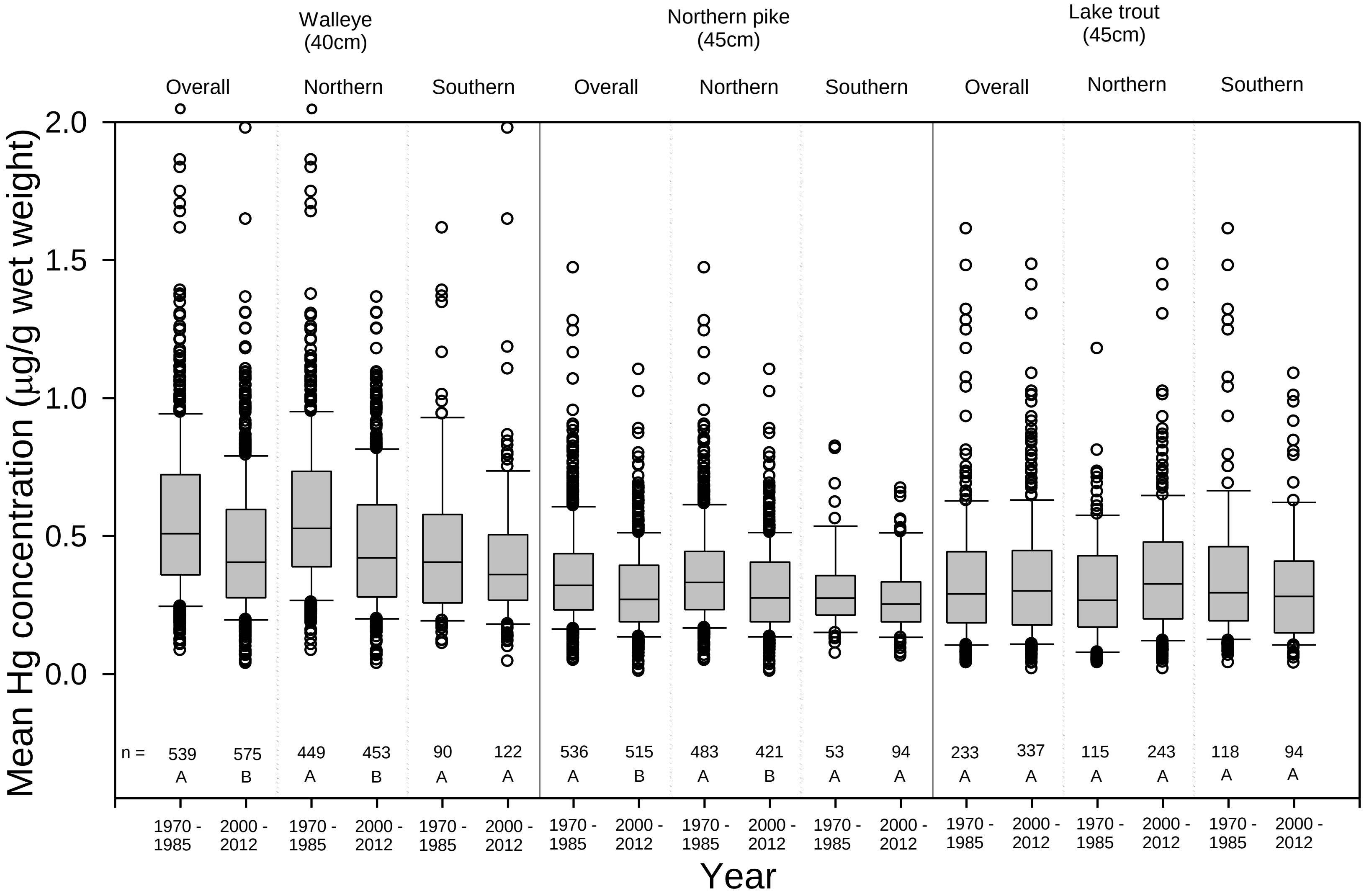

**Figure S2b:** Mercury concentrations (µg/g wet weight) for two time periods (historical 1970-1985, recent 2000-2012) in skin-off fillets of **medium sized** overall Ontario, Northern Ontario and Southern Ontario walleye, northern pike and lake trout. *n* represents number of locations. For each species, identical letters for each region belong to the same statistical group, where group A is significantly different (*p*<0.05) from group B.

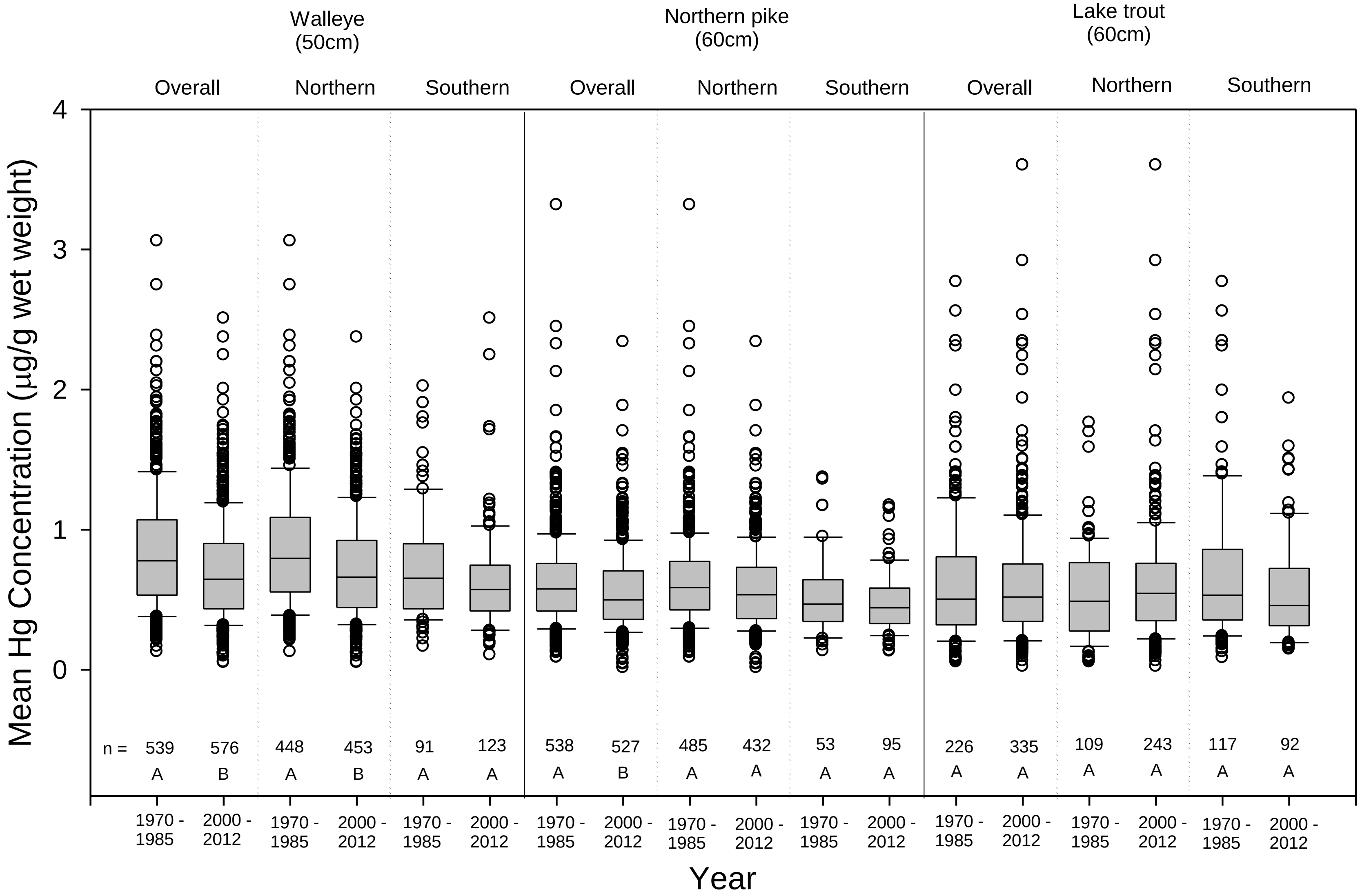

**Figure S2c:** Mercury concentrations (µg/g wet weight) for two time periods (historical 1970-1985, recent 2000-2012) in skin-off fillets of **large sized** overall Ontario, Northern Ontario and Southern Ontario walleye, northern pike and lake trout. *n* represents the number of locations. For each species, identical letters for each region belong to the same statistical group, where group A is significantly different ($p$<0.05) from group B. Measurements plotted outside the chart area represent values outside the range of Y-Axis.

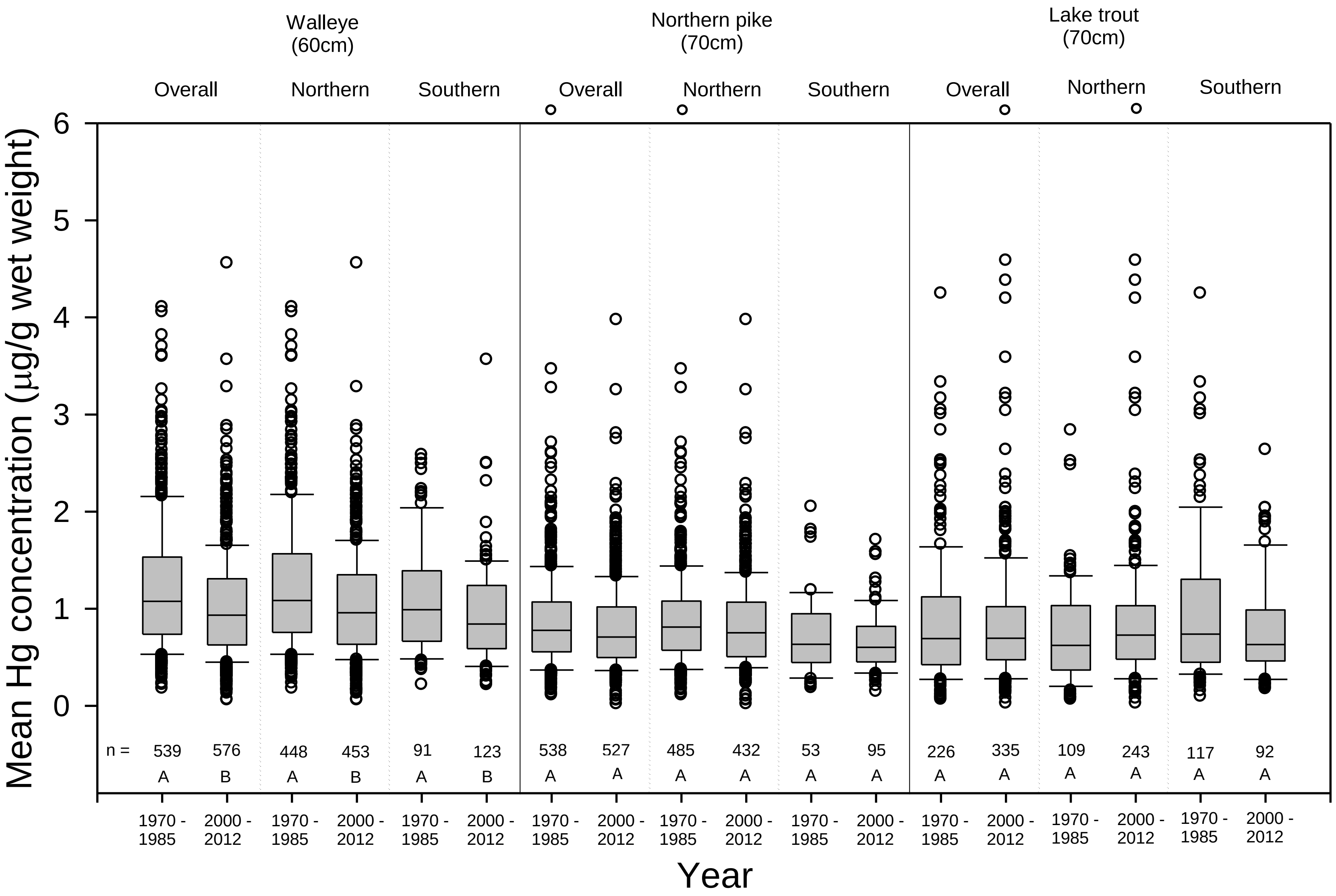

**Figure S3a:** Box plots of rates of mercury change (µg/g wet weight per decade) for different time periods in skin-off fillets of small, medium and large sized **Northern Ontario** walleye, northern pike and lake trout. The dotted line represents no change. Average values of mercury change (avg.Δ) are also presented. *n* represents the number of locations. For each species/std-length and the 1970-1990 and 1995-2012 time periods, ES (effect size) indicates the difference (with 95% confidence intervals), and identical letters belong to the same statistical group, where, for example, group A is significantly different ($p$<0.05) from group B. Measurements plotted outside the chart area represent values outside the range of Y-Axis.

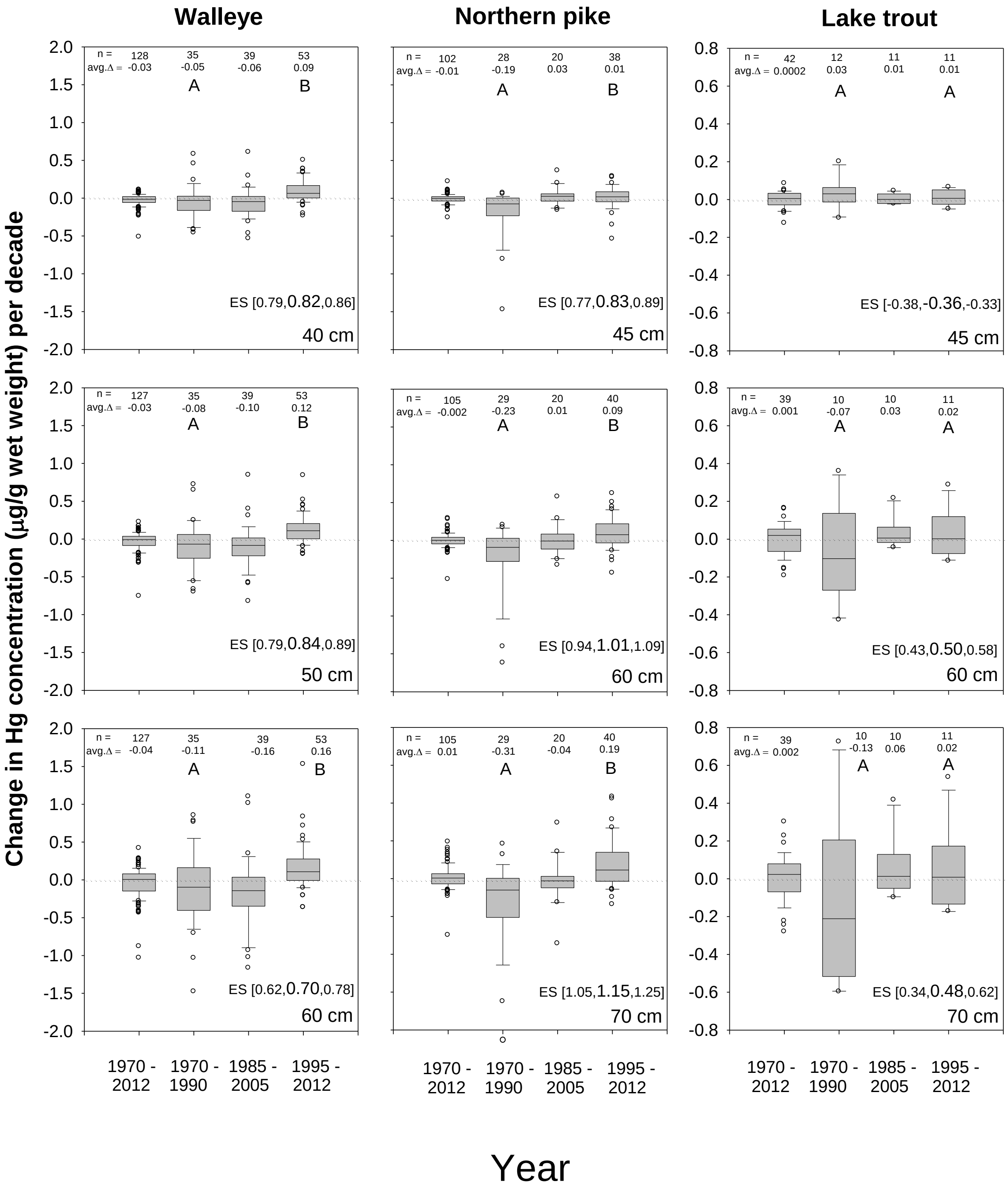

**Figure S3b:** Box plots of rates of mercury change (µg/g wet weight per decade) for different time periods in skin-off fillets of small, medium and large sized **Southern Ontario** walleye, northern pike and lake trout. The dotted line represents no change. Average values of mercury change (avg.Δ) are also presented. *n* represents the number of locations. For each species/std-length and the 1970-1990 and 1995-2012 time periods, ES (effect size) indicates the difference (with 95% confidence intervals), and identical letters belong to the same statistical group, where, for example, group A is significantly different ($p$<0.05) from group B. Measurements plotted outside the chart area represent values outside the range of Y-Axis.

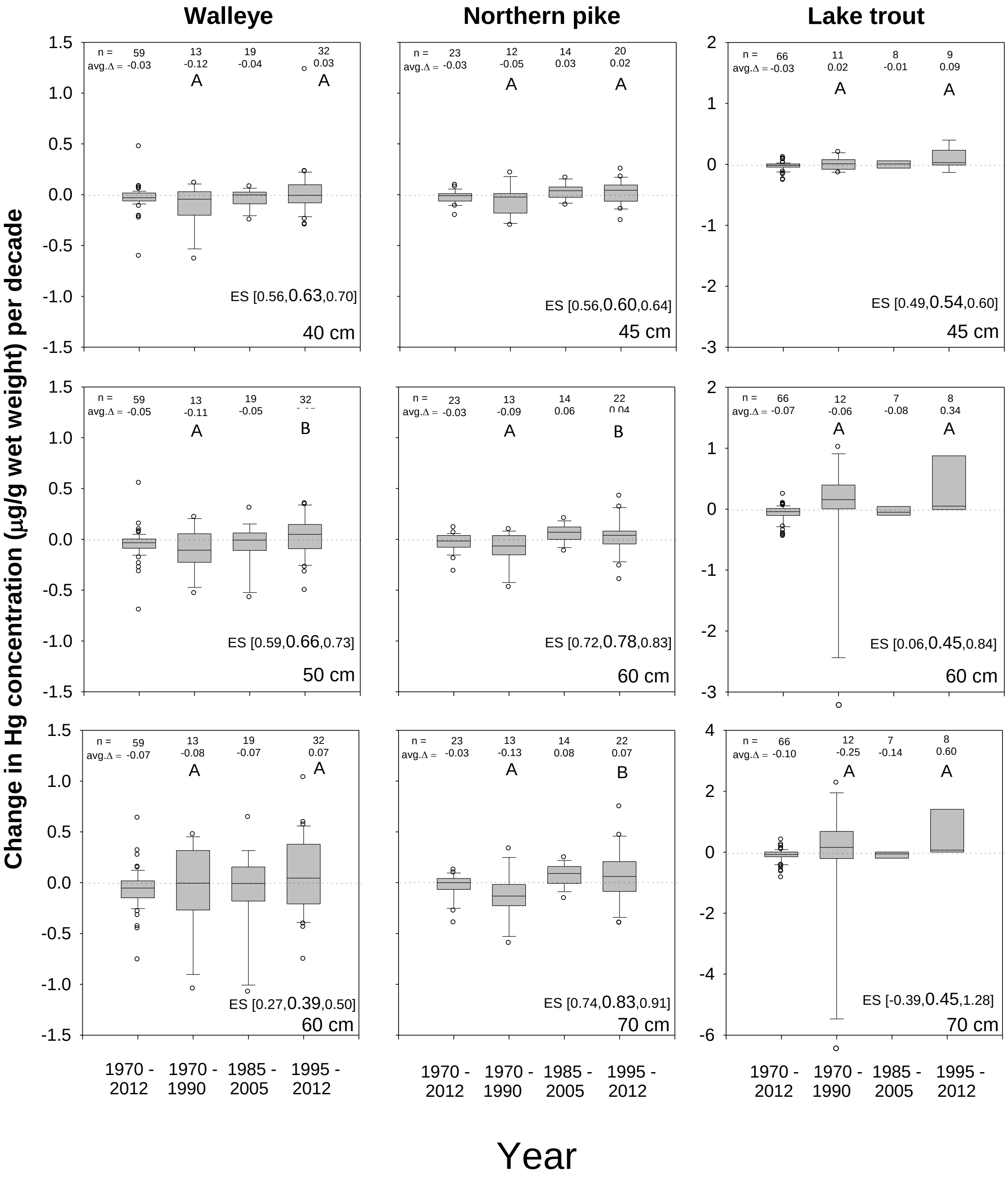

**Figure S4a:** Spatial distribution of rates of mercury change (µg/g wet weight per decade) for different time periods in skin-off fillets of small sized Ontario walleye, northern pike and lake trout. The concentrations have been grouped into various categories. Percentage of total locations within each category is also presented. *n* represents the number of locations for each time period and species.

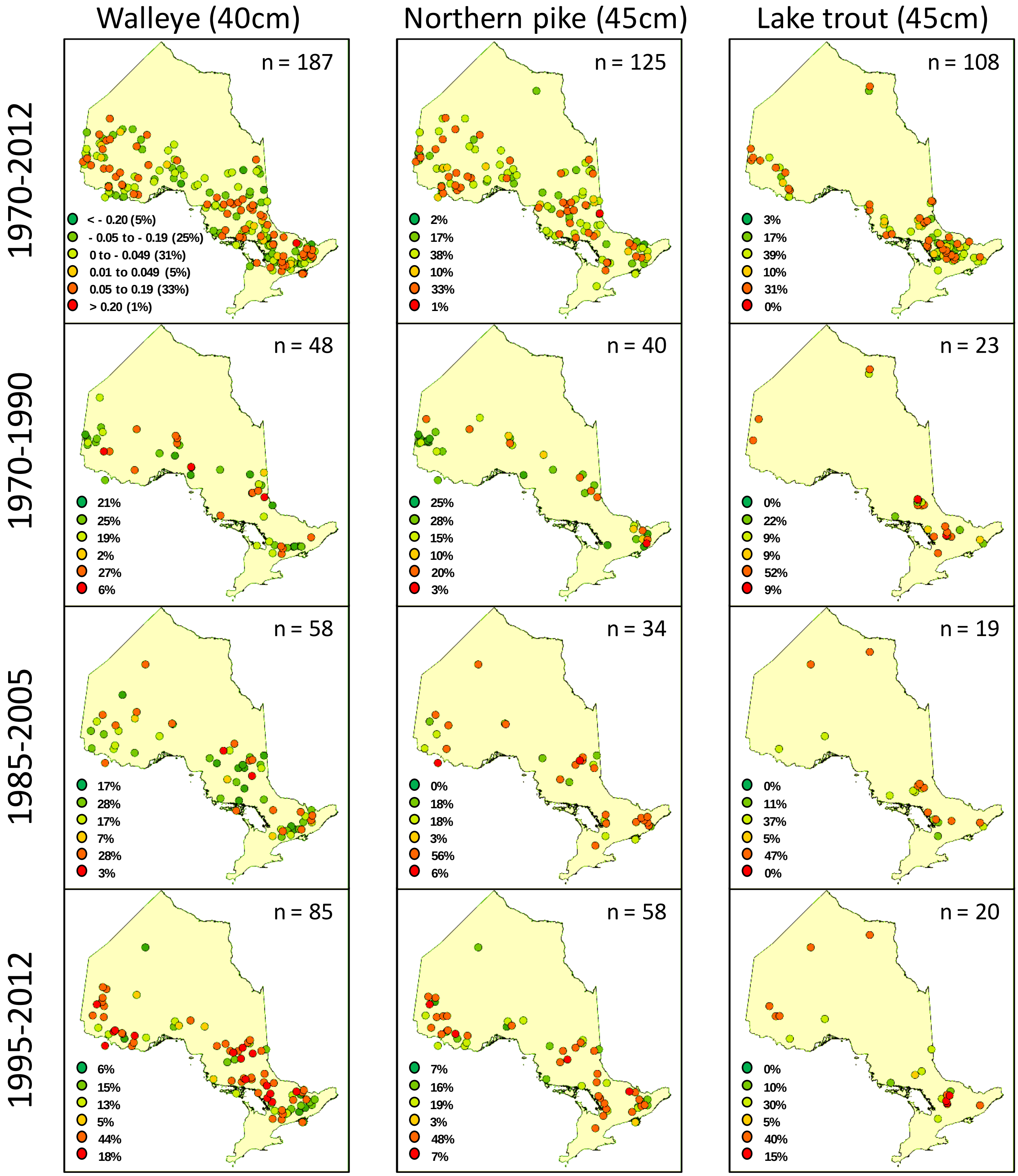

**Figure S4b:** Spatial distribution of rates of mercury change (µg/g wet weight per decade) for different time periods in skin-off fillets of large sized Ontario walleye, northern pike and lake trout. The concentrations have been grouped into various categories. Percentage of total locations within each category is also presented. *n* represents the number of locations for each time period and species.

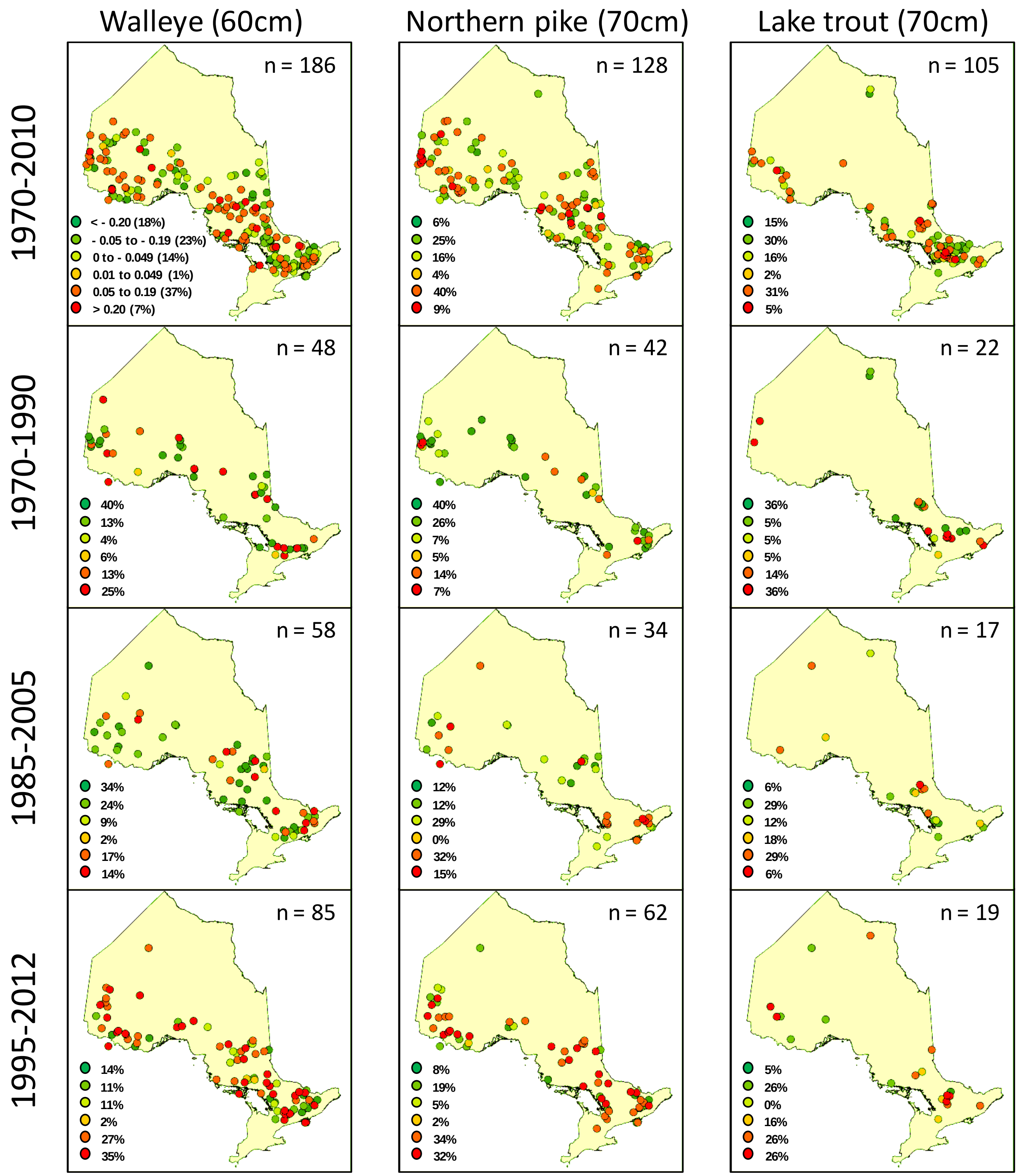

**Figure S5:** Box plots of rates of mercury change (µg/g wet weight per decade) for different time periods in skin-off fillets of small, medium and large sized Ontario walleye, northern pike and lake trout. The locations considered required to have minimum 3 time point measurements and were constant for all time periods. The dotted line represents no change. *n* represents the number of locations. For each species/std-length and the 1970-1990 and 1995-2012 time periods, identical letters belong to the same statistical group, where, for example, group A is significantly different ($p<0.05$) from group B.

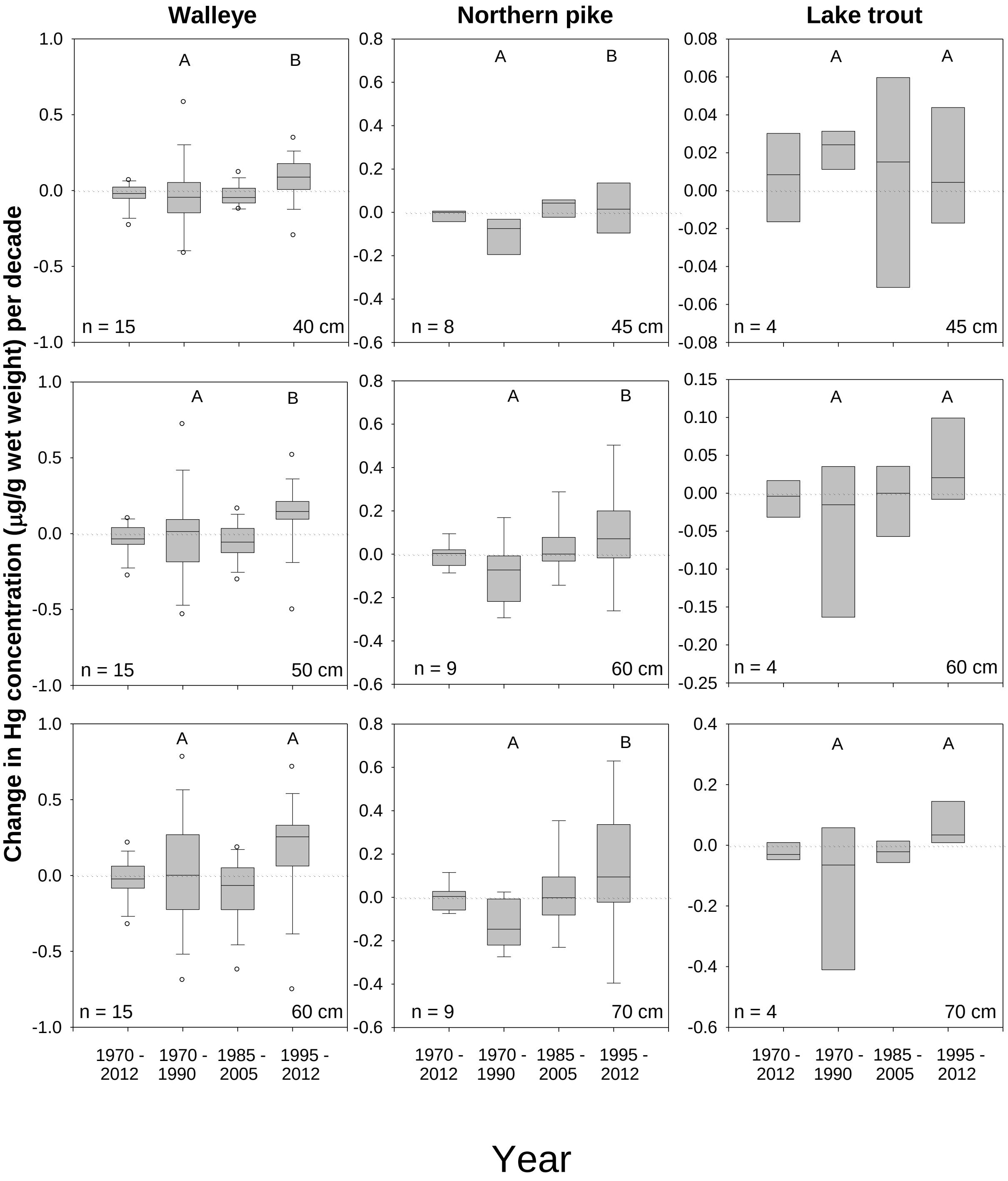

**Figure S6:** Geological Map of Canada showing U-shaped (almost semi-circular) Canadian Shield Region in Red/Orange (from Wikipedia).