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Assessment of fish mercury levels in the upper St. Lawrence River, Canada

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ABSTRACT

The stretch of the St. Lawrence River near Cornwall, Ontario, Canada, between the Moses–Saunders power dam to the eastern outlet of Lake St. Francis in Québec, is currently listed as an Area of Concern (AOC), and has restrictive fish consumption advisories in place, largely due to high mercury levels. This study examined long-term temporal trends of mercury concentrations in northern pike, smallmouth bass, walleye and yellow perch from the St. Lawrence River, including the Cornwall AOC. In addition, differences in fish mercury concentrations among river sections were compared for each species using historical (1975–1979) and recent (2000–2011) data. Statistically significant declines in mercury concentrations were observed in all river sections for yellow perch (~40%), as well as in northern pike and walleye in at least one river section. For the river section encompassing the AOC, recent mercury concentrations are 33–59% lower than historical mercury concentrations for all four species. Further, a comparison of recent mercury concentrations between the AOC river section and other Ontario lakes/rivers suggests that AOC mercury values are within the 75th percentile of values for other Ontario water bodies in three of the four species considered. However, current fish mercury values for the AOC river section remain higher than one or more upstream river sections. These results indicate that recovery is still ongoing in this AOC.

Introduction

Mercury is a well-known global pollutant, and is present in trace amounts in all freshwater fish in the United States (Stahl et al., 2009) and likely worldwide. Elevated fish mercury levels can be attributed to anthropogenic activities that release mercury emissions both directly and indirectly into the environment (Engstrom et al., 2007). Despite efforts to eliminate point-source mercury inputs to water bodies, the inherent persistence of mercury can result in a continued elevated exposure to fish that inhabit mercury-polluted areas.

Mercury bioaccumulates within the tissues of fish and continues to be a major concern for the health of human fish consumers. High consumption of mercury-contaminated fish is known to have negative effects on human health (Health Canada, 2007). The regular consumption of fish with elevated mercury could result in a significant accumulation of mercury within the body at which health concerns would become prevalent. As the developing brain is particularly at risk of negative effects from mercury, children under age 15 and women of child-bearing age who can transfer mercury to a developing fetus are considered to be a sensitive population, and are therefore advised to greatly minimize their exposure to mercury. In order to protect human health from mercury exposures through fish consumption, various regulatory agencies release fish consumption advisories (e.g., OMOE, 2013; US EPA, 2012).

Mercury contamination is a well-documented issue in the Great Lakes and St. Lawrence region (Bhavsar et al., 2010, 2011; Goulet et al., 2008; Ridal et al., 2010). Mercury contamination of the St. Lawrence River began from the development of chemical, pulp and paper, and aluminum production facilities in the late 1950s (Ridal et al., 2010). Due to a variety of factors, primarily elevated fish mercury levels, an 80 km stretch of the St. Lawrence River near Cornwall was designated as an Area of Concern (AOC) in 1985 by the International Joint Commission (St. Lawrence Remdial Action Plan Team, 1991). A Remedial Action Plan (RAP) was implemented for this AOC in 1987 to restore the quality and health of this aquatic ecosystem (Environment Canada, 2010). By the mid-1990s, three of the major contributors of contaminated discharges were closed (International Joint Commission, 2003), and overall the RAP has successfully achieved elimination of all significant sources of mercury within the Cornwall area (Environment Canada, 2010). However, despite this achievement, restrictive advisories for sport fish consumption remain in effect (OMOE, 2013).

The Ontario Ministry of Environment (OMOE), in partnership with the Ontario Ministry of Natural Resources (OMNR), has been monitoring sport fish contamination levels in this region since 1975, and therefore, long-term mercury monitoring data sets for a variety of fish species exist. A previous study by Goulet et al. (2008)

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examined fish mercury trends within the St. Lawrence River over a 20-year (1975–1995) period; however, recent temporal trend data are lacking in the published literature.

In this study, temporal trends in fish mercury concentrations in the upper St. Lawrence River, including the AOC region, are assessed over ~35 years (1975–2011). In addition, spatial patterns in fish mercury concentrations among different river sections are assessed in both a historical (1975–1979) and recent (2000–2011) context. Assessment of temporal and spatial dynamics in fish mercury concentrations will allow for further insight into both the current status of mercury contamination in St. Lawrence fishes, as well as the ongoing recovery of the St. Lawrence AOC. Finally, a comparison of fish mercury concentrations within limited size (i.e., length) ranges between the AOC river section and other Ontario lakes/rivers is presented for the first time to understand relative significance of the mercury problem in the AOC.

Materials and methods

OMOE has conducted extensive long-term sampling efforts in the upper St. Lawrence River, which is defined in this study as the stretch of the river from the outlet of Lake Ontario to the Beauharnois Hydroelectric Power Dam in Québec. OMOE has spatially divided the upper St. Lawrence River into smaller sections for effective monitoring of fish contaminants and issue fish consumption advisories (Fig. 1, Bhavsar et al., 2011). Sampling of a variety of sport fish species from all St. Lawrence River sections has been conducted from ~1975–present, with varying frequency depending on need and resources.

The Cornwall AOC is located within block 15 (Fig. 1, inset). Blocks 12, 13 and 14 are upstream of block 15, and are physically separated from block 15 by the Moses Saunders Power Dam. Block 16 corresponds to the Raisin River, a tributary of the St. Lawrence River with an outlet downstream of block 15 (Fig. 1). For the purposes of this study, sampling blocks 12, 13, 14, and 16 were considered reference sites, because these areas of the St. Lawrence River were not exposed to any known major point-source mercury contamination. This study assumes that individuals collected in each block are resident to that section and not transient, although it should be noted that some species are considered to be migratory and thus their mercury burden may be not be entirely reflective of the river section in which they were captured. However, Goulet et al. (2008), based on calculations by Minns (1995), concluded that yellow perch collected from a specific river section are likely representative of that river section. Further, the Moses–Saunders dam, located between block 15 and the upstream reference blocks (Fig. 1), is a physical barrier minimizing fish movement between these two regions.

Species with data covering a broad range of years were selected to assess temporal trends in mercury concentrations. Based on data availability and temporal extent of sampling, long-term trends for northern pike (*Esox lucius*) and yellow perch (*Perca flavescens*) were assessed for blocks 12, 13, 14 and 15 (Fig. 1). In addition, temporal trends for smallmouth bass (*Micropterus dolomieu*) were assessed for blocks 13 and 14, and walleye (*Stizostedion vitreum*) was assessed for blocks 14, 15 and 16 (Fig. 1). All of these species are commonly found in the St. Lawrence AOC, and currently have consumption restrictions (OMOE, 2013). Temporal patterns of fish mercury concentrations within the Great Lakes region have varied by species (Bhavsar et al., 2011), and therefore more than one species were considered to ensure a proper assessment.

Laboratory analysis

Skinless, boneless fish filets were stored at −20 °C prior to chemical analysis at the OMOE Laboratory in Toronto, Ontario. Fish tissue

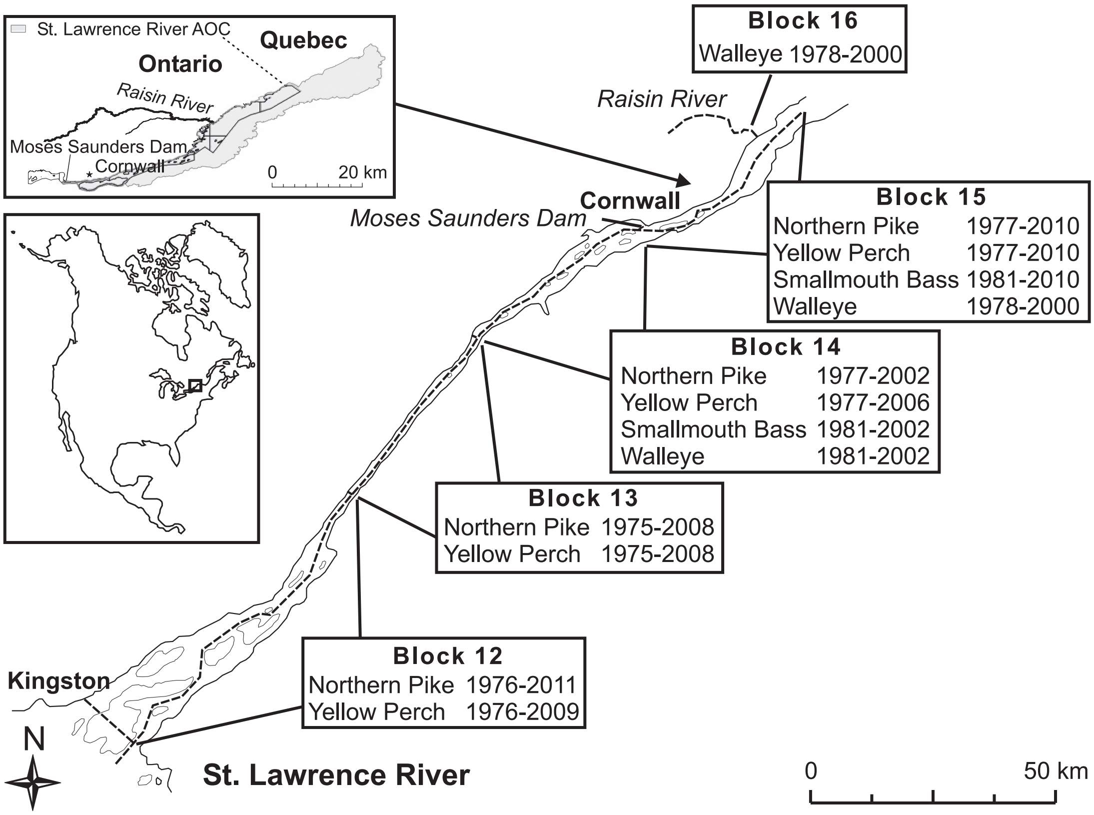

Fig. 1. Map of the St. Lawrence River, from Lake Ontario at Kingston, Ontario to the Raisin River. Dashed lines indicate OMOE sampling block boundaries. Sampling block numbers are indicated in boxes, with species and temporal extent of sampling represented in each block. The inset for block 15 indicates the spatial extent of the St. Lawrence River area of concern (AOC), with the Raisin River and the Moses–Saunders Dam west of Cornwall labeled for context.

mercury levels were determined as wet weight µg/g, using protocols as in OMOE method HGBIO-E3057 (OMOE 2006), and described by Bhavsar et al. (2010).

Statistical analysis

The relationship between mercury concentration and fish length is well-established, where logarithmically transformed mercury concentration generally increases linearly with fish length (Gewurtz et al., 2011; Sonesten, 2003). In this study, within-year relationships between logarithmically-transformed mercury concentration and fish length were not consistently parallel and statistically significant across years, thereby rendering ANCOVA with length as a covariate an inappropriate approach (Somers and Jackson, 1993). Instead, a restricted size range for each species was used to account for the effect of fish length on mercury concentration. Appropriate size ranges were estimated using the grand mean fish length across all years for each species/block, and then applying a 10 cm range around that value. The selected size ranges are similar to those used by similar studies (e.g., Bhavsar et al., 2010; Neff et al., 2012): northern pike, 55–65 cm; smallmouth bass, 32–42 cm; walleye, 45–55 cm; and yellow perch, 15–25 cm. No significant relationship existed between the restricted fish length and the mercury concentration within each year ($p > 0.05$), and the mean length of the restricted data set did not significantly differ across years ($p > 0.05$).

After the size restriction was applied to all data for each species, years with less than two samples were eliminated. Yearly mean mercury concentrations were then calculated for each species in each block. Sample sizes for each year of data varied, as sampling frequency was not consistent over the examined time period (Fig. S1). Long-term temporal trends in mercury concentrations were assessed using Mann–Kendall tests on annual means of logarithmically-transformed mercury values. The non-parametric Mann–Kendall analysis tests for the presence of a monotonic increasing or decreasing trend. The slope of the linear trend can then be estimated using the non-parametric Sen's slope estimate (Salmi et al., 2002). The percent decline over time for each species/block was calculated using the following formula:

$$\frac{(Hg_i - Hg_f)}{Hg_i} \times 100$$

where Hg_i is the initial fish mercury concentration in the first year of sampling for that species and block, and Hg_f is the final measured fish mercury concentration in the last year of sampling for that species and block.

Differences in historical (1975–1979) and recent (2000–2011) fish mercury concentrations within block 15 were assessed for each species using the Mann–Whitney test. In addition, historical and recent spatial patterns in fish mercury concentrations among river blocks were examined using ANOVA with pairwise post-hoc comparison tests. Time periods were selected to represent the extreme temporal ends of the available data. The historical time period (1975–1979) reflects the period of initial fish contaminant monitoring in this region by the OMOE, and includes the era when mercury was being discharged into the river system. As sampling frequency decreased over time, a wider range of years was selected for the “recent” time period (2000–2011) in order to retain robust sample sizes between time periods.

Recent fish mercury concentrations in the St. Lawrence River were also compared to regional fish mercury values. For all water bodies in the OMOE Sport Fish Contaminant Monitoring Program, 2000–2011 size-restricted northern pike, smallmouth bass, walleye and yellow perch mercury records were used to generate a mean fish mercury value for each water body with data for that species. These values were then compared to mean mercury concentrations from 2000–2011 in each species and in each of the listed St. Lawrence River blocks. However, not all St. Lawrence River blocks are identical in physical and chemical characteristics — most notably, portions of the river flow over Precambrian Shield bedrock (Fig. S2). As the physicochemical characteristics of Shield waters may influence fish mercury dynamics – such as increased propensity for acidification (e.g., Suns and Hitchin, 1990) – the Ontario-wide dataset was further subdivided according to a water body's location on- or off-Shield.

In addition to sport fish, mercury values for two forage fish species, spottail shiner (*Notropis hudsonius*) and young-of-year (YOY) yellow perch are also presented. Data for these two species were collected as part of the OMOE Sport Fish Contaminant Monitoring Program from various locations throughout block 15. The data used for these two species was restricted to sampling sites in the Cornwall, ON area, as patterns in fish mercury concentrations in these species can vary greatly over small spatial scales (Choy et al., 2008).

Mann–Kendall tests and Sen's slope estimates were calculated with the MAKESENS 1.0 Excel template (Salmi et al., 2002), and ANOVA comparisons were performed in SigmaStat v. 3.11 (2004). Statistical significance for all analyses was set at $p < 0.05$.

Results

Temporal trends

Temporal trends of fish mercury concentrations varied among species and river blocks examined (Fig. 2, Fig. S3). Mercury levels significantly declined over time by 31–53% in yellow perch from blocks 12, 13, 14 and 15 (Table 1, Fig. 2). For northern pike, mercury concentrations declined over time by 48% in block 12 ($p < 0.05$) and 46% in block 15 ($p < 0.1$) (Table 1). Smallmouth bass mercury concentrations did not significantly decline in any block, but had a non-significant ($p < 0.1$) decline of 47% in block 15 (Table 1). Walleye mercury concentrations significantly declined by 61% over time in block 15, but showed no trend in block 16 (Table 1). For northern pike, smallmouth bass and walleye, mean 2010 mercury concentrations in block 15 are above the first consumption advisory guideline of 0.26 µg/g (Fig. 2).

While Mann–Kendall analysis indicated that only walleye and yellow perch mercury had significant declines in block 15 over time, comparisons of historical (1975–1979) and recent (2000–2011) block 15 values for each species show that historical concentrations were significantly higher (33–59%) than recent concentrations for all four species within this block (Mann–Whitney test, $p < 0.001$). Similarly, mercury concentrations in spottail shiner and YOY yellow perch, two forage fish species, have also declined over time in block 15 (82% and 60%, respectively) (Fig. S4).

Despite overall long-term declines, trends for some species suggest that concentrations may be increasing in recent years. For example, yellow perch mercury concentrations in block 15 and 12 appear to increase from 2000 onwards (Fig. 2d). Mann–Kendall analysis on data from these blocks for 2000–2011 revealed a significant increasing trend for block 15 (2000–2010, $p < 0.05$) and an insignificant increasing trend for block 12 (2000–2009, $p < 0.1$). Trends in walleye mercury concentrations from blocks 14 and 16 also suggest increasing trends from the early 1990s–2002 (Fig. 2c); however, this is based on very few data points and more data is needed in order to determine if this is a real trend.

Spatial patterns

Spatial patterns in fish mercury concentrations were assessed both historically (1975–1979) and recently (2000–2011). Historical spatial fish mercury patterns show significant differences in mercury concentrations among blocks for northern pike (blocks 12/15 > 13/14), smallmouth bass (block 15 > 14), walleye (block 15 > 14), and yellow

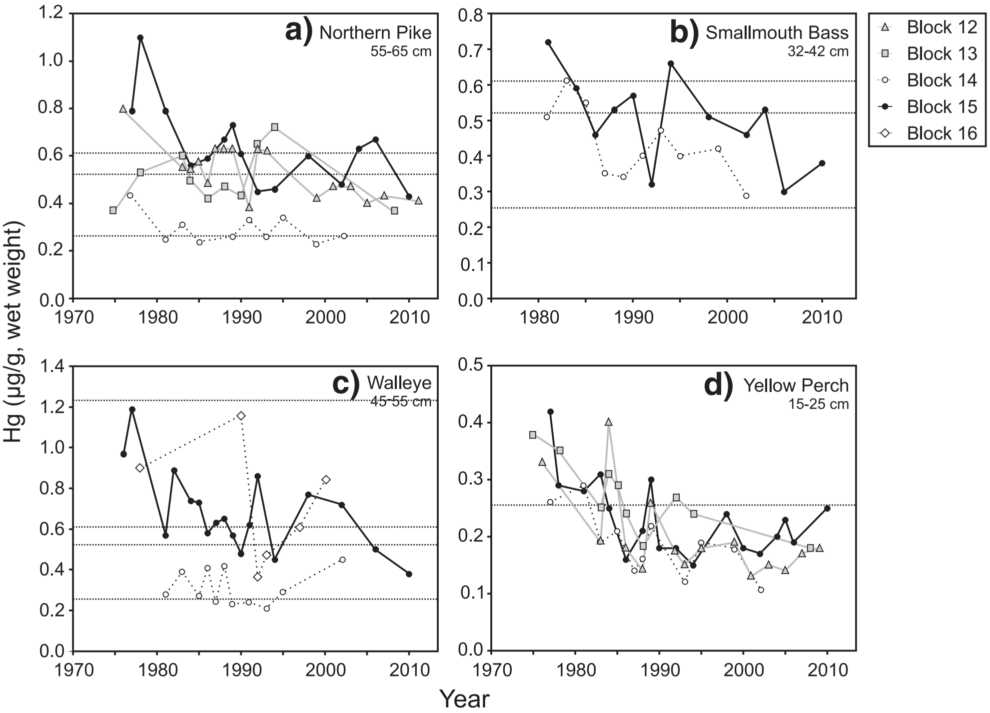

Fig. 2. Yearly mean mercury concentrations (µg/g wet weight) for blocks 12, 13, 14, 15 and 16 for a) northern pike (55–65 cm), b) smallmouth bass (32–42 cm), c) walleye (45–55 cm) and d) yellow perch (15–25 cm). Horizontal lines indicate consumption advisory guidelines as used in the Guide to Eating Ontario Sport Fish (OMOE, 2013), where 0.26 and 0.52 µg/g wet weight reflect concentrations for 4 and 0 meals/month, respectively, for the sensitive population, and 0.61, 1.23 and 1.84 µg/g wet weight reflect concentrations for 4, 2 and 0 meals/month, respectively, for the general population.

perch (blocks 13/15 > 14) (Fig. 3). Spatial patterns in recent mercury concentrations for each species are similar, despite generally lower concentrations overall, where block 15 > 14 for northern pike and smallmouth bass and blocks 13/15 > 14 for yellow perch (Fig. 3). Walleye did not show any significant differences between blocks 14 and 15 for the recent scenario (Fig. 3c).

Comparison with other Ontario water bodies

Parts of the St. Lawrence River (block 12) flow over the Precambrian Shield (Fig. S1), a geological feature that can influence physical habitat, water chemistry and species assemblages in overlaying waters (e.g., Neff and Jackson, 2012). Lakes and rivers located on the Precambrian Shield have elevated fish mercury levels compared to off-Shield locations (Fig. 4), likely due to differences in water chemistry between the two areas. Mean 2000–2011 block 15 concentrations are close to or greater than the 75th percentile of off-Shield lakes and rivers for all four species (Fig. 4). In contrast, compared to Shield lakes and rivers, the block 15 mercury levels are close to median values for northern pike and smallmouth bass, near 25th percentile for walleye and <75th percentile for yellow perch (Fig. 4). Since the overall Ontario scenario was largely represented by Shield lakes, a comparison with all Ontario lakes/rivers is similar to that for the Shield lakes (Fig. 4). Block 12, which is located over Shield geology, had recent mean fish mercury concentrations within the range for Shield locations within Ontario for both species with data for this block (i.e., northern pike and yellow perch) (Fig. 4).

Discussion

This study investigated long-term temporal trends (1975–2011) in fish mercury concentrations within the St. Lawrence Area of

Table 1
Summary of temporal trends in fish mercury content in the St. Lawrence River, for each sampling block and each species with available data. Block 15 includes the Cornwall, Ontario AOC. Negative Z-values coupled with large, negative Q values (i.e., Sen's slope estimate) indicate strongly declining trends. Where n < 10, S-values are calculated instead of Z-values. Significance are as follows: ** = p < 0.01; * = p < 0.05, and † = p < 0.1.

Block	Species	Size	Years	n	M–K test		Sen's slope	Sig. level
					S	Z	Q	
12	Northern pike	55–65 cm	1976–2011	19		−2.80	−0.014	**
	Yellow perch	15–25 cm	1976–2009	16		−2.12	−0.014	*
13	Northern pike	55–65 cm	1975–2008	10		−0.18	−0.002	
	Yellow perch	15–25 cm	1975–2008	11		−2.65	−0.023	**
14	Northern pike	55–65 cm	1977–2002	11		−0.78	−0.006	
	Smallmouth bass	32–42 cm	1981–2002	10		−1.43	−0.019	
	Walleye	45–55 cm	1981–2002	10		−0.47	−0.014	
	Yellow perch	15–25 cm	1977–2006	12		−2.26	−0.028	*
15	Northern pike	55–65 cm	1977–2010	15		−1.68	−0.014	†
	Smallmouth bass	32–42 cm	1981–2010	12		−1.85	−0.016	†
	Walleye	45–55 cm	1976–2010	18		−2.50	−0.021	*
	Yellow perch	15–25 cm	1977–2010	18		−2.05	−0.017	*
16	Walleye	45–55 cm	1978–2000	7	−3		−0.017	

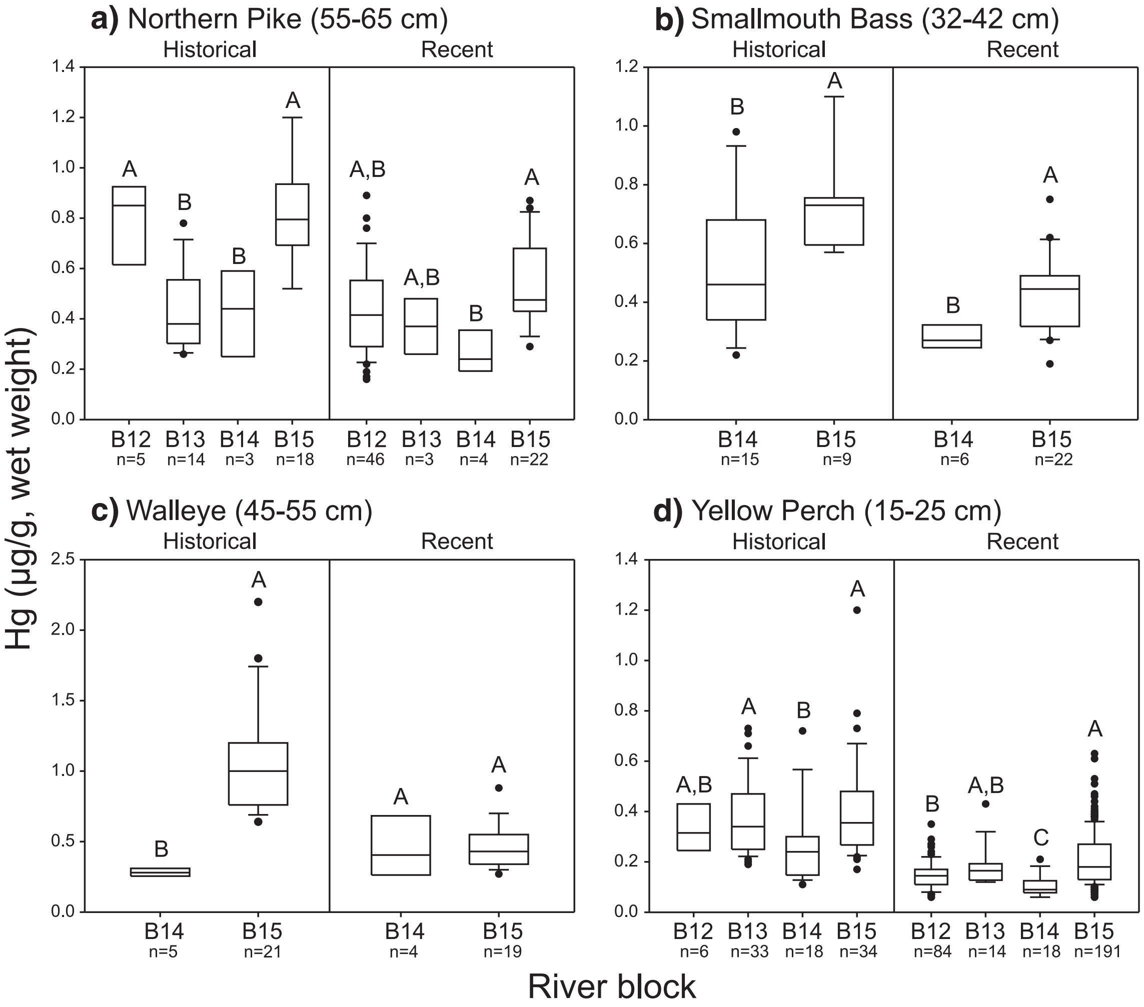

Fig. 3. Boxplots of historical (1975–1979) and recent (2000–2011) mercury concentrations for a) northern pike (55–65 cm), b) smallmouth bass (32–42 cm), c) walleye (45–55 cm) and d) yellow perch (15–25 cm), for each sampling block. Within each time period, boxes with identical letters belong to the same statistical group, where group A is significantly different ($p < 0.05$) from group B, and so on. Sample sizes for each box are depicted below the x-axis. Lines in each box represent the median, boxes indicate the 25th and 75th quartile values, and whiskers indicate the upper and lower values not classified as statistical outliers or extremes.

Concern at Cornwall, Ontario in comparison with upstream reference sections of the St. Lawrence River, as well as historical (1975–1979) and recent (2000–2011) spatial trends among river sections. While designated an Area of Concern in part due to high fish contaminant levels, fish mercury levels in the block 15 river segment at the start of monitoring were generally lower than a number of other well-known contaminated sites worldwide, as summarized by Neff et al. (2012). For example, mean values in the first year of monitoring for northern pike (1976) and walleye (1977) for block 15 are considerably lower (85–86% and 88–93%, respectively) than values recorded for northern pike (1976) and walleye (1970) in Clay Lake of the English–Wabigoon River in northwestern Ontario (Neff et al., 2012).

The temporal patterns observed are similar to other historically contaminated waterways in the region (e.g., Bhavsar et al., 2010; Gewurtz et al., 2010; Neff et al., 2012). Overall, significant declines in mercury concentrations were observed in the AOC river block for walleye and yellow perch, and non-significant declines for northern pike and smallmouth bass. Similar trends were also observed in two forage fish species (Fig. S4). These results are supported by sediment data for Lake St. Francis (i.e., block 15), where the median sediment mercury concentrations for this section of the river declined by 56% (from 0.14 µg/g to 0.07 µg/g) since 1979 (Pelletier, 2010). Significant temporal declines were also documented in upstream reference blocks for both northern pike (block 12) and yellow perch (blocks 12, 13 and 14).

Despite the declines observed in block 15, recent fish mercury concentrations in this section remain elevated compared to upstream blocks. Currently, fish consumption advisories are in place for the sensitive population (i.e., women of childbearing age and children under the age of 15) for all four species within block 15 examined in this study (OMOE, 2013). From these results, it is clear that recovery from historical mercury contamination within the St. Lawrence AOC is ongoing, and varies by species. Further, recent trends for yellow perch in blocks 15 and 12 suggest that mercury concentrations may be increasing, a pattern which has been previously documented in the Great Lakes (Bhavsar et al., 2010; Monson et al., 2011; Sadraddini et al., 2011). Likewise, trends in sediment mercury concentrations for Lake St. Francis suggest that mercury levels have stabilized since 1999 (Pelletier, 2010). However, these results should be interpreted with caution, as there was considerable variation in mean mercury concentrations over time in all species and blocks, and the detection of a true increasing trend will require additional data for future years.

Previous studies focusing on Lake Ontario and the St. Lawrence River have documented mercury declines over time in a number of fish species (e.g., Bhavsar et al., 2010; Borgmann and Whittle, 1991, 1992; French et al., 2006). Results from this study for yellow perch in particular support this trend, as concentrations significantly declined in river sections not known to have major historical point-source mercury inputs (e.g., blocks 12–14). A previous study by Goulet et al. (2008) also used data from the OMOE Sport Fish

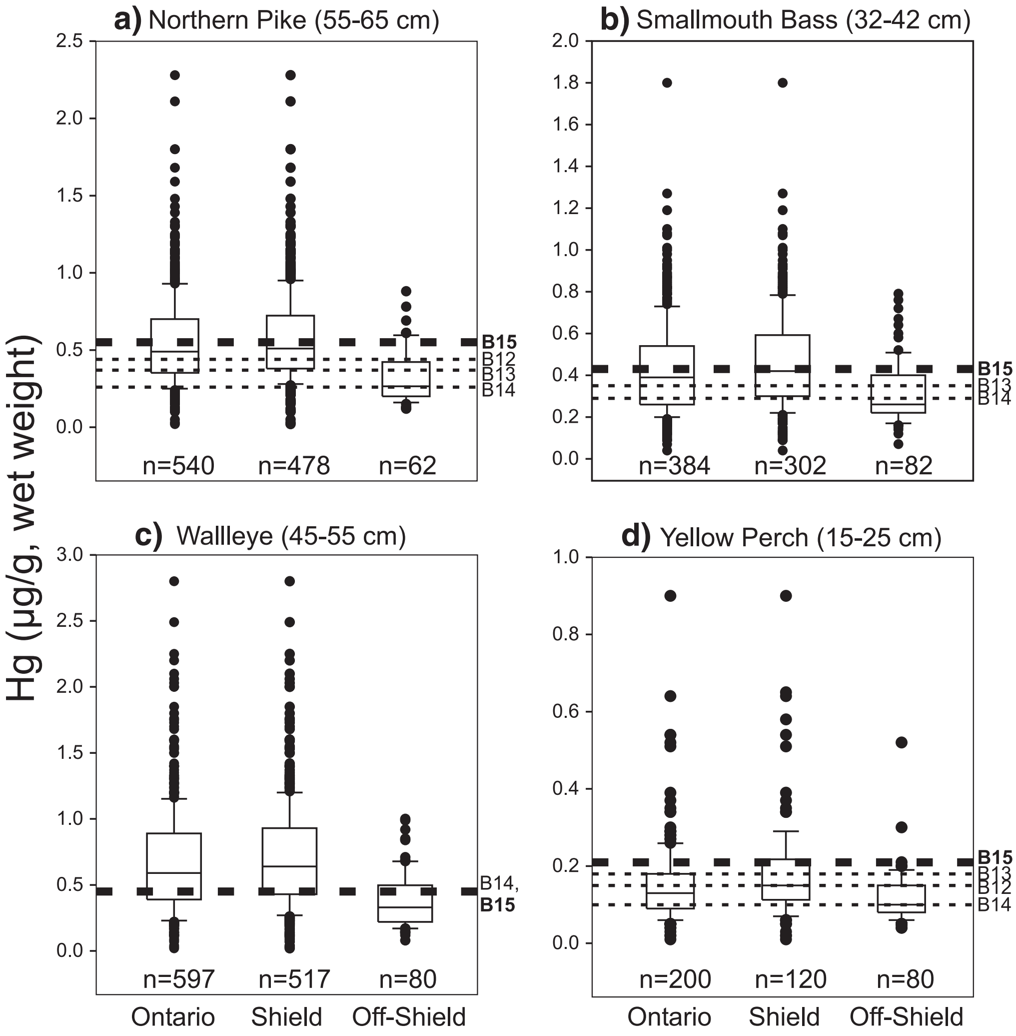

Fig. 4. Boxplots of recent (2000–2011) mean mercury values for lakes and rivers within Ontario for a) northern pike, b) smallmouth bass, c) walleye and d) yellow perch, including separate boxes for locations located on the Precambrian Shield ("Shield"), and those not located on the Precambrian Shield ("off-Shield"). Lines representing 2000–2011 mean fish mercury concentrations for St. Lawrence River blocks are overlaid for comparison. Sample sizes (i.e., the number of unique water bodies) for each box are depicted below each block. Lines in each box represent the median, boxes indicate the 25th and 75th quartile values, and whiskers indicate the upper and lower values not classified as statistical outliers or extremes.

Contaminant Monitoring Program and examined temporal and spatial trends in mercury concentrations in northern pike, walleye, white sucker (*Catostomus commersoni*) and yellow perch from four sections of the St. Lawrence River — Thousand Islands, Brockville, Lake St. Lawrence and Lake St. Francis, which correspond to the river blocks used in this study (blocks 12, 13, 14 and 15, respectively). However, instead of limiting data to a restricted size range, Goulet et al. (2008) used general linear models with fish length as an independent variable. The trends reported by Goulet et al. (2008) cover 1975–1995, and are generally in agreement with the trends observed in this study, which extended the temporal period to 2011. Significant declines in mercury concentrations were observed both in Goulet et al. (2008) (1975–1995) and in this study (1975–2011) for northern pike from the Thousand Islands region (block 12), yellow perch in the Brockville (block 13), Lake St. Lawrence (block 14) and Lake St. Francis (block 15) regions, and walleye in the Lake St. Francis (block 15) region.

In contrast, conclusions regarding the spatial patterns of fish mercury concentrations in the St. Lawrence River have been mixed. Ion et al. (1997) investigated spatial differences in contaminant concentrations in yellow perch collected in 1991–1992 from five sections of the St. Lawrence River downstream of the Cornwall AOC. Yellow perch mercury concentrations varied among river sections (0.12–0.15 µg/g wet weight), but there were no significant differences between the Québec portion of fluvial Lake St-Francois, immediately downstream of block 15 as designated in this study, and other downstream fluvial lakes (Ion et al., 1997). Similarly, Goulet et al. (2008), using data from 1991–1995, did not find any region in the river, including the Lake St. Francis (block 15), to be consistently more contaminated than the other for northern pike, walleye, white sucker and yellow perch. In contrast, we observed significantly higher fish mercury concentrations in the AOC river block compared to upstream blocks for both historical (1975–1979) and recent (2000–2011) scenarios for nearly all species considered.

The following could explain differing findings from this study and Goulet et al. (2008), which are both based on the same source of monitoring data. Our study considered a longer time period, which included observations for the last 15 years not considered by Goulet et al. (2008). Recent fish mercury values for northern pike, smallmouth bass and yellow perch indicate that block 15 has generally the highest fish mercury levels compared to all upstream blocks (Fig. 3). Further, historical mercury values for the AOC river block were statistically indistinguishable from the upstream river blocks in some cases. For example, historical mercury concentrations in northern pike from the AOC river block 15 and block 12 were not significantly different, but collectively were significantly greater (51%) than blocks 13 and 14. In addition, historical yellow perch block 15 mercury values were not significantly different from the corresponding block 13 values, but together were significantly greater (31%) than block 14 values.

It is possible that variations in fish mercury concentrations within block 15 are at least in part responsible for the lack of a consistent spatial pattern across sampling regions. Recent studies have examined fish mercury dynamics within the river AOC block by further delineating this section into "contaminated" and "reference" areas. Fowlie et al. (2008) found that mercury levels were higher (2.3–3.6×) in yellow perch from one contaminated zone, especially during the summer months, than in fish from other contaminated and reference zones within block 15. In a similar study, Choy et al. (2008) examined mercury concentrations in YOY spottail shiners (*Notropis hudsonius*) from contaminated and reference zones within the AOC block, and found that mercury concentrations were significantly higher in a contaminated zone (0.07 μg/g wet weight) compared to reference zones (0.03–0.05 μg/g wet weight). Further, Choy et al. (2008) also reported significant differences in fish mercury concentrations for sites within contaminated zone, despite being only 500 m apart. These studies suggested that internal sources and processes of mercury may be influencing local mercury dynamics within the St. Lawrence AOC. Unfortunately, the resolution of the data in the OMOE Sport Fish Contaminant Monitoring Program is unable to examine fish mercury variation within block 15. Considering the evidence for localized fish mercury dynamics within this river section, future studies on this region would likely be strengthened by sampling over smaller spatial scales.

To our knowledge, there is no current or historical point-source discharge of mercury into the river upstream of the AOC block, aside from high sediment concentrations reported for the Inner Harbour of Kingston, Ontario, just upstream of block 12 (Manion et al., 2010). However, 2010 sport fish tissue concentrations for this area are relatively low (0.03–0.47 μg/g wet weight) (OMOE, unpublished data), and are not the cause of current consumption advisories in this area (OMOE, 2013). As such, major sources of mercury in the river upstream of the AOC would be export from Lake Ontario, atmospheric deposition and runoff from the catchment basin. In addition, the drainage area for the upstream section of the river includes the Frontenac Axis portion of the Precambrian Shield, which bisects through the river. This runoff could be naturally higher in mercury, due to the geological composition of the underlying bedrock. Recent (2000–2011) fish mercury concentrations indicate that all species in block 12, are within the range of values for these species from other Ontario lakes and rivers located on the Precambrian Shield, and thus fish mercury concentrations in this river section are within the natural range of variation for this region. These elevated concentrations in block 12 decline in downstream blocks 13 and 14, especially for northern pike and yellow perch (Fig. 3). Fish mercury concentrations for blocks 13 and 14 (off-Shield), with the exception of yellow perch from block 13, are generally within the range of values for other Ontario lakes and rivers also located off-Shield. In contrast, block 15, also off-Shield, is outside the range of variation for other off-Shield Ontario lakes and rivers, suggesting that fish mercury levels in this river section remained elevated beyond what would be expected for the region. Further, recovery in block 15 is likely also influenced by the presence of contaminated sediment as well as input from upstream blocks, Lake Ontario, and tributaries. For example, the tributaries draining the Canadian agricultural lands adjacent to the AOC have water mercury levels 2–8× greater than uncontaminated zones in the St. Lawrence River and the fractions of methyl mercury in the total mercury in the tributary waters are 2–5× higher than normal values for most uncontaminated freshwater systems (unpublished data, Jeff Ridal, St. Lawrence River Institute of Environmental Sciences).

Fish mercury concentrations can be influenced by a number of factors, including water chemistry (e.g., Bodaly et al., 1993; Rasmussen et al., 2007), productivity (e.g., Cizdziel et al., 2002; Pickhardt et al., 2002), sediment composition (e.g., Rudd et al., 1983), and climate (e.g., French et al., 2006; Rennie et al., 2010), as well as fish growth (e.g., Simoneau et al., 2005), condition (e.g., Cizdziel et al., 2002; Rennie et al., 2010; Suns and Hitchin, 1990), gender (e.g., Simoneau et al., 2005) and trophic position (e.g., Bodaly et al., 1993; Cizdziel et al., 2002; Swanson et al., 2006). Patterns over time may also be influenced by certain environmental changes, such as climate change, species invasions and/or changes to atmospheric deposition of mercury (Bhavsar et al., 2010; French et al., 2006; Neff et al., 2012). Within the St. Lawrence River, it has been shown that fish mercury dynamics can vary by species and location, even at small scales (Choy et al., 2008; Fowlie et al., 2008; Goulet et al., 2008; Ridal et al., 2010). It is not surprising, then, that not all species had significant trends in all blocks in this study, or that some blocks had species with significant temporal trends while others did not. Teasing apart the abiotic and biotic factors accountable for these differences is not within the scope of this study, although it is important to consider that a number of factors may be at play in this system, ultimately influencing temporal and spatial patterns of fish mercury concentrations. However, this study clearly shows that recent fish mercury values from the St. Lawrence River AOC are significantly lower than they were historically, indicating that some degree of recovery has occurred in this historically contaminated section of the St. Lawrence River.

Conclusion

Fish mercury concentrations have significantly declined in some sections of the St. Lawrence River, particularly in block 15 that includes the region designated as an Area of Concern. In the absence of a point source discharge within block 15 and the upstream blocks, further declines in AOC fish mercury levels can be expected as recent fish mercury levels from this river section remain elevated compared to the upstream reference blocks. This is also supported by the finding that the recent fish mercury concentrations for the off-Shield AOC river section remain elevated compared to other off-Shield Ontario lakes and rivers. Fish consumption advisories should still be followed when consuming fish from the St. Lawrence River. Continued monitoring of fish mercury concentrations within this system will be necessary to update the risk of fish consumption and ensure that the recovery of the AOC continues in future.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.jglr.2013.03.005.

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