Improvements in fish polychlorinated biphenyl and other contaminant levels in response to remedial actions in Hamilton Harbour, Ontario, Canada

Margaret R. Neff,1 Samantha Stefanoff,2 and Satyendra P. Bhavsar1,3*
1Ontario Ministry of the Environment and Climate Change, Sport Fish Contaminant Monitoring Program, Environmental Monitoring and Reporting Branch, 125 Resources Road, Toronto, Ontario M9P 3V6, Canada
2Department of Science, York University, 4700 Keele Street, Toronto, Ontario M3J 1P3, Canada
3School of the Environment, University of Toronto, 33 Willcocks Street, Suite 1016V, Toronto, Ontario M5S 3E8, Canada
*Corresponding author: satyendra.bhavsar@ontario.ca; s.bhavsar@utoronto.ca

Hamilton Harbour, located in Ontario, Canada at the western end of Lake Ontario, is recognized as one of the most anthropogenically-impacted regions within the Great Lakes and is currently listed as an Area of Concern. One of the Beneficial Use Impairments for the harbour has been restrictions on fish consumption due to elevated contaminant levels. In this study, we examined past and recent fish contaminant data collected by the Ontario Ministry of the Environment and Climate Change in partnership with other agencies to evaluate temporal trends in fish contaminant concentrations. Measurements for both resident and migratory sport fish as well as juvenile/forage fish were considered, with analysis focused on polychlorinated biphenyls, the group of chemicals identified as the major contaminant of concern. Current contaminant levels were evaluated against fish consumption advisory benchmarks used by Ontario Ministry of the Environment and Climate Change, and compared with corresponding observations for other locations across the Great Lakes, including other Areas of Concern. The results show statistically significant improvements in fish contaminant levels within Hamilton Harbour, with recent fish mercury concentrations below the first advisory benchmarks for all species included in this study. Polychlorinated biphenyl concentrations declined by 59–82% from historical levels, although this decline was not statistically significant in Brown Trout, Common Carp, Freshwater Drum and White Sucker. Further, all species exhibit recent polychlorinated biphenyl concentrations above the first consumption advisory benchmark of 105 ng g⁻¹. Compared to other Great Lakes locations, including other areas of concern, Hamilton Harbour polychlorinated biphenyl concentrations remain amongst the highest. The results suggest that recovery of Hamilton Harbour is still on-going.

Keywords: Area of Concern, mercury, Great Lakes, temporal trends, fish consumption

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**Introduction**

Hamilton Harbour, situated at the western end of Lake Ontario, Canada, was identified as one of the most anthropogenically-impacted regions of the Great Lakes by the International Joint Commission (IJC) in 1985 and consequently listed as an Area of Concern (AOC) in 1987. This area has been impacted by rigorous industrial (e.g., iron, steel manufacturing) and urban development around its shoreline for several decades, and subsequently, the water and sediments of the harbour have become contaminated by metals, pesticides, polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs) (Environment Canada and Ontario Ministry of the Environment, 2010). In 1992, a Remedial Action Plan (RAP) was implemented to protect and restore the AOC, with a focus on remediation/improvement of fish and wildlife populations, toxic substances and sediments, water quality and bacterial contamination, urbanization and land management, and public access and aesthetics (Hall et al., 2006).

One of the fourteen Beneficial Use Impairments (BUIs) of Hamilton Harbour is restrictions on fish and wildlife consumption. At present, fish consumption advisories for Hamilton Harbour are mainly due to high levels of PCBs (Bhavsar et al., 2011; Gandhi et al., 2014; OMOE, 2013). PCBs were first manufactured in 1929 and used widely for several decades in various industrial materials as well as in coolants and lubricants for electrical equipment, but were effectively banned in North America by 1977 due to human and environmental health concerns (Health Canada, 2005). PCBs have been found in fish throughout the Great Lakes, often in concentrations high enough to warrant consumption advisories (OMOE, 2013); however, current levels in Great Lakes fishes are much lower compared to historical peak values (Bhavsar et al., 2007). Within Hamilton Harbour, anthropogenic activities have resulted in very high levels of PCBs in both sediment and water compared to Lake Ontario and the rest of the Great Lakes (Environment Canada and Ontario Ministry of the Environment, 2010), with historical loadings of PCBs to the harbour estimated at 50 kg yr$^{-1}$ (Labencki, 2008). The major sources of PCBs were identified as wastewater treatment plants, inputs of urban runoff, combined sewer overflows and storm sewers, and unidentified industrial sources (Labencki, 2008). Recent work indicates that sediment PCB concentrations within the harbour remain high enough to cause restrictive consumption advisories in fish (Bhavsar et al., 2010a; Labencki, 2008).

In many cases, fish contaminant concentrations, a result of pelagic and benthic exposure, can reflect both water and sediment concentrations as well as indicate the overall condition of the aquatic system (Bhavsar et al., 2007; Morrison et al., 2002). As various remedial actions have been enacted since the harbour’s designation as an AOC, assessment of fish contaminant concentrations will help determine the success of these projects in reducing overall contaminant concentrations within the harbour. In this study, we use a 35-year data series (1976–2013) of PCB and mercury concentrations for various fish species in the Hamilton Harbour AOC to assess how levels of PCBs and mercury have changed over this time period. Further, we compare current fish contaminant levels within the harbour to other locations within the Great Lakes, including other AOCs. This analysis is expected to provide an insight into the comparative current status of PCBs and mercury contamination in Hamilton Harbour, as well as the recovery of the Hamilton Harbour AOC by remediation efforts.

**Materials and methods**

**Sample collection and processing**

Since the early 1970s, the OMOECC, in partnership with Ontario Ministry of Natural Resources and Forestry (OMNRF), has collected a wide range of fish species in water bodies throughout the province of Ontario, including the Great Lakes and Hamilton Harbour, with the frequency being variable depending on requirement and resources. Department of Fisheries and Oceans Canada and Environment Canada have also helped the OMOECC in collection of fish samples from Hamilton Harbour and certain other locations. Typically, skinless, boneless dorsal muscle fish fillets are used to monitor contaminants for the purpose of assessing environmental condition as well as establishing consumption advisories for sport fisheries (OMOE, 2013). Due to their large surface area and variation in within-lake environmental conditions, fish contaminant levels within the Great Lakes fishes are assessed on a localized basis, with each lake split into several sampling
blocks (Figure 1). These blocks have been determined by the OMOECC Fish Contaminant Monitoring Program, in consultation with OMNRF biologists, based on differences in fish contaminant levels and/or fish populations. Fish collections from the Hamilton Harbour area, or Lake Ontario block 3 (LO3), began in 1976 and cover a wide variety of species, with varying sampling frequency over the years. At each sampling event, fish selected for contaminant analysis were based on data gaps within OMOECC’s database; generally, attempts were made to analyze all species possible over large size ranges. For this analysis, species were selected based on data availability over a broad temporal range. Multiple species were examined in order to encompass any among-species variation in contaminant concentrations (Bhavsar et al., 2010b, 2007).

Skinless, boneless fillets were stored at $-20\degree C$ until chemical analysis at OMOECC laboratories in Toronto, Ontario. The majority of samples collected from Hamilton Harbour have been analyzed for mercury, total PCBs, mirex, toxaphene and photomirex; a smaller subset of samples have been analyzed for dioxins/furans, and, in recent years, perfluorooctanesulfonic acid (PFOS). Chemical analysis of fish samples follow standard protocols developed by OMOECC laboratories, including HGBIO-E3057 (OMOE, 2006; mercury), PFAOC-E3136 (OMOE, 2005; PCBs), DFPCB-E3481 (OMOE, 2012; dioxins/furans) and PFAS (OMOE, 2014); details have been described in previous studies (Awad et al., 2011; Bhavsar et al., 2014, 2010b, 2008, 2007; Gewurtz et al., 2014).

**Figure 1.** Ontario Ministry of Environment Climate Change (OMOECC) fish consumption advisory blocks within the Canadian waters of Lakes Superior, Huron, Erie and Ontario. Advisory blocks associated with Areas of Concern considered in this study are highlighted in grey. Inset depicts Hamilton Harbour (Lake Ontario block 3, LO3), with forage fish sampling stations A, B, C, D, E and F.

**Statistical analysis**

The influence of fish size on contaminant concentrations is typically addressed in one of two ways: the exclusion of data outside of a limited size range (Bhavsar et al., 2010b; Neff et al., 2013), or using standardized contaminant values for specific fish lengths (Gewurtz et al., 2011). For this analysis, the restricted size range approach was deemed most appropriate, due to poor relationships between fish length and contaminant concentrations for some species in some years. For all species included in the analysis, a 10-cm size range was selected, based on the grand mean fish length across all years for each species. If a
significant, statistical relationship (power series regression, p < 0.05) between concentration and fish length remained, the size range was reduced until the relationship was no longer present.

Temporal trends in Brown Bullhead (*Ameiurus nebulosus*, 25–35 cm), Common Carp (*Cyprinus carpio*, 50–60 cm) and White Perch (*Morone americana*, 16–26 cm) contaminant concentrations over time within Hamilton Harbour were analysed with the non-parametric Mann-Kendall test with Sen’s slope estimate. Within each species, the sample size for each year varied, but in all cases, n ≥ 2. Additionally, there were no significant differences in mean length among years for any species (ANOVA, p > 0.05). As many species did not have adequate temporal coverage for trend analysis, historical (1976–1985) and recent (2005–2013) concentrations within Hamilton Harbour were compared by pooling all samples within each time period and examining for statistical differences with a t-test. Species used for this comparison included Brown Bullhead (25–35 cm), Brown Trout (*Salmo trutta*, 49–59 cm), Common Carp (50–60 cm), Freshwater Drum (*Aplodinotus grunniens*, 40–50 cm), Northern Pike (*Esox lucius*, 60–70 cm), White Perch (16–26 cm), White Sucker (*Catostomus commersonii*, 35–45 cm) and Yellow Perch (*Perca flavescens*, 15–25 cm). The impact of fillet percent lipid content on PCB measurements was assessed using a lipid normalization procedure, where the PCB value of each individual fish sample was divided by its total muscle lipid content (%). Temporal trends and changes from historical to recent PCB concentrations were then re-assessed using the lipid-normalized PCB values.

To compare fish PCB concentrations in Hamilton Harbour to reference locations within Lake Ontario, recent (2005–2013) data from sampling blocks LO2a and LO4 were considered (Figure 1). These sampling areas at the western end of Lake Ontario represent both open-water (LO4) and harbour (LO2a) environments and have not been exposed to localized PCB contamination. Comparisons to LO2a were made for Bluegill (*Lepomis macrochirus*, 10–20 cm), Brown Bullhead (25–35 cm), Common Carp (50–60 cm), Largemouth Bass (*Micropterus salmoides*, 30–40 cm), and White Perch (20–25 cm). Comparisons to LO4 were made for Rock Bass (*Ambloplites rupestris*, 15–20 cm), White Sucker (35–45 cm), and Yellow Perch (15–20 cm). Recent (2005–2013) Hamilton Harbour data were also compared to recent data for other Great Lakes sampling blocks, including those associated with AOCs which have Beneficial Use Impairments for fish consumption: Thunder Bay (Lake Superior 3), Peninsula Harbour (Lake Superior 8a), St. Marys River (SMR), Spanish Harbour (North Channel NC2), St. Clair River (Lake Erie 7a, 7b, 7c), Detroit River (Lake Erie 5b), Niagara River (Lake Ontario 1b), Toronto (Lake Ontario 4a), Bay of Quinte (Lake Ontario 9, 9a, 9b), and St. Lawrence River (Lake Ontario 15) (Figure 1). Comparisons were made with Brown Bullhead (25–35 cm), Brown Trout (50–60 cm), Common Carp (60–70 cm), Freshwater Drum (50–60 cm), White Perch (15–25 cm), White Sucker (40–50 cm) and Yellow Perch (15–25 cm).

Forage fish contaminant measurements have also been collected from various locations within Hamilton Harbour, although with less frequency. The data include samples collected between 1984 and 2010, from Carroll’s Point (2009, 2010), Cootes Paradise (1987), offshore the Burlington Golf & Country Club (1984, 1986, 1987, 1991), Grindstone Creek (1989, 2006), Willow Cover (1989, 1991), and most frequently, near the Queen Elizabeth Way (QEW, 1977–2006) (Figure 1). The majority of samples collected consisted of Emerald Shiner (*Notropis atherinoides*) and Spottail Shiner (*Notropis hudsonius*). Lack of data prevented any statistical analysis for forage fish; however, mean PCB concentrations for each year of collection of Emerald Shiner and Spottail Shiner were calculated for the QEWS site and have been presented here. In addition, pooled data for all Hamilton Harbour forage fish were compared to forage fish data for nearby nearshore locations within LO2 and LO4 (Figure 1), near the cities of Burlington, Mississauga, Oakville, Scarborough, St. Catherines and Toronto, as well as the Niagara River.

Where relevant, results are presented with demarcation of MOECC’s consumption advisory benchmarks. For mercury, benchmarks are at 0.26 and 0.5 μg g⁻¹; for PCBs, benchmarks are at 105, 211, 422 and 844 ng g⁻¹. Mann-Kendall tests and Sen’s slope estimates were calculated with the MAKESENS 1.0 Excel template (Salmi et al., 2002); all other statistical analyses were calculated with R (R Core Team, 2013). Statistical significance for all analyses was set at p < 0.05.

**Results**

Brown Bullhead mercury concentrations in Hamilton Harbour significantly declined over
time, while Common Carp and White Perch did not show any significant trends (Figure 2). However, recent (2005–2013) mercury concentrations for Brown Bullhead, Freshwater Drum and Yellow Perch were significantly lower than historical concentrations (Figure S1, available in the online supplementary information). Overall, however, mercury concentrations have been and continue to be low in Hamilton Harbour, and recent data indicate that all species are below the first

Figure 2. Trends in mercury and PCB fish concentrations in (a) Brown Bullhead, (b) Common Carp and (c) White Perch in Hamilton Harbour. Data points indicate mean concentrations for each year, ± standard deviation. Solid lines indicate Sen’s slope estimate from the MAKESENS analysis. The dashed line(s) indicate mercury and PCB consumption advisory benchmarks. Brown Bullhead mercury concentrations and lipid-normalized PCB concentrations (not shown) significantly declined over time ($p < 0.05$, Mann-Kendall test); no other trends were statistically significant.
consumption advisory benchmark (0.26 μg g⁻¹) for mercury.

There were no significant temporal trends in PCB concentrations for Brown Bullhead, Common Carp and White Perch (Mann-Kendall test, p > 0.05) (Figure 2). PCB concentrations did decline by 59–83% between the first and last sampling years in Brown Bullhead, Brown Trout, Common Carp, Northern Pike, White Perch and Yellow Perch, but increased by 33% and 42% for Freshwater Drum and White Sucker, respectively (Figure 3). When PCB values were lipid-normalized, there was a significant, declining trend for Brown Bullhead (Mann-Kendall test, p < 0.05). Recent (2005–2013) PCB concentrations were significantly lower than historical (1976–1985) concentrations in Brown Bullhead, Northern Pike, White Perch and Yellow Perch, for both raw (Figure 3) and lipid-normalized PCB values (not shown) (t-test or Mann-Whitney test, p < 0.05); however, concentrations remain above certain consumption advisory benchmarks for all species (Figure 3).

Recent (2005–2013) PCB concentrations in all examined Hamilton Harbour species were significantly greater than corresponding concentrations in the reference blocks LO2a and LO4 (t-test or Mann-Whitney test, p < 0.05; Figure 4). Recent (2005–2013) fish PCB concentrations in Hamilton Harbour are also among the highest of all other AOCs with comparable data (Figure 5). PCB levels in Brown Trout and White Sucker from Hamilton Harbour were similar to those from the Toronto AOC, and for White Perch, Hamilton Harbour levels were similar to those from the Detroit River (Figure 5). Further comparisons to non-AOC Great Lakes locations revealed that fish PCB concentrations from within the harbour were among the highest for all species (Figure S2, available in the online supplementary information). However, Brown Trout PCB concentrations were similar across all locations with applicable data, and fish PCB concentrations in Whitby Harbour (LO6b) were higher than or similar to concentrations in Hamilton Harbour for all species with applicable data except for Freshwater Drum (Figure S2). Whitby Harbour is a known hotspot for dioxins and furans, and current fish consumption advisories for Brown Bullhead and Common Carp are due to restrictive levels of these
contaminants (Ontario Ministry of the Environment, 2013).

Recent (2005–2013) PCB concentrations in Emerald Shiner and Spottail Shiner from Hamilton Harbour were the highest compared to other nearby open-water locations outside of the AOC (Figure 6a). Temporal trends in Hamilton Harbour forage fish were assessed with Spottail Shiner (1987, 1991, 1994, 2000) and Emerald Shiner (2003, 2006) collected at the QEW sampling station. The relationship between Emerald Shiner and Spottail Shiner PCB concentrations is nearly 1:1 (French et al., unpublished data), and thus the more recent Emerald Shiner data is presented here as a good estimate of Spottail Shiner PCB concentrations from 2003 and 2006. The resulting temporal trend suggests relatively stable but variable PCB concentrations up to 2003, with a higher value for 2006 (Figure 6b). The most recent PCB values for Hamilton Harbour forage fish remain above the IJC guideline for the protection of fish-eating wildlife (International Joint Commission, 2012).

Figure 4. Comparisons of recent (2005–2013) fish PCB values from Hamilton Harbour to (a) Lake Ontario block 4 (LO4) and (b) Lake Ontario block 2a (LO2a). The dashed line(s) indicate PCB consumption advisory benchmarks.

Figure 5. Comparisons of historical (1975–1985) recent (2005–2013) PCB concentrations in fish from Hamilton Harbour (HH) to other AOCs, including Bay of Quinte (BOQ), Detroit River (DR), Niagara River (NR), Spanish Harbour (SH), St. Clair River (SCR), St. Lawrence River (SLR), St. Marys River (SMR), Thunder Bay (TB), and Toronto and Region (TOR). The dashed line(s) indicate PCB consumption advisory benchmarks.
Discussion

While there were no statistically significant temporal trends in PCBs, the main contaminant of concern for the harbour, comparison of historical data to recent data showed that PCB concentrations in four sport fish species are now significantly (p < 0.05) lower than previous years. However, this improvement is tempered by evidence that PCB concentrations in sport fish from Hamilton Harbour remain elevated compared to both nearby reference areas in Lake Ontario, as well as other AOC and non-AOC locations in the Great Lakes, and are greater than consumption advisory benchmarks. Thus, while conditions are improving in this AOC, recovery is still ongoing.

The observed declines are likely at least in part the result of a number of remedial actions addressed by the AOC’s Remedial Action Plan (RAP), including implementation of abatement regulations to eliminate issues associated with industrial discharges entering the harbour, dredging of Windermere Basin, containment of landfill leachate, and updated sewer system bylaws and enhanced enforcement of adjacent municipalities (Environment Canada and Ontario Ministry of the Environment, 2010). However, sediment PCB concentrations remain high, particularly in Windermere Arm (Labencki, 2008), and PCB source track-down investigations suggest ongoing sources of PCBs to the Strathearn Slip. Further, water and sediment exchange with Lake Ontario is limited to a narrow shipping channel at the east end of the harbour (Figure 1), and dredging/shipping activities in the harbour continue, which likely influence sediment resuspension and PCB dynamics within the harbour.

Aside from fish PCB contamination via sediments, there is some evidence to suggest that there may also be exposure via the water column. This is supported by high PCB concentrations in forage fish, particularly when compared to other nearby areas of Lake Ontario (Figure 6). PCBs measured via semipermeable membrane devices (SPMDs) are thought to be indicative of pelagic fish exposure, and concentrations measured via these devices within the harbour are many times greater than other nearshore areas of the Great Lakes (e.g., ~400 ng ml\(^{-1}\) triolein in the main basin, ~1,900 ng ml\(^{-1}\) triolein in Windermere Arm; Labencki, 2008; Tanya Long, OMOECC, personal communication).

It is likely that differences in both feeding ecology and migratory behaviour are contributing to the wide variation in PCB concentrations among species within Hamilton Harbour. Recent PCB concentrations in the two top-predator species included in this study, Brown Trout and Northern Pike, were substantially lower than historical levels (77% and 76%, respectively). In contrast, with the exception of Brown Bullhead, bottom-feeder species (Common Carp, Freshwater Drum and White Sucker) had no significant PCB declines, and in the case of Freshwater Drum, PCBs increased over time by 33%. This increase may be at least partially explained by the propensity of Freshwater Drum to

Figure 6. (a) Recent (2005–2013) mean PCB ± standard deviation concentrations in Emerald Shiner and Spottail Shiner in Hamilton Harbour and other nearby nearshore locations in Lake Ontario. Mean length of each location is depicted above each bar. Values for Hamilton Harbour are pooled data from all Hamilton Harbour forage fish sampling locations (Figure 1, inset). (b) Temporal trends in Spottail Shiner and Emerald Shiner mean PCB concentrations from the QEW sampling site (Figure 1, inset). The IJC PCB guideline for the protection of fish-eating wildlife (100 ng g\(^{-1}\)) is depicted with a dashed line.
consume zebra mussels – an early study by Morrison et al. (1998) projected that PCB burdens would increase by 14% in Freshwater Drum with the inclusion of zebra mussels in their diet. Zebra and quagga mussels have been found in Hamilton Harbour, although Dermott et al. (2007) reported that average Dreissena biomass was considerably lower than in Lake Erie or Bay of Quinte. While also a bottom-feeder, Brown Bullhead consume a large amount of aquatic vegetation while feeding for benthic organisms compared to Common Carp and White Sucker (S. Petro, personal communication), which may have limited PCB uptake in Brown Bullhead relative to these species. In contrast, recent PCB concentrations in White Perch significantly declined from historical levels – while this species also consumes insects and small fishes, it feeds both in deeper waters and as well as near the surface at night (Holm et al., 2009), and thus has less contact with bottom sediments.

Another contributing factor to variations in PCB concentrations among species may be their propensity to move in and out of the harbour. Given the proximity of other PCB-contaminated AOCs, such as Toronto and the Niagara River, it is possible that the PCB concentrations observed in some species collected from Hamilton Harbour are the product of exposure at several different sites in Lake Ontario. Of the species included in this study, Brown Trout are migratory, and will move in and out of the harbour. All other species should be able to find suitable habitat within the harbour, but have no barrier to prevent them from moving in or out (S. Petro, OMOECC, personal communication). At this stage, there little is known about this aspect of fish behaviour in Hamilton Harbour.

Conclusions

Overall, temporal changes in fish PCB concentrations in Hamilton Harbour do suggest improvement, indicating that remedial actions have been successful in reducing contaminant inputs to the harbour. However, the impact of historical contamination, as well as any impact of ongoing PCB inputs to Windermere Arm, is not insignificant, and recent (2005–2013) fish PCB levels remain above consumption advisory benchmarks in all sport fish species examined. Continued monitoring is suggested to assess fish PCB levels within the harbour in the future.

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Supplemental materials

Supplementary data for this article can be accessed on the publisher’s website.

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