Sub-Indicator: Toxic Chemicals in Sediment

The Toxic Chemicals in Sediment sub-indicator is used to report long-term trends of contaminants in sediment, rather than short-term change. The delays on the data collection and analysis of sediment in the Great Lakes due to the Covid-19 pandemic have been reduced and monitoring schedules have largely returned to normal. These delays as a result of the pandemic are not likely to affect trend analysis over the reporting timeframe. Many of the Chemicals of Mutual Concern (CMCs) are persistent in the environment and bio-accumulate in the food chain. Legacy contaminants such as Polychlorinated Biphenyls (PCBs) also have first-order half-lives significantly longer than the timeframe at which this indicator is applied (Li et al. 2009, Table 3). Given these factors, the assessments presented in the 2019 Toxic Chemicals in Sediment sub-indicator report are still valid and continue to represent the status of the Great Lakes in the 2025 report. Polycyclic Aromatic Hydrocarbons (PAHs) data for Lake Ontario (from 2018 and 2019) has been added to the 2019 State of the Great Lakes sub-indicator report.

Overall Assessment

Status: Fair

Trends:

10-Year Comparison: Unchanging

Long-term Trend (1970 – 2015): Improving

Rationale: In general, legacy contaminants that are persistent, bioaccumulative and/or toxic have decreased in Great Lakes sediment. Long-term trends for many legacy contaminants including mercury are exhibiting declines or no change. Legacy compounds including PCBs and dichloro-diphenyl-tricholoroethane (DDT) are generally below Canadian Council of Ministers of the Environment (CCME) sediment quality guideline values, while other contaminants including mercury, arsenic, polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) and polybrominated diphenyl ethers (PBDEs) exhibit some exceedances of guidelines, particularly in Lake Ontario. Emerging and new contaminants are of interest as many exhibit trends toward increasing concentrations and need to be studied further to determine acceptable limits.

Lake-by-Lake Assessment

Lake Superior

Status: Good

10-Year Comparison: Unchanging

Long-term Trend (1970 – 2015): Improving

Rationale: Lake Superior is the largest, coldest and deepest of the Great Lakes. Physical processes such as longer contaminant cycling times and lower rates of volatilization have resulted in lower rates of decrease in concentrations for some legacy contaminants, compared to the other Great Lakes. However, typical offshore deep-water sediment contaminant concentrations are very low, with atmospheric deposition as the primary source. While still exhibiting the highest toxaphene concentrations in the Great Lakes, levels have declined by an order of magnitude since their peak in the 1980s. Concentrations of copper and lead often exceed the strictest sediment quality guidelines due to the geochemistry of the watershed (pre-Cambrian shield) while historical regional sources associated with mining and smelting are the likely cause of arsenic guideline exceedances. However,

concentrations of metals in Lake Superior can be influenced by geochemistry in watersheds that represent naturally-occurring sources. While the concentration of some of the brominated flame retardants (BFRs) including decabromodiphenyl ether (BDE 209), Dechlorane 604 and decabromodiphenylethane (DBDPE) are the lowest in the Great Lakes, they are increasing in concentration with doubling times of 7-24 years, 5-38 years and 5-16 years, respectively (Guo 2015), as a result of the same physical processes influencing concentrations of legacy contaminants. There is an apparent overall reduction of PAH concentrations in surficial sediment between 2001 and present in Lake Huron, Georgian Bay, and Lake St. Clair, and an overall increase across the same time period in Lake Superior.

Lake Michigan

Status: Fair

10-Year Comparison: Unchanging

Long-term Trend (1970 – 2015): Improving

Rationale*:* Lake Michigan consists of a cold, deep and forested northern basin, and a more urbanized and agriculture-influenced southern basin. Atmospheric deposition is a primary source of most contaminants in sediments due to the lake's large surface area; however, inputs from tributaries and other local sources are also important (Lepak et al. 2015, Zhang et al. 2009, Eisenreich and Strachan 1992). Some chemicals exhibit elevated concentrations in sediment, in areas such as Green Bay, at sites on the eastern shores of the lake, and/or in the southern basin. Mercury concentrations are highest in Green Bay with higher contributions from industrial and watershed-derived sources (Lepak et al. 2015). Concentrations of some flame retardants are highest in Lake Michigan, compared to the upper Great Lakes (lower Great Lakes not assessed), with the highest levels in the southeast portion of the lake and near Sleeping Bear Dunes (Guo 2015). PCBs concentrations are declining – albeit very slowly. In Lake Michigan, halving times in sediments are between 32 and 179 years (Li et al. 2009). Other Perfluoroalkyl Compounds (PFCs) that have replaced the more well-known Perfluorooctane sulfonate (PFOS) and Perfluorooctanoic acid (PFOA) are now being found at comparable levels to PFOS and PFOA in Lake Michigan sediments (Codling et al. 2014).

Lake Huron

Status: Good

10-Year Comparison: Unchanging

Long-term Trend (1970 – 2015): Improving

Rationale: Lake Huron is similar to Lake Superior from a sediment contamination viewpoint, as the lake is large, cold and deep. There are few sources of contamination to the lake with atmospheric deposition as the primary source of most contaminants. Typical sediment contaminant concentrations are very low; however PCDD/Fs, nickel and copper concentrations are above guidelines in areas of Spanish Harbour and the Whalesback Channel due to local historical industrial/mining activity. However, concentrations of metals in Lake Huron can be influenced by geochemistry in watersheds that represent naturally-occurring sources. Arsenic concentrations are above guidelines across a third of the lake and may be increasing (Data source: ECCC). Very low sedimentation rates negatively impact natural recovery in the lake.

Lake Erie

Status: Fair

10-Year Comparison: Improving

Long-term Trend (1970 – 2015): Improving

Rationale: Lake Erie exhibits a definitive spatial gradient in contamination with decreasing concentrations from the western basin to the eastern basin, and from the southern area to the northern area of the central basin. This spatial distribution in Lake Erie is influenced by industrial activities in the watersheds of major tributaries, including the Detroit and St. Clair Rivers which, along with the Maumee River, hydrodynamically impact the southern shoreline. Sediment quality in the eastern basin continues to be classified as Good. Lake wide decreases for legacy contaminants in sediment are impressive with declines of greater than 50% for mercury, PCBs, hexachlorobenzene (HCB), DDT and lead (Table 1). Government initiatives and remedial actions have effectively diminished point sources across the Great Lakes basin. Lake Erie has the highest sedimentation rate of the Great Lakes, and as a result exhibits the largest declines in bottom sediment legacy contaminant concentrations. Mean trace metal concentrations are no longer above the CCME federal threshold effects level (TEL) for all three basins of the lake; however, some exceedances in the probable effects level (PEL) still occur in the western basin (Table 2). Lake Erie and Lake Ontario contain the most highly PAH contaminated sediment, as reflected in average sum PAH values being at least double the next highest average result from Lake Superior. Urbanized/industrialized areas of the lower lakes are significant contributors.

Lake Ontario

Status: Fair

10-Year Comparison: Improving

Long-term Trend (1970 – 2018/2019): Improving

Rationale: Lake Ontario continues to exhibit the poorest sediment quality of all the Great Lakes. The greatest frequency and magnitude of exceedances of the CCME sediment quality guidelines is for PCDD/Fs and mercury (Table 2). This legacy contamination issue is the result of historical industrial activities in the Niagara River watershed and the influence of sources in the upstream lakes resulting in contaminants ultimately accumulating in the sedimentary record of Lake Ontario. However, current levels of PCDD/Fs contamination represent a ~70-80% decline from peak levels in the 1970s. Mercury continues to have PEL exceedances in offshore depositional areas, however, declines of as much as 90% are evident lake wide. Trends in most legacy chemicals in Lake Ontario point toward improvements in sediment quality over time with levels of DDT, Dieldrin, dioxin-like PCBs and Mirex/Photomirex declining from 60-95% since their maximum accumulations. While most BFR concentrations are low, dechlorane plus, also a result of historical industrial activity in the Niagara River watershed, is several orders of magnitude higher in Lake Ontario, compared to the other Great Lakes. From data collected in 2018 and 2019, Lake Ontario contains some of the most highly contaminated sediment for PAHs of all of the Great Lakes, with Hamilton Harbour being one of the most contaminated PAH sites. Concentrations of bisphenol-A (BPA) are 5-10 times higher in Hamilton and Toronto Harbours, relative to the open lake sediments, indicating urbanized areas as primary sources and wastewater treatment plants as primary vectors (Data source: ECCC).

Status Assessment Definitions

Good: The metrics show that toxic chemical concentrations are meeting the ecosystem objectives or they are otherwise in an acceptable condition.

Fair: The metrics show that toxic chemical concentrations are not meeting the ecosystem objectives, but they are exhibiting minimally acceptable conditions.

Poor: The metrics show that toxic chemical concentrations are not displaying minimally acceptable conditions and are severely impacted.

Undetermined: Data are not available or are insufficient to assess condition of the ecosystem components.

Trend Assessment Definitions

Improving: Decrease in concentration or frequency of detection of toxic chemicals.

Unchanging: No change in the concentration or frequency of detection of toxic chemicals.

No Trend: Data are available for a 10-year or longer time period but the different groups of toxic chemicals are not trending in the same direction or display high variability in the data and an expert opinion of the overall direction of the trend cannot be determined.

Deteriorating: Increased concentration or frequency of detection of toxic chemicals.

Undetermined: Data are not available or are insufficient to assess the trends (including cases with low frequency of detection).

For the purposes of this sub-indicator report, trend is represented by a comparison of a limited number of data points over the 10-year period. This **comparison** is not representative of a statistical **trend** analysis.

Endpoints and/or Targets

The target or endpoint for this sub-indicator will have been met when the sediments of the Great Lakes are free from pollutants in quantities or concentrations that could be harmful to human health, wildlife or aquatic organisms, through direct exposure or indirect exposure through the food chain.

Status of surficial sediment will be determined by comparison with sediment quality criteria (e.g., CCME Probable Effect Level), or on a case-by-case basis when no guidelines exist. A weight-of-evidence approach will be taken in assessments, using factors including number of chemicals detected, comparison with sediment quality guidelines, or relative toxicity, if known. Status of temporal trends will be assessed using concentration profiles in sediment cores. Progress will be based on whether trends in chemicals are increasing or decreasing, the rate of change in concentrations, and the number of chemicals exhibiting changing trends.

Sub-Indicator Purpose

The purpose of this sub-indicator is to assess the concentrations of toxic chemicals in sediments throughout the Great Lakes; to infer the potential for impairment to the quality of sediment of the Great Lakes by harmful pollutants; to infer progress toward virtual elimination of Chemicals of Mutual Concern; to inform the risk assessment of toxic chemicals and the development of risk management strategies; to inform the development of environmental quality guidelines; and to report on environmental response (i.e., progress) toward the achievement of targets identified in action plans and risk management strategies for toxic chemicals in the Great Lakes basin.

Ecosystem Objective

This sub-indicator best supports work towards General Objective #4 of the 2012 Great Lakes Water Quality Agreement (GLWQA) which states that the Waters of the Great Lakes should "be free from pollutants in quantities or concentrations that could be harmful to human health, wildlife, or aquatic organisms, through direct exposure or indirect exposure through the food chain."

Methods

Each Great Lake will have a selection of chemicals assessed representing several chemical classes including Chemicals of Mutual Concern (CMCs). The chemicals that will be assessed may include hexachlorobenzene (HCB), polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethane (DDT), dioxins, lead and mercury as well as PBDEs and non-brominated flame retardants and chlorinated paraffins. The sub-indicator report will include results of monitoring and surveillance activities for toxic chemicals of current and future concern. The monitoring data will be used to inform the selection of chemicals of mutual concern for Annex 3 of the GLWQA as well as monitor to assess the progress of pollution prevention and control measures for those compounds.

Sediment sampling in the Great Lakes takes place under the Great Lakes Sediment Surveillance Program (GLSSP) coordinated by the Environmental Protection Agency (EPA) and the United States Geological Survey (USGS), and the Great Lakes Sediment Monitoring Program (GLSMP) of Environment and Climate Change Canada (ECCC). The two programs sample on alternating 5-year cycles, following the schedule of the Cooperative Science and Monitoring Initiative (CSMI). However, GLSSP sampling in Lake Michigan was suspended in 2020. Sampling took place in Lake Superior in 2021, Lake Huron in 2022, Lake Ontario in 2023 and Lake Erie in 2024. Lake Michigan sediment samples will be collected in 2025. Lake Erie was sampled by both the EPA and USGS, as well as ECCC during the 2024 season, as ECCC began their 5-year sediment sampling cycle as part of CSMI.

New data from these monitoring efforts will be included in subsequent State of the Great Lakes (SOGL) reports as samples continue to undergo analysis.

As a sub-indicator of long-term *temporal trends* the concentrations of toxic chemicals in sediment cores at selected sites within the Great Lakes will be measured. Sampling for each lake will follow the CSMI schedule. The chemicals of interest include CMCs, other chemicals of interest and emerging chemicals which may be harmful to the Great Lakes ecosystem.

The sediment concentrations would be depicted using the standard tables and figures showing the change in concentration at different depths. Temporal trends may also be depicted using estimated fluxes to sediments for each core section.

Short-term trends (10 years or less) cannot be measured in the Great Lakes using this sub-indicator. Short term (10 years or less) changes will be determined by comparing surficial sediment concentration measured over a 10-year period, or where this data does not exist, a comparison of the top two (1 cm) slices of a sediment core.

Assessment of temporal trends and concentrations of contaminants in sediment are influenced by a number of factors, including physical processes (e.g., current regime, sedimentation rate) and physical/chemical properties of chemicals (e.g., solubility, persistence). These same factors result in sediment having an overall slow response to changes in contaminant loadings; as a result, changes in concentrations may not be realized for a number of years.

As a sub-indicator of occurrence and *spatial distribution*, the concentrations of toxic chemicals in surficial sediments will be measured. Sampling will usually follow the CSMI schedule. Sampling locations will include not only the

depositional zones of the lakes, but also near shore locations. Surficial sediments may either represent the top three centimeters in Lakes Michigan, Huron, Erie and Ontario and Lake Superior; or a homogenized sample collected with a ponar.

The number of stations sampled varies by lake, based on historical stations, sedimentology and bathymetry. A set of maps showing locations and concentrations would serve to illustrate the sub-indicator and will be considered in the final version of the sub-indicator report.

Because basin-wide information on bioaccumulative contaminants is not collected every year, the sub-indicator report will be a partial update every second State of the Great Lakes reporting cycle, i.e. every 6 years.

Discussion

Status of Contaminants in Sediment

Sediments in the Great Lakes generally represent a primary sink for contaminants but can also act as a source through resuspension and subsequent redistribution. Burial in sediments also represents a primary mechanism by which contaminants are sequestered and prevented from re-entering the water column. An Environment and Climate Change Canada initiative which samples Great Lakes sediment according to the CSMI schedule enables assessment of sediment quality in the connecting channels and the Great Lakes for current and future State of the Great Lakes reports. For the purposes of this sub-indicator report, only open lake data are included in the status and trend analysis.

Comparisons of surficial sediment contaminant concentrations with sub-surface maximums indicate that contaminant concentrations have generally decreased by more than 50 percent, and, in some cases, by as much as 90 percent in the lower Great Lakes over the past four decades (Table 1).

Sediment concentrations were also be assessed against guideline values established for the protection of aquatic biota, e.g., Canadian Sediment Quality Guidelines Probable Effect Level (PEL, CCME 1999). These guidelines were applied as a screening tool in the assessment of potential risk, and for the determination of relative sediment quality concerns.

Status of Chemicals of Mutual Concern (CMCs)

The Parties to the GLWQA designated the first set of Chemicals of Mutual Concern (CMCs) in May 2016. For State of the Great Lakes reporting in 2019 and moving forward, the toxic chemical sub-indicators, where possible, will support reporting on CMCs in a more fulsome, consistent and transparent way. Information on additional chemicals of interest for the Great Lakes (non-CMCs) is valuable for inclusion in the report and will be included in a separate section below, as appropriate. For the 2025 Toxic Chemicals in Sediment sub-indicator report, the assessment is not based solely on CMCs, with additional chemical information being presented below.

The following chemicals were identified as the first set of CMCs:

- Hexabromocyclododecane (HBCD)
- Long-Chain Perfluorinated carboxylic acids (LC-PFCAs)
- Mercury (Hg)
- Perfluorooctanoic acid (PFOA)
- Perfluorooctane sulfonate (PFOS)

- Polybrominated Diphenyl Ethers (PBDEs)
- Polychlorinated Biphenyls (PCBs)
- Short-Chain Chlorinated Paraffins (SCCPs)

Mercury and Metals

The spatial distribution of mercury contamination in Great Lakes sediments generally represents those of other legacy toxic compounds, including other metals and organics such as PCBs, as accumulation of a broad range of contaminants on a lake-by-lake basis can be the result of common sources. The highest concentrations of mercury in sediments of Lakes Michigan, Erie and Ontario are observed in offshore depositional areas characterized by fine-grained sediments. Contaminant concentrations are generally correlated with particle size; hence the distribution of mercury is not only a function of loadings and proximity to sources, but of substrate type and bathymetry. Research by ECCC confirms earlier work by Marvin et al. 2004 that mercury contamination in sediment is generally lower in Lakes Huron, Michigan, Superior, and Lake St. Clair, compared with higher concentrations in Lake Ontario and the western basin of Lake Erie (Figure 1). Levels of mercury in the Great Lakes indicate that most of Lake Ontario, western Lake Erie and areas in the north of Lake Superior still exceed the PEL (Figure 1). There is a gradient in contamination in Lake Erie toward decreasing concentrations from the western basin (mean 410 ng/g) to the central basin (190 ng/g) to the eastern basin (62 ng/g) (Data source: ECCC). The spatial distribution in Lake Erie is influenced by industrial activities in the watersheds of major tributaries, including the Detroit River, and areas along the southern shoreline (Marvin et al. 2004). Sources and loadings of mercury to Lake Huron appear to have been reduced to the point that no apparent spatial pattern exists. Current sediment contamination is substantially lower than peak levels that occurred in the mid – 1950s through the early 1970s for all of the lakes with concomitant reductions in the connecting channels including the Niagara, lower Detroit and upper St. Clair Rivers, all of which are associated with historical mercury contamination as a result of chlor-alkali production. These areas were also intensively industrialized and were primary sources of a variety of persistent toxic chemicals to the open lakes, including PCBs. Studies conducted from 2012 through 2014 (Lepak et al. 2015) and 2013-2019 (Figure 1; Data source: ECCC) are consistent with earlier studies showing:

- a wide total mercury concentration range across Great Lakes sediments;
- lowest total mercury concentrations observed offshore in Lakes Huron and Superior and higher concentrations in western Lake Erie and in Lake Ontario; and
- regional increases in mercury concentration relative to those offshore in Lake Michigan (Green Bay) and Lake Superior sediment (Thunder Bay).

For metals, PEL guideline exceedances were frequent in Lake Ontario for lead, cadmium, zinc and mercury. Guideline exceedances (PEL) were infrequent in all of the other lakes, with the exception of lead in Lake Michigan where the PEL (91.3 µg/g) was exceeded at over half of the sites, and arsenic in Lakes Huron and Ontario (Table 2).

PCBs

PCB results from Li et al. (2009), conducted during a similar time period to the study by Burniston et al. (2011), found a 30% reduction in PCB concentration across the Great Lakes, compared to results from Eisenreich (1987), with the greatest decrease occurring in Lake Ontario. The comparison of PCB totals to historical studies is confounded by changes in analytical methodology. In this assessment, comparing surficial sediment with sub-surface maxima using similar analytical techniques may provide more reliable results. Reductions for PCBs across the Great Lakes, comparing lakewide average of surficial sediment with sub-surface maxima, ranged from 5% in Lake Michigan to 94% in Lake Ontario. For PCBs, while decreased production contributes to this reduction, the

decreased concentrations may also be the result of the loss of light congeners due to repeated resuspension of surficial sediment, desorption of light congeners and subsequent evaporation (in Lake Michigan; (Li et al. 2009)) or by anaerobic reductive dechlorination (in Lake Ontario; (Li et al. 2009)). Because of differences in toxicity between congeners, the latter process could reduce the toxicity of the PCBs (Li et al. 2009). First order half-lives ($t_{1/2}$) vary from 44.9 years (Lake Huron) to 9.7 years (Lake Superior), see Table 3, with shorter half-lives found in lakes with sites closer to tributary sources (Ontario, Erie, Superior) and thus more responsive to PCB source reductions (Li et al. 2009). While PEL (277 ng/g total PCBs) guideline exceedances for PCBs are currently rare in any of the Great Lakes sediments, levels still remain elevated in areas of the St. Clair-Detroit River Ecosystem.

Polycyclic Aromatic Hydrocarbons (PAHs)

Sediment monitoring for PAHs (Polycyclic Aromatic Hydrocarbons) was conducted between 2014 and 2019 in the Great Lakes. Surface sediment and sediment core samples were collected from 290 stations in Lake Superior, Lake Huron, Georgian Bay, Lake St. Clair, Lake Erie, Lake Ontario, Hamilton Harbour, and the St. Marys and St. Clair Rivers. Confidence intervals of yearly PAH means were calculated using the Regression on Order Statistics (ROS) function from the NADA package in R (Helsel, 2012). This function allowed for the incorporation of censored data into calculations of summary statistics, thus avoiding bias in analysis arising from the removal, or substitution of values in the place of results below detection limit (i.e., censored data). Confidence intervals for each available parameter were compared to sediment guidelines for PAHs published by the Canadian Council of Ministers of the Environment (CCME, 1999), where available. The CCME has published two sets of guidelines: the Interim Sediment Quality Guidelines (ISQGs), and the more stringent Probable Effect Levels (PELs). PELs are the concentration of PAHs in sediment where adverse environmental effects are nearly always observed. ISQGs, based on toxicological data, are the threshold PAH concentrations in sediment where adverse effects to aquatic biota are almost never observed (Marvin, 2021). Of the four Great Lakes included in this assessment, Lake Erie and Lake Ontario contain the most highly PAH contaminated sediment, as reflected in average sum PAH values being at least double to the next highest average result from Lake Superior. Higher levels of PAH contamination in the lower Great Lakes could be a result of legacy industrial activity, and dense urbanization resulting in greater vehicular traffic, and proliferation of impervious surfaces surrounding these water bodies that promote PAH transport through runoff (Marvin, 2021).

Results were compared with historical analyses, including assessments of PAHs in surface sediments of Lake Superior, Lake Huron and Georgian Bay from 2001/2002 (Gewurtz et al., 2008), and Lake St. Clair from 2001 (Gewurtz et al., 2007). For Lake Superior, Lake Huron, and Georgian Bay, results for concentrations of individual PAHs differ significantly between 2001/2002 and 2016/2017. Maximum individual PAH values from 2016/2017 were found to be generally higher for Lake Superior, and lower for Lake Huron and Georgian Bay. However, the overall pattern of ISQG and PEL guideline exceedances between the two studies is roughly complementary. Gewurtz et al. (2008) reported no PEL guideline exceedances, and in this assessment, only a single PEL exceedance for phenanthrene was observed in the port of Duluth. In Lake St. Clair, maximum individual PAH values were significantly higher for each parameter measured in 2001 by Gewurtz et al. (2007) compared to the 2014 study. Results suggest an overall reduction of PAH concentrations in surficial sediment between 2001 and present in Lake Huron, Georgian Bay, and Lake St. Clair, and an overall increase across the same time period in Lake Superior.

Figure 5 illustrates the significant differences in EPA16 PAH means between certain Great Lakes Areas of Concern and their surrounding regions. The proportion of individual samples exceeding each of the relevant PELs and ISQGs (represented graphically for ISQGs and PELs) are included in **Figures 6 and 7,** respectively. The highest PAH concentrations are observed in Lake Superior's port of Duluth (mouth of the St. Louis River), the St. Marys, St. Clair and Detroit Rivers, Lake Erie's port of Cleveland (mouth of the Cuyahoga River), and Lake

Ontario's Hamilton Harbour. Lake Erie and Lake Ontario exhibit open lake PAH concentrations of a higher range than the other Great Lakes, with some exceedances of the ISQGs, and limited exceedances of the PEL in Lakes Superior and Erie, see **Figures 6 & 7**. As suggested by Marvin et al. (2021), increased levels of PAHs in the open water environments of the lower Great Lakes could be a result of deposition from densely urbanized regions immediately adjacent.

Flame Retardants (FR)

Flame retardants (FRs) are heavily used globally in the manufacturing of a wide range of consumer products and building materials. FRs have been found to be bioaccumulating in Great Lakes fish and in breast milk of North American women (Schecter et. al. 2003). While industrial discharges may not be responsible for ongoing contamination, modern urban/industrial centers can act as diffuse sources. Studies of sediment core profiles of PBDEs in Lake Ontario suggest accumulation of these chemicals peaked, or continues to increase (Marvin et al. 2007; Shen et al. 2010). The Lake Ontario total PBDE profile indicates a leveling off of accumulation in the past decade, presumably as a result of voluntary cessation of production of these compounds in North America. Core studies (Data source: ECCC) indicate total PBDE surficial sediment maximums (lakewide average 3.4 ng/g) in Lake Superior, while in Lake Huron, levels have started to decline (Figure 2). Surficial sediment concentrations in Lake Huron are low and range from 0.1-14 ng/g and <0.1-12 ng/g for total PBDEs and BDE 209, respectively. While other contemporary studies (Guo, 2015; Zhu and Hites, 2005) have shown total PBDEs, and in particular the deca-substituted BDE 209, are continuing to increase across all five Great Lakes with doubling times ranging from 4 years to 74 years. BDE209 was produced in the U.S. as late as 2014, but still remains in many products and is the predominant congener in sediment, accounting for over 90% of total PBDEs. This is of concern because BDE209 can degrade in biota and sediment to less substituted BDEs that are more toxic and more bioaccumulative (Gauthier et al, 2008). A study of the upper lakes by Guo (2015) found the highest surficial concentrations for both total PBDE and BDE209 concentrations were in Lake Michigan (especially southeast and Sleeping Bear Dunes), and Lake Huron (especially Saginaw Bay and North Channel), and were comparable to Lake Erie concentrations, but lower than Lake Ontario.

Other FRs such as dechlorane plus (*anti* and *syn*) and related compounds Dec604 and Dec602 are found at low levels throughout the upper Great Lakes, but are more elevated in Lake Erie and an order of magnitude higher in Lake Ontario (Figure 3.). Most FRs increased significantly after 1920 and have leveled off or decreased since 2000, but Dec604 and DBDPE may still be increasing in Lake Ontario. For the upper Great Lakes, PBDEs and 1,2-Bis(2,4,6-tribromophenoxy) ethane (BTBPE) dominate in both southern and northern Lake Michigan, especially the southeast portion of the lake and the sites near Sleeping Bear Dune. Despite these trends, maximum concentrations of many FRs remain well below maximum concentrations of contaminants such as DDT and PCBs observed historically. In the lower Great Lakes, levels have shown a leveling off in recent years (Letcher et al., 2015, Lu et al., 2015). An ECCC study detected few halogenated flame retardants in Lake Erie (n=10), including HBCD, even though detections of 2-Ethylhexyl-2,3,4,5-tetrabromobenzoate (EHTBB) and bis(2-ethyl-1-hexyl)tetrabromophthalate (BEHTBP) were frequent, albeit low, in the Detroit River (n=12).

Perfluoroalkyl Compounds (PFCs)

Perfluoroalkyl Compounds (PFCs) have attracted scientific and regulatory interest in recent years as a result of their detection globally in humans and wildlife. They are routinely detected in precipitation and air in urban and rural environments. These compounds have a myriad of applications but have been primarily used as soil and liquid repellents for papers, textiles and carpeting. Two classes of PFCs, the perfluoroalkyl sulfonate acids (PFSAs), particularly perfluorooctane sulfonate (PFOS), and the perfluorocarboxylates (PFCAs), particularly

perfluorooctanoic acid (PFOA), are the most commonly measured PFCs in sediment; these compounds are highly stable and persistent in the environment, and are potentially toxic. In surficial sediments, concentrations of perfluorobutane sulfonate (PFBS) and perfluoro-n-butanoic acid (PFBA) are now occurring at concentrations comparable to those of the PFCs which they replaced (PFOS and PFOA) (Codling et al. 2014). PFCs have been detected in environmental samples in remote areas such as the Canadian Arctic. The physical and chemical properties of PFCs are different from many other semi-volatile pollutants as they have both hydrophilic and hydrophobic properties. While persistent and bioaccumulative, PFCs are transported in both the aqueous and non-aqueous phase. As well, PFCs in bottom sediment may diffuse to the surface and become bioavailable.

There is a gradient toward increasing PFC contamination from the upper Great Lakes (Superior and Huron) to the lower Great Lakes (Erie and Ontario). This spatial trend is reflected in the concentrations of PFOS in surficial sediment, which range from 0.24 – 6.26 ng/g and 0.19 – 5.29 ng/g for Lakes Superior and Huron, respectively, to 0.66 – 15.3 ng/g and 0.65 – 46.0 ng/g for Lakes Erie and Ontario, respectively (Data Source, ECCC). When concentrations of PFCs in Lake St. Clair were compared with sediments (Marvin et al., 2004) and fishes (McGoldrick and Murphy, 2016) in other Great Lakes, concentrations of PFCs in Lakes Ontario and Erie were greater. Concentrations of PFCs in sediments are influenced not only by proximity to sources, but physical processes and bathymetry. The watersheds of Superior and Huron are less densely populated, and PFC concentrations are typically less. Most locations have concentrations that indicate non-point source contamination; however, concentrations of PFCs in sediments at a few sites are greater, which indicates influences from more local sources. In Lake Superior, the deeper sediment typically contained lesser concentrations of PFCs. Given the longer residence time of water in Lake Superior, there might be some breakdown or uptake of these compounds before reaching the sediment layer.

Concentrations of surface sediment are dominated by PFBA and perfluoro-n-hexanoic acid (PFHxA), indicating these compounds might not be bound to the solid phase of sediments. Concentrations of PFBA in core samples were greater in deeper sediment than would have been predicted based on how much was being manufactured and used. Concentrations indicate contamination may enter the food chain since they are not bound to sediment particles (Codling et al, 2018 b). In general, PFC concentrations have increased over time, which largely corresponds to increased use. However, PFCs that have been subjected to targeted action have seen declines in concentrations in sediment. For example, concentrations of PFOS in Lake Ontario peaked in 2001 (Myers et al., 2012). PFBA and PFHxA were frequently detected in surface sediment and upper core samples indicating a shift in use patterns. Distributions of PFCs within dated cores largely corresponded with increase in use, but with physiochemical characteristics also affecting distribution. PFCs with chain lengths >7 that include perfluoro-n-octane sulfonate (PFOS) bind more strongly to sediment, which resulted in more accurate analyses of temporal trends. Shorter-chain PFCs, such as PFBA, which is the primary replacement for C8 compounds that were phased out, are more soluble and were identified in some core layers at depths corresponding to pre-production periods, which indicates movement through the sediment cores. Thus, analyses of temporal trends of these more soluble compounds in cores of sediments are less accurate. Based upon their physical-chemical properties, temporal analysis in sediment cores is challenging (Codling et al, 2018 a). Research and monitoring programs continue to collect and analyze data for a suite of perfluorinated substances in various media. Lake Michigan sediment data will be included in future reporting as samples are analyzed by the U.S. EPA GLSSP.

Linkages

Sediment contamination affects both water quality and aquatic dependent life. Sediment can be a source of mercury and other toxic chemicals to enter the water column. These chemicals are components of the Toxic Chemicals and the Habitat and Species indicators including “Toxic Chemicals in Water” and “Toxic Chemicals in the

Atmosphere".

Linkages to other sub-indicators in the indicator suite include:

- Benthos – sediments serve as habitat for many benthic species and communities. However, sediments also serve as a reservoir for bioaccumulation and trophic transfer of contaminants, resulting in potential effects to the bottom-dwelling communities as well as implications for fish and other species in the food web.

Assessing Data Quality

Data Characteristics	Agree	Neutral or Unknown	Disagree	Not Applicable
Data are documented, validated, or quality-assured by a recognized agency or organization	X			
Data are from a known, reliable and respected generator of data and are traceable to original sources	X			
Geographic coverage and scale of data are appropriate to the Great Lakes Basin	X			
Data obtained from sources within the U.S. are comparable to those from Canada	X			
Uncertainty and variability in the data are documented and within acceptable limits for this sub-indicator report	X			
Data used in assessment are openly available and accessible	Yes	Great Lakes Sediment Monitoring and Surveillance Data - Open Government Portal (canada.ca) Great Lakes Sediment Archive Database (1960-1975) - Open Government Portal (canada.ca)		

Data Limitations

Sediment cores reflect a long-term temporal record of contaminants in the Great Lakes. Relatively low sedimentation rates mean that short time-frames between core collections will show relatively little change in this record, and thus provide limited benefits to 3-year updates. Sampling on the Great Lakes is conducted on the CSMI cycle in partnership with the USA, and this means that cores are typically collected every ten years for a given lake.

However, the updates of the sub-indicator when new information arises are applicable to past years (i.e., sediment cores will fill in the history for the previous decade). The thinnest increments in sediment cores from lakes with low sedimentation rates (e.g., Lake Superior) do not afford adequate temporal resolution in order to determine year-to-year changes in contaminant concentrations.

Additional Information

This sub-indicator will track whether concentrations of the toxic chemicals are, as a group, decreasing, staying the same, or increasing in Great Lake sediment and waters over time. The sub-indicator data will also demonstrate the magnitudes of the trends of the various chemicals. The magnitudes of the trends are expressed as halving time, or time to which the concentration of the chemical is decreased by a factor of two. The time which is most relevant to virtual elimination is the longest halving time of the measured chemicals.

Assessment of nearshore sediment quality will be done using surficial sediment collected every 5-10 years from sites either previously monitored for contaminants in sediment, water and/or fish, or determined by resource managers to be a high priority for surficial sediment information (e.g. tributary input). Sites would also be chosen based on sediment type, expected sedimentation rates, and proximity to potential sources. Cores would be sectioned, dated and analyzed for the toxic chemicals.

Efforts to control inputs of historical contaminants have resulted in decreasing contaminant concentrations in the Great Lakes open-water sediments for many of the legacy chemicals. However, chemicals such as FRs and current-use pesticides may represent emerging issues and potential future stressors to the ecosystem. These results corroborate observations made globally, which indicate that large urban centers act as diffuse sources of chemicals that are heavily used to support our modern societal lifestyle.

Long-term research and monitoring programs are valuable tools for demonstrating effectiveness of remedial actions and management initiatives, as well as acting as indicators of emerging issues. Enhanced Canadian Great Lakes studies now include the regular sampling of sediment to be collected following the CSMI schedule. The Great Lakes Sediment Surveillance Program is a complimentary program in the U.S. Comparison of contaminant results between studies and across lakes is currently difficult because of differences in sampling designs, sampling locations, and analytical procedures. Changes in contaminant deposition cannot be detected over time frames less than the temporal resolution of the surficial sediment samples, which can be from 3 to 220 years.

The selection of Chemicals of Mutual Concern (CMCs) has been completed by the Parties to the GLWQA. These identified CMCs and any additional CMCs added in the future, will be included in future reporting where possible in the sub-indicators included in the Toxic Chemicals Indicator category of the Great Lakes indicator suite.

Report Authors

Chris Marvin, Aquatic Contaminants Research Division, Environment and Climate Change Canada, 867 Lakeshore Rd., Burlington, ON L7S 1A1

Tim Pascoe, Water Quality Monitoring and Surveillance; Environment and Climate Change Canada, 867 Lakeshore Rd., Burlington, ON L7S 1A1

Brian Lenell, U.S. Environmental Protection Agency, Great Lakes National Program Office, Chicago, IL

Tyler Lake, Water Quality Monitoring and Surveillance; Environment and Climate Change Canada, 867 Lakeshore Rd., Burlington, ON L7S 1A1

Debbie Burniston, Water Quality Monitoring and Surveillance; Environment and Climate Change Canada, 867 Lakeshore Rd., Burlington, ON L7S 1A1

Meg Corcoran, Oak Ridge Institute for Science and Education / Research Fellow appointed to the U.S. Environmental Protection Agency, Great Lakes National Program Office, Chicago, IL

Todd Nettesheim, U.S. Environmental Protection Agency, Great Lakes National Program Office, Chicago, IL

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Last Updated

State of the Great Lakes 2025 Report

Table 1. Estimated percentage declines in sediment contamination in the Great Lakes (1970 – 2015) based on comparison of surface sediment concentrations with sub surface maximum concentration.

Source: Environment and Climate Change Canada; Li (2006); Marvin (2004)

	Lake Superior	Lake Huron	Lake Michigan	Lake Erie	Lake Ontario
Mercury	4.5	11	49	60	94
PCBs	45	9	5	51	85
PCDD/Fs	NA	NA	NA	NA	53
HCB	NA	NA	NA	78	40
DDT	NA	93	NA	60	60
Lead	10	43	NA	71	65

Table 2. Occurrence of lead, arsenic and mercury in Great Lakes sediments in comparison to CCME guideline values. Sites with no sediment assigned "0" for all contaminants. TEL = Threshold Effect Level, PEL = Probable Effect Level, LEL = Lethal Effect Level, SEL = Severe Effect Level.

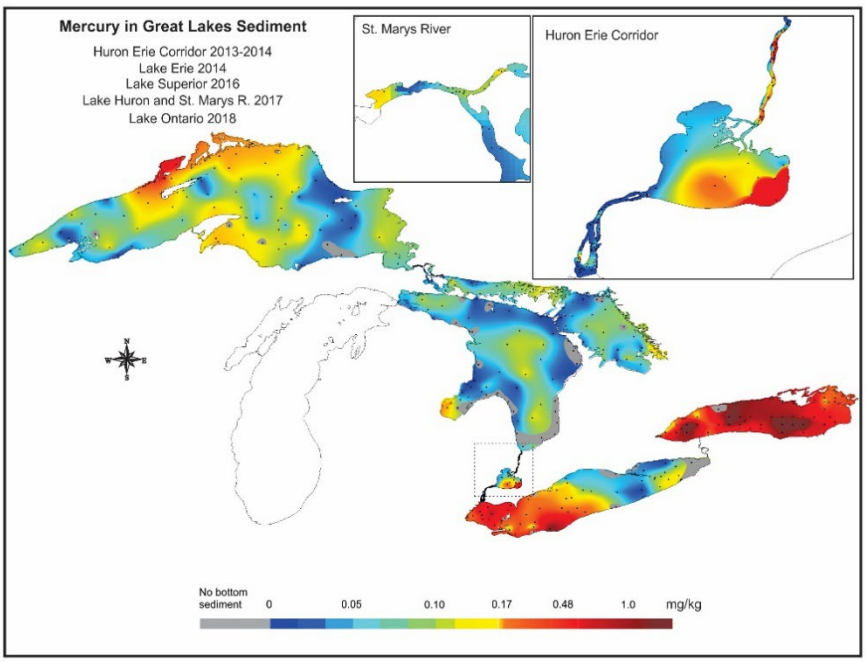
Data Source: ECCC.

% of sites above guideline. Guideline values expressed as µg/g		**Lead**				**Arsenic**				**Mercury**			
	N	TEL (35)	PEL (91.3)	LEL (31)	SEL (250)	TEL (5.9)	PEL (17)	LEL (6)	SEL (33)	TEL (0.17)	PEL (0.486)	LEL (0.2)	SEL (2)
Lake Superior 2016	76	53	6.6	57	0	73.7	16	72	2.6	12	0	3.95	0
Lake Huron 2017	93	42	6.5	44	0	59	28	56	5.4	0	0	0	0
Lake Erie 2014	54	48	1.9	50	0	76	3.7	65	0	35	11.1	31	0
Lake Ontario 2018	42	95	45	100	0	100	71	100	4.8	86	57	86	0

Table 3. First order half-life (t1/2) of PCBs in sediments of the Great Lakes[a] . [a] The first order $t_{1/2}$ values at other sampling locations cannot be obtained due to insufficient numbers (<3) of data points (SU08, SU12, SU16, HU38, HU48) or severe sediment mixing (ER09).[b] The top segment was excluded in $t_{1/2}$ calculation at these sites.

Source: Li et al. 2009

Lake	Location	Peak year	Half-life ($t_{1/2}$), Years
Superior	SU22	1993	9.7±7.9
Michigan	LM41[b]	1979	31.7±14.3
Huron	HU12[b]	1981	44.9±1.0
Erie	ER37	1981	16.6±2.2
Ontario	ON-30	1973	11.0±1.0
Ontario	ON-40	1963	17.0±4.4



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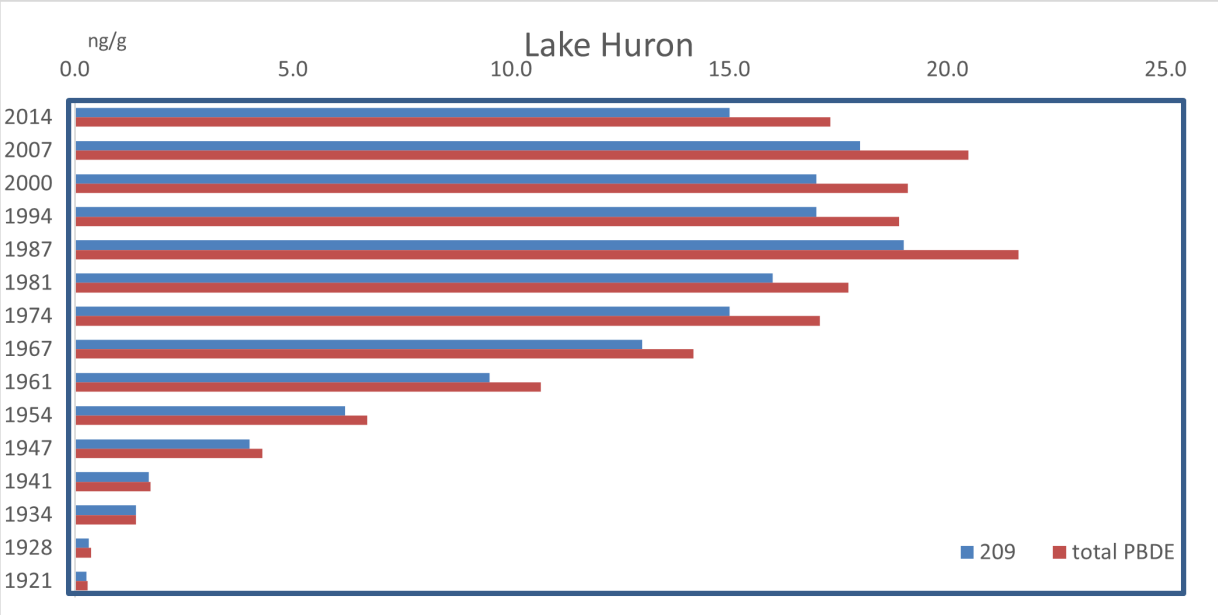
Source: Environment and Climate Change Canada



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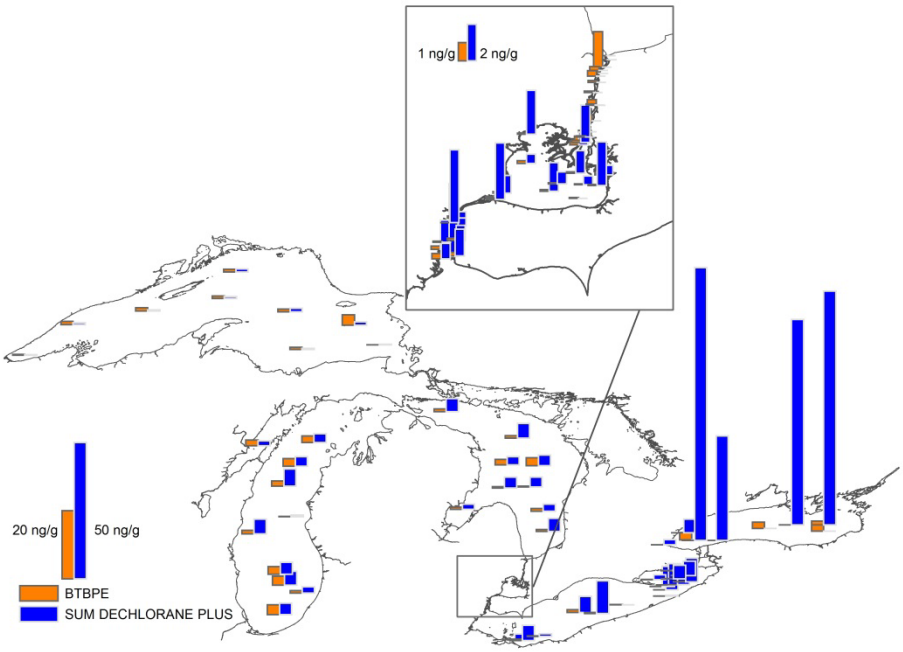

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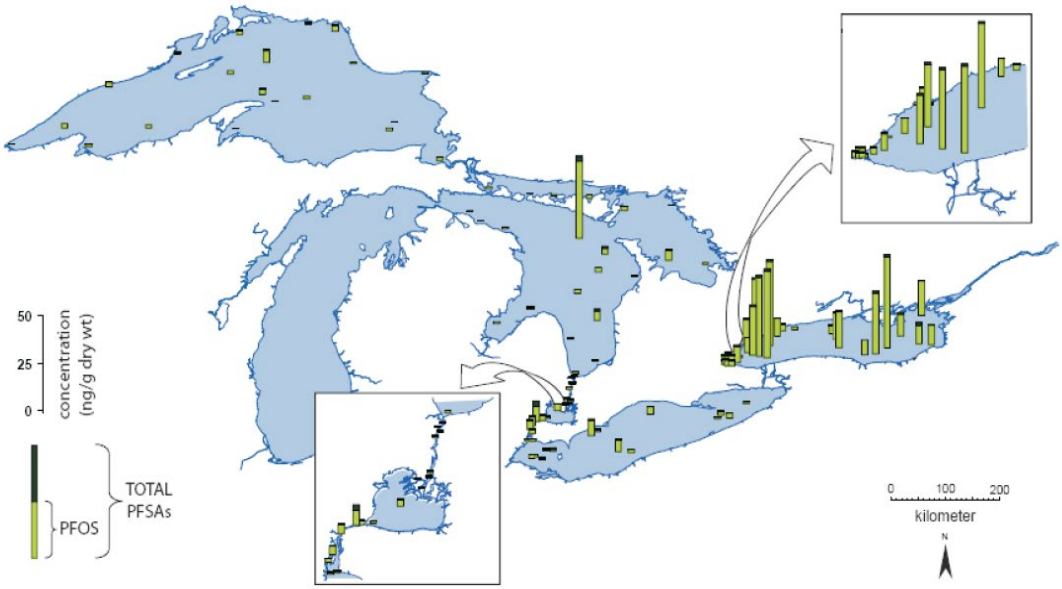

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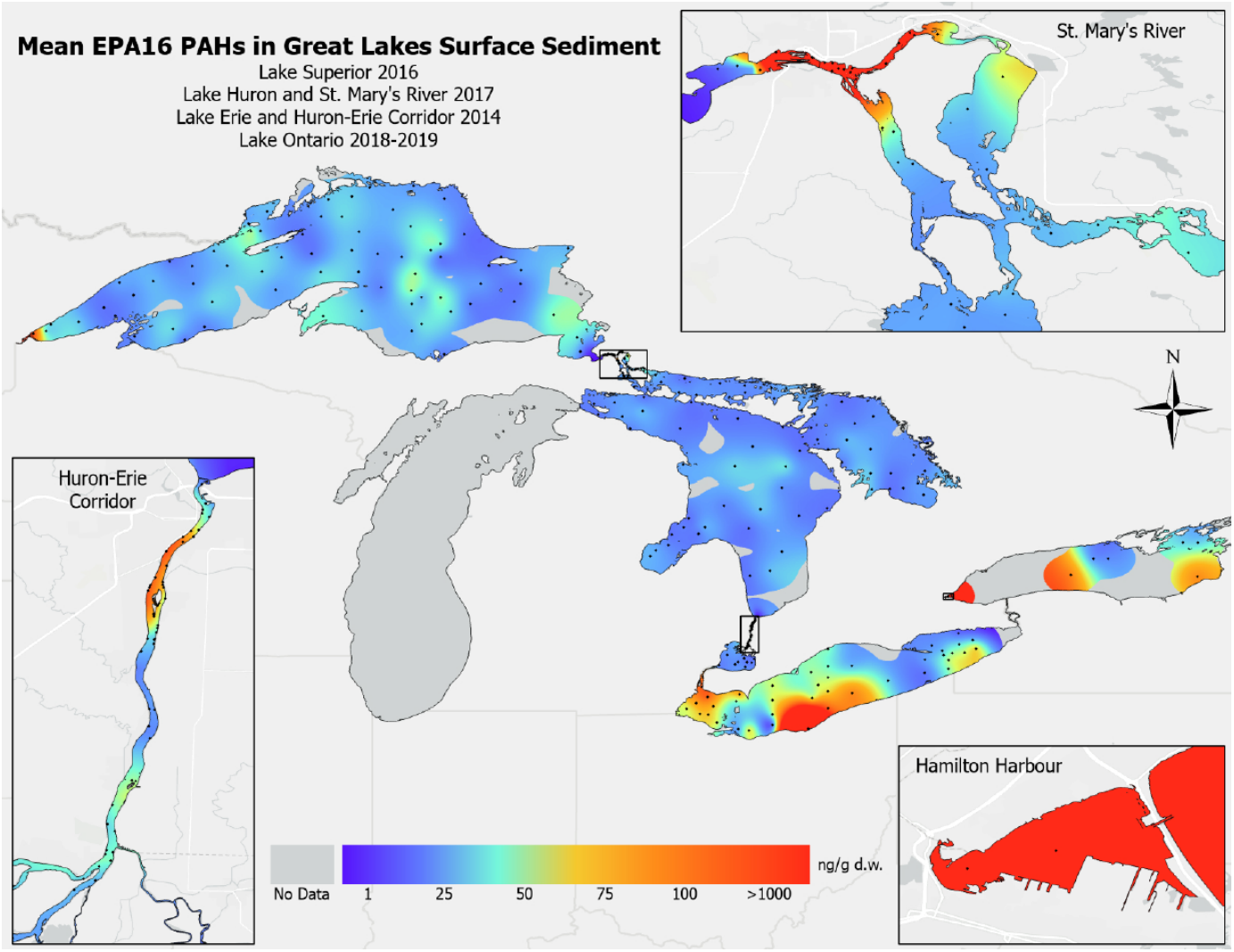

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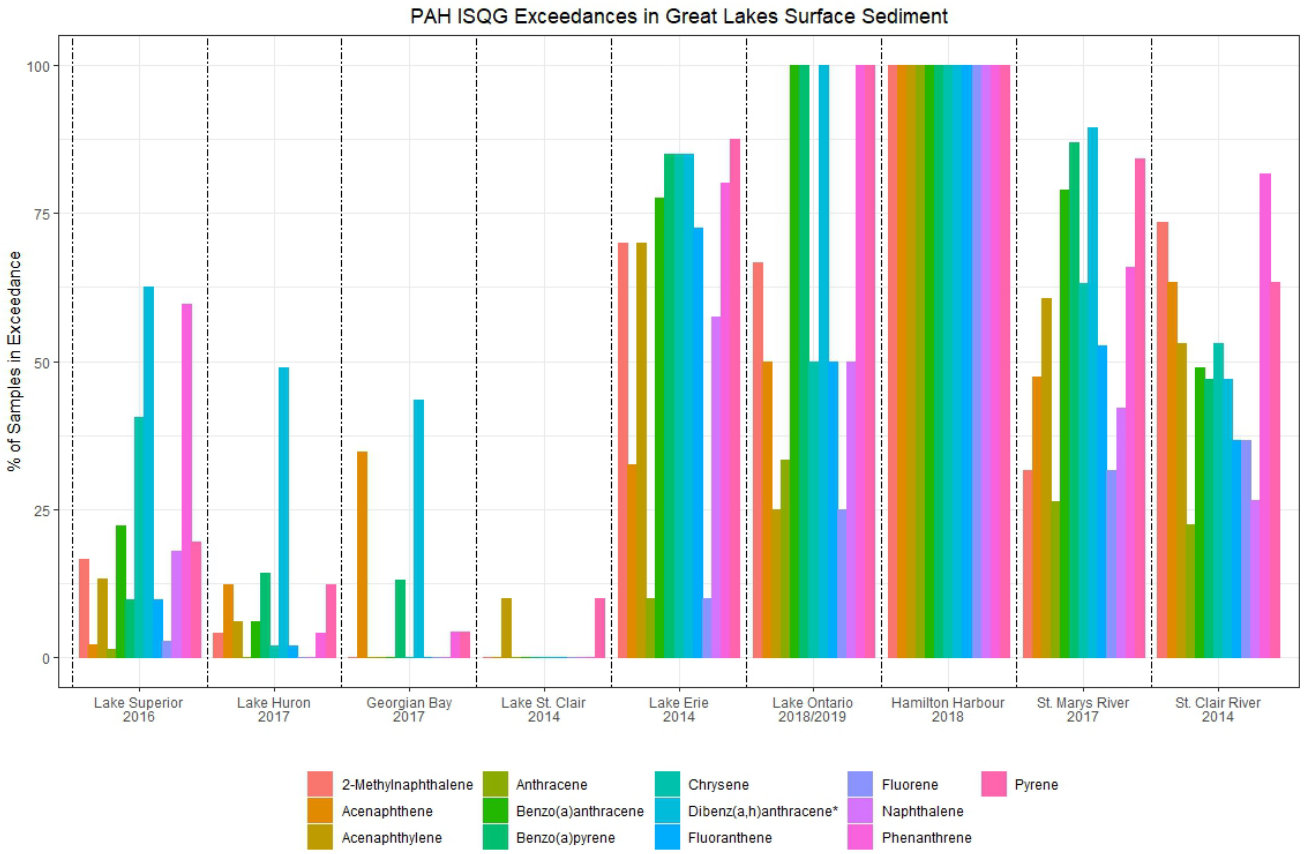

Figure 6. PAH ISQG Exceedances in Great Lakes surface sediment

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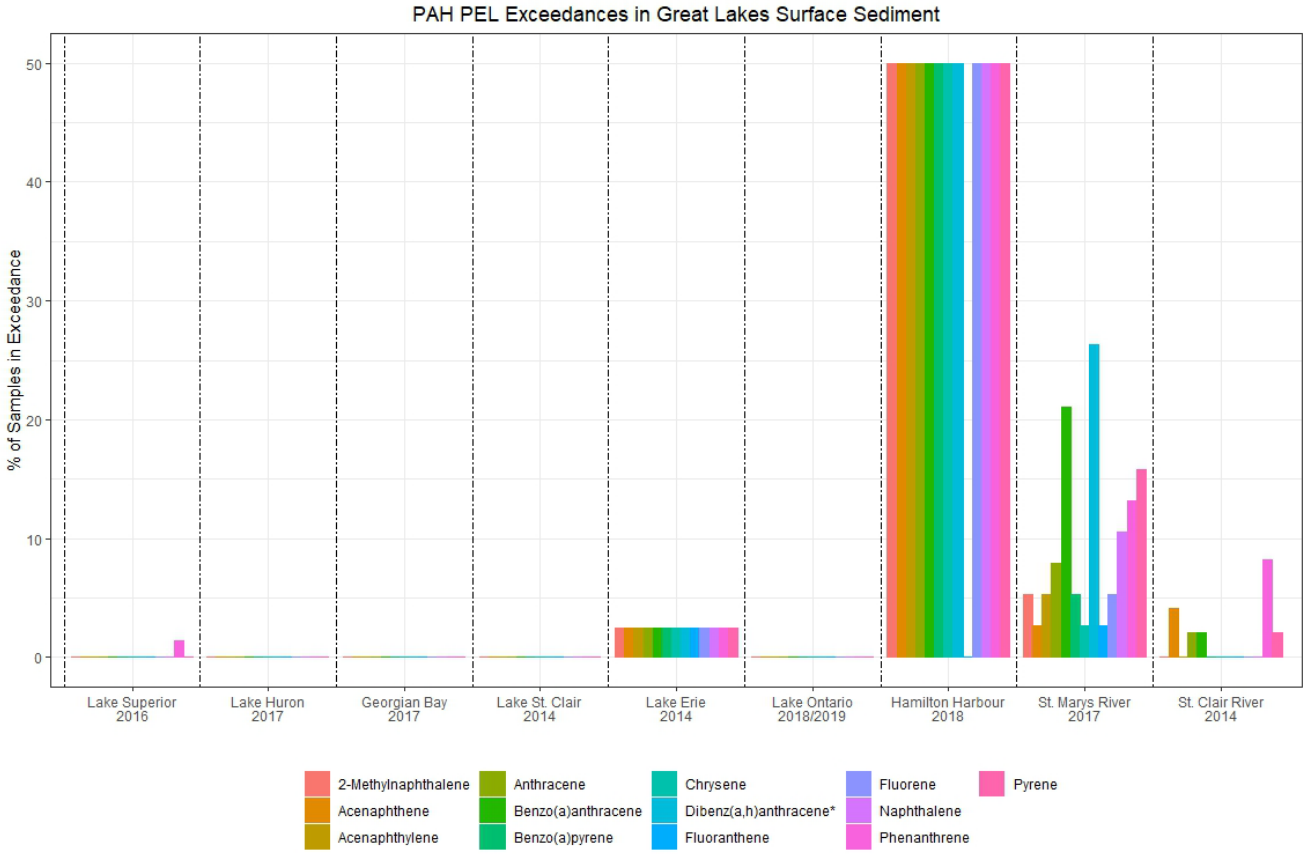

Figure 7. PAH PEL Exceedances in Great Lakes surface sediment

Data Source: Water Quality Monitoring and Surveillance Analysis 2024 for Draft PME Report on PAHs, ECCC