Sub-Indicator: Toxic Chemicals in Whole Fish

Overall Assessment

Status: Fair

Trends:

10-Year Trend (2011 - 2020): No Trend

Long-term Trend (1977 – 2020): Improving

Rationale: While concentrations of some chemicals of mutual concern (CMCs) continue to exceed environmental guidelines or targets, on average the levels of CMCs are near their guidelines. The average Mean Deviation Ratio (MDR) for CMCs in whole fish remains in the yellow zone of the MDR chart, meaning the MDR is not significantly greater or less than zero and the overall status of the Great Lakes Basin is assessed as Fair (Figure 1). The MDR is a unit-less composite measure used to identify if contaminant concentrations in fish exceed guidelines and is utilized as an indicator of the state of toxic chemicals in whole fish. The calculation of MDR considers concentrations of Tetra-, Penta-, and Hexa- brominated diphenyl ethers (TeBDE, PeBDE, HxBDE), total mercury (Hg), total polychlorinated biphenyls (Total PCBs), hexabromocyclododecane (HBCD), and perfluorooctane sulphonate (PFOS). Long term conditions have IMPROVED since monitoring began in 1977 due mostly to declines in PCBs, Hg, and Polybrominated Diphenyl Ethers (PBDEs). A statistically significant trend in the MDR is not present over the last 10-years of monitoring, indicating that the condition has remained UNCHANGED. Concentrations of PCBs, PBDEs, and PFOS still remain above environmental guidelines in all five Great Lakes. Because of the shift to focus on CMCs, this sub-indicator will no longer report out on legacy pollutants that are not CMCs (such as dichlorodiphenyltrichloroethane (DDT)). Through the Annex 3 process, new chemicals designated as CMCs will be further incorporated into monitoring and surveillance programs and reported via this sub-indicator report.

Lake-by-Lake Assessment

The COVID 19 pandemic in 2020 had a negative impact on the availability of contaminant data from the Canadian waters of the Great Lakes as normally collected by Environment and Climate Change Canada (ECCC). Due to operational restrictions, ECCC was not able to collect fish for contaminants analysis in the Great Lakes in 2020, creating a data gap for many of the Canadian sites on the Great Lakes. This data gap on the Canadian side continued at many sites in 2021 and the lack of data will be reflected in future sub-indicator reports. This data gap may have had an impact on the status indicator assessment for Lakes Erie, Ontario, Huron and Superior in 2020, as the Canadian sites on the north shore tend to be more removed from point sources of contaminants. A lack of Canadian data may make the indicator change.

Lake Superior

Status: Fair

10-Year Trend (2011 - 2020): No Trend

Long-term Trend (1977 – 2020): Improving

Rationale: In Lake Superior, the MDR indicates that on average the concentrations of CMCs are below targets or guidelines. While this is true for mercury, TeBDE, HxBDE and HBCD, concentrations of the remaining CMCs used in this calculation (PCBs, PeBDE, and PFOS) are still above environmental guidelines or targets (Table 1) in Lake

Superior. MDR values for Lake Superior fall within the yellow zone of the MDR chart indicating that the MDR is not significantly different from zero and that overall conditions remain as FAIR (Figure 2). Despite the increase in the MDR in ~2000, which was a result of the introduction of PBDEs into our monitoring programs, conditions have IMPROVED since monitoring began in 1977. A statistically significant trend was not present in the last 10-years of monitoring indicating that the condition has remained UNCHANGED.

Lake Michigan

Status: Fair

10-Year Trend (2011 - 2020): No Trend

Long-term Trend (1977 – 2020): Improving

Rationale: In Lake Michigan, the MDR indicates that on average the concentrations of CMCs are above environmental targets or guidelines (Table 1). Concentrations of mercury, TeBDE, and HBCD are below environmental guidelines or targets while those of PCBs, PeBDE, and PFOS are above guidelines or targets. The MDR values for Lake Michigan fall within the yellow zone of the MDR chart meaning that the MDR is not significantly different from zero and that overall conditions are FAIR (Figure 3). Since monitoring began, conditions have IMPROVED in Lake Michigan due to the continued declines of PCBs as well as Te- and PeBDEs. A statistically significant trend was not present in the last 10-years of monitoring indicating that the condition has remained UNCHANGED.

Lake Huron

Status: Fair

10-Year Trend (2011 - 2020): No Trend

Long-term Trend (1977 – 2020): Improving

Rationale: In Lake Huron, the MDR indicates that on average the concentrations of CMCs are below environmental targets or guidelines (Table 1). Mercury, TeBDE, HxBDE and HBCD concentrations are below guidelines or targets while those of PCBs, PeBDE, and PFOS were above environmental guidelines or targets. The MDR values for Lake Huron fall within the yellow zone of the MDR chart indicating that the MDR is not significantly different from zero and that overall conditions are FAIR (Figure 4). Since monitoring began, conditions have IMPROVED in Lake Huron due largely to declines of PCBs and TeBDE. A statistically significant trend was not present for the MDR in the last 10-years of monitoring indicating that the condition has remained UNCHANGED.

Lake Erie

Status: Good

10-Year Trend (2011 - 2020): Improving

Long-term Trend (1977 – 2020): Improving

Rationale: In Lake Erie, the MDR indicates that on average the concentrations of CMCs are below environmental targets or guidelines (Table 1). Mercury, TeBDE, HxBDE, and HBCD concentrations are below guidelines or targets while those of PCBs, PeBDE, and PFOS were above environmental guidelines or target. The MDR values for Lake Erie fall within the green zone of the MDR chart indicating that the MDR is significantly different from zero and that overall conditions are GOOD (Figure 5). Since monitoring began, conditions have IMPROVED in Lake Erie due to drop in some CMCs in recent years. A statistically significant trend was present for the MDR in the last 10-years of monitoring indicating that the condition has changed to IMPROVING. However, data from 2019 and 2020 is limited

by a reduction in sample size due to restrictions on field activities for the Canadian program. It is possible that this smaller sample size is influencing the lake-wide trend. Further observation of this trend is warranted as future data is collected.

Lake Ontario

Status: Fair

10-Year Trend (2011 - 2020): No Trend

Long-term Trend (1977 – 2020): Improving

Rationale: In Lake Ontario, the MDR indicates that on average the concentrations of CMCs are higher than environmental targets or guidelines. PCBs, PeBDE, HxBDE, and PFOS concentrations are above guidelines or targets while those for mercury, TeBDE, and HBCD were below environmental guidelines or targets. The MDR values for Lake Ontario fall within the yellow zone of the MDR chart indicating that the MDR is not significantly different from zero and that overall conditions are FAIR (Figure 6). Since monitoring began, conditions have IMPROVED in Lake Ontario due largely to declines of PCBs, mercury and PBDEs. A statistically significant trend was not present for the MDR in the last 10-years of monitoring indicating that the condition has remained UNCHANGED.

Status Assessment Definitions

Good: The MDR shows that, on average, levels of CMCs in fish are below ecosystem objectives or targets (Table 1) with 95% confidence or they are otherwise in an acceptable condition, in the green zone of the MDR chart.

Fair: The MDR shows that, on average, levels of CMCs in fish are at or near, but not significantly above or below, ecosystem objectives or targets (Table 1) with 95% confidence, in the yellow zone of the MDR chart

Poor: The MDRs show that on average, levels of CMCs in fish are above ecosystem objective or targets (Table 1) with 95% confidence, in the red zone of the MDR chart.

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

Trend Assessment Definitions

Improving: Concentrations of contaminants are declining with statistical significance according to the Mann-Kendall test

Unchanging: Concentrations of contaminants are not declining or increasing with statistical significance.

No Trend: Data are available but the trend is not statistically improving or deteriorating due to high variability in the data.

Deteriorating: Concentrations of contaminants are increasing with statistical significance

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

Endpoints and/or Targets

The purpose of this sub-indicator is to identify the presence of CMCs and new and emerging chemicals of concern

in Great Lakes whole fish and provide an interpretation and an explanation of what the presence of those chemicals means to the health of the Great Lakes ecosystem. In the absence of binational targets in the 2012 Great Lakes Water Quality Agreement (GLWQA), contaminant concentrations will be compared to environmental quality guidelines (ex. Canadian Federal Environmental Quality Guidelines - FEQGs) or other published ecotoxicological thresholds. The criteria used in this report are provided in Table 1. These criteria were selected by the authors based on availability, status (draft vs. final), appropriateness, and best professional judgement to be most protective of the Great Lakes ecosystem. Criteria can be draft or interim for a while in the absence of a decision or concurrence or an alternative.

Sub-Indicator Purpose

The purpose of this sub-indicator is to describe temporal and spatial trends of bioavailable contaminants in representative open water fish species from throughout the Great Lakes; to infer the effectiveness of remedial actions related to the management of critical pollutants; and to identify the nature and describe the trends of new and emerging chemicals of concern. The Toxic Chemicals in Whole Fish sub-indicator includes contaminant from whole body fish, which includes bones, organs, and blood of the fish. Concentrations presented in this report do not necessarily reflect contaminant concentrations in edible portions of fish tissue, please see Contaminant in Fish Fillets sub-indicator for this information.

Ecosystem Objective

Great Lakes waters should be free of toxic substances that are harmful to fish and wildlife populations. 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.” The programs used to develop this indicator directly impact and collaborate with Annex 3, Chemicals of Mutual Concern, and Annex 10, Science, of the 2012 Great Lakes Water Quality Agreement.

Methods

The assessment of status and trend incorporates chemical concentrations of multiple CMCs, from multiple fish species, in all 5 of the Great Lakes over time. The MDR is used to estimate the overall status and trends based on multiple variables. The overall and lake specific assessments in the sub-indicator are limited to the CMCs monitored in whole fish and include: Tetra-, Penta-, and Hexa-brominated diphenyl ethers (TeBDE, PeBDE, HxBDE), total mercury (Hg), total polychlorinated biphenyls (Total PCBs), hexabromocyclododecane (HBCD), and perfluorooctane sulphonate (PFOS). Data and analyses for the sub-indicator report were provided by the United States Environmental Protection Agency (USEPA) and Environment and Climate Change Canada (ECCC) generated from existing and long-term fish contaminant monitoring and surveillance programs operating in the Great Lakes basin. Individual and composite samples of homogenized whole fish are analyzed to provide data on priority contaminants present in Great Lakes aquatic ecosystems. Data are statistically analysed to determine mean and variance for each fish species, chemical, lake and year as well as temporal trends. The 10-year trend assessments are based on data from 2011-2020, and the long-term trend assessments are based on data from 1977-2020. The status assessments are based on a 3-year moving average (2018-2020).

Mean Deviation Ratio (MDR)

The MDR is a unit-less ratio that represents the average distance the contaminants included in the calculation are from the desired condition (ecosystem objective or targets). If all contaminants were present at their target or guideline the MDR calculation would result in a value of zero. MDR values significantly greater than zero indicate that on average one or more contaminants are above the targets or guidelines, which for toxic chemicals would be interpreted as Poor condition. MDR value that are significantly less than zero indicate that conditions are below the targets or guidelines and interpreted as Good condition. The MDR is calculated as presented in equation 1:

$$MDR = \frac{\sum_{j=1}^{P}\left(\frac{\sum_{i=1}^{n}\log\left(\frac{C_{ij}}{T_j}\right)}{n_j}\right)}{P} \quad (1)$$

Where P is the number of parameters measured, nj is the number of measurements in a time period for each parameter j, Cij is the ith measured concentration of each parameter j, and T is the target or environmental guideline for parameter (Table 1). Over 95% of measured concentrations were above the detection limit; half the detection limit was substituted in for the small number of values which were below the detection limit. Briefly, each measured concentration is divided by its corresponding guideline, the resulting ratio is averaged for each contaminant in each collection year, and the overall mean of the individual contaminant means within each year is the annual MDR. The MDR was calculated annually for the entire period of record for each of the Great Lakes using the parameters and guidelines listed in Table 1. The variance in the MDR calculation for each lake was carried through all steps of the calculation in order to estimate the variance of each annual MDR value (SOGL 2019). The variance of the 10 years including and preceding the most current value was used to characterize the contemporary standard deviation of the MDR. A region above and below a value of zero bounded by 1.96 x standard deviation was deemed to represent a value of Fair ecological condition. This region would approximate the 95% confidence interval of the MDR and any value within this region would have an interval that straddles the boundary between Good and Poor ecological condition (zero).

Temporal trends of the status of this sub-indicator are assessed for each contaminant and for the MDR for the entire period of record (long-term) and for the last 10 years (recent). Trends are assessed using non-parametric Mann-Kendall test with α=0.05 using the package “trend” in r-stats (Pohlert 2018, trend: Non-Parametric Trend Tests and Change-Point Detection. R package version 1.1.0.), which is a commonly used technique to statistically analyze the significance of monotonic contaminant time trends. Mann-Kendall tests cannot be performed on time series with missing values hence, any missing values (i.e. years where concentrations of a parameter were not measured) were imputed using the “na.kalman” function from the package “imputeTS” in r-stats (Moritz and Bartz-Beielstein 2017). Add sentence on bias in long term trend

This imputes annual means for each parameter for years where data is missing by fitting the existing data to a structural model and then imputing the missing means using a kalman smoothing function.

Discussion

Background and Methods

Long-term (greater than 25 years), basin-wide monitoring programs that measure whole body concentrations of contaminants in top predator fish (Lake Trout (*Salvelinus namaycush*) and/or Walleye (*Sander vitreus*)) are conducted by both the U.S. EPA Great Lakes National Program Office (GLNPO) through the Great Lakes Fish Monitoring and Surveillance Program (GLFMSP), and the ECCC Fish Contaminants Monitoring Program (FCMP).

These monitoring programs aim to identify risks posed from contaminants to fish and their wildlife consumers as well as to monitor trends in time as a measure of progress towards the Ecosystem Objectives of the 2012 GLWQA. The Toxic Chemicals in Whole Fish sub-indicator is included in the Toxic Chemicals indicator assessment for the Great Lakes since long-term trends of contaminants in biota provide valuable insights into the relative abundance of CMCs and other bioaccumulative contaminants in the environment. Fish integrate their exposure to contaminants over time and across their range and thus provide a broader assessment of environmental exposure than would a water sample taken at a single location at a point in time. Bioaccumulative contaminants are also found at higher concentrations in biota than they are in water, allowing for more accurate and cost-effective determination of levels in the environment. It is important to note, however, that contaminant levels in biota are not just influenced by contaminant concentrations in the water, but are the result of the integration of many biological, chemical and physical interactions (e.g. bioaccumulation and biomagnification processes, variations in diet and growth rates, food web changes as well as direct and indirect (via food items consumed) exposures to contaminated sediments).

The GLFMSP completed an interlaboratory comparison study of multiple age structures to identify the most appropriate age estimation structure for the Program. The Lake Trout maxilla was selected, over the otolith, as the more precise, accurate, and rapidly assessed structure for the Program when compared between laboratories and against the known age from coded wire tags (CWTs). Age-normalization practices can now be implemented when assessing contaminant concentrations and trends for the GLFMSP (Murphy et. al 2018). The FCMP program uses scales as the main age structure for the Great Lakes although some fish are aged using otoliths. Note, that data used to assess the MDR and trends in this report are not age or lipid corrected. Therefore, trends that appear in this report may differ from those published in the literature using the same data sources, which may include age correction in trend analysis.

Fish Collection and Program Design

ECCC reports annually on contaminant burdens in similarly aged Lake Trout (4+ through 6+ year range) and Walleye (Lake Erie) as well as in Rainbow Smelt (*Osmerus mordax*), a common forage species. ECCC analyzes and reports chemical concentration on individual fish. The U.S. EPA monitors contaminant burdens in similarly sized Lake Trout (600-700 mm total length) and Walleye (western Lake Erie, 400-500 mm total length) annually from alternating locations by year in each lake. U.S. EPA collects fish in a standard size range and determines individual fish age prior to homogenizing samples into 5 fish composites. Both programs monitor contaminants in known age fish, so we are able to account for effects of age on contaminant body burden. Monitoring stations for both ECCC and USEPA are shown in Figure 7. Environment and Climate Change Canada does not collect samples in Lake Michigan, and individual program contaminant lists are not identical (Table 2). Despite these differences in collection and analysis, the interpretation of results and trends by both programs are very similar.

Basin Wide Summary

Since the late 1970s, concentrations of legacy persistent organic pollutants such as polychlorinated biphenyls (PCBs) and organochlorine pesticides (OC_{pest}) in most monitored fish species have declined. Long-term mercury trends show varied results across the basin. Concentrations of mercury continue to be below the target established in the 1987 GLWQA. In general, the levels of regulated compounds are slowly declining or have stabilized in the tissues of Great Lakes top predatory fish. Basin wide, the changes are often lake-specific as they are dependent, in part, on the physio-chemical characteristics of the contaminants, hydrological characteristics of the lake, and the biological composition of the fish community and associated food webs. Despite declines, some compounds such as PCBs and PBDEs continue to exceed environmental quality objectives at some sites. It is important to note that

both the U.S. and Canadian monitoring programs continue to monitor and report out on these chemicals to track trends and assess the health of the ecosystem. A section on "Other Chemicals of Interest" is included in this sub-indicator to help inform readers of the current state of the science and to aid the Annex 3 process in reviewing other chemicals in the future.

Status of Chemicals of Mutual Concern (CMCs)

The 2012 Great Lakes Water Quality Agreement requires the United States and Canada to identify Chemicals of Mutual Concern (CMCs) that are potentially harmful to human health or the environment and that originate from anthropogenic sources. Pursuant to the Agreement, these substances will be targeted for binational action. In May of 2016, the first group of Chemicals of Mutual Concern was identified through a binationally agreed upon multi-stakeholder process. This effort was led by the Chemicals of Mutual Concern Annex Sub-committee, based on advice from experts from government, industry, environmental non-government organizations, and academia. The following 8 chemicals were identified as the first set of CMCs:

- Polychlorinated Biphenyls (PCBs)
- Mercury (Hg)
- Polybrominated Diphenyl Ethers (PBDEs)
- Perfluorooctanoic acid (PFOA)
- Perfluorooctane sulfonate (PFOS)
- Hexabromocyclododecane (HBCDD/HBCD)
- Short-Chain Chlorinated Paraffins (SCCPs)
- Long-Chain Perfluorinated carboxylic acids (LC-PFCAs)

This toxic chemical sub-indicator reports on Chemicals of Mutual Concern (CMCs) in a fulsome, consistent and transparent way. Information on additional chemicals of interest for the Great Lakes (not CMCs) is valuable for inclusion in the report and will be included in a separate section below, as appropriate, but is not part of the formal assessment.

Total polychlorinated biphenyls (PCBs)

Historically, PCBs were used in many things such as electrical equipment, capacitors, oils, and insulation. They were banned in the U.S. in 1979. In general, total PCB (Arochlor 1254) concentrations in Great Lakes top predator fish have continuously declined since their phase-out in the 1970s (Figure 8) and remain above the GLWQA 1987 Amendment target of 100 ng/g wet weight (ww) (Table 1). Long-term concentration trends are declining for all lakes with significance (Table 3). Short-term concentration trends are generally declining but without significance (Table 4). The exceptions are Lake Huron which is showing a very small, non-significant short-term increase and Lake Erie and Michigan which have seen significant declines over the past 10 years. Biological factors such as shifts in food webs, resulting in changes to growth rates of top predator species may be affecting concentrations and trends of PCB. To account for changes in growth rates, the GLFMSP has revised its compositing scheme to be based on age, rather than size, and age normalization is being incorporated into trend publications in peer reviewed literature (Zhou et. al 2018). Age is an important factor when analyzing contaminants in whole fish, since older fish usually have higher contaminant burdens due to longer life exposure. Studies have been conducted which compare temporal trends of PCBs in Great Lakes fish and air (Hites and Holsen, 2019). GLNPO also published a technical report which evaluates long term trends of PCBs, PBDEs, and mercury in top predator fish through 2019 (U.S. EPA, 2024). Results from these studies show statistically significant declines in PCB concentrations at each long-term

sampling site since 1991. These significant declines in PCB concentrations were also seen in age adjusted data at U.S. EPA long term sites between 2004 and 2020 (Pagano and Garner, 2024).

Total Mercury (Hg)

Mercury is used in chemical production, dental amalgam, fluorescent lighting, batteries, automotive switches and thermometers although some of these uses are being phased out. Concentration trends for total mercury in Great Lakes top predator fish have been variable. Their rates of decline and statistical significance have varied spatially across the basin and between the monitoring programs operated by ECCC and the U.S. EPA over the last few reporting cycles (Figure 9). Methyl mercury is expected to constitute over 90% of the total mercury body burden in top predator fish (Westöö G, 1973, Barone et al 2021). Concentrations of mercury continue to be below the environmental target of 0.3 mg/kg ww which is set based on the ecological risk of methylmercury to piscivorous fish in the Great Lakes (Sandheinrich et al. 2011). Long-term concentration trends are declining in Lakes Ontario, Superior, Huron, Erie, and Michigan. Lake Ontario is the only lake with a statistically significant declining trend (Table 3). Short term concentration trends are mixed with no significant changes except in Lake Erie which experienced a significant decline (Table 4). As with PCBs, the GLFMSP is incorporating age normalization practices into its trend publications in peer reviewed literature (Zhou et al. 2017).

The GLFMSP has been collaborating with the USGS Wisconsin Water Science Center and U.S. EPA Office of Research & Development, to identify sources of mercury from stable mercury isotope ratios in Great Lakes fish and sediment. Results indicate atmospheric sources dominate in Lakes Huron, Superior, and Michigan sediment while watershed derived and industrial sources dominate in Lakes Erie and Ontario sediment. Comparison of Δ^{200} mercury signatures in predatory fish from the upper Great Lakes reveals that bioaccumulated mercury is more similar to atmospherically derived mercury than a lake's sediment. These data suggest that in some cases, atmospherically derived mercury may be a more important source of methylmercury to higher trophic levels than legacy sediments in the Great Lakes (Lepak et al. 2015).

Further investigation into distinct mercury signatures driven by both food-web and water-quality characteristics has been completed in GLFMSP samples. Results indicate that sources of mercury, as assessed by stable mercury isotopes, vary by lake and indicate variability in the depth of the euphotic zone, where mercury is most likely incorporated into the food web. Mercury from precipitation, appears both disconnected from lake sedimentary sources and comparable in fish among the five lakes. Similar to the open ocean, water-column methylation occurs in the Great Lakes, possibly transforming recently deposited atmospheric mercury. The degree of photochemical processing of mercury is likely controlled by phytoplankton uptake rather than by dissolved organic carbon quantity among lakes (Lepak et al. 2018). Additional work was also done to identify changes in dietary mercury sources being altered by dreissenid mussel invasions in Lake Michigan. It was identified that mercury concentrations in fish can be affected by shifts in trophic structures and dietary preferences (Lepak et al. 2019).

Similar temporal patterns in mercury concentrations are also observed in Rainbow Smelt, a common forage fish for many fish and birds in the Great Lakes basin (Figure 10). Concentration of mercury in smelt is highest in Lake Superior and have been highly variable in all lakes since the last report. Continued monitoring of mercury levels in fish from all the lakes is warranted to adequately assess the future risk to wildlife consumers of fish in the Great Lakes basin, especially in areas where levels appear to be increasing.

Polybrominated Diphenyl Ethers (PBDEs)

PBDEs are commonly used in commercial formulations for fire retardants, plastics, consumer electronics, and coatings. In a national survey of PBDE concentrations in top predator fish from lakes across Canada, the highest concentrations were observed in fish from the Great Lakes and > 95% of the PBDE compounds in the fish were tetra-, penta-, or hexa-BDEs (Gewurtz et al. 2011). Concentrations of the tetra BDE mixture (BDE-47) in all five

Great Lakes continue to remain below the Federal Environmental Quality Guideline (FEQG) target of 44 ng/g ww (Figure 11). Concentrations of the penta BDE mixture (BDE-99 + 100) in all five Great Lakes continue to remain above the FEQG of 1.0 ng/g ww (Figure 12). Concentrations of the hexa BDE mixture (BDE-153 + 154) in all five Great Lakes are generally at or below the FEQG of 4 ng/g ww (Figure 13). Long-term concentration trends for the 3 BDE mixtures are declining in all lakes with mixed significance (Table 3). In general, short term concentration trends for the 3 BDE mixtures are declining with limited significance although non-significant increases were seen in Lake Michigan for Hexa and Penta BDE (Table 4). As with PCBs, the GLFMSP is incorporating age and age normalization practices into its trend publications in peer reviewed literature. Trends of total 5 PBDE congener concentrations using age normalization were evaluated at all GLFMSP sites from 2004-2020. Results show declines of Total-5 PBDEs (congeners 47, 99, 100, 153, and 154) in each lake. The largest percent decline (84.1%) was at the Lake Huron Rockport site (Pagano & Garner, 2024).

The FCMP collaborated with researchers from Environment and Climate Change Canada to examine trends in Halogenated flame retardants (HFRs) such as PBDEs in air, wet precipitation, fish and colonial water birds in Lake Ontario. Declining trends in PBDEs seen in air and wet precipitation were mirrored in Lake trout from Lake Ontario (li et al 2023). The halving time ranged from 6.1 years for BDE209 to 16 years for BDE154 in Lake Ontario trout. Using probabilistic models they found that tributaries and waste water were the main source of inputs of HFRs to Lake Ontario while transformation of HFRs in water and sediment were the main output pathways.

Recent work on temporal trends of these 5 major PBDE congeners in age normalized Great Lakes fish showed that concentrations increased from 1979 to 2000 with subsequent declines until 2007 in response to phase-outs in production. Since that time the decreasing trends have leveled off and recently have become unchanging, as declines in BDE 47 are offset by increasing trends in the other four BDEs, potentially from the breakdown of higher brominated congeners (Zhou et al. 2019).

Perfluorooctane Sulfonate (PFOS)

Perfluorooctane sulfonate (PFOS) is a synthetic substance belonging to a larger class of organic fluorochemicals that are either partially or completely saturated with fluorine. These compounds have been used as water repellants and flame retardants. Perfluorooctanoic acid (PFOA) is generally not found in Great Lakes whole fish, since it does not easily biomagnify through the aquatic food web and will not be reported on through this sub-indicator. Concentration trends for PFOS in Great Lakes top predator fish have been variable spatially across the basin and between the monitoring programs (Figure 14). Both long- and short-term concentrations in all 5 Great Lakes remain above the FEQG of 4.6 ng/g ww for mammalian diet. Long term significant declines in PFOS were seen in Lake Ontario and Lake Huron but not other lakes, (Tables 3 & 4).

Recent work has been done to evaluate temporal trends of perfluoroalkyl acids in top predator fish from 2005-2016. Results show that generally analyte concentrations declined in fish basin-wide from 2005 to 2016, however increasing trends were observed at the odd-year GLFMSP sampling site in Lake Superior, and the two GLFMSP sampling sites in Lake Erie (Point et al. 2021).

Additionally, studies were done on bioaccumulation and biomagnification of PFAS compounds within the food web in Lakes Huron, Erie, and Ontario. Samples of top-predator fish, forage fish, zooplankton, mysis, and benthos were collected from each GLFMSP sampling site in the three lakes listed above. Results show that PFOS is the dominant compound found in top-predator fish. Comparing concentrations of PFAS with $\delta^{15}N$ sample values indicate biomagnification with increasing trophic level (Ren et al., 2022a, Ren et al., 2022b, Ren et al., 2023).

Hexabromocyclododecane (HBCD)

HBCD is a high production flame retardant used mainly in polystyrene foams and is believed to have been used as a replacement alternative to PBDEs. HBCD was added as a routine analyte to both the U.S. and Canadian

monitoring and surveillance programs in 2012 as a result of it being designated as a CMC, however retrospective analysis resulted in data as far back as 2006. Current and retrospective analyses for this compound were completed in both fresh and archived fish tissues to allow for a more robust data set. In general, concentration trends are variable spatially across the basin and are well below the European Union's Environmental Quality Standard (EQS) for fish tissue of 167 ng/g lipid (Figure 15). Long- and short-term concentration trends, and their significance, are varied across the five Great Lakes with an increasing long-term trend seen in Lake Michigan and a decreasing long-term trend seen in Lake Ontario (Tables 3 & 4).

A study of HBCD in top predator fish (Lake Trout, Walleye, or Brook Trout) from across Canada identified the spatial distribution of HBCD in 2013 (n = 165) from 19 sampling sites and in 2015 (n= 145) from 20 sites across Canada. HBCD was measurable in at least one sample at each sampling site regardless of sampling year with the exception of Walleye from the south basin of Lake Winnipeg (2013). Sampling sites in or near the Laurentian Great Lakes had greater ∑HBCD concentrations compared to locations to the west or east. HBCD has 16 possible stereo-isomers with different biological activities. The greatest mean ∑HBCD concentration was 72.6 ng/g lipid weight in fish from Goderich (Lake Huron) (2015). Regardless of the sampling sites, α-HBCD was the dominant congener followed by γ-HBCD, whereas β-HBCD was barely detectable. In fish from the same waterbody there were comparable α/γ isomer concentration ratios. The greatest ratio was 20.8 in fish from Lake Ontario, whereas the lowest ratio was 6.3 for fish from Lac Memphrémagog (Québec) likely related to more recent emissions of a technical HBCD mixture (primarily composed of γ-HBCD but also contains measurable α- and β-HBCD). Temporal trends of HBCD in Lake Trout from Lake Ontario showed a significant decreasing trend for γ-HBCD with a half-life estimate of 10 years over a 36-year period (1979–2015), and for α-HBCD with a half-life of 11 years over the years of 2008 to 2015. The proportion of α-HBCD to ∑HBCD increased significantly during 1979 to 2015. The study provided novel information on the isomer-specific HBCDs in Canada freshwater fish. (Su et al. 2018). Li et al (2023) found non-monotonic trends in air and wet precipitation in HBCD in the Lake Ontario basin with significant rapid increasing trends post 2011. The lack of consistent declining trends in Lake Ontario trout may be reflective of increasing loadings of HBCD through wet deposition.

Long-term HBCD trends have been evaluated in recent studies. Researchers noted that trends of ∑HBCD (2004-2016) show concentration increases in Lakes Erie and Ontario in Great Lakes fish. They also observed concentration decreases in Lakes Superior and Michigan and found that concentrations in Lake Huron have been generally unchanged during this time period (2004-2016) (Parvizian et al. 2020).

Short-Chain Chlorinated Paraffins (SCCPs) & Medium Chain Chlorinated Paraffins (MCCPs)

These classes of compounds have been used as lubricants and coolants for metal cutting. Older data monitored by the FCMP from 2011 and 2013 showed concentrations of total SCCPs in lake trout ranged from non-detect (ND) to 9 n/g wet weight (ND to 80 ng/g lipid weight). Concentrations of total SCCPs in walleye from Western Lake Erie ranged from 0.06 ng/g to 4.8 ng/g wet wt (18 to 28 ng/g lipid wt). All were well below the FEQG for SCCPs of 2,700 ng/g lipid. Levels of chlorinated paraffins in fish from Canadian lakes, showed that fish from the Great Lakes had higher levels of the medium chain chlorinated paraffins (MCCPs) (C14-C17) than SCCPs (C10-C13) (Saborido Basconcillo et al. 2015). The levels of MCCPs ranged from 1.8 to 46 ng/g wet wt (11 to 351 ng/g lipid wt) in Lake Trout to 3.5 to 25 ng/g wet wt (14 to 126 ng/g lipid wt) in walleye from Lake Erie. More recent data from 2018 showed measurable concentrations of SCCPs, MCCPs and long chain chlorinated paraffins LCCPs in Lake trout from Lake Superior and Lake Ontario. The highest detection frequency and highest concentrations were for MCCPs (Simonnet-Laprade et al 2022). Low chlorinated SCCPs were more likely to be detected in trout and were found at higher concentrations (DF 90%, concentration: 2.8 ng/g wet wt) than higher chlorinated SCCPs (average. All trout had measurable concentrations of MCCPs (average 41 ng/g wet wt) and low chlorinated LCCPs (13 ng/g wet wt). GLFMSP cooperators have began analyzing top-predator fish samples from 2010-2020. Results show no obvious

trends in Lakes Ontario, Michigan, Huron, and Superior. However, Lake Erie did exhibit a possible increasing trend in MCCPs. (Parvizian et al. 2024).

Other Chemicals of Interest

Both the U.S. and Canadian monitoring and surveillance programs have invested in the identification and quantification of non-CMCs and emerging chemicals through the Great Lakes Restoration Initiative and Canada's Chemicals Management Plan. Some, such as dioxins and furans or polychlorinated napthalenes, are considered legacy compounds but may still be at concentrations harmful to fish or consumers of fish, or trends may have changed. There is also a need to survey emerging or unknown contaminants in order to gather enough data to determine if they may be causing harm, or have the potential to become harmful. Summaries of recent publications on identification, quantification and/or methods of non-CMCs and emerging chemicals are provided below. Because many of these chemicals are newly identified, or data have only recently become available, status and trend statements are not possible at this time. However, the authors feel it is important to highlight this work and if warranted, these compounds may appear in future State of the Great Lakes sub-indicator reports. It is also important to note that surveillance for emerging chemicals and continued tracking of legacy chemicals is an important part of both countries' programs.

Dioxins and Furans (PCDD/F)

Dioxins and furans are primarily unintended byproducts of combustion or industrial processes such as herbicide production or the bleaching of wood pulp. The U.S. and Canadian Fish Monitoring and Surveillance Programs completed a trend concentration assessment of age corrected Lake Trout and Walleye for polychlorinated dibenzo-pdioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), and coplanar polychlorinated biphenyls (CP-PCBs) from samples collected between 2004 and 2014. Age–contaminant corrections were developed to accurately report contaminant trends due to significant Lake Trout age structure changes, using the age-trend model (ATM). The ATM used a lake-specific age–contaminant regression to mitigate the effect of a fluctuating Lake Trout age structure to directly improve the log–linear regression models. ATM results indicated that half-life (t1/2) and percent decreases for PCDD/ Fs, CP-PCBs, and toxic equivalence (TEQ) (average –56 to 70%) were fairly uniform and consistent across the Great Lakes over the 2004–2014 period. The vast majority of TEQ associated with all Great Lakes Lake Trout and Walleye samples is due to the non-ortho CP-PCBs (average = 79%) as compared with PCDD/Fs (average = 21%). On average, CP-PCB-126 individually accounted for over 95% of the total CP-PCB TEQ. A retrospective analysis (1977–2014) of 2378-TCDF and 2378-TCDD raw concentrations in Lake Ontario Lake Trout revealed decreases of 94% and 96%, respectively. Tissue residue guidelines for wildlife protection based on Lake Trout and Walleye total TEQ were uniformly exceeded in all the Great Lakes (Pagano et al. 2018).

Novel Halogenated Organic Contaminants

The GLFMSP has developed non-targeted screening methodologies to identify new and emerging chemicals in whole fish samples. Halogenated chemicals were identified using a combination of authentic standards and library spectral matching, with molecular formula estimations provided by exact mass spectral interpretation. Halogenated organic compounds represent one of the largest groups of chemicals found in the environment and have been studied extensively over the past four decades due to their persistent, bioaccumulative, and toxic (PBT) properties. In addition to the halogenated chemicals currently being targeted by the GLFMSP, more than 60 non-targeted halogenated species were identified. Most appear to be metabolites or breakdown products of larger halogenated organics. The most abundant compound class was halomethoxyphenols accounting for more than 60% of the total concentration of halogenated compounds in top predator fish from all five Great Lakes. These results illustrate the need and utility of non-targeted halogenated screening of aquatic systems using this platform (Fernando et al. 2018).

More recently, non-targeted methodologies were used to compare novel halogenated features in Lake Trout between two time periods a decade apart, 2005/2006 and 2015/2016. Greater than 2000 unknown halogenated features were detected. Lake Superior had the lowest number of unknown halogen features while fish from Lake Ontario had the highest number. Most of these halogenated features changed in the Great Lakes between the two time periods, at a rate higher than changes in total PCBs, meaning generally they have decreased at a faster rate (Fakouri Baygi et al. 2020).

National Research Institute for Agriculture, Food, and Environment (INRAE) in France, McGill University and the FCMP developed a non-targeted screening approach for persistent organohalogen compounds using liquid and gas phase chromatography coupled to high resolution mass spectrometry (Simonnet-Larade et al 2022). This was used to compare the complementarity of targeted and non-targeted analysis of halogenated flame retardants in Lake trout in Lake Ontario. A total of 195 halogenated mass spectral features were characterized including well known POPs such as PBDEs, HBCD, DDT, and emerging brominated flame retardants. Some of the unidentified compounds corresponded to an oxygenated form of dichlorodiphenyldichloroethylene (DDE) and were among the most abundant halogenated compounds in Lake Ontario trout. There was a lack of correspondence between targeted and non-targeted analysis with only 20% of the halogenated compounds detected using targeted screening also detected in the non-targeted approach.

Additional Per- and Polyfluoroalkyl Substances (PFAS) compounds

Lake Michigan Lake Trout from the GLFMSP were analyzed for polyfluoroalkyl acids using a computer algorithm that would reference compounds against 3570 possible compounds including C4-C10 perfluoro- and polyfluoroalkyl, polyfluorochloroalkyl acids and sulfonates, and potential ether forms from chemical libraries. The results suggested the presence of 30 polyfluorinated chemical formulas which have not been previously reported in the literature. Little is known, at this time, regarding the toxicity of these novel PFAS compounds. As PFAS compounds are identified via this virtual library, and as they are quantified via neat standards (certified reference material), they will be considered for incorporation into the GLFMSP routine analyte list (Fakouri Baygi et al. 2016).

Polyhalogenated carbazoles (PHCZs)

PHCZs were investigated in archived GLFMSP Lake Trout collected between 2004 and 2016. Median concentrations of Total PHCZs by lake ranged from 54.7 to 154 ng/g lipid weight (6.8-28.0 ng/g wet weight). Dominant congeners included 3,6-dichlorocarbazole, 1,3,6-tribromocarbazole, and 1,3,6,8-tetrachlorocarbazole. The highest Total PHCZs concentrations were found in Lakes Michigan and Ontario fish, followed by Lake Huron, whereas Lakes Erie and Superior fish contained the lowest concentrations. Congener profiles of PHCZs also exhibited spatial variations. After age normalization to minimize fish age influence on bioaccumulation rates, Total PHCZ concentrations declined significantly over time in all lakes except Lake Erie, with slopes ranging from -10.24% to -3.85% per year. The median toxic equivalent (TEQ) of PHCZs due to their dioxin-like activity was determined to range from 8.7 to 25.7 pg/g lipid weight (lw) in Great Lakes fish. This study provides the first insight into the bioaccumulation and spatiotemporal trends of PHCZs in Great Lakes and suggests the need for further research on this group of chemicals (Wu et al.2018)

Halogenated Flame Retardants

The identification, persistence, accumulation and trophic transfer of 25 polybrominated diphenyl ether (PBDE) congeners, 23 non-PBDE halogenated flame retardants (NPHFRs), 4 polybrominated-diphenoxybenzenes (PB-DiPhOBzs) and 6 methoxylated (MeO–) PB-DiPhOBzs were investigated in predator and prey fish collected in 2010 from sites in Lakes Ontario (n = 26) and Erie (n = 39). Regardless of locations or species, 20 PBDEs and 12 NPHFRs were quantifiable in at least one of the 65 analyzed samples, and polybrominated-1,4-diphenoxybenzenes (PB-DiPhOBzs) and MeO-PB-DiPhOBzs were not detectable in any of analyzed samples.

Among the Flame Retardants (FRs), the greatest concentrations were the ∑PBDE, ranging from 1.06 (Rainbow Smelt, Lake Erie) to 162 (Lake Trout, Lake Ontario) ng/g wet weight (ww), which was followed by mean HBCDD concentrations ranging ND to 17.3 (Lake Trout, Lake Ontario) ng/g ww. The remaining FRs were generally not detectable or at sub-ppb levels. In most of cases, FR concentrations in samples from Lake Ontario were greater than those from Lake Erie. Strong and significant positive linear relationships occurred between log-normalized FR concentrations (ww or lw) and ages of the top predator Lake Trout (n = 16, from Lake Ontario), and the estimated FR doubling ages (T2) were 2.9–6.4 years. For Walleye from Lake Erie, significantly positive linear relationships were also observed for some FRs, but the linear relationships generally became negative after FR concentrations were normalized with lipid weight. This study provides novel information on FR accumulation in aquatic organisms, and for the first time, significant positive linear relationships are reported between log-normalized FR concentrations (lw or ww) and ages of Lake Trout from the Great Lakes (Su et al. 2017).

Spatial and temporal trends in alternative flame retardants, including 18 dechlorane analogues and 20 alternative brominated FRs (ABFRs) were investigated in mega-composites of 50 Lake Trout or walleye from the Great Lakes, between 2004 and 2016. Concentration of sum dechlorane analogues ranged from 0.33 to 31.9 ng/g lipid weight, with highest concentrations in Lake Ontario and profiles there may be indicative of point sources. Other ABFRs measured included hexabromobenzene, pentabromotoluene, and tetrabromo-o-chlorotoluene. The total ABFRs ranged from 0.91 to 54.7 ng/g lipid weight, and were lowest in Lake Erie. The concentration of total dechloranes and total ABFRs declined in all lakes, with the exception of Lake Erie (Wu et al 2019).

Polychlorinated Naphthalenes (PCNs)

Polychlorinated naphthalenes (PCNs) are legacy contaminants, produced primarily as flame retardants and dielectrics until phased-out in Europe and North America in the 1970s. PCNs have been studied in whole fish, herring gull eggs, and sediment. Spatial and temporal trends (1979–2013) of PCN concentrations were studied throughout the Great Lakes and St. Lawrence River, whereas sediments were analyzed for 2011–2013 only. For both fish and gull eggs, concentrations of PCNs were highest in Western Lake Erie (7660 & 3020 pg/g ww respectively) and declined downstream to St. Lawrence River (range: 34–2370 pg/g ww). For sediments, concentrations were highest in suspended sediments from the Detroit River (264,000 pg/g), and were lower in surficial sediments downstream to the St. Lawrence River (range: 440–19,300 pg/g). PCNs declined at all sites from ~1980 to 1995, but in Lake Erie concentrations of PCNs increased in gulls and fish from 1995 until 2005. The resurgence in PCNs in biota corresponded to the timing of remedial dredging of sediment highly contaminated with PCNs in the Detroit River, and the effects of this dredging appeared to be manifested downstream to Lake Ontario. Congener profiles of PCNs differed between Lake Erie and Lake Ontario until post-dredging, where PCN profiles of fish in both lakes became increasingly more similar. PCNs in gull eggs were mostly hepta-PCNs, whereas fish had higher concentrations of lower chlorinated PCNs. Patterns of PCNs in gulls and fish appear to be influenced by differences in not only routes of exposure and differential metabolic ability, but also resuspension of PCN contaminated sediments from dredging-related activities (McGoldrick et al. 2018).

Trends analysis for PCNs were performed from 2004-2018 in Lake Trout and Walleye in each of the Great Lakes. Trends generally showed significant decreases in total PCNs over this period, except for fish from Lake Erie, where concentrations increased. Total PCN concentrations in Great Lakes fish ranged from 5701 and 100 pg/g ww. In addition, researchers observed a prominent PCN concentration trend break point in Lake Ontario lake trout over the 2012-2016 period likely associated with hazard waste clean ups, channel dredging and spoil disposal in Detroit River and the western basin of Lake Erie (Pagano and Garner, 2021).

Substituted Diphenylamine Antioxidants (SDPAs)

SDPAs and benzotriazole UV stabilizers (BZT-UVs) are industrial additives of emerging environmental concern. However, the bioaccumulation, biomagnification, and spatial distribution of these contaminants in the Great Lakes

are unknown. A 2018 study addressed these knowledge gaps by reporting SDPAs and BZT-UVs in Herring Gull eggs, Lake Trout, and their food web in the Great Lakes for the first time. Herring Gull eggs showed much higher detection frequency and concentrations of target SDPAs and 2-(2H-benzotriazol-2- yl)-4,6-di-tert-pentylphenol (UV328) than that of the whole-body fish homogenate. Interestingly, the predominant SDPAs in Herring Gull eggs were dinonyl- (C9C9) and monononyl-diphenylamine (C9) which were previously shown to be less bioaccumulative than other SDPAs in fish. In contrast, dioctyl-diphenylamine (C8C8) was the major SDPA in Lake Trout, and biodilution of C8C8 was observed in a Lake Superior Lake Trout food web. Such variations in Herring Gull eggs and fish indicate the differences in accumulation and elimination pathways of SDPAs and BZT-UVs and require further clarification of these mechanisms (Lu et al. 2018).

Linkages

Contaminant levels in Lake Trout and Walleye are dependent on complex biological and physiochemical interactions both within and outside of the Great Lakes basin as these apex predators integrate contaminant inputs from water, air, sediment, and their food sources. Long-term changes in precipitation and wind currents could alter the influx of contaminants from sources outside of the basin and may alter food webs and the contaminant transfer through them. Aquatic invasive species also alter food webs and change energy and contaminant dynamics in the lakes. They also may introduce new pathways by which sediment contaminant pools could be mobilized and transferred to fish. Many new contaminants of concern are components of consumer products, personal care products, or pharmaceuticals. As a result, wastewater treatment effluents may be an important source of contamination. Inferences on the effects of invasive species and changes in water temperature on the accumulation of contaminants in aquatic biota are not well understood. However, changes in contaminant trends in whole fish, as they relate to changes in the ecosystem, are considered in interpreting results.

It is important to note how chemical concentrations are behaving in the environment in various media. For example, a recent publication from the US Air and Fish Monitoring programs recently evaluated reported levels of PCBs and DDTs over time and calculated the rates at which age adjusted concentrations have decreased over time for comparison. In general, the halving times (9–17 years for PCBs and 7–10 years for DDTs) estimated from the full fish dataset are similar to those estimated from the atmospheric data, suggesting that the atmospheric and the fish levels are coupled. The more recent, age-adjusted rates are sometimes significantly faster than those from the full fish and atmospheric datasets, suggesting that the air-water dynamic may now be changing (Hites et al. 2019). Similar correspondence was found for trends in PBDEs in Lake Ontario between lake trout and both air and wet precipitation (Li et al 2023). These evaluations are important both to confirm observed trends across media but also to identify how chemicals can behave differently across media for a more accurate depiction of the health of the Great Lakes total environment.

Changes in the food webs of the Great Lakes are becoming much more important to understand and quantify. For example, the decline in age and older Alewife (*Alosa pseudoharengus*) abundance in Lake Huron is primarily responsible for slower growing Lake Trout (He et al. 2016), resulting in higher chemical concentrations of Persistent Bioaccumulative Toxics in the Lake Trout. Prior to the complete collapse of the Alewife population during 2003-2004, Alewives had been the mainstay of the diet of Lake Trout in Lake Huron. The decline in abundance of age and older Rainbow Smelt, which represented another component of Lake Trout diet, may have also contributed to the decrease in Lake Trout growth and condition. Food web assessments for chemical transfer fatty acid content, fatty acid ratios, and stable carbon and nitrogen isotopes are ongoing in the U.S. to assist with the interpretation of chemical results and trends.

Changes in water temperature and precipitation patterns may have an impact on contaminant body burdens in Great Lakes fish and may accelerate or exacerbating existing processes. Increased temperatures may change the movement of contaminants within the ecosystem, their transformation into more harmful compounds and their availability to aquatic biota (Noyes et al 2009). For instance, mercury body burdens in fish may be impacted by warmer temperatures, fluctuating water levels and increased run-off that may effect the inputs of mercury to lakes, the methylation of mercury in the aquatic system and the rate of mercury uptake in fish (Grieb et al 2019). Lake levels, which fluctuate in response to long term meteorological cycles can also have implications for mercury cycles that are evidenced in fluctuations in mercury concentrations in fish and other wildlife (Watras et al 2020). The impact of lake level fluctuations on Hg concentrations may mask declining trends of Hg burdens in fish associated with declines external sources such as atmospheric deposition and may be even more impactful in the future if ecosystem conditions in the Great Lakes continue to change. Rising surface water temperatures may also increase or change the impact of contaminants to fish. As fish are exothermic animals, their ability to metabolize contaminants may change with increasing water temperatures, which may reduce or increase the toxicity of the compound, depending on the mechanism of action. For instance, the toxicity of some pesticides has been shown to increase with temperature in fish (Noyes et al. 2009).

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	ECCC: https://data-donnees.az.ec.gc.ca/data/substances/monitor/great-lakes-fish-contaminants-monitoring-and-surveillance-data/ U.S. EPA: https://cdx.epa.gov/		

Notes on Accessing Data

U.S. EPA fish data in this report is stored in the Great Lakes Environmental Database (GLENDA). In order to access this data, users need to register a free account to begin querying data in GLENDA.

Data Limitations

This sub-indicator relies on U.S. and Canadian government monitoring programs for the majority of the information reported. The list of contaminants which require monitoring continues to grow in number which can be challenging for operating budgets and staffing resources. These limitations will require changes to the frequency and intensity of monitoring of some compounds as newer compounds are added to the priority list. As monitoring priorities for chemicals have shifted over time, not all CMCs have been measured in every lake, in every year or over the entire time span of ECCC and USEPA monitoring and surveillance programs. We acknowledge that adding additional CMCs to the indicator can lead to a shift in values and that this may have an impact on long-term trends in the MDR. It is also important to note that the ten-year time trend assessment may not be long enough to detect trends, especially when it is compared to long-term trend assessments. Trend detection may be more difficult if there are insufficient staffing or budgetary resources to measure CMCs in every lake, each year.

Additional Information

Reductions in contaminant levels in whole fish will reflect environmental change in the overall water quality of the Great Lakes. Reductions in contaminant loading with subsequent reductions in the concentration of contaminants in the water will pose less risk of harm to fish communities and fish-eating wildlife.

Identification of and revisions to environmental targets and guidelines will result in revised MDR development for status and trend evaluation. Additionally, the incorporation of new CMCs, should this be applicable, and additional existing CMC trend data will affect how the MDR is developed and interpreted.

Ancillary data collections have been established that include fish age, length, weight, sex, and lipid content of the fish collected. Detailed data on sample size, location and the complete suite of ancillary measurements is available to be sourced.

Environmental specimen banks containing tissue samples are a key component of both the U.S. and Canadian monitoring programs, allowing for retrospective analyses of newly identified chemicals of concern to be able to develop long-term trends in the short-term.

The authors have made efforts to improve the statistical rigor of this sub-indicator report through the inclusion of error bounds on estimated concentrations and trends through time. The authors have also focused on contaminants with defined environmental targets, guidelines and/or thresholds to put observed concentrations in context with risk to the environment. Other improvements to statistical rigor, such as, better methods to characterize dataset with censored values (i.e. non-detects) should be investigated and incorporated in future reports on this sub-indicator.

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Data available upon request from authors or U.S. data can be accessed at: https://cdx.epa.gov/

Some Canadian data can be accessed at: https://open.canada.ca/data/

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Source: Environment and Climate Change Canada and U.S. Environmental Protection Agency

Source: Canadian Federal Environmental Quality Guidelines - https://www.canada.ca/en/health-canada/services/chemical-substances/fact-sheets/federal-environmental-quality-guidelines.html

Sandheinrich, M.B., Bhavsar, S.P., Bodaly, R.A., Drevnick, P.E., Paul, E.A., 2011. Ecological risk of methylmercury to piscivorous fish of the Great Lakes region. Ecotoxicology 20, 1577-1587

European Union Environmental Quality Standards - https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX:32013L0039

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Source: Environment and Climate Change Canada and U.S. Environmental Protection Agency

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Source: Environment and Climate Change Canada and U.S. Environmental Protection Agency

Figure 14. Mean annual PFOS concentrations (wet weight) for individual (Environment and Climate Change Canada) and composited (U.S. Environmental Protection Agency, Point et al. 2021.) whole body Lake Trout or Walleye (Lake Erie, EPA only, western basin) collected from each of the Great Lakes. Bars represent standard deviations around the mean. Canadian guidelines for avian diet are shown for context.

Source: Environment and Climate Change Canada, U.S. Environmental Protection Agency

Figure 15. Mean annual total HBCD concentrations (wet weight) for individual (Environment and Climate Change

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Source: Environment and Climate Change Canada and U.S. Environmental Protection Agency

Last Updated

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Sandheinrich, M.B., Bhavsar, S.P., Bodaly, R.A., Drevnick, P.E., Paul, E.A., 2011. Ecological risk of methylmercury to piscivorous fish of the Great Lakes region. Ecotoxicology 20, 1577-1587
European Union Environmental Quality Standards - https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX:32013L0039

Contaminant	Criteria Source	Value (ng/g ww)
Sum-tetrabrominated diphenyl ethers (TeBDE) (BDE47)	FEQG* (wildlife diet)	44
Sum-pentabrominated diphenyl ethers (PeBDE) (BDE 99 + 100)	FEQG* (fish tissue)	1
Sum-hexabrominated diphenyl ethers (HxBDE) (BDE 153+154)	FEQG* (wildlife diet)	4
Perfluorooctane sulphonate (PFOS)	FEQG* (mammalian diet)	4.6
Total polychlorinated biphenyls (PCBs)	GLWQA 1987 Amendment	100
Sum DDT (DDT+DDD+DDE)	GLWQA 1987 Amendment	1000.0
Short Chain Chlorinated Alkanes (SCCA)	FEQG* (ng/g lipid)	2700.0
Total Hexabromocyclododecane (HBCDD)	EQS European Union (EQS)	167
Total Mercury	Sandeinrich et al. 2011 (LOER)	300

Contaminant Criteria for Environmental Monitoring and Surveillance

* Canadian Federal Environmental Quality Guidelines
Sandheinrich, M.B., Bhavsar, S.P., Bodaly, R.A., Drevnick, P.E., Paul, E.A., 2011. Ecological risk of methylmercury to piscivorous fish of the Great Lakes region. Ecotoxicology 20, 1577-1587
Lowest Observed Effect Residue (LOER)
http://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX:32013L0039
Environmental Quality Standard (EQS)

Table 2. Chemicals detected greater than 10 % frequency identified through monitoring and surveillance programs. Source: Environment and Climate Change Canada and U.S Environmental Protection Agency.

Compound or Class	Agency		Great Lake				
	Environment and Climate Change Canada	US EPA	Ontario	Erie	Huron	Michigan	Superior
Hexabromocyclododecane (α-, γ-HBCD)	X	X	X	X	X	X	X
Polychlorinated naphthalenes (PCN)[1]		X	X	X	X	X	X
tris(2-butoxyethyl) phosphate (TBOEP)							
Chlorinated alkanes (short and medium chain)	X		X	X	X		X
Decamethylcyclopentasiloxane (D5)	X		X	X	X		X
Dodecamethylcyclohexasiloxane (D6)	X		X	X	X		X
Octamethylcyclotetrasiloxane (D4)	X		X	X	X		X
Perfluorooctane sulfonamide (PFOSA)	X		X	X	X		X
Perfluorooctanoic acid (PFOA)	X						
Chlordane (α-, γ-)	X	X	X	X	X	X	X
Dieldrin	X	X	X	X	X	X	X
Heptachlor epoxide	X	X	X	X	X	X	X
Hexachlorobenzene (HCB)	X	X	X	X	X	X	X
Hexachlorocyclohexane (α-, γ-HCH)	X	X	X	X	X	X	X
Mercury	X	X	X	X	X	X	X
Mirex	X	X	X	X	X	X	X
p,p'-dichlorodiphenyldichloroethane (DDD)	X	X	X	X	X	X	X
p,p'-dichlorodiphenyldichloroethylene (DDE)	X	X	X	X	X	X	X
p,p'-dichlorodiphenyltrichloroethane (DDT)	X	X	X	X	X	X	X
Perfluorodecanesulfonate (PFDS)	X	X	X	X	X	X	X
Perfluorodecanoic acid (PFDA)	X	X	X	X	X	X	X
Perfluorododecanoic acid (PFDoA)	X	X	X	X	X	X	X
Perfluorononanoic acid (PFNA)	X	X	X	X	X	X	X
Perfluorooctanesulfonate (PFOS)	X	X	X	X	X	X	X
Perfluorotridecanoic acid (PFTrA)	X	X	X	X	X	X	X
Perfluoroundecanoic acid (PFUnA)	X	X	X	X	X	X	X
Polybrominated diphenyl ethers (PBDE)[1]	X	X	X	X	X	X	X
Polychlorinated biphenyls (PCB)[2]	X	X	X	X	X	X	X
Endrin		X	X	X	X	X	X
cis-nonachlor		X	X	X	X	X	X
Endosuflan (I, II)		X	X	X	X	X	X
Endosulfan sulfate		X	X	X	X	X	X
Hexachlorocyclohexane (β-, δ-HCH)		X	X	X	X	X	X
Octachlorostyrene		X	X	X	X	X	X
Oxychlordane		X	X	X	X	X	X
Total Dioxin TEQ (Mammal)		X	X	X	X	X	X
Toxaphene (Camphechlor)		X	X	X	X	X	X
trans-nonachlor		X	X	X	X	X	X

Chemicals detected greater than 10 % frequency identified through Monitoring and Surveillance

Table 3. Sen's slope and p-values from Mann-Kendall trend analysis of long term concentrations of CMCs in fish from each of the Great Lakes. Source: Environment and Climate Change Canada and U.S. Environmental Protection Agency

Mercury	Ontario	Huron	Erie	Superior	Michigan
P value	**<0.001**	0.930	0.317	0.137	1.000
Sen's slope (%)	-0.52%	-0.03%	-0.12%	-0.26%	-0.02%

HxBDE	Ontario	Huron	Erie	Superior	Michigan
P value	**<0.001**	0.464	0.786	**0.050**	0.487
Sen's slope (%)	-1.77%	-0.27%	-0.31%	-1.82%	-0.33%

PeBDE	Ontario	Huron	Erie	Superior	Michigan
P value	**<0.001**	**0.042**	0.349	**0.004**	**0.017**
Sen's slope (%)	-2.39%	-0.82%	-0.63%	-2.82%	-2.17%

TeBDE	Ontario	Huron	Erie	Superior	Michigan
P value	**<0.001**	**<0.001**	0.139	**0.009**	**<0.001**
Sen's slope (%)	-3.24%	-3.40%	-0.85%	-2.10%	-4.61%

HBCDD	Ontario	Huron	Erie	Superior	Michigan
P value	0.387	0.235	0.166	**0.038**	**0.008**
Sen's slope (%)	-0.41%	-1.53%	-1.78%	-3.13%	1.40%

PFOS	Ontario	Huron	Erie	Superior	Michigan
P value	**0.049**	**0.004**	0.161	0.322	1.000
Sen's slope (%)	-1.39%	-1.68%	-3.61%	1.41%	1.43%

PCB	Ontario	Huron	Erie	Superior	Michigan
P value	**<0.001**	**<0.001**	**<0.001**	**<0.001**	**<0.001**
Sen's slope (%)	-3.03%	-2.64%	-1.46%	-2.07%	-2.44%

LEGEND

P value	**Significant**	Not Sig
Sen's slope (%)	Increasing	Decreasing

Table 4. Sen's slope and p-values from Mann-Kendall trend analysis of short term (10yr) concentrations of CMCs in fish from each of the Great Lakes. Source: Environment and Climate Change Canada and U.S. Environmental Protection Agency

Mercury	Ontario	Huron	Erie	Superior	Michigan
P value	0.474	0.283	**0.032**	0.474	0.592
Sen's slope (%)	0.26%	-1.20%	-2.96%	1.39%	0.67%

HxBDE	Ontario	Huron	Erie	Superior	Michigan
P value	0.283	0.211	0.371	0.858	0.592
Sen's slope (%)	-0.93%	-1.07%	-3.60%	-1.51%	0.96%

PeBDE	Ontario	Huron	Erie	Superior	Michigan
P value	**0.049**	0.858	0.474	0.858	0.721
Sen's slope (%)	-1.86%	-0.20%	-1.75%	-0.63%	0.46%

TeBDE	Ontario	Huron	Erie	Superior	Michigan
P value	0.074	0.073	**0.049**	1.000	0.592
Sen's slope (%)	-5.90%	-3.50%	-3.29%	-0.22%	-1.28%

HBCDD	Ontario	Huron	Erie	Superior	Michigan
P value	0.371	0.592	0.371	0.592	0.371
Sen's slope (%)	-0.41%	-1.86%	-6.03%	-2.16%	0.75%

PFOS	Ontario	Huron	Erie	Superior	Michigan
P value	0.858	0.721	0.474	0.721	1.000
Sen's slope (%)	-0.08%	-1.33%	-4.19%	1.17%	1.43%

PCB	Ontario	Huron	Erie	Superior	Michigan
P value	0.211	0.592	**0.004**	1.000	**0.012**
Sen's slope (%)	-1.23%	0.85%	-5.61%	-0.71%	-4.44%

LEGEND

P value	**Significant**	Not Sig	
Sen's slope (%)	Increasing	Decreasing	Unchanged

Figures:

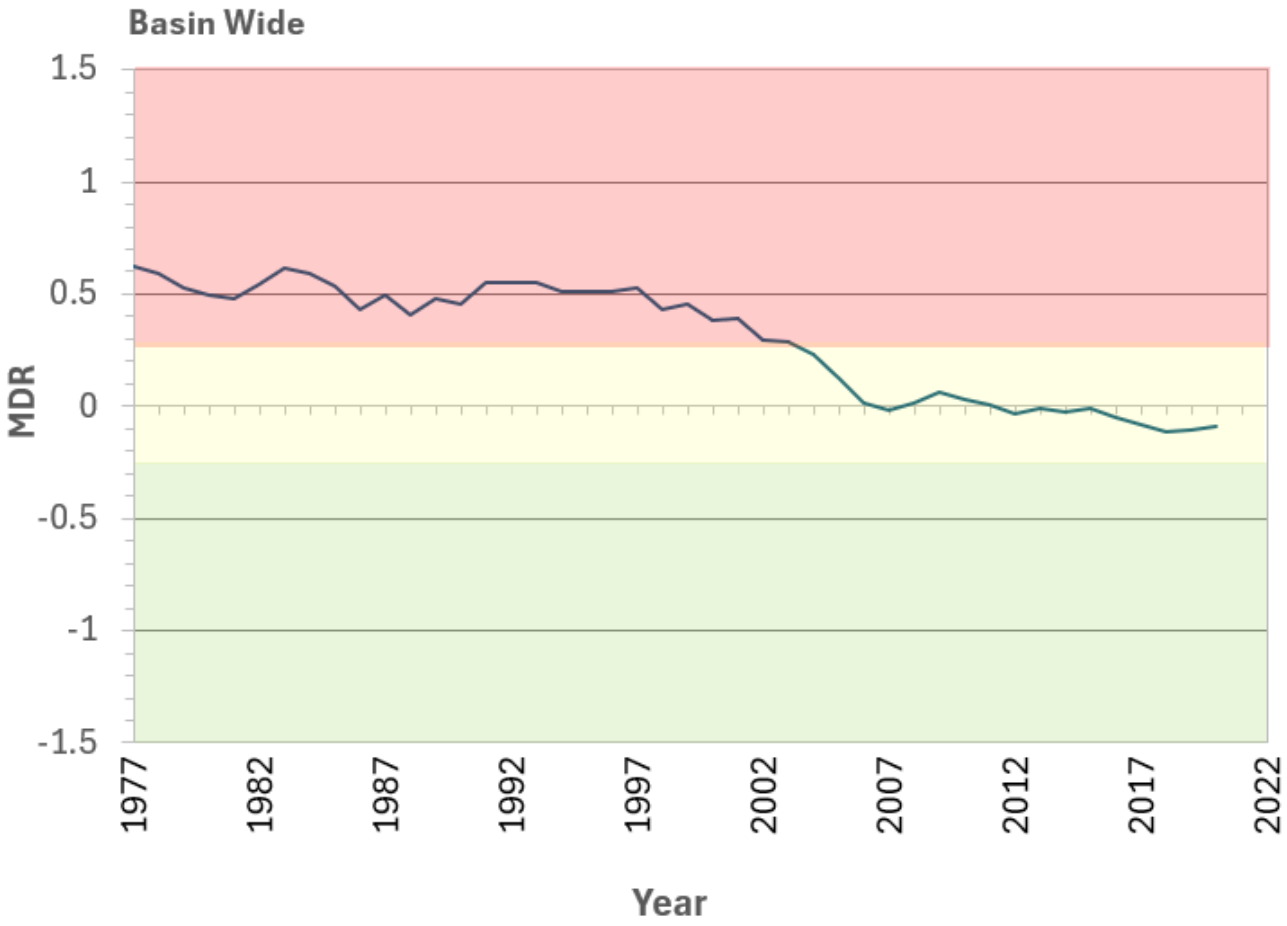


Figure 1. Mean Deviation Ratio (MDR) values (blue line) for the Great Lakes basin. Source: Environment and Climate Change Canada and U.S. Environmental Protection Agency

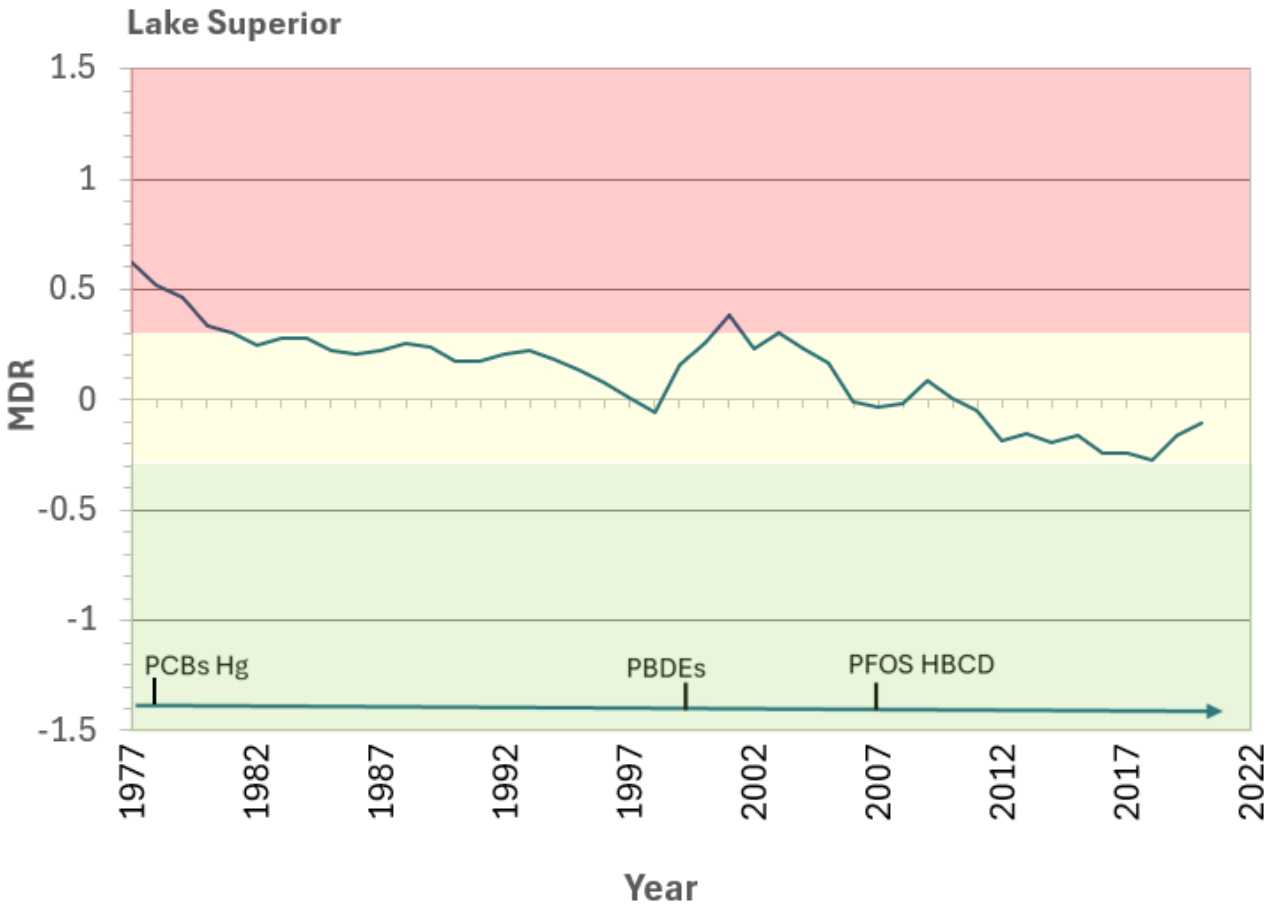


Figure 2. Mean Deviation Ratio (MDR) for Lake Superior. The timeline indicates the year in which different contaminants were added to the MDR calculation. Source: Environment and Climate Change Canada and U.S Environmental Protection Agency.

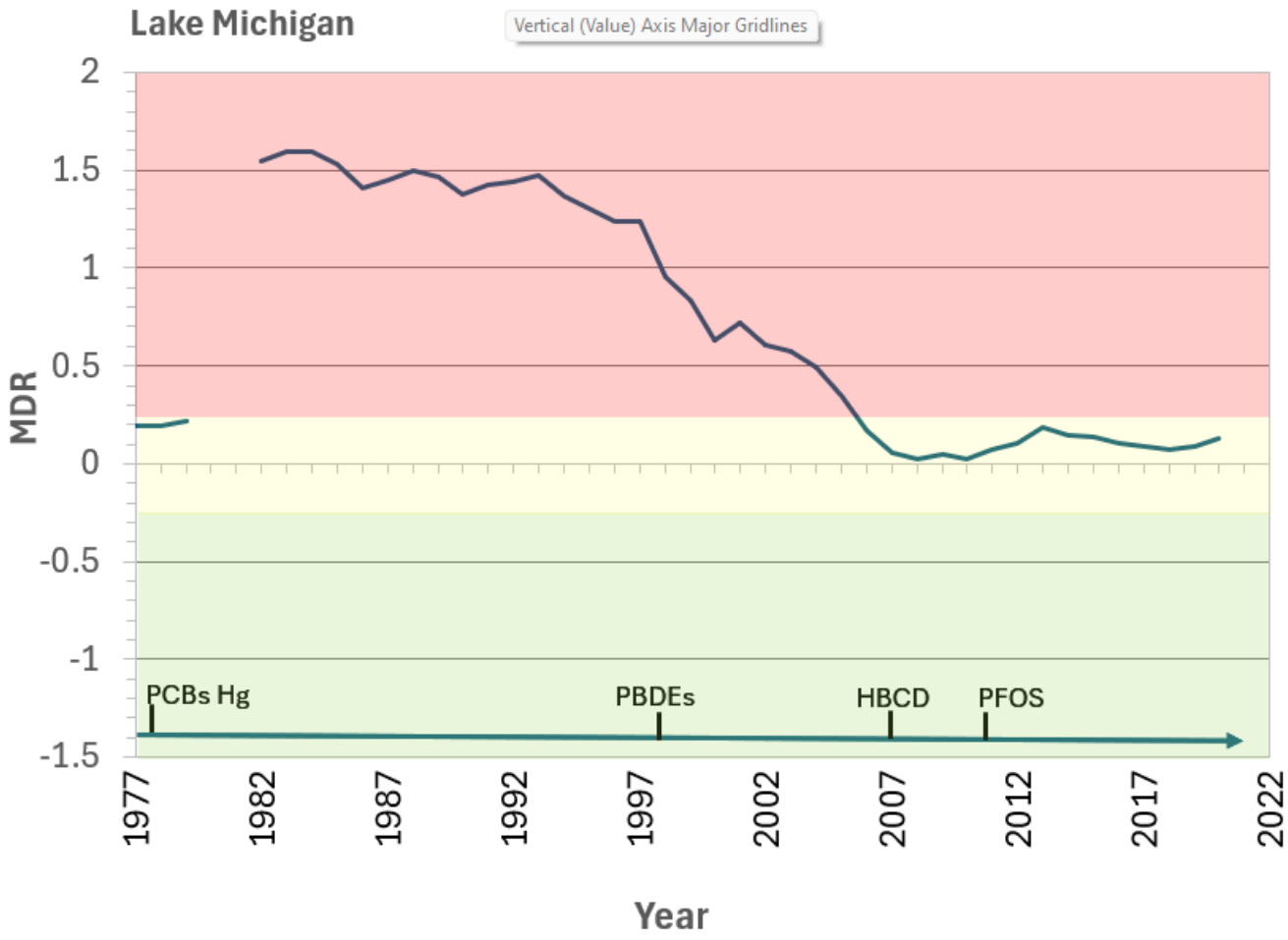


Figure 3. Mean Deviation Ratio (MDR) for Lake Michigan. The timeline indicates the year in which different contaminants were added to the MDR calculation. Source: Environment and Climate Change Canada and U.S. Environmental Protection Agency

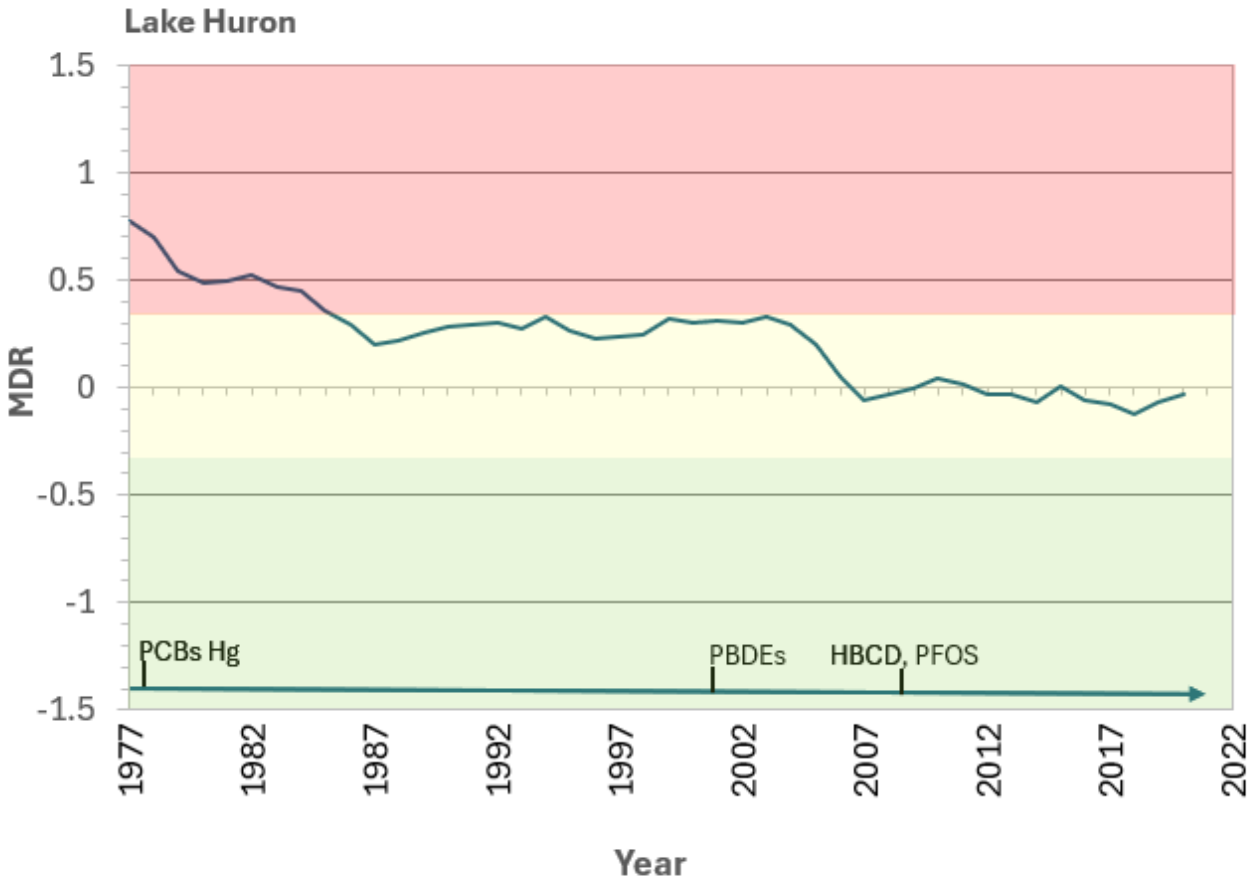


Figure 4. Mean Deviation Ratio (MDR) for Lake Huron. The timeline indicates the year in which different contaminants were added to the MDR calculation. Source: Environment and Climate Change Canada and U.S. Environmental Protection Agency

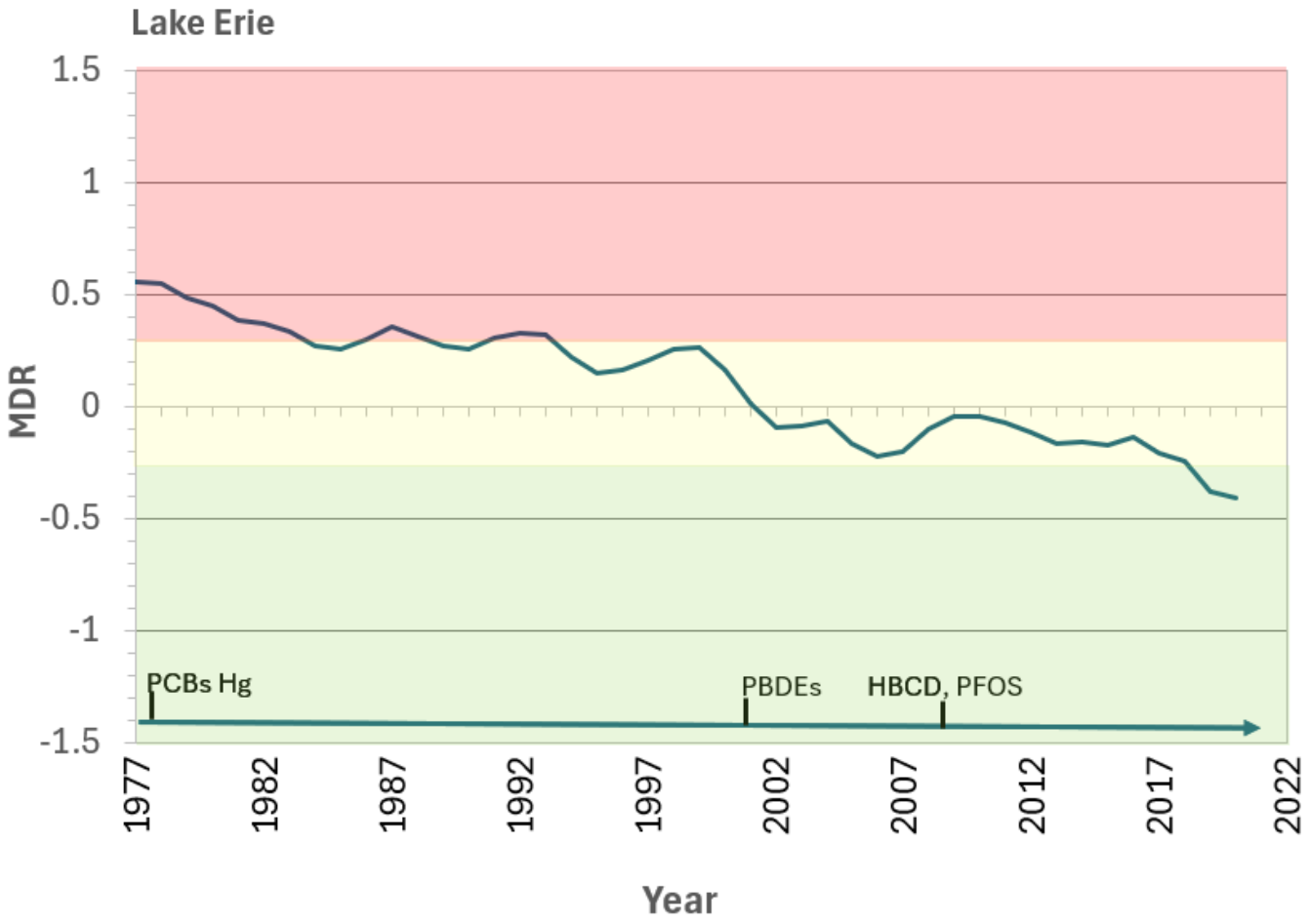
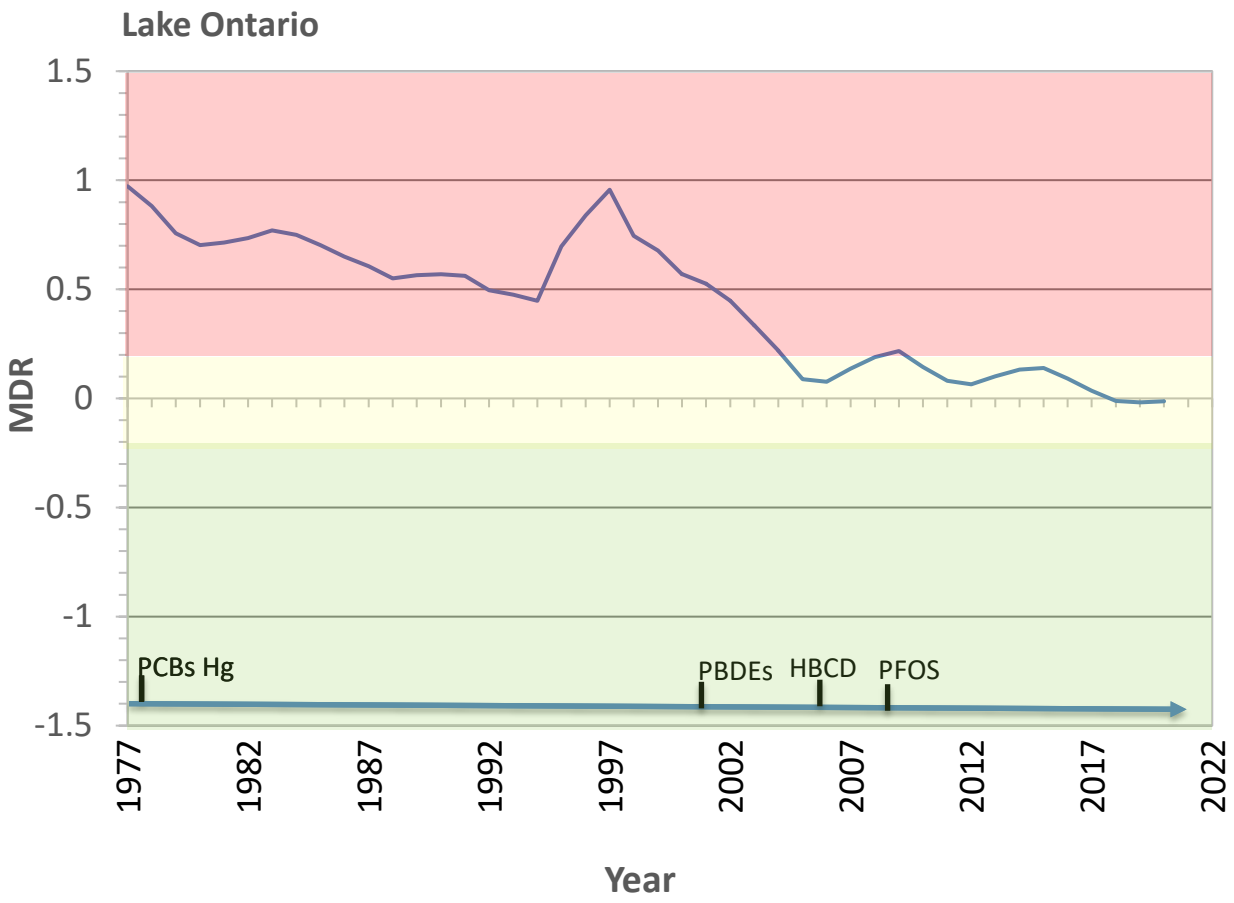


Figure 5. Mean Deviation Ratio (MDR) for Lake Erie. The timeline indicates the year in which different contaminants were added to the MDR calculation. Source: Environment and Climate Change Canada and U.S. Environmental Protection Agency



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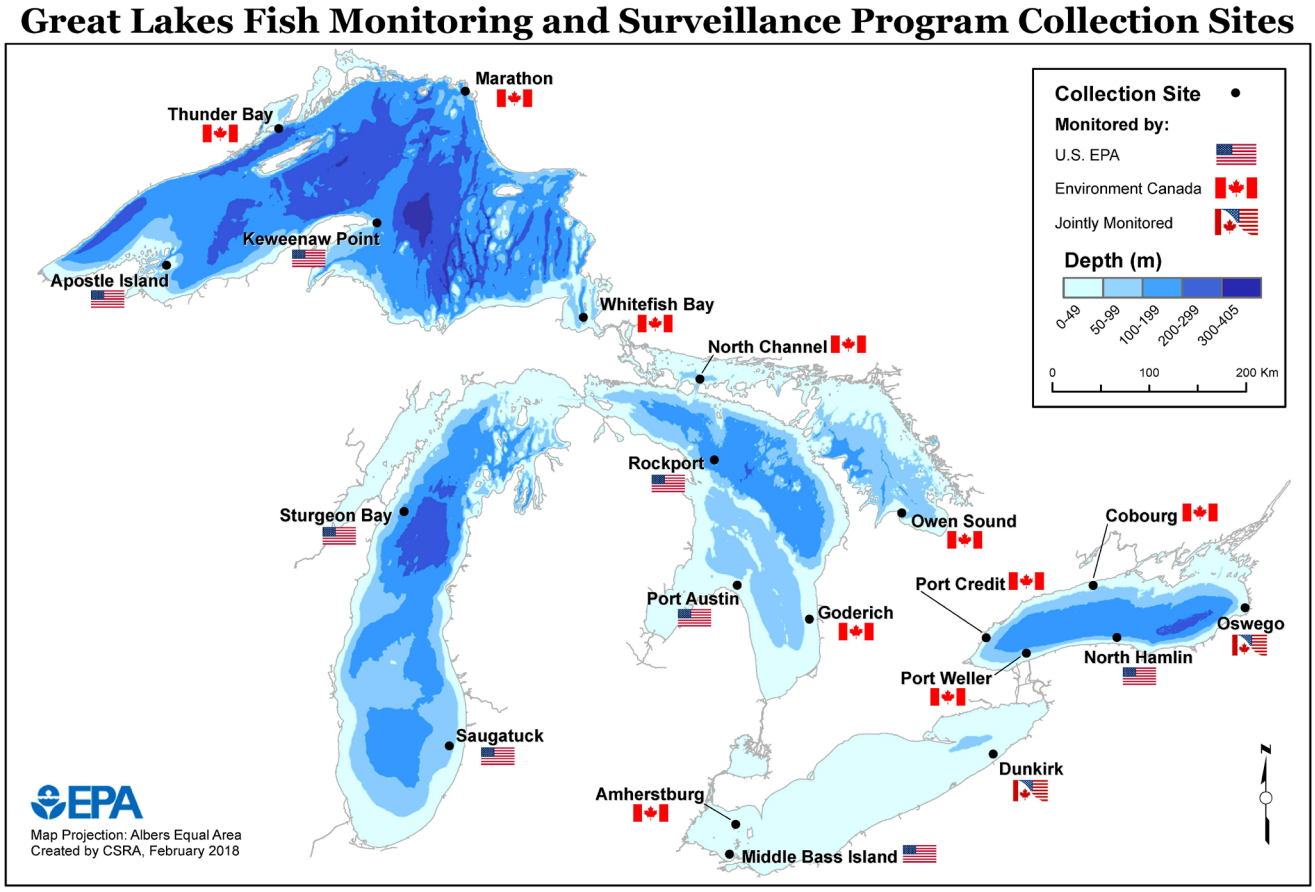

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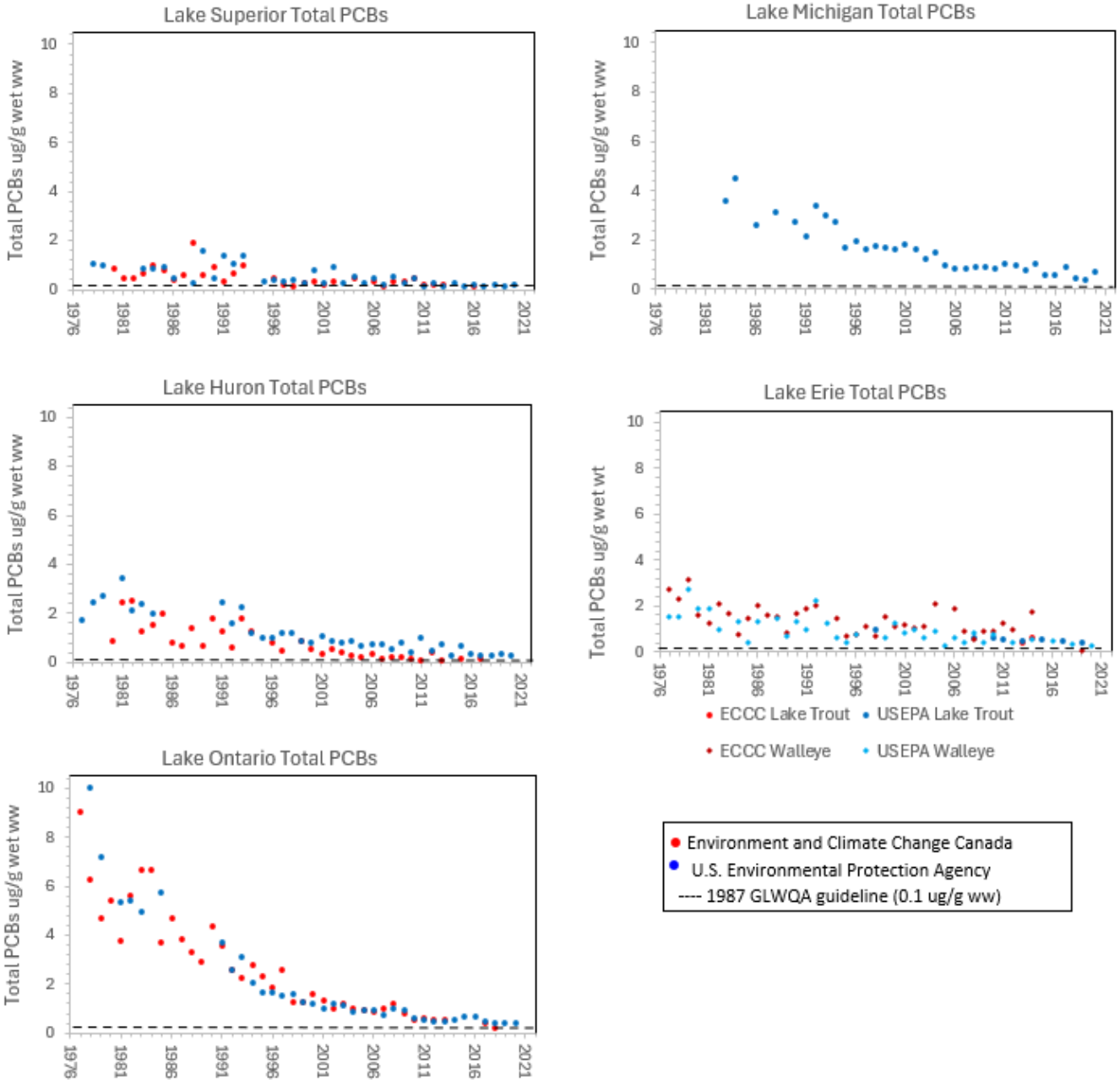

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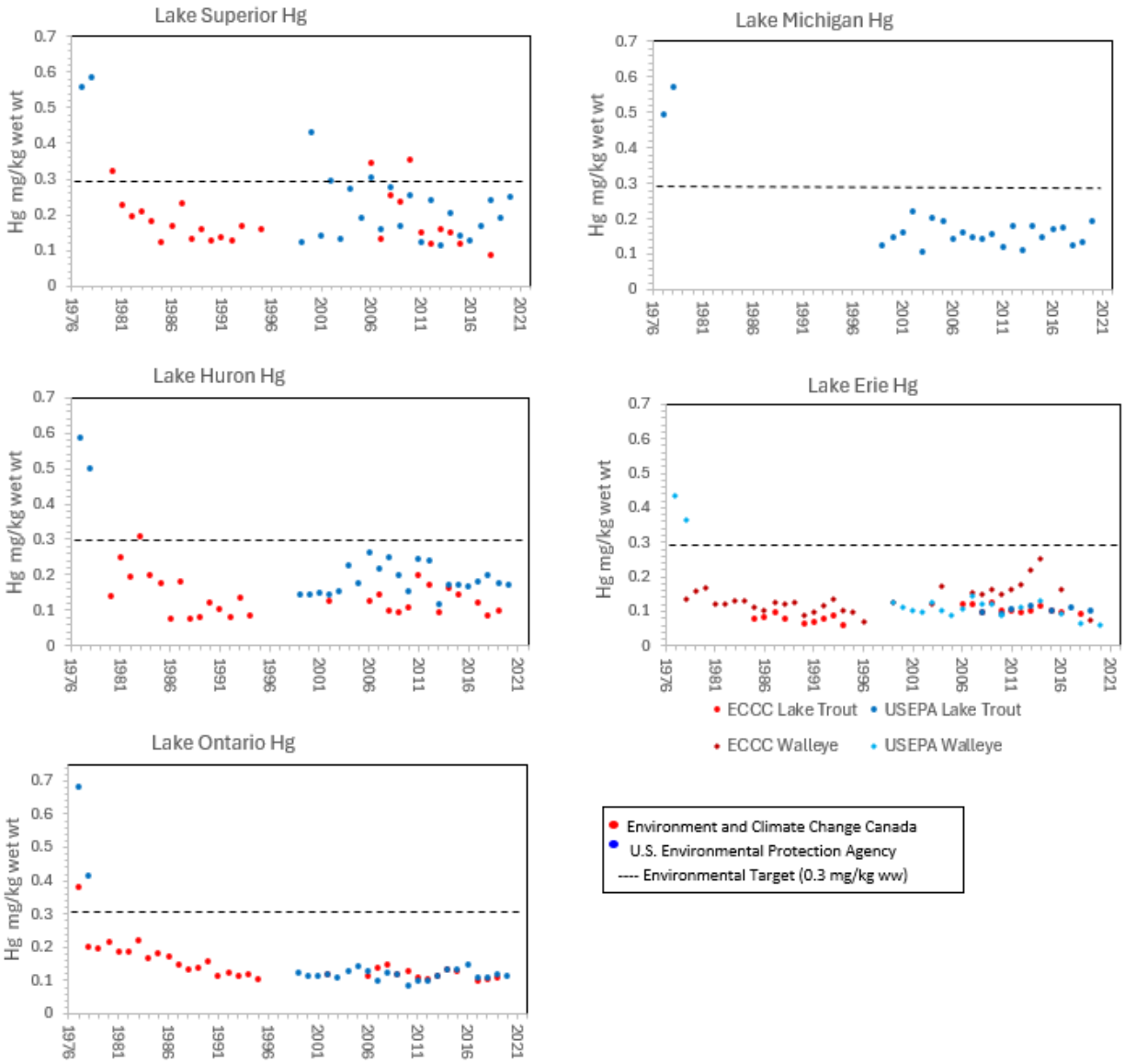

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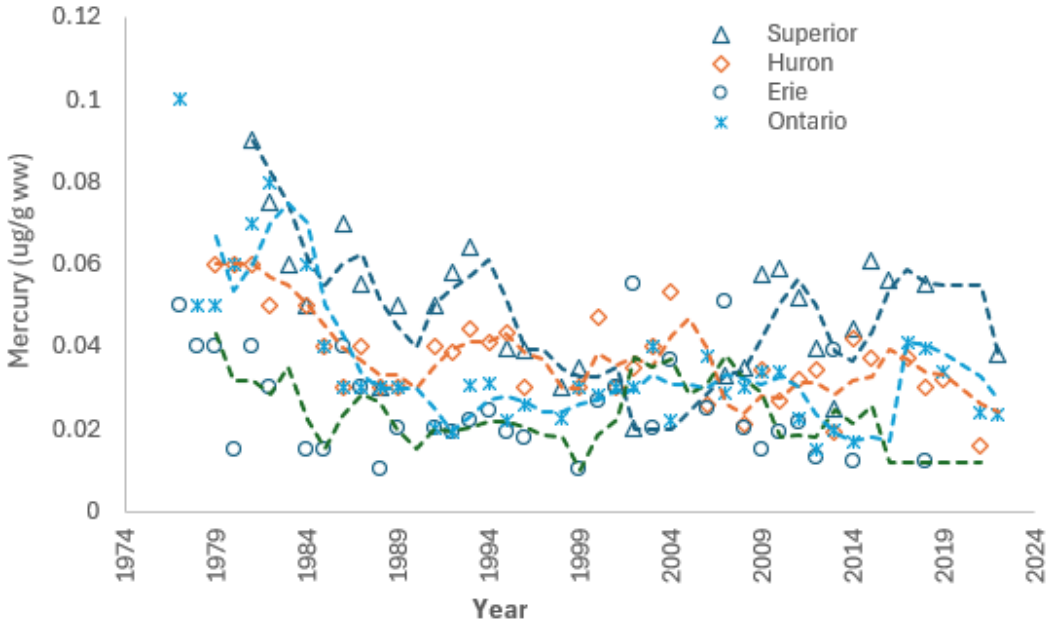

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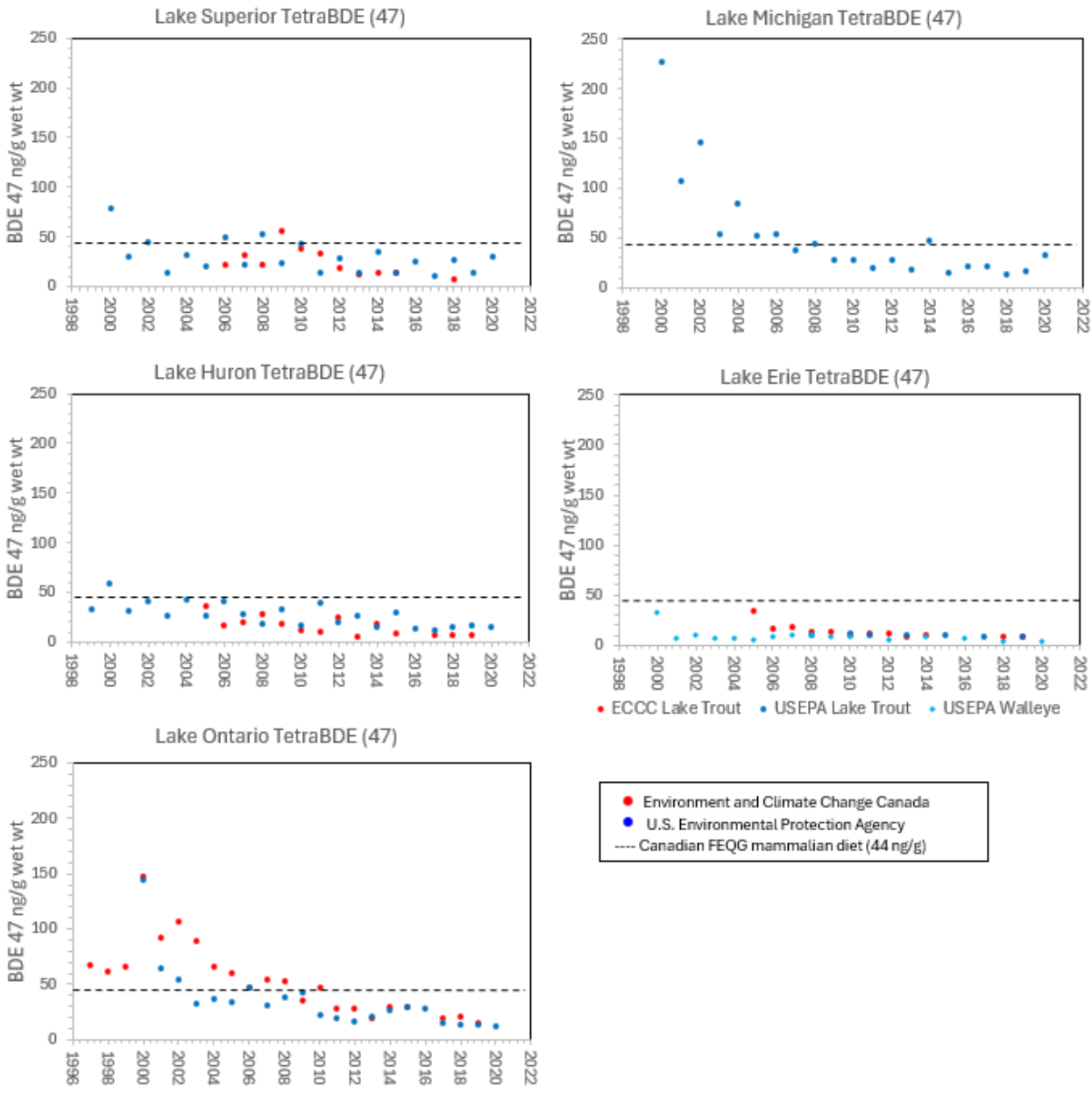

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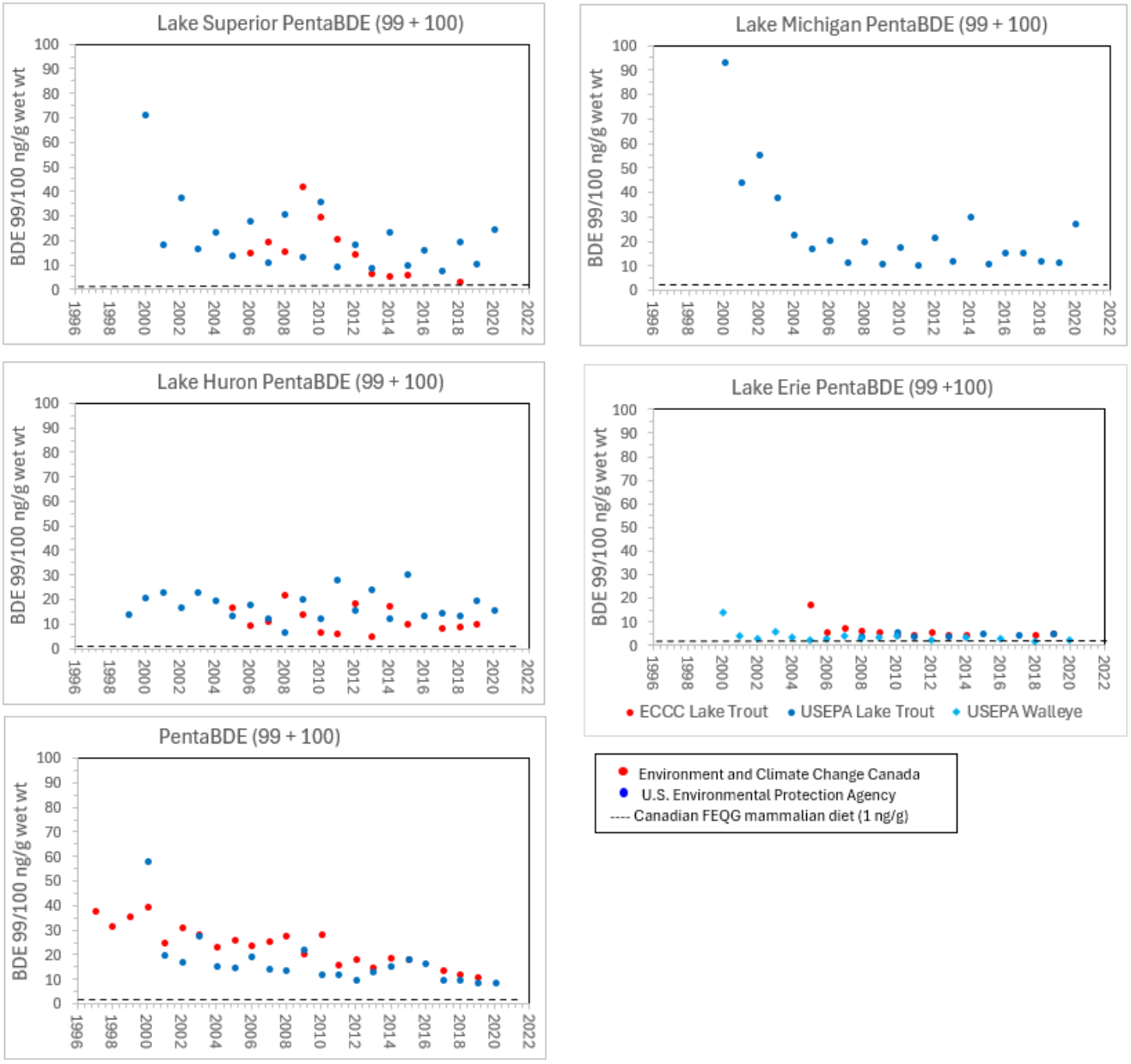

Figure 12. Mean annual penta BDE concentrations (wet weight) for individual (Environment and Climate Change Canada) and composited (U.S. Environmental Protection Agency) whole body Lake Trout or Walleye (Lake Erie, EPA only, western basin) collected from each of the Great Lakes. Canadian guidelines for fish tissue are given for context. Source: Environment and Climate Change Canada and U.S. Environmental Protection Agency

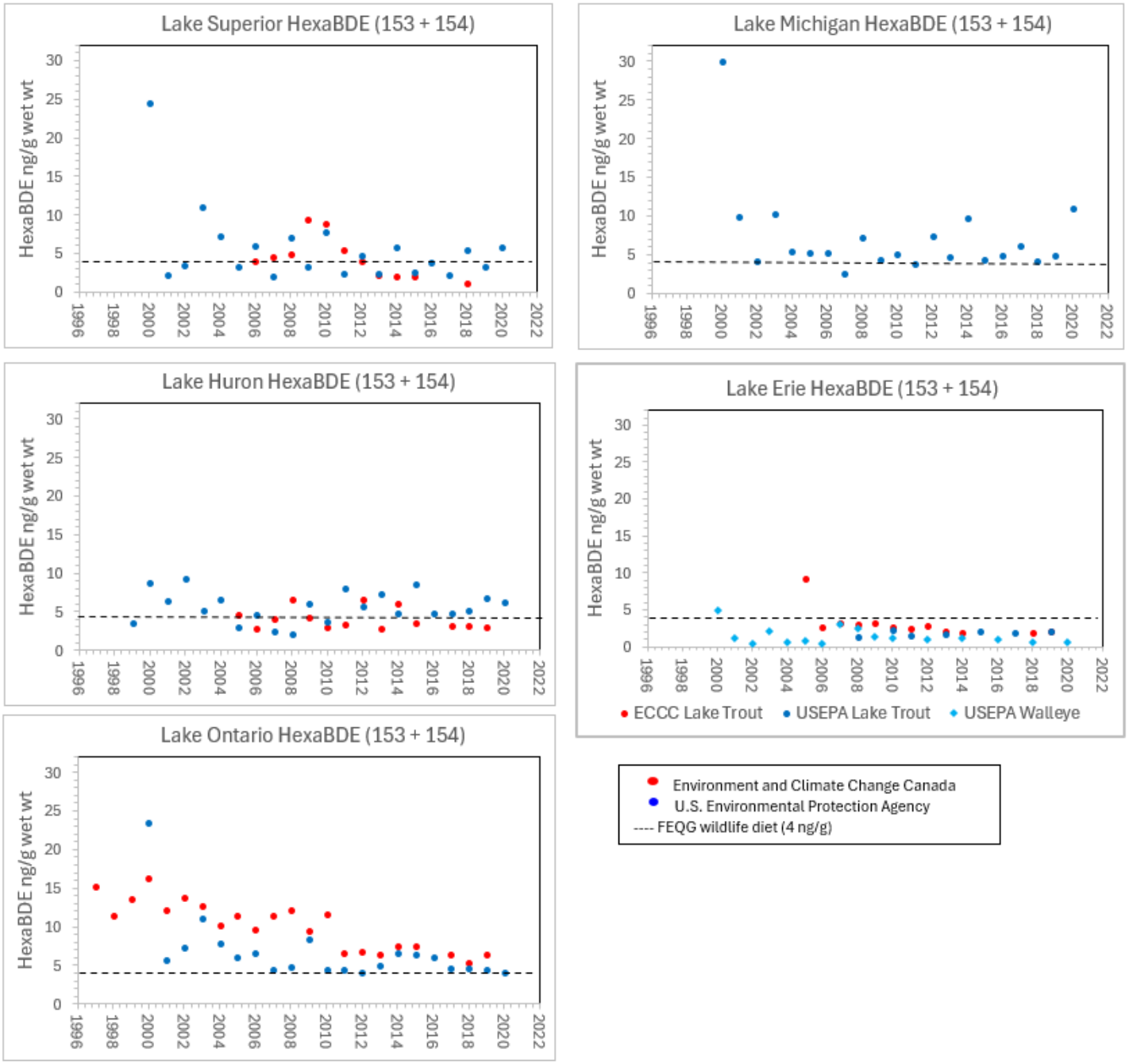

Figure 13. Mean annual HxBDE concentrations (wet weight) for individual (Environment and Climate Change Canada) and composited (U.S. Environmental Protection Agency) whole body Lake Trout or Walleye (Lake Erie, EPA only, western basin) collected from each of the Great Lakes. Canadian guidelines for avian diet are shown for context. Source: Environment and Climate Change Canada and U.S. Environmental Protection Agency

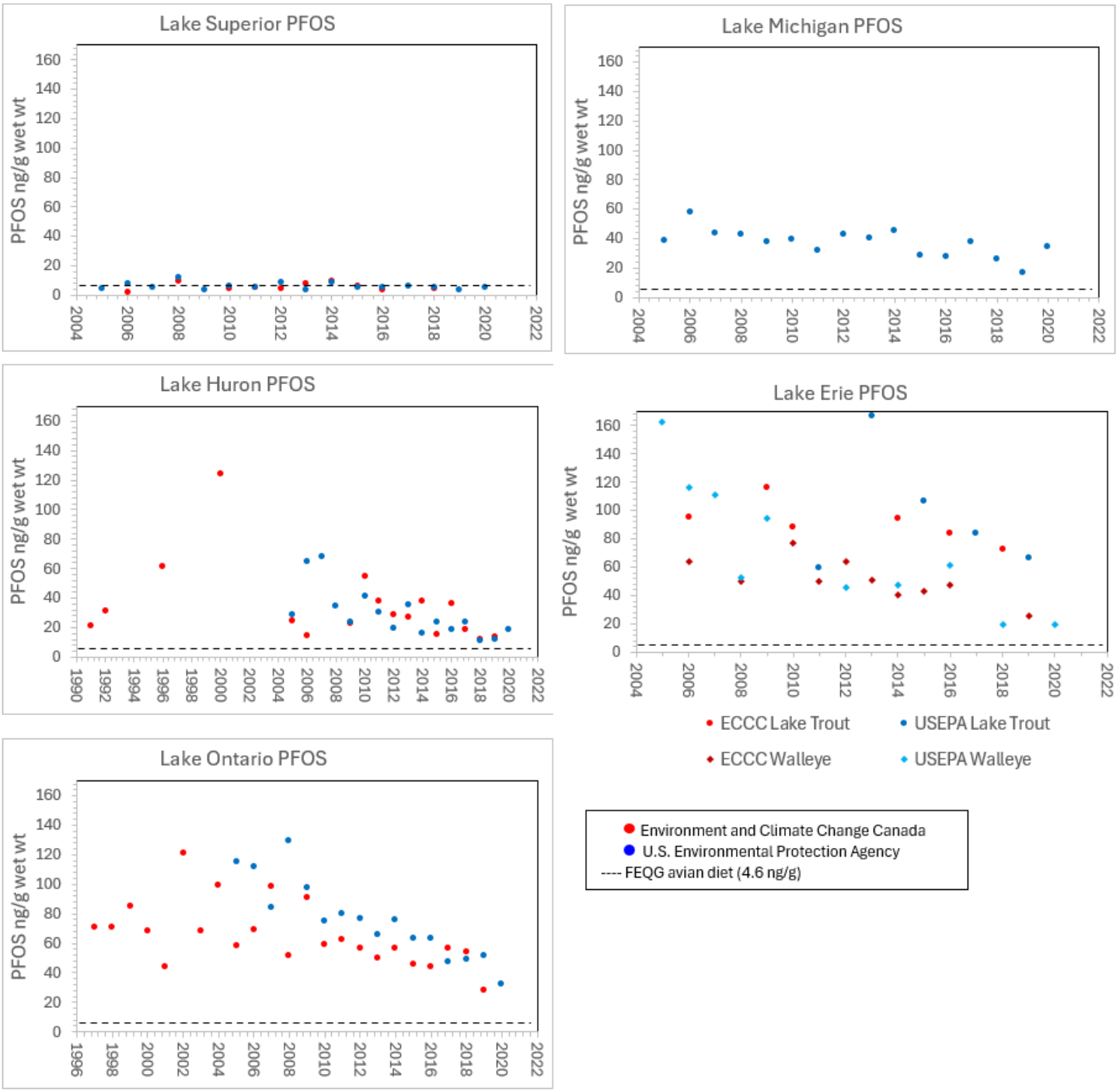

Figure 14. Mean annual PFOS concentrations (wet weight) for individual (Environment and Climate Change Canada) and composited (U.S. Environmental Protection Agency, Point et al. 2021.) whole body Lake Trout or Walleye (Lake Erie, EPA only, western basin) collected from each of the Great Lakes. Bars represent standard deviations around the mean. Canadian guidelines for avian diet are shown for context. Source: Environment and Climate Change Canada, U.S. Environmental Protection Agency

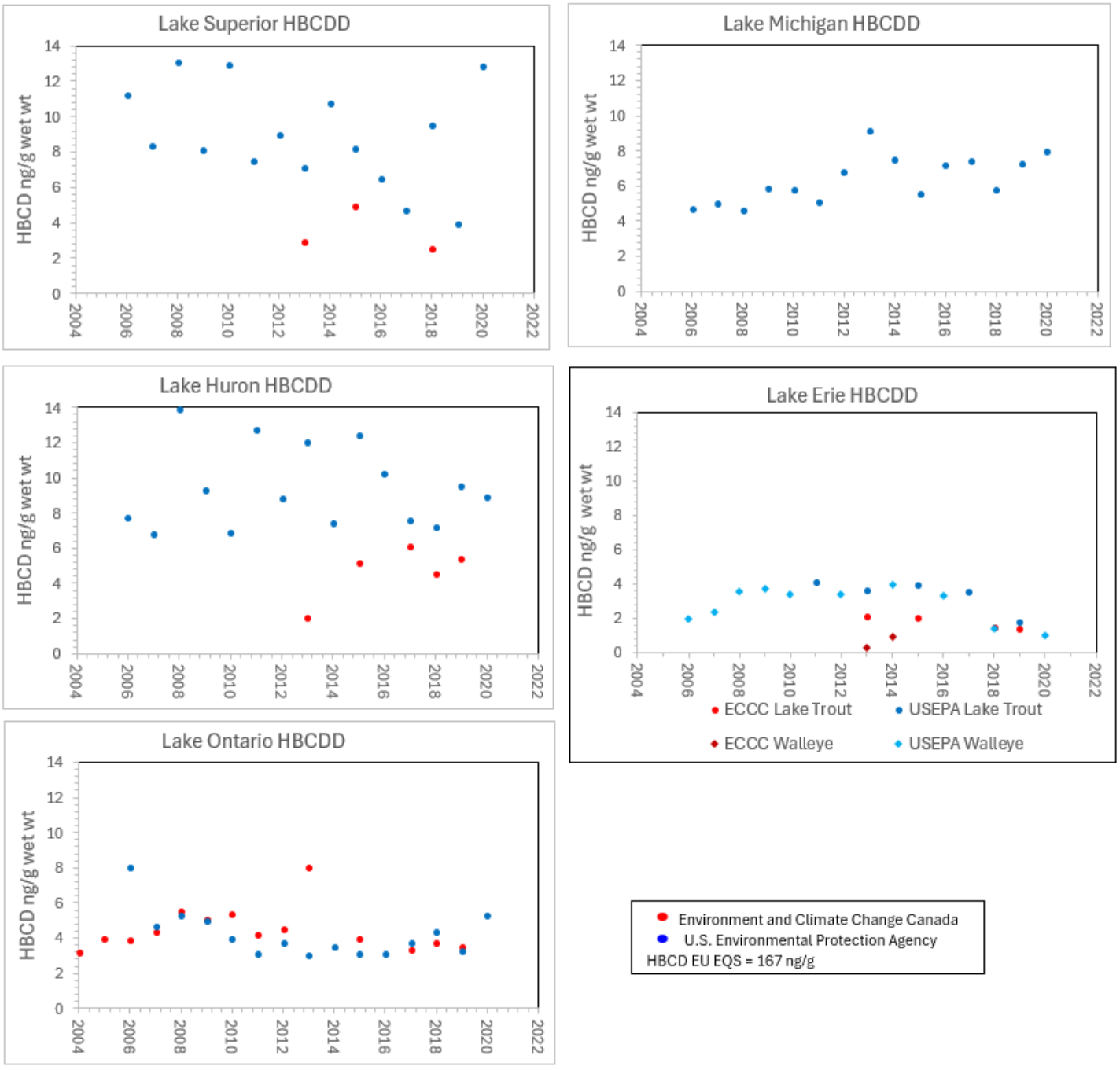

Figure 15. Mean annual total HBCD concentrations (wet weight) for individual (Environment and Climate Change Canada) and composited (U.S. Environmental Protection Agency) whole body Lake Trout or Walleye (Lake Erie open dots) collected from each of the Great Lakes.
Source: Environment and Climate Change Canada and U.S. Environmental Protection Agency