GREAT LAKES FISH MONITORING AND SURVEILLANCE PROGRAM

TECHNICAL REPORT

Status and Trends of Contaminants in Whole Fish through 2017


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1 EXECUTIVE SUMMARY

The U.S. Environmental Protection Agency (EPA) Great Lakes National Program Office (GLNPO) Great Lakes Fish Monitoring and Surveillance Program (GLFMSP) is a long-term monitoring program designed to: 1) collect, analyze, and report contaminant concentrations in Great Lakes top-predator fish (Lake Trout and Walleye), 2) improve understanding of contaminant cycling throughout food webs in the Great Lakes, and 3) screen for emerging chemicals in fish tissue to identify priority chemicals warranting future trend analysis and study. Samples collected for the GLFMSP are screened for emerging chemicals and analyzed for several different classes of contaminants including polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), mercury, hexabromocyclododecane (HBCDD), per- and polyfluoroalkyl substances (PFAS), toxaphene, chlordanes, and other organochlorine pesticides (OCPs).

This report presents summarized data and trends for PCBs, PBDEs, mercury, HBCDD, and PFAS in Lake Trout (*Salvelinus namaycush*) and contaminants of emerging concern (CEC) screening analyses in Lake Trout for the five GLFMSP sites sampled in odd years. The analytical results from 2017 are placed into the context of long-term trends beginning when each contaminant was first subjected to routine monitoring, with the exception of the Dunkirk, Lake Erie eastern basin site. Collection of Lake Trout in the eastern basin of Lake Erie began in 2008; therefore, trends from 2008-2017 are reported for Lake Erie. Trends in the 2017 technical report may differ from trends reported in the 2016 technical report due to local factors at the different sampling sites within each lake.

An assessment of data through 2017 shows that concentrations of several contaminants are decreasing in Lake Trout. Key highlights of the concentration trends include:

- Mean total PCB concentrations in Lake Trout have declined at the odd-year sampling sites in Lakes Huron, Michigan, Superior, and Ontario from 1991 to 2017. Concentrations have also declined in the eastern basin of Lake Erie since monitoring of Lake Trout began in 2008.
- Mean total PBDE concentrations in Lake Trout have declined at the odd-year sampling sites in lakes Michigan, Ontario, and Superior since 2001. No significant changes in Lake Trout at the Port Austin sampling site were found in this timeframe. Concentrations have also declined in eastern basin of Lake Erie since monitoring of Lake Trout began in 2008.
- Mercury concentrations in Lake Trout have declined at the Port Austin sampling site in Lake Huron since 2007 and increased at the odd-year sampling site in Lake Ontario and Erie since 2007 and 2008, respectively. Lake Trout collected at the odd-year sampling sites in lakes Superior and Michigan did not show statistically significant changes in mercury concentrations from 1999 to 2017. No statistically significant changes occurred at any sampling site since 1999.

The most abundant CEC compound class detected in Lake Trout in 2017 in all Lakes was halomethoxyphenols. Only 2017 CEC screening results are presented, as there are currently not enough years of data to evaluate temporal trends for CECs. In 2017, mean total HBCDD in Lake Trout was highest at Port Austin sampling site in Lake Huron and lowest in eastern basin of Lake Erie. In 2017, mean concentrations of total PFAS and perfluorooctanesulfonic acid (PFOS) (a PFAS compound) tended to be highest in Lake Trout in the eastern basin of Lake Erie and lowest at Keweenaw Point in Lake Superior.

Field and biological data collection results are presented for Lake Trout, forage fish, and invertebrates that were collected by the GLFMSP in support of the 2017 Lake Huron Cooperative Science and Monitoring Initiative (CSMI) studies of contaminant cycling in the Lake Huron food web. Analytical results of the CSMI studies will be presented in future GLFMSP reports.

2 INTRODUCTION

The U.S. Environmental Protection Agency (EPA) Great Lakes National Program Office (GLNPO) Great Lakes Fish Monitoring and Surveillance Program (GLFMSP) is a long-term monitoring program that was initiated in 1977 and designed to: 1) collect, analyze, and report contaminant concentrations in Great Lakes top-predator fish (Lake Trout and Walleye), 2) improve understanding of contaminant cycling throughout food webs in the Great Lakes, and 3) screen for emerging chemicals in fish tissue to identify priority chemicals warranting future trend analysis and study. Lake Trout and Walleye are targeted by the GLFMSP for biomonitoring because these top predator fish occupy the highest trophic levels in the Great Lakes aquatic food web and as such, tend to accumulate higher levels of persistent and bioaccumulative contaminants (McGoldrick and Murphy, 2016).

The present design of the GLFMSP includes two components: 1) Base Monitoring Program and 2) Cooperative Science and Monitoring Initiative (CSMI)/Special Studies.

The GLFMSP helps EPA satisfy its statutory requirements under Section 118 of the Clean Water Act to establish a Great Lakes system-wide surveillance network to monitor the water quality of the Great Lakes (33 U.S.C. § 1268 *et seq.*) with a specific emphasis on the monitoring of toxic pollutants. It also helps satisfy the Agency's obligations under the Great Lakes Water Quality Agreement (GLWQA) to "monitor environmental conditions so that the Parties may determine the extent to which General Objectives, Lake Ecosystem Objectives, and Substance Objectives are being achieved," and "undertake monitoring and surveillance to anticipate the need for further science activities and to address emerging environmental concerns" (GLWQA 2012). Further, this program allows EPA to meet commitments in the Great Lakes Restoration Initiative (GLRI) Action Plan III to "assess the overall health of the Great Lakes ecosystem and identify the most significant remaining problems" (GLRI 2019).

This report summarizes chemical and biological data collection results for the 2017 Base Monitoring Program and CSMI/Special Studies collection efforts and presents the 2017 Base Monitoring Program analytical results in context with long-term trends.

3 DESCRIPTION OF METHODS

This section summarizes methods for sample collection, biological data collection, homogenization, and analysis.

3.1 SAMPLE COLLECTION

Field sampling teams perform sample collections every year in the late summer to fall according to sample collection standard operating procedures (SOPs) (EPA 2012a) and deliver fish to a homogenization laboratory after collection. A total of seven sampling teams collected fish for the Base Monitoring Program and CSMI/Special Studies components in 2017 between June and November:

- Great Lakes Indian Fish and Wildlife Commission
- Michigan Department of Natural Resources Alpena Fisheries Research Station
- New York State Department of Environmental Conservation Lake Erie Fisheries Research Unit
- U.S. Fish and Wildlife Service (USFWS) Green Bay Fish and Wildlife Conservation Office
- U.S. Geological Survey (USGS) Great Lakes Science Center
- USGS Lake Ontario Biological Station

Detailed information on collection methods can be found in the subsections below.

3.1.1 Base Monitoring Program

Top predator fish are collected at two sites in each of the Great Lakes with sites alternating within each lake annually (Figure 1) for the Base Monitoring Program. Collection sites are intended to be representative of offshore conditions in each Lake. Lake Trout (*Salvelinus namaycush*) are collected in all lakes and Walleye (*Sander vitreus*) are collected at one site located in the western basin of Lake Erie which is too shallow to support Lake Trout. As the western basin of Lake Erie is sampled during even years, Walleye data are not included in this 2017 report. In 2011, after two years (2008 and 2010) of comparison of contaminant body burden in Lake Trout and Walleye, Lake Trout replaced Walleye as the GLFMSP target species in the eastern basin of Lake Erie. Lake Trout were found to be more readily available for collection at the eastern basin site (Dunkirk), and had comparable contaminant burdens to Walleye. Additionally, this change allowed the GLFMSP to compare contaminants in Lake Trout across all five Great Lakes. Lake Trout data collected in 2008 and 2010 at Dunkirk for the comparison study are included in this 2017 report. Lake Trout in the size range of 600-700 mm are targeted and Walleye in the size range of 400-500 mm are targeted for collection (target number of fish per site = 50). Fish size ranges were determined with the assumption that they represented specific age ranges, 6-8 years for Lake Trout and 4-5 years for Walleye. Detailed collection and site information for the GLFMSP Base Monitoring Program is located in the GLFMSP Quality Assurance Project Plan (QAPP) (EPA 2012a).

3.1.2 Cooperative Science and Monitoring Initiative (CSMI) / Special Studies

The Cooperative Science and Monitoring Initiative (CSMI) is a binational effort instituted under the 2012 GLWQA to coordinate science and monitoring activities in one of the five Great Lakes each year to generate data and information for environmental management agencies. The GLFMSP supports the CSMI via additional sample collection efforts and analyses to gather information regarding contaminant cycling throughout food webs in the Great Lakes. During the CSMI field year, fish are collected at both GLFMSP sites within the CSMI lake; in 2017, the CSMI lake was Lake Huron. Lake Trout in the size and age range collected as part of the Base Monitoring Program are targeted (target number of fish per site = 10). The top five most abundant species of forage fish in the CSMI lake are also collected at both sites when available (total target number of fish per site = 110). The GLFMSP cooperators collect sediment, benthic invertebrates, phytoplankton, zooplankton/seston and water samples in the CSMI lake aboard the Research Vessel (R/V) *Lake Guardian*. Detailed collection and site information for the GLFMSP CSMI/Special Studies component is provided in the GLFMSP QAPP (EPA 2012a).

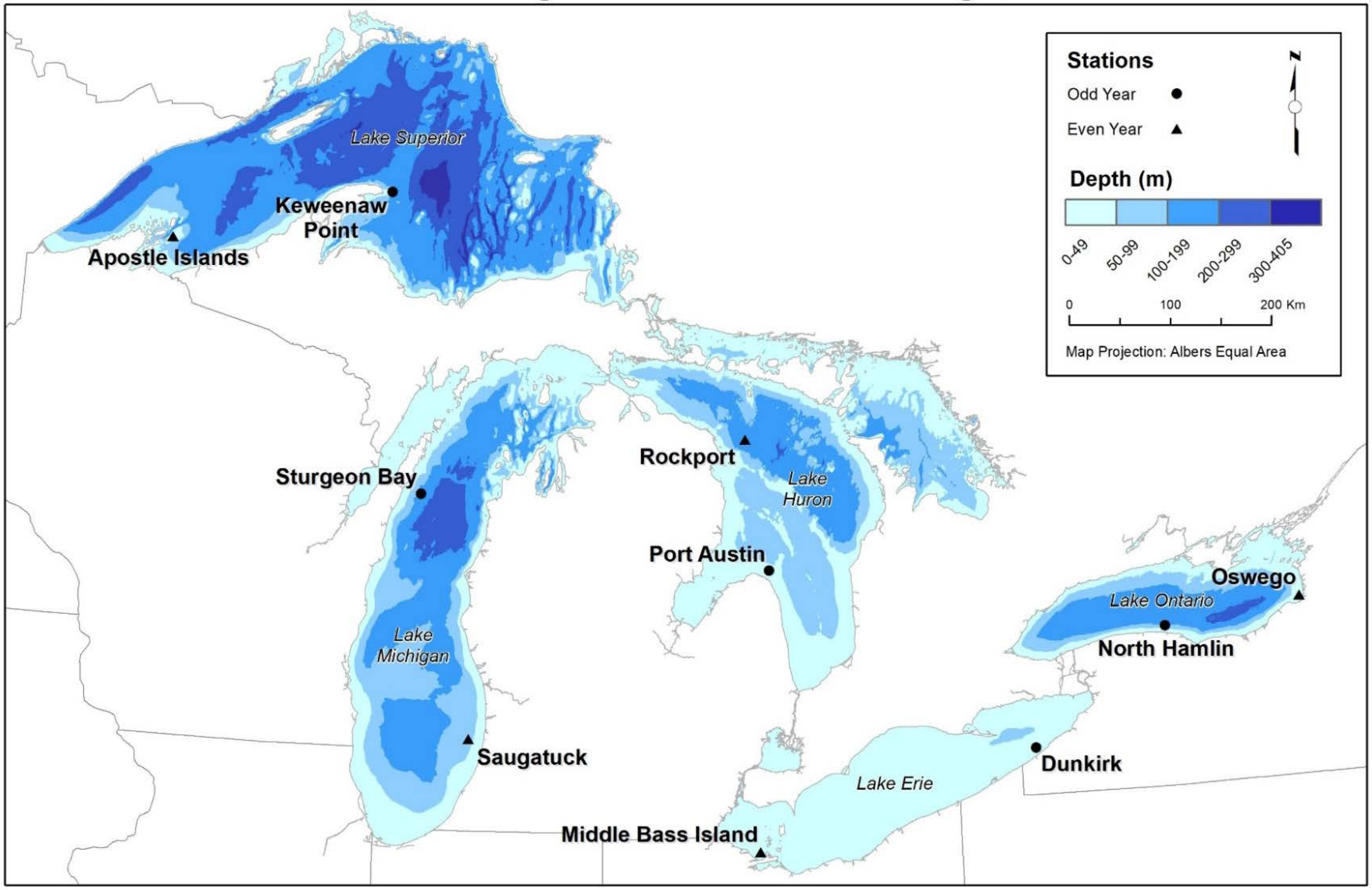

Figure 1: GLFMSP Collection Sites.

3.2 BIOLOGICAL DATA COLLECTION AND HOMOGENIZATION

The homogenization laboratory receives fish from the field sampling teams and processes these fish in the winter to spring time period. In 2017, the homogenization laboratory was Aquatec Environmental, Inc. (Aquatec). Aquatec follows approved GLFMSP-specific SOPs (Aquatec 2016) when processing samples.

The homogenization laboratory recorded biological data (e.g. length, width, weight) and any abnormalities (e.g., tumors, fins missing, wounds), collected samples for aging purposes (e.g., scales, maxillae, coded wire tags [CWTs]), and aged the fish. In 2017, Lake Trout age was determined based on annuli enumeration of maxillae. CWTs were also used to age Lake Trout when available. Fish age is an important variable when assessing contaminant trends and as such, the GLFMSP compositing scheme was amended in 2013 to group fish according to age (rather than by length) prior to homogenization and chemical analysis. More information on this change can be found in the *Journal of Great Lakes Research* publication "Revised fish aging techniques improve fish contaminant trend analyses in the face of changing Great Lakes food webs" (Murphy et al. 2018) and in the Great Lakes Fish Monitoring and Surveillance Program Technical Report: Status and Trends through 2016 (EPA 2020). EPA reviewed the ages for 2017 Lake Trout and assigned fish into five fish per composites (target number of composites per site = 10) based on age for sites where the target 50 fish were collected. At the Keweenaw Point site, a total of 42 Lake Trout were collected and one Lake Trout was accidentally discarded prior to being sent to the homogenization laboratory, so eight composites of five fish and a homogenized sample of one fish were created. At the Sturgeon Bay site, a total of 49 Lake Trout were collected, so nine composites of five fish and one composite of four fish were created.

After grouping fish into composites based on EPA's criteria noted above, the homogenization laboratory processed the whole fish and prepared composites of these samples. In addition, a mega-composite was prepared (i.e., tissue from all composites from a single site) where applicable for screening of contaminants of emerging concern. The single fish homogenate from Keweenaw Point was not included in the mega-composite for this site in 2017. The homogenization laboratory created tissue aliquots and delivered them to the analytical laboratory cooperator and to EPA's archival facility.

3.3 ANALYSIS

The analytical laboratory cooperator receives fish tissue aliquots from the homogenization laboratory in the spring of the year following the collection year. The analytical laboratory cooperators that analyzed the 2017 collected fish tissue were Clarkson University, State University of New York (SUNY) Oswego, SUNY Fredonia and AEACS, LLC. The 2017 Base Monitoring Program analytical data sets are presented in Table 1. All analytical data generated to support the GLFMSP are prepared in accordance with an approved QAPP and SOPs (Clarkson University 2016).

Upon sample receipt, the analytical laboratory cooperator analyzed the homogenized tissue for different classes of contaminants including PCBs, PBDEs, mercury, HBCDD, PFAS, toxaphene, chlordanes, and other organochlorine pesticides (OCPs). The analytical laboratory cooperator also utilized mega-composite samples collected for the Base Monitoring Program to determine the presence of CECs. Following data review by EPA, the data are used for reporting and made available to the public in the Great Lakes Environmental Database (GLENDA), and can also be requested from EPA (contact information is provided on page ii of this report).

Table 1: 2017 Base Monitoring Program Analytical Data Sets

Collection Effort	Analytes
Composites and mega-composites	• Percent Moisture • Mercury • PCBs/OCPs/PBDEs/Lipids/Mirex • Toxaphene
Composites only	• PFAS
Mega-composites only	• Dioxins / Furans & Coplanar PCB congeners • HBCDD • CECs

Results generated by all analytical methods were reported on a wet weight basis in accordance with SOPs (Clarkson University 2016). No mathematical adjustments based on lipid content or fish age were performed on the 2017 results or as part of the trend analyses presented in this report. Long-term analytical data in the GLFMSP presented in this report have not been corrected to adjust for fish age the reason being that fish have only been aged since 2003 as part of the sampling process and historically were grouped into estimated age composites according to length measurements. To ensure consistency in how data are reported, publicly available data for GLFMSP are reported as contaminant concentrations for each composite for a given sampling year at each collection site. Age-corrected data from the 2017 GLFMSP collected fish are presented in Pagano et al. (2018), Zhou et al. (2018), Zhou et al. (2019), Pagano et al. (2019), Parvizian et al. (2020), and Pagano et al. (2020).

4 QUALITY ASSURANCE AND CONTROL

The GLFMSP operates under a quality management plan (QMP), a QAPP, and numerous SOPs. The GLFMSP quality management system is defined in the GLFMSP QMP (EPA 2012b). Quality assurance/quality control (QA/QC) activities and procedures associated with the sample collection, biological data collection, homogenization, and analysis of fish samples are described in the QAPPs and SOPs identified in Section 3.

Several types of laboratory QC measures including equipment blanks, standard reference materials, blind duplicates, method blanks, replicate samples and surrogate spikes, are implemented at both the homogenization laboratory and the analytical laboratory to monitor data quality. These measures assist in identifying and correcting problems as they occur. They also define the quality of data generated by the program. QC metrics are tailored to specific sample and analytical processes. The analytical laboratory cooperator’s QAPP provides specific QC requirements to identify background contamination and extraction efficiency and ensure accurate identification and quantification of targeted analytes. If any QC criteria are not met, the data are reviewed carefully to identify the cause of the problem and determine the appropriate corrective action. If reanalysis is not warranted, the data are submitted with QC flags to indicate the nature of the failure.

To date, no major QA/QC issues have been identified through 2017.

5 RESULTS

This section summarizes results from 2017 sample collection, biological data collection, and analysis, and presents the 2017 Base Monitoring Program analytical results in context with long-term trends.

5.1 SAMPLE COLLECTION

5.1.1 Base Monitoring Program

A total of 241 Lake Trout were collected in Lakes Erie, Huron, Michigan, Ontario, and Superior in 2017 (Table 2). Due to low availability of Lake Trout in the target size range at two collection sites, a total of 42 Lake Trout were collected at Keweenaw Point and a total of 49 Lake Trout were collected at Sturgeon Bay instead of the target 50.

Table 2: 2017 Base Monitoring Program Field Data

Lake	Site	Species	Date	Sampling Depth (m)	Collection Method	Field Length Range (mm)	Field Weight Range (g)
Erie (n=50)	Dunkirk	Lake Trout	August 2017	30.5-39.6	Gillnet	582-719	2100-5300
Huron (n=50)	Port Austin	Lake Trout	September, October 2017	40	Trap Net	570-715	1630-3768
Michigan (n=49)	Sturgeon Bay	Lake Trout	September, October, November 2017	6.1-16.2	Gillnet	618-892	2010-6630
Ontario (n=50)	North Hamlin	Lake Trout	September 2017	25-35	Gillnet	550-762	1525-5450
Superior (n=42)	Keweenaw Point	Lake Trout	October, November 2017	7.3-9.1	Gillnet	554-846	1400-5500

5.1.2 Cooperative Science and Monitoring Initiative (CSMI) / Special Studies

In 2017, 20 additional Lake Trout were collected in Lake Huron, from Rockport and Port Austin (Table 3). A total of 490 forage fish were collected from Rockport and Port Austin (Table 4). Sediment, benthic invertebrates, zooplankton, *Mysis*, and water samples were also collected from both Lake Huron sites during a dedicated R/V *Lake Guardian* CSMI survey (Table 5).

Table 3: 2017 CSMI Lake Trout Field Data

Lake	Site	Date	Depth (m)	Collection Method	Field Length Range (mm)	Field Weight Range (g)
Huron (n=10)	Rockport	October 2017	10	Gillnet	585-820	2315-4850
Huron (n=10)	Port Austin	September, October 2017	40	Trap Net	555-805	1636-5344

Table 4: 2017 CSMI Forage Fish Field Data

Lake	Site	Species Collected	Date	Depth (m)	Collection Method
Huron	**Rockport**	• Rainbow Smelt (n=230) • Bloater (n=39) • Deepwater Sculpin (n=28) • Round Goby (n=30) • Yellow Perch (n=3)	October 2017	9-64	Bottom Trawl
Huron	**Port Austin**	• Rainbow Smelt (n=90) • Bloater (n=60) • Deepwater Sculpin (n=6) • Round Goby (n=4)	October 2017	18-73	Bottom Trawl

Table 5: 2017 CSMI R/V *Lake Guardian* Collected Field Data

Lake	Site	Sample Type and Sampling Depth (m)	Date	Collection Method
Huron	**Rockport**	Water (2 m)	June 2017	Surface water (~1000L) was collected using a submersible and peristaltic pump in series. The water was then passed through two pentaplates fitted with nylon (10µm) and glass fiber filters (0.7µm), respectively, to remove particulate matter. A tertiary downstream pump was then used to pass filtered water through resin (Porapak) columns to collect dissolved phase contaminants.
		Zooplankton (60 m for vertical/horizontal tows and 28 m for Tucker Trawl)	June 2017	Bulk material was size fractionated on the boat using different mesh size screens (500, 243, 118, 63 µm). All samples from vertical and horizontal tows for a specific size class were combined to maximize the mass for analysis.
		Mysis (63 m)	June 2017	Benthic sled (500 µm net) and Vertical Tow (500 µm net and 250 µm cod end)
		Sediment (60 m)	June 2017	Ponar
		Mussels (63 m)	June 2017	Benthic sled (500 µm net)
Huron	**Port Austin**	Water (2 m)	June 2017	Pump (see Rockport description)
		Zooplankton (35 m for vertical/horizontal tows and 28 m for Tucker Trawl)	June 2017	Bulk material was size fractionated on the boat using different mesh size screens (500, 243, 118, 63 µm). All samples from vertical and horizontal tows for a specific size class were combined to maximize the mass for analysis. Samples were also collected using a Tucker Trawl (500 µm).
		Mysis (36 m)	June 2017	Benthic sled (500 µm net) and Vertical Tow (500 µm net and 250 µm cod end)
		Sediment (36 m)	June 2017	Ponar
Huron	**Rockport and Port Austin (Combined)**	Benthic Invertebrates (36-60m)	June 2017	Benthic sled (500 µm net) and Ponar

5.2 BIOLOGICAL DATA COLLECTION AND HOMOGENIZATION

Tables 6 and 7 provide a summary of biological data measurements (excluding age results which are included in Table 8) as recorded by the homogenization laboratory for the 2017 Base Monitoring Program and CSMI/Special Studies samples.

Table 6: 2017 Base Monitoring Program Biological Data

Lake	Site	Species	Lab Length Range (mm)	Lab Weight Range (g)	Gender Count (M, F)	Dominant Maturity Stage [b, c]
Erie (n=50)	Dunkirk	Lake Trout	565-694	2202-5324	25, 25	Gravid (48%), Mature (50%)
Huron (n=50)	Port Austin	Lake Trout	553-688	1588-3659	25, 25	Gravid (46%), Mature (50%)
Michigan (n=49)	Sturgeon Bay	Lake Trout	594-889	1994-6524	41, 8	Mature (84%)
Ontario (n=50)	North Hamlin	Lake Trout	510-736	1513-5405	32, 18	Mature (58%)
Superior (n=41) [a]	Keweenaw Point	Lake Trout	532-822	1512-4354	37, 4	Mature (90%)

[a] *One age-17 Lake Trout was accidentally discarded by the field sampling team prior to being sent to the homogenization laboratory and therefore not used in the analysis.*
[b] *Mature = maturity stage in which fish is sexually mature (egg deposition status is either unknown, unimportant, or nonapplicable); Gravid = maturity stage in which ovary is full of eggs that are not yet ready for deposition or fertilization (eggs still contained within ovary wall structure)*
[c] *% = percentage out of total number of fish collected at each site*

Table 7: 2017 CSMI/Special Studies Lake Trout Biological Data

Lake	Site	Species	Lab Length Range (mm)	Lab Weight Range (g)	Gender Count (M, F)	Dominant Maturity Stage [a, b]
Huron (n=10)	Rockport	Lake Trout	575-788	2244-4618	6, 4	Mature (60%)
Huron (n=10)	Port Austin	Lake Trout	544-776	1603-5179	5, 5	Gravid (40%), Mature (50%)

[a] *Mature = maturity stage in which fish is sexually mature (egg deposition status is either unknown, unimportant, or nonapplicable); Gravid = maturity stage in which ovary is full of eggs that are not yet ready for deposition or fertilization (eggs still contained within ovary wall structure)*
[b] *% = percentage out of total number of fish collected at each site*

Table 8 provides a summary of age data for 2017 Base Monitoring Program and CSMI/Special Studies Lake Trout samples. Age results included in the table were determined based on annuli enumeration of maxillae and on CWTs. The dominant aging method used to obtain the final age for each fish is listed. Final age was determined based on CWT where available and then based on annuli enumeration of maxilla if no CWT was present. The majority of Lake Trout exceeded the target age range of 6-8 years at Rockport (80%) and Sturgeon Bay (63%), while 45% exceeded the age range at Keweenaw Point, 26% exceeded the age range at Dunkirk, 18% exceeded the age range at Port Austin, and 6% exceeded the age range at North Hamlin. It would be expected that fish exceeding the age range may have higher contaminant concentrations due to longer exposure times (i.e. bioaccumulation) of the environmental contaminants.

Table 8: 2017 Age Data (Base Monitoring Program and CSMI/Special Studies Lake Trout)

Lake	Site	Species	Age Range (years)	Dominant Aging Method	% Fish Exceeding Target Age Range
Erie (n=50)	Dunkirk	Lake Trout	4-10	Maxilla	26%
Huron (n=60)	Port Austin	Lake Trout	5-20	CWT, Maxilla	18%
Huron (n=10)	Rockport	Lake Trout	6-19	Maxilla	80%
Michigan (n=49)	Sturgeon Bay	Lake Trout	5-22	Maxilla	63%
Ontario (n=50)	North Hamlin	Lake Trout	3-9	CWT	6%
Superior (n=41) [a]	Keweenaw Point	Lake Trout	7-15	Maxilla	44%

[a] One age-17 Lake Trout was accidentally discarded by the field sampling team prior to being sent to the homogenization laboratory and therefore not used in the analysis.

5.3 ANALYSIS

The sections below summarize results for five contaminants (PCBs, PBDEs, mercury, HBCDD, and PFAS) in fish collected for the Base Monitoring Program in 2017, places these results in context with long-term trends for the odd-year sampling sites, and present results from the CEC screening analyses performed on these samples. The 2017 CSMI/Special Studies Program analytical results will be presented in future GLFMSP reports.

Ten-year (2007-2017) trends as well as longer term trends for contaminants at each collection site are presented in the sections below, with the exception of the Dunkirk collection site in Lake Erie. When the GLFMSP was designed, Walleye were selected to be collected in Lake Erie due to limited availability of Lake Trout at both collection sites (EPA 2012a). Walleye were collected exclusively at both collection sites through 2007. The abundance of Lake Trout in the eastern basin of Lake Erie (where the Dunkirk site is located) slowly began to increase starting in 2000 (NYSDEC 2009) and increased dramatically in 2011 (NYSDEC 2012). The GLFMSP had Lake Trout collected at Dunkirk in 2008 and 2010, and then switched to collecting Lake Trout at Dunkirk in odd years starting in 2011 (EPA 2012a). The GLFMSP has Dunkirk Lake Trout data from 2008, 2010, and odd years 2011-present; therefore, the sections below only present data from the 2008-2017 time frames for Dunkirk. Because Dunkirk trends are only summarized over this nine-year period, these estimated changes are not directly comparable to those from the other sites for which the change is summarized over a ten-year period.

As stated in Section 3.2, one homogenate from Keweenaw Point contained only one fish instead of the target of five fish per composite sample. This single fish homogenate was analyzed for all contaminants but was not included in site means or mega-composite presented in this report.

5.3.1 PCBs

The GLFMSP provides long-term data trends for PCBs in Lake Trout and Walleye from the 1970s - present. Prior to 1991, methods and target congeners varied. In this report, PCB trends for odd-year sites from 1991– 2017 (at all sites except Dunkirk as explained in Section 5.3) are presented as these are the date ranges for which the current sampling design (i.e., 10 composites of five fish with sites alternating within each lake annually) has been implemented.

Site mean total PCB concentrations ranged from 109 to 885 ng/g across the five sites (Table 9) in 2017. Mean total PCB concentrations were calculated based on 142 out of 209 individual PCB congeners. Measured results were not censored based on reporting or detection limits and all reported results were included in the totals. In general, mean total PCB concentrations have exhibited a decreasing trend at all sites over the 2007-2017 time frame (2008-2017 time frame for Dunkirk) (Table 9). Mean total PCB concentrations have also exhibited a decreasing trend at all sites over the 1991-2017 (Table 9 and Figure 2) time series, excluding Dunkirk for which we do not have Lake Trout data prior to 2008.

Estimated declines since 2007 (2008 for Dunkirk) are statistically significant at all sites. The 2007-2017 declines range from 24% at Sturgeon Bay to 59% at Port Austin, and the 2008-2017 decline for Dunkirk was 28%. Estimated PCB declines since 1991 at all non-Lake Erie sites are statistically significant and range from 72% at Sturgeon Bay to 91% at Keweenaw Point.

Table 9: Summary of 2017 Total PCB Site Means and Temporal Trends

Lake	Site	# Composites	Species	2017 Site Means Total PCB Concentration (standard error) (ng/g)	Estimated % Decline 1991-2017 (95% CI LL- UL)[c]	Estimated % Decline 2007-2017 (95% CI LL-UL)
Erie	Dunkirk	10	Lake Trout	450 (32.8)	N/A[d]	28 (18 - 36)[e]
Huron	Port Austin	10	Lake Trout	278 (24.0)	76 (71 - 81)	59 (41 - 72)
Michigan	Sturgeon Bay	10[a]	Lake Trout	885 (114)	72 (66 - 76)	24 (3 - 40)
Ontario	North Hamlin	10	Lake Trout	491 (57.2)	85 (82 - 87)	39 (24 - 51)
Superior	Keweenaw Point	8[b]	Lake Trout	109 (38.3)	91 (88 - 93)	58 (38 - 72)

[a] *Based on 9 composites of 5 fish and 1 composite of 4 fish*
[b] *Based on 8 composites of 5 fish (single fish homogenate not included)*
[c] *CI LL-UL indicates confidence interval lower level-upper level*
[d] *Lake Trout were not collected at the Dunkirk collection site until 2008*
[e] *Dunkirk estimated % decline is for the 2008-2017 period*

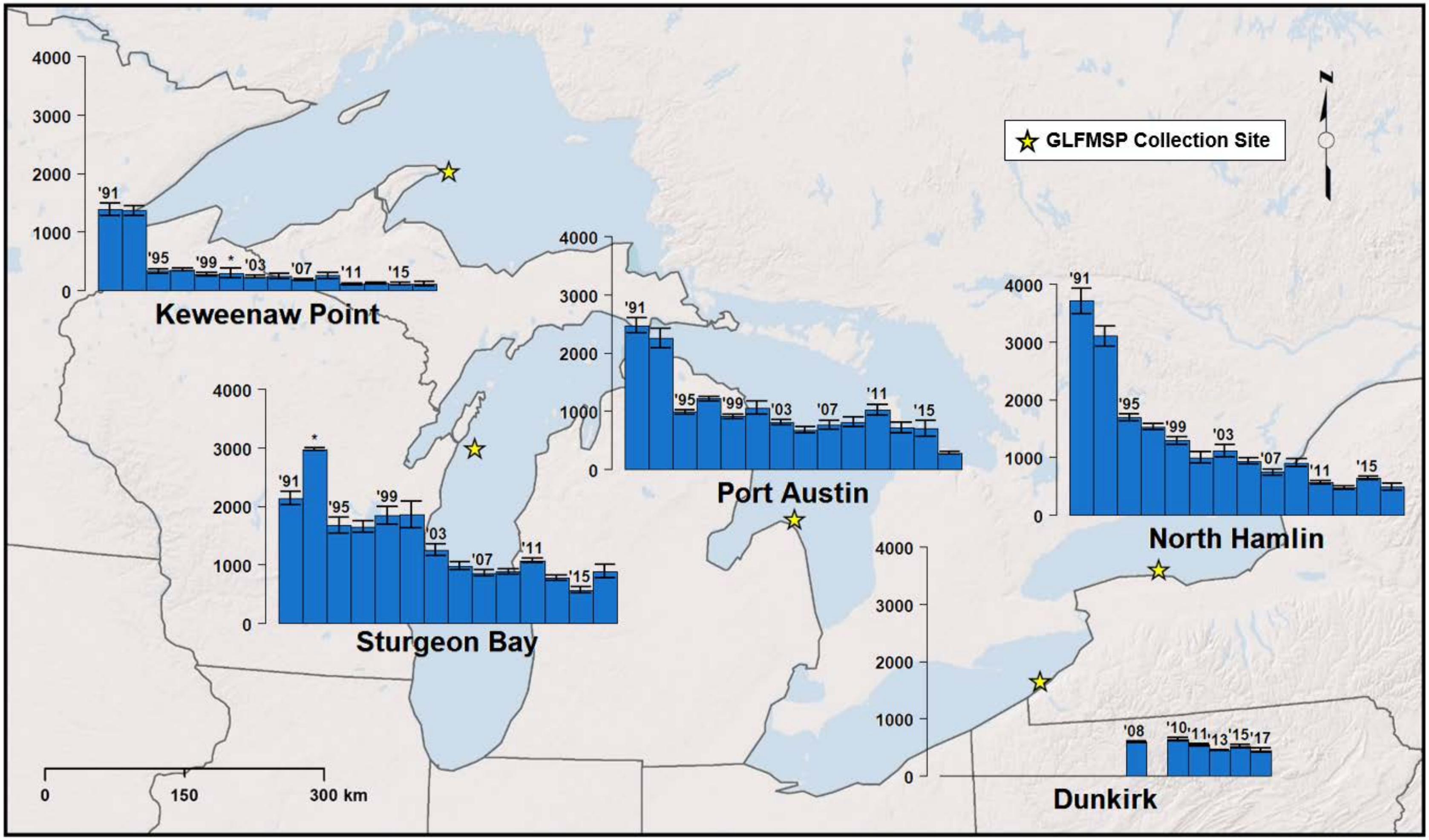

Figure 2. Mean Total PCB Concentration (ppb) in Lake Trout 1991-2017.

Notes: 1) *Stations are not representative of the entire lake.*
2) *A missing bar = samples not collected for that site/year.*
3) *An asterisk (*) indicates less than 5 composites are included in the sampling period.*
4) *The last two digits of collection years are displayed above corresponding bars as 'XX.*
5) *The Dunkirk bar graph is not directly comparable to those from other sites because there is one year of consecutive data for Dunkirk, while all other sites include data for odd years only (every other year).*

5.3.2 PBDEs

The GLFMSP began monitoring for PBDEs using congener-specific analyses in 2000, with a complete set of analyses for most lakes available beginning in 2001. Lake Trout were not collected at the Dunkirk collection site until 2008 as explained in Section 5.3, so PBDE trends are only presented for the 2008-2017 time period for Dunkirk.

Site mean total PBDE concentrations ranged from 15.3 to 42.9 ng/g across the five sites (Table 10) in 2017. Mean total PBDE concentrations were calculated based on five congeners (47, 99, 100, 153, and 154) that have been analyzed consistently across all years. These are the only PBDE congeners that have been consistently measured by GLFMSP and are the PBDE congeners found in the highest concentrations in Great Lakes fish (Zhou et al. 2018). Measured results were not censored based on reporting or detection limits and all reported results were included in the totals. In general, mean total PBDE concentrations showed a statistically significant decline at Sturgeon Bay, North Hamlin, and Keweenaw Point over the 2007-2017 time series (Table 10), with ten-year declines ranging from 24% at Sturgeon Bay to 43% at Keweenaw Point. While Dunkirk and Port Austin also exhibited a decrease in mean total PBDE concentrations since 2008 and 2007 respectively, neither were statistically significant.

Estimated total PBDE concentration declines over the 2001-2017 time series (Table 10 and Figure 3) are statistically significant at Sturgeon Bay, North Hamlin, and Keweenaw Point, and range from 44% at Keweenaw Point to 76% at Sturgeon Bay. The 5% decline in PBDE concentration at Port Austin since 2001 is not statistically significant.

Table 10: Summary of 2017 Total PBDE (5 congeners) Site Means and Temporal Trends

Lake	Site	# Composites	Species	2017 Total PBDE Site Mean Concentration (standard error) (ng/g)	Estimated % Decline [c] 2001-2017 (95% CI LL-UL) [d]	Estimated % Decline [c] 2007-2017 (95% CI LL-UL)
Erie	Dunkirk	10	Lake Trout	15.3 (1.32)	N/A [e]	3 (-13 - 17) [f]
Huron	Port Austin	10	Lake Trout	30.2 (2.78)	5 (-32 - 32)	21 (-12 - 45)
Michigan	Sturgeon Bay	10 [a]	Lake Trout	42.9 (5.56)	76 (68 - 81)	24 (3 - 41)
Ontario	North Hamlin	10	Lake Trout	29.9 (3.95)	47 (26 - 62)	40 (20 - 54)
Superior	Keweenaw Point	8 [b]	Lake Trout	20.0 (4.35)	44 (17 - 62)	43 (15 - 61)

[a] *Based on 9 composites of 5 fish and 1 composite of 4 fish*
[b] *Based on 8 composites of 5 fish (single fish homogenate not included)*
[c] *A negative percent decline of –X% corresponds to a percent increase of X%*
[d] *CI LL-UL indicates confidence interval lower level-upper level*
[e] *Lake Trout were not collected at the Dunkirk collection site until 2008*
[f] *Dunkirk estimated % decline is for the 2008-2017 period*

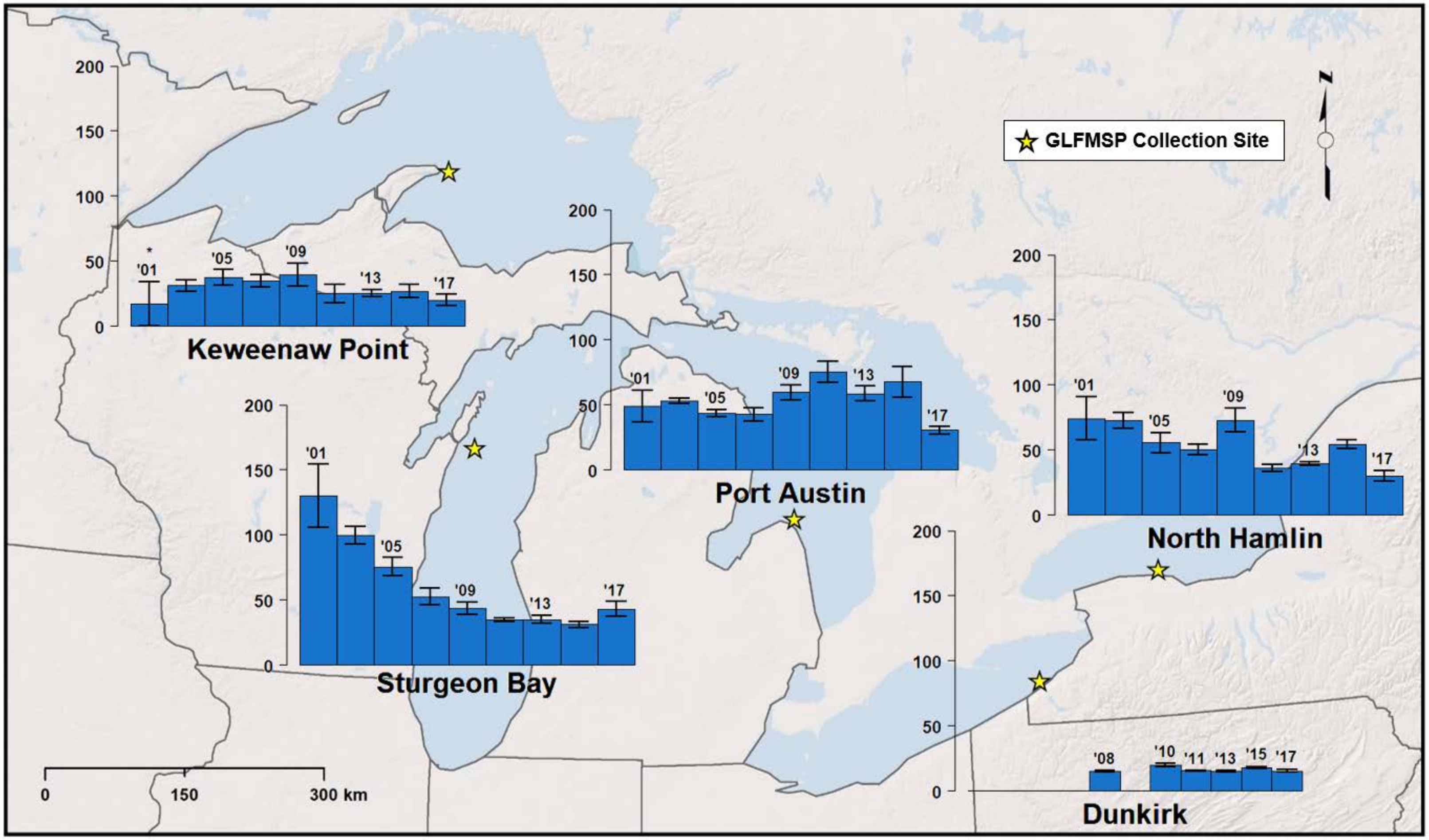

Figure 3. Mean Total PBDE (5 Congeners) Concentration (ppb) in Lake Trout 2001-2017.

Notes: 1) Stations are not representative of the entire lake.
2) A missing bar = samples not collected for that site/year.
3) An asterisk (*) indicates less than 5 composites are included in the sampling period.
4) The last two digits of collection years are displayed above corresponding bars as 'XX.
5) Total PBDE = sum of congeners 47, 99, 100, 153, and 154.
6) The Dunkirk bar graph is not directly comparable to those from other sites because there is one year of consecutive data for Dunkirk, while all other sites include data for odd years only (every other year).

5.3.3 Mercury

The GLFMSP began monitoring for total mercury in 1999. Mean total mercury concentrations are shown at all odd-year sampling sites from 1999-2017 (except Dunkirk as explained in Section 5.3) in Figure 4.

Site mean mercury concentrations ranged from 110 to 180 ng/g across the five sites (Table 11) in 2017. Mean mercury concentrations showed a statistically significant decline over the 2007-2017 time series (Table 11) at Port Austin. Keweenaw Point exhibited a decrease in mean mercury concentrations since 2007 as well, although it was not statistically significant. At North Hamlin, a statistically significant increase of 15% was shown from 2007-2017 and at Dunkirk, a statistically significant increase of 13% was exhibited from 2008-2017. The increasing age of the Lake Trout collected at Dunkirk in 2015 and 2017 could explain the increasing trend observed at Dunkirk for the 2008-2017 time series. Sturgeon Bay also exhibited an increase in mean mercury concentrations since 2007, although it was not statistically significant.

Since 1999, no statistically significant changes in mercury concentrations have been detected at any of the non-Lake Erie sites. While North Hamlin did exhibit a decrease in mercury concentrations (4%), it was not statistically significant. The other three non-Lake Erie sites exhibited an increase in mercury concentrations (ranging from 3% at Keweenaw Point to 16% at Port Austin), although none of these increases were statistically significant (Table 11).

Table 11: Summary of 2017 Total Mercury Site Means and Temporal Trends

Lake	Site	# Composites	Species	2017 Total Mercury Site Mean Concentration (standard error) (ng/g)	Estimated % Decline [c] 1999-2017 (95% CI LL-UL) [d]	Estimated % Decline [c] 2007-2017 (95% CI LL-UL)
Erie	Dunkirk	10	Lake Trout	111 (5.28)	N/A [e]	-13 (-23 to -3)
Huron	Port Austin	10	Lake Trout	180 (9.78)	-16 (-36 to 2)	26 (9 to 40)
Michigan	Sturgeon Bay	10 [a]	Lake Trout	177 (9.91)	-8 (-27 to 8)	-8 (-28 to 8)
Ontario	North Hamlin	10	Lake Trout	110 (7.59)	4 (-6 to 14)	-15 (-29 to -2)
Superior	Keweenaw Point	8 [b]	Lake Trout	155 (26.5)	-3 (-29 to 17)	11 (-16 to 32)

[a] *Based on 9 composites of 5 fish and 1 composite of 4 fish*
[b] *Based on 8 composites of 5 fish (single fish homogenate not included)*
[c] *A negative percent decline of –X% corresponds to a percent increase of X%*
[d] *CI LL-UL indicates confidence interval lower level-upper level*
[e] *Lake Trout were not collected at the Dunkirk collection site until 2008*
[f] *Dunkirk estimated % decline is for the 2008-2017 period*

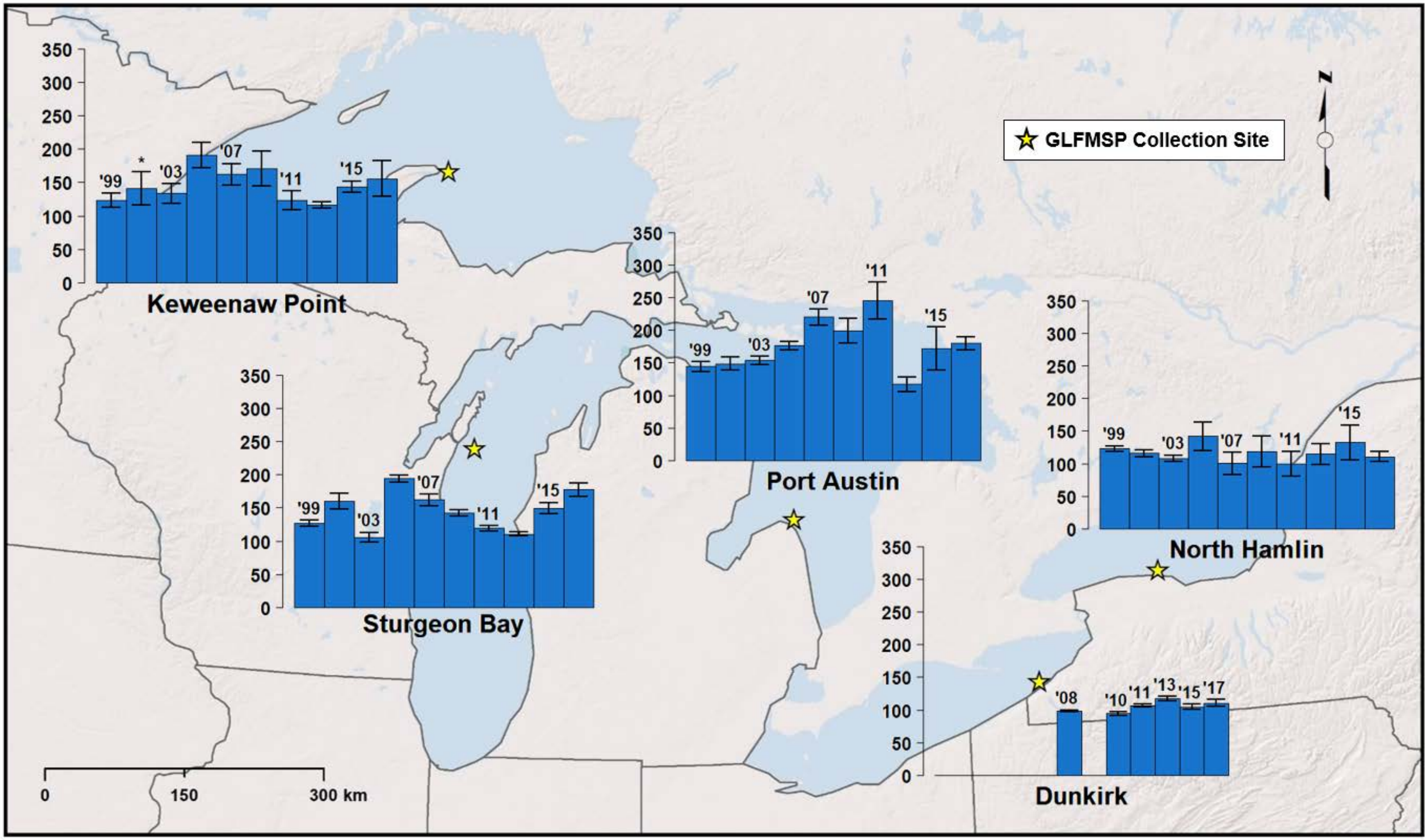

Figure 4. Mean Total Mercury Concentration (ppb) in Lake Trout 1999-2017.

Notes: 1) *Stations are not representative of the entire lake.*
2) *A missing bar = samples not collected for that site/year.*
3) *An asterisk (*) indicates less than 5 composites are included in the sampling period.*
4) *The last two digits of collection years are displayed above corresponding bars as 'XX.*
5) *The Dunkirk bar graph is not directly comparable to those from other sites because there is one year of consecutive data for Dunkirk, while all other sites include data for odd years only (every other year).*

5.3.4 HBCDD

The GLFMSP added analysis of three HBCDD isomers in mega-composite samples to the program in 2012, beginning with analysis of samples collected in 2010. HBCDD was added to the GLFMSP due to its designation as a chemical of mutual concern under the GLWQA. Four years of data (2011, 2013, 2015, and 2017) are available for odd-year sites. Because this time period is not sufficient to allow for a meaningful evaluation of trends, temporal trends for total HBCDD concentration are not evaluated in this report. However, each mega-composite sample was analyzed for three HBCDD isomers in triplicate, such that site means and associated analytical variability could be calculated. Total HBCDD mega-composite means range from 3.48 ng/g at Dunkirk to 8.63 ng/g at Port Austin (Table 12) in 2017. Mean total HBCDD concentrations were calculated based on the three analyzed HBCDD isomers. Measured results were not censored based on reporting or detection limits and all reported results were included in the totals.

Table 12: Summary of 2017 Total HBCDD Mega-composite Means

Lake	Site	# Replicates [a]	Species	2017 Total HBCDD Mega-composite Mean Concentration (standard error) (ng/g)
Erie	Dunkirk	3	Lake Trout	3.48 (0.49)
Huron	Port Austin	3	Lake Trout	8.63 (0.79)
Michigan	Sturgeon Bay	3	Lake Trout	7.46 (0.05)
Ontario	North Hamlin	3	Lake Trout	3.65 (0.04)
Superior	Keweenaw Point	3	Lake Trout	4.83 (0.11)

[a] *Single mega-composite samples were analyzed in triplicate (so variability estimates include analytical variability but not sampling variability, which is included in the calculated standard errors for other analyte classes presented in this report)*

5.3.5 PFAS

The GLFMSP began monitoring PFAS compounds in 2011. The list of analyzed PFAS compounds has varied since 2011. In 2017, monitored PFAS compounds included 26 perfluorinated carboxylic acids and sulfonates with 4 to 13 carbons, including 10 branched isomers. In recent years, including 2017, the method used to quantify PFAS was modified to improve reproducibility in complex biological tissues (Point et al. 2019). This method utilizes ultra-high-performance liquid chromatography with tandem mass spectrometry (UPLC-MSMS). Due to the evolving analytical methodology and smaller number of composites analyzed, it is not appropriate at this time to assess temporal trends for PFAS compounds. Table 13 and Figure 5 show total PFAS and perfluorooctanesulfonic acid (PFOS) site mean concentrations and their associated standard errors for the composites that were analyzed at each site. Because the PFAS analysis scheme was generally consistent across sites, the mean concentrations can be compared to each other. As seen in Table 13 and Figure 5, total PFAS and PFOS concentrations are generally highest at Dunkirk and lowest at Keweenaw Point. Mean total PFAS concentrations were calculated based on the 16 PFAS compounds that were analyzed, excluding branched isomers. Measured results were not censored based on reporting or detection limits and all reported results were included in the totals.

Table 13: Summary of 2017 Total PFAS and PFOS Composite Means

Lake	Site	# Composites	Species	2017 Total PFAS Composite Mean (standard error) (ng/g)	2017 PFOS Composite Mean (standard error) (ng/g)
Erie	Dunkirk	5	Lake Trout	96.5 (8.2)	84.3 (7.5)
Huron	Port Austin	5	Lake Trout	35.3 (1.1)	23.5 (0.79)
Michigan	Sturgeon Bay	5	Lake Trout	44.3 (3.2)	37.5 (2.9)
Ontario	North Hamlin	5	Lake Trout	52.0 (4.2)	47.3 (3.9)
Superior	Keweenaw Point	5	Lake Trout	15.0 (0.94)	6.1 (0.40)

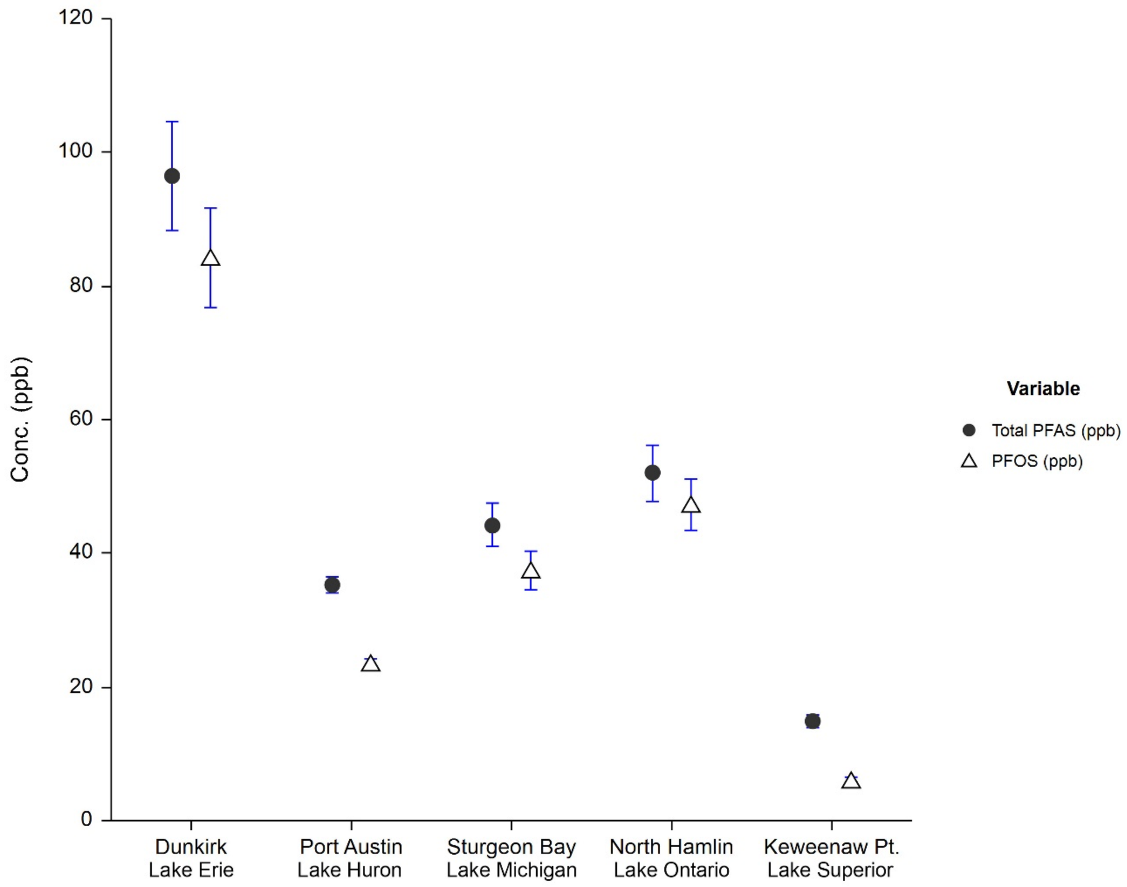

Figure 5. 2017 Mean Total PFAS and PFOS Concentrations Per Site (±1 standard error).

5.3.6 Contaminants of Emerging Concern (CECs)

Since 2014, Base Monitoring Program mega-composites samples have been screened for CECs. Initial screening studies have been focused on detecting organic compounds that contain one or more chlorine or bromine atom. Historically, organic chemicals containing carbon bonded to chlorine or bromine have been found to be bioaccumulative and potentially exhibit adverse effects on lake biota (e.g., PCBs, OCPs, PBDEs) (Howard and Muir 2010).

Figure 6 summarizes the total concentration of halogenated organic chemicals observed in Lake Trout collected from Lakes Superior (Keweenaw Point), Huron (Port Austin), Erie (Dunkirk), Michigan (Sturgeon Bay), and Ontario (North Hamlin). Sturgeon Bay exhibited the highest total concentration followed by North Hamlin, Dunkirk, and Keweenaw Point, respectively. Port Austin exhibited the lowest total concentration of halogenated chemicals. Similar to observations in the Great Lakes Fish Monitoring and Surveillance Program Technical Report: Status and Trends through 2016 (EPA 2020) for even year GLFMSP collection sites, halomethoxyphenols were the dominant class of compounds observed in all of the lakes, followed by PCBs and other halogenated components on the routine monitoring schedule (i.e., organochlorine pesticides).

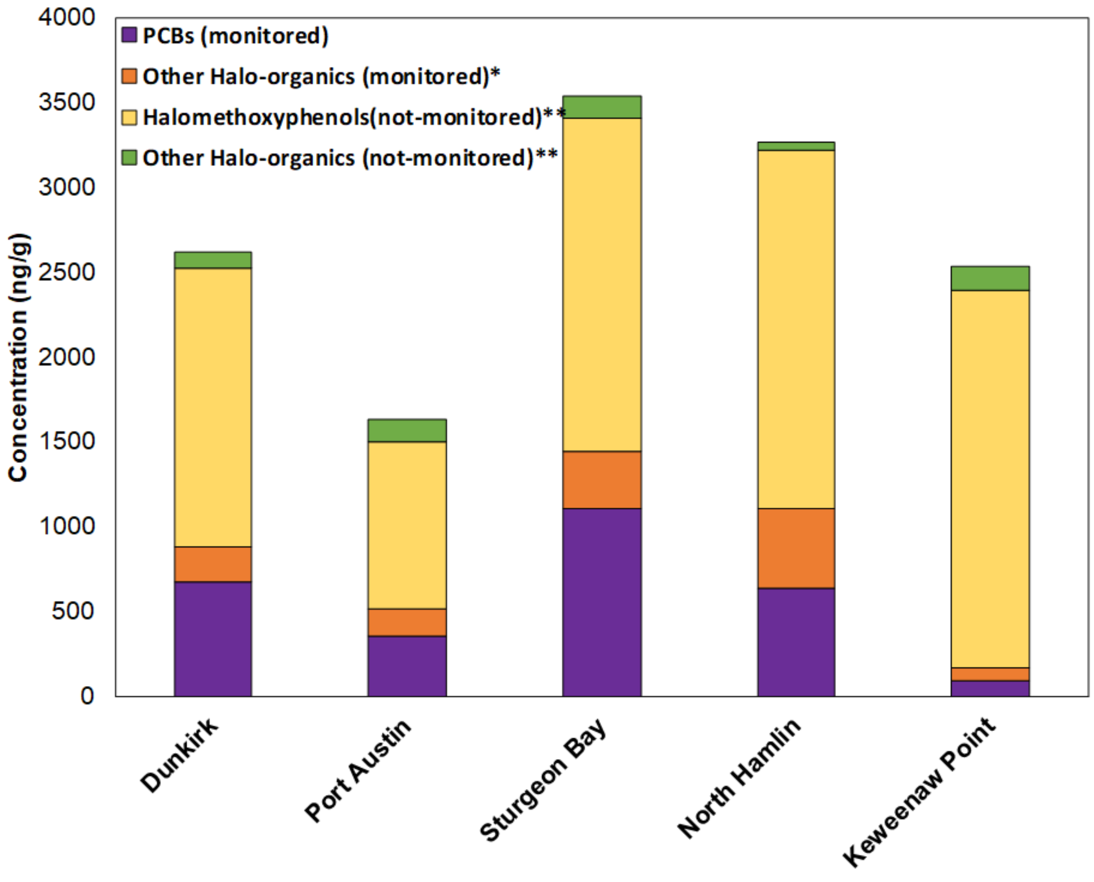

Figure 6. Concentrations of Halogenated Compounds and PCBs in GLFMSP Mega-composite Samples from 2017. * Includes PBDEs and OCPs. ** Concentrations were determined using reference standards where available or structurally similar compound.

6 SUMMARY

The 2017 GLFMSP Technical Report details sampling information of the Base Monitoring Program and CSMI, assesses data and trends through 2017, and shows that various legacy contaminant concentrations are decreasing in Great Lakes top predator fish. Key highlights include:

- Mean total PCB concentrations in Lake Trout have declined at the odd-year sampling sites at Port Austin (Lake Huron), Sturgeon Bay (Lake Michigan), Keweenaw Point (Lake Superior), and North Hamlin (Lake Ontario) from 1991 to 2017. Concentrations have also declined in the eastern basin of Lake Erie since monitoring of Lake Trout began in 2008 at the Dunkirk site.
- Mean total PBDE concentrations in Lake Trout have declined at the odd-year sampling sites in Lakes Michigan, Ontario, and Superior since 2001. No significant changes in Lake Trout at the Port Austin sampling site were found in this timeframe. Concentrations have also declined in the eastern basin of Lake Erie since monitoring of Lake Trout began in 2008.
- Mercury concentrations in Lake Trout have declined at the Port Austin sampling site in Lake Huron since 2007 and increased at the odd-year sampling sites in North Hamlin (Lake Ontario) and Dunkirk (Lake Erie) since 2007 and 2008, respectively. Lake Trout collected at the odd-year sampling sites in lakes Superior and Michigan did not show statistically significant changes in mercury concentrations from 1999 to 2017. No statistically significant changes occurred at any sampling site since 1999.

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APPENDIX A – LIST OF RECENT GLFMSP PUBLICATIONS

The following is a list of GLFMSP publications produced between 2017 and 2021.

Crimmins, B.S., Holsen, T.M., 2019. Non-targeted Screening in Environmental Monitoring Programs. Advances in Experimental Medicine and Biology: Advancements of Mass Spectrometry in Biomedical Research. 1140, 731-741.

Crimmins, B.S., McCarty, H.B., Fernando, S., Milligan, M.S., Pagano, J.J., Holsen, T.M., Hopke, P.K., 2018. Commentary: Integrating Non-targeted and Targeted Screening in Great Lakes Fish Monitoring Programs. Journal of Great Lakes Research. 44, 1127-1135.

Dupree, E.J., Crimmins, B.S., Holsen, T.M., Darie, C.C., 2019. Developing Well-Annotated Species-Specific Protein Databases Using Comparative Proteogenomics. Advances in Experimental Medicine and Biology: Advancements of Mass Spectrometry in Biomedical Research. 1140, 389-400.

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