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NATIONAL STATUS AND TRENDS, MUSSEL WATCH PROGRAM

A 2022 Assessment of Legacy Organic Contaminants in Dreissenid Mussels in the Great Lakes Basin









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A 2022 Assessment of Legacy Organic Contaminants in Dreissenid Mussels in the Great Lakes Basin

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Authors:

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EXECUTIVE SUMMARY

In 2022, the National Mussel Watch Program (MWP) conducted an assessment of the presence, distribution, and magnitude of legacy organic contaminants in dreissenid mussels (zebra/quagga; *Dreissena spp*.) from the Laurentian Great Lakes. To achieve this, the MWP employed a sentinel-based monitoring approach by collecting and analyzing dreissenid mussels as surrogates for assessing water pollutant levels in the Great Lakes. Mussels and oysters are sessile organisms that filter and accumulate particles from water; therefore, measuring contaminant levels in their tissue is a good indicator of local chemical contamination. Mussel tissue samples from 66 monitoring sites within the Great Lakes Basin were analyzed in this study. In 2022, tissue samples were analyzed for nine legacy compound groups including butyltins, chlordanes, chlorobenzenes, dichlorodiphenyltrichloroethane (DDTs), dieldrins, endosulfans, hexachlorocyclohexane (HCHs), polycyclic aromatic hydrocarbons (PAHs), and polychlorinated biphenyls (PCBs), in addition to 2 individual contaminants, chlorpyrifos and mirex.

The MWP provides unique data that is essential for assessing the health of the Nation's coastal waters through temporal and spatial evaluations of chemical contamination. Studies such as this provide needed data and information for the MWP and also address contamination data gaps that are relevant for coastal management within the Great Lakes. Results from the 2022 dreissenid mussel tissue samples indicated that legacy organic contaminants are present at varying concentrations in the Great Lakes Basin. No sites within this analysis were devoid of chemical contamination, emphasizing the ubiquity of these contaminants in coastal waters, even after some of these organic pollutants have been phased out some two decades ago. Overall, when compared to national MWP historic concentrations, legacy contaminants concentrations measured in Great Lakes dreissenid mussels were generally low. The accumulation of legacy organic contaminants in organisms is often analyte and location-dependent. This study found that the presence and concentration of a given contaminant are strongly influenced by several key factors, notably its chemical properties, sources, environmental fate, and transport mechanisms. The information presented here is useful to regional managers because of the long-term nature and geographic spread of the dataset, which could further complement data from other regional monitoring programs in the Great Lakes. Therefore, coastal, state, and local managers within the Great Lakes Basin would benefit from this study as they develop long-term policies to abate legacy organic contaminants in an attempt to protect their managed ecosystems.

Given that Total PAHs and Total PCBs exceeded US EPA screening values for both recreational and subsistence fishers at certain MWP sites, the combined findings from our 2022 Great Lakes legacy contaminants assessment suggest these compounds may present a significant risk, including the potential for sub-lethal and harmful biological effects. In addition, despite some legacy contaminants being detected at environmentally low concentrations, they are often part of complex mixtures of compounds and the cumulative risk of long-term exposure, even at low concentrations, should not be ignored. Therefore, from a management perspective, with bivalves shown to act as transfer vectors of various contaminants to higher trophic-level organisms (e.g., fish) across the Great Lakes Basin food-web, the toxicological effects and endpoints of these contaminants on reproduction, development, and growth warrants additional monitoring and assessment of these contaminants. The findings of this study will help identify and contextualize the relationship between detected legacy compounds and their environmental pathways, as well as “sinks” and “hot spots” in the Great Lakes Basin. These efforts will further aid in the chemical prioritization of which legacy compounds to be monitored, thus making the most efficient use of resources and funds that can be used to support measures in reducing and abating the continued occurrence of these legacy compounds in the Great Lakes Basin.

KEY FINDINGS

- Nine (Total Butyltins, Total Chlorobenzenes, Total Chlordanes, Total DDTs, Total Endosulfans, Total HCHs, mirex, Total PAHs, and Total PCBs) of the eleven legacy contaminant groups targeted in this study were detected at least once in dreissenid mussels above method detection limit (> MDLs).

- Chlorpyrifos and Total Dieldrins were not detected in dreissenid mussel tissue at any sites assessed in this 2022 study.

- Legacy organic contaminants measured in dreissenid mussel tissue were detected at low concentrations compared to historical Great Lakes MWP legacy contaminant data.

- Elevated legacy contaminant concentration levels (> MDLs) measured in dreissenid mussel tissue were typically associated with Total DDTs, Total PAHs, and Total PCBs.

- On average, individual contaminant groups concentration levels measured in dreissenid mussel tissue were found from lowest to highest concentrations: Total Endosulfans > Total Chlorobenzenes > Mirex > Total HCHs > Total Butyltins > Total Chlordanes > Total DDTs > Total PCBs > Total PAHs.

- Basin-wide, the highest legacy contaminant concentration levels were measured in mussels from sites sampled in Lake Michigan, followed by sites sampled in Lake Erie, Lake Ontario, and Lake Huron/Lake St. Clair.

- Elevated mussel legacy contaminant levels closely matched sites adjacent to urban rivers and tributaries, with larger population densities and industrial centers in Lakes Michigan, Erie, Lake Huron/Lake St. Clair, and Ontario.

- Despite declining temporal trends, contaminant groups such as Total Chlordanes, Total DDTs, Total PCBs, and Total PAHs are still among the most abundant contaminants observed in mussel tissue.

- Total DDTs, Total PAHs, and Total PCBs were detected at 78.5% - 98.5% of sites assessed in this study.

- Mirex, Total Butyltins, Total Chlorobenzenes, and Total Chlordanes, were detected in 4.6% - 46.9% of sites.

- For long-term sites assessed in this study, the majority of legacy pollutants detected in our 2022 study are either significantly declining (e.g. Total Chlordanes, Total DDTs, Total PAHs, Total PCBs) or showing no directional change over time in mussel tissue.

- The only sites with significant increasing site-specific trends were LHTB for Total Chlordanes, LMNC for Total DDTs, LMHM and LHTB for Total PAHs, and LMCB, LMNC, LHTB, LELR, and LERB for Total PCBs.

- The 2022 study results confirm that some legacy contaminant groups assessed in mussel tissue have persisted at background levels, suggesting further significant reductions in contaminant concentrations across the Great Lakes Basin may be challenging.

Table of Contents

COMMONLY USED ACRONYMS. ii

1.0 HISTORY OF MUSSEL WATCH PROGRAM. 1

2.0 INTRODUCTION. 3

3.0 METHODS. 4

4.0 RESULTS - TOTAL BUTYLTINS. 10

5.0 RESULTS - TOTAL CHLORDANES. 17

6.0 RESULTS - TOTAL CHLOROBENZENES. 24

7.0 RESULTS - CHLORPYRIFOS. 31

8.0 RESULTS - TOTAL DDTs. 37

9.0 RESULTS - TOTAL DIELDRINS. 44

10.0 RESULTS - TOTAL ENDOSULFANS. 50

11.0 RESULTS - TOTAL HCHs. 57

12.0 RESULTS - MIREX. 64

13.0 RESULTS - TOTAL PAHs. 71

14.0 RESULTS - TOTAL PCBs. 79

15.0 SUMMARY. 88

REFERENCES. 99

APPENDICES. 107

COMMONLY USED ACRONYMS

AOC	Areas of Concern
CECs	Contaminants of Emerging Concern
DDT	dichlorodiphenyltrichloroethane
dw	dry weight
EPA	Environmental Protection Agency
FDA	Food and Drug Administration
g	gram
HCH	hexachlorocyclohexane
MDL	method detection limit
MWP	Mussel Watch Program
NCCOS	National Centers for Coastal Ocean Science
ng	nanogram
NOAA	National Oceanic and Atmospheric Administration
NS&T	National Status and Trends
OCs	organic compounds
OCPs	organochlorine pesticides
OPs	organic pollutants
PAHs	polycyclic aromatic hydrocarbons
PCBs	polychlorinated biphenyls
SD	standard deviation
SV	Screening Values
US	United States
ww	wet weight

Introduction

1.0 HISTORY OF MUSSEL WATCH PROGRAM

The National Mussel Watch Program (MWP), which began in 1986, was designed by the National Oceanic and Atmospheric Administration (NOAA) to monitor the nation's coastal waters for chemical contaminants and biological indicators of water quality (Apeti et al., 2018). The MWP was established in response to a legislative mandate under Section 202 of Title II of the Marine Protection, Research and Sanctuaries Act (MPRSA) (33 USC 1442), which called on the Secretary of Commerce to initiate a continuous monitoring program, among other activities. The MWP design is based on the periodic collection and analysis of bivalves (oysters and mussels) and sediment from a network of monitoring sites located throughout the nation's coastal zones. To date, NOAA's MWP is one of the longest running, continuous coastal monitoring programs (Swam et al., 2024).

The MWP monitoring sites are found along all of the U.S. coastlines, including Alaska, the Great Lakes, Hawaii, and in territories such as Puerto Rico. As a result of this large spatial coverage, different target bivalves are used as sentinel species. Mussels and oysters are sessile organisms that filter and accumulate particles from water and their body burden reflects ambient concentrations; therefore, measuring contaminant levels in their tissue is a good indicator of local chemical contamination (Farrington, 1983). Mussels (*Mytilus* species) are collected from the North Atlantic and Pacific coasts, oysters (*Crassostrea virginica*) are collected from the Mid-Atlantic (Delaware Bay) southward and along the Gulf Coast, the invasive zebra and quagga mussels (*Dreissena spp*.) are collected from the Great Lakes, mangrove oysters (*Crassostrea rhizophorae*) are collected from Puerto Rico, and Hawaiian oysters (*Dendostrea sandvicensis*) are collected from Hawaii.

A fundamental challenge faced by any long-term environmental monitoring program is how to evolve in response to changing conditions and technologies. In 2013, due to budgetary constraints, the National Centers for Coastal Ocean Science (NCCOS) undertook the task of re-designing the MWP, moving from a nationwide annual monitoring approach to the rotating regional monitoring model that is currently employed. The regional approach allows the program to improve its presence in coastal communities by increasing interaction with local stakeholders, integrating inputs from coastal resource managers, and providing specific data needs to help fill local data gaps. By making adaptive changes and leveraging regional partnerships, the program has increased its scientific relevance and reputation and has evolved to include more than 300 monitoring sites (Figure 1) and nearly 600 chemical contaminants, including metals, legacy organic compounds, and contaminants of emerging concern (CECs; Apeti et al., 2018).

The MWP provides unique data that is vital to evaluating the health of the nation's estuarine and coastal waters, particularly describing the levels of chemical contamination. The MWP dataset allows for temporal and spatial evaluation of regional and national changes in chemical distribution, including CECs as their potential risks are identified (Edwards et al., 2024a). The program's long-term data supports the assessment of impacts of unforeseen events such as oil spills and hurricanes, the evaluation of sanctuary statuses, the analysis of resource and ecosystem service trends, and the evaluation of the effectiveness of regulations that ban toxic chemicals or support legislation such as the Clean Air and Clean Water Acts.

Introduction



Figure 1. National Mussel Watch sites.

Introduction

2.0 INTRODUCTION

In 2022, the MWP conducted a comprehensive assessment of legacy organic contaminants in the Laurentian Great Lakes (Figure 2). The study was designed within the framework of the NOAA/NCCOS MWP regional monitoring approach, which balances flexibility in study design with the cost of broad contaminant surveys. With this framework in mind, and realizing that challenges still remain in accurately tracking (e.g., spatially and temporally) and monitoring various organic pollutants across the Great Lakes Basin, the objectives of this study were to 1) assess the magnitude and distribution of legacy organic contaminants historically assessed by the MWP; 2) compare contamination in the Great Lakes in 2022 to previous studies in the same and other regions across the United States as well as to established thresholds and guidelines; and 3) make data electronically available to watershed and coastal resource managers in the Great Lakes.

The continued ubiquitous detection of legacy organic contaminants, including industrial and agricultural chemicals, in aquatic environments is cause for concern (Fuller et al., 2025; Legrand et al., 2023; Maloney et al., 2025). Studies have shown the physicochemical properties (e.g., limited solubility in water, and resist degradation in aquatic and terrestrial environments) of legacy organic pollutants including polychlorinated biphenyls (PCBs; industrial chemicals), chlordane (pesticides), dichloro-diphenyl-trichloroethanes (DDTs; pesticides), dieldrin (pesticides), and mirex (pesticides), render these contaminants highly lipophilic, extremely persistent (extended half-life in aquatic and terrestrial matrices), bioaccumulative, and toxic to wildlife and humans (Cornwell et al., 2015; Tudi et al., 2021; Rafferty et al., 2009). Observed apical end-points associated with legacy organic contaminants include, but are not limited to, decreased reproductive potential, behavioral and neurologic changes, endocrine disruption, reduced growth and adverse health effects, to name a few (Cornwell et al., 2014; De Rosa et al., 2006; Fuller et al., 2024; Hammond et al., 2023). Several legacy organic contaminants including PCBs, PAHs, DDTs, dieldrin, and mirex were earmarked by the International Joint Commission (IJC) in 1985 as critical Great Lakes pollutants (De Rosa et al., 2006; Johnson et al., 1999; Thomas, 1995). As such, these contaminants are subjected to binational monitoring and prevention efforts within the Great Lakes Basin. Despite the wealth of information that surrounds these organic contaminants in upper trophic-level organisms (e.g., fish), data and information on the environmental occurrence, magnitude, and spatial distribution of legacy organic contaminants in Great Lakes lower trophic-level aquatic organisms and food-webs remain limited (Kimbrough et al., 2014).

As part of the NOAA/NCCOS MWP, a suite of nearly 150 chemical contaminants including total butyltins, total PCBs, total PAHs, total DDTs, total chlordanes, total dieldrins, mirex, total chlorobenzenes, total HCHs, total endosulfans, and chlorpyrifos), is measured on an annual basis since 1986 (Apeti et al., 2018; Kimbrough et al., 2013). Since 1992, the MWP has used dreissenid mussels to monitor the environmental occurrence and magnitude of a wide suite of organic pollutants in the Laurentian Great Lakes, establishing one of the most spatially robust biomonitoring data sets in the region (Edwards et al., 2024a; Edwards et al., 2024b; Kimbrough et al., 2018). Dreissenid mussels are extensively used as bioindicator organisms due to their sessile and sedentary nature, high population density (≥700,000 specimens/m^2; Carvalho et al., 2021), and their quick response to contamination changes in aquatic systems. Other attributes which enable dreissenid mussels as excellent bioindicators include their ability to sequester and bioaccumulate contaminants from water and suspended sediments, thus reflecting ambient contamination levels in surrounding aquatic environments (Edwards et al., 2024a; Edwards et al., 2024b; Kimbrough et al., 2014).

The results summarized in this report are intended to inform MWP management, Great Lakes resource managers, stakeholders, water/watershed managers, and the larger Great Lakes community on the magnitude and environmental occurrence of legacy organic contaminants detected in dreissenid mussels within the Great Lakes Basin. This information is important in developing integrated approaches for legacy contaminants prioritization, mitigation strategies, ecological risk assessments, and suitable mechanisms for source identification of detected legacy compounds that are likely “hotspots” within the Great Lakes Basin.

3.0 METHODS

3.1 Study Area and Sampling Design

The Laurentian Great Lakes constitute one of the largest freshwater systems in the world, with an estimated 21% of the Earth's surface freshwater (~ 90% of United States freshwater; Fields, 2005; Sponberg, 2009). The Great Lakes covers a drainage basin of over 244,000 km^2 (94,000 mi^2), and is home to over 35 million people (Breffle et al., 2013; Danz et al., 2007). The region's watershed, which drain almost 518,000 km^2 (200,000 mi^2), supports numerous economic industries, including manufacturing, agriculture, and commercial fisheries, with areas of intense urbanization and industrialization occurring along its coastal zone (Breffle et al., 2013; Wolter et al., 2006). Land-use and land cover (LULC) differs between eco-regions and eco-provinces, with predominantly forested areas in the northern and southeastern sections, and agricultural activities more pronounced in the western and central sections of the Basin (Morrice et al., 2007). Increased environmental stressors from agricultural (an estimated one-third of the Great Lake's land area [17.3 million ha] is used for agricultural activities; Murphy et al., 2012), residential, municipal, and industrial activities have led to increased water quality impairment within the Basin (Muir et al., 2006; Kiesling et al., 2022). Specifically, continuous loading of organic pollutants from point and diffuse sources has been identified as an important drivers behind coastal water quality and ecosystem impairment within the Great Lakes Basin (Hull et al., 2015; Kiesling et al., 2019).

In this study, dreissenid mussels from 18 long-term monitoring sites and 48 recently established sites representing inshore (tributary, rivers, harbors), nearshore (i.e., shoreline-related region reaching a 30-m depth contour or a distance of 5-km from shore; Yurista et al., 2016), and offshore sites were collected and analyzed for 11 legacy organic contaminant groups in 2022. Collectively, 30 sites were sampled in Lake Michigan, 7 sites were sampled in Lake Huron, 2 sites were sampled in Lake St. Clair, 18 sites were sampled in Lake Erie, 1 site was sampled in Niagara River, and 8 sites were sampled in Lake Ontario and St. Lawrence River between August – September 2022. Long-term or historical Great Lakes monitoring sites were selected in locations with abundant bivalve populations to allow for repetitive sampling and to convey information about the degree of chemical contamination in the Great Lakes Basin area over time. Equally important, MWP sites were purposefully selected in EPA's designated areas of concern (e.g., Clinton River AOC, Niagara River AOC, Milwaukee Estuary AOC) to assess the status and trends of chemical stressors in the nearshore areas, and evaluate the impacts of land-use categories on the distribution of chemical stressors.

In 2022, dreissenid mussel sample collection followed standard protocols utilized by the MWP including site navigation, recording site conditions, and measuring water quality parameter (salinity, temperature, and dissolved oxygen; Apeti et al., 2012). When available, divers harvested *in situ* dreissenid mussels from established populations at outer harbor stone breakwaters and at offshore lake zones. Inshore mussel sampling locations included sites sampled at harbor, river, and tributary locations. Sampling locations previously established by the MWP in Great Lakes Areas of Concern (AOC; areas designated for restoration due in part to historical environmental contamination; Hartig et al., 2020), smaller harbor sampling locations (e.g., Ashtabula River, Cuyahoga River, Presque Isle Bay, and Black River), and other priority urban areas (e.g., Milwaukee, Chicago, and the Buffalo/Niagara River region) were also targeted in 2022, with the objective of providing a more robust measure of bioavailable legacy contaminants that would be generated from municipal, industrial, and fugitive sources.

Dreissenid mussels were rinsed with site water to remove debris, placed in labeled freezer bags, packed in ice containers (0 - 5°C) with sample bags properly sealed and free of freshwater contact. All collected mussel samples were shipped to an analytical laboratory (TDI-Brooks International Inc., College Station, TX) within two days of collection for contaminant analysis. Tissue sample collection and processing were consistent with NOAA methods and procedures for bivalve tissue assessment (Apeti et al., 2012; Kimbrough et al., 2013).

Methods

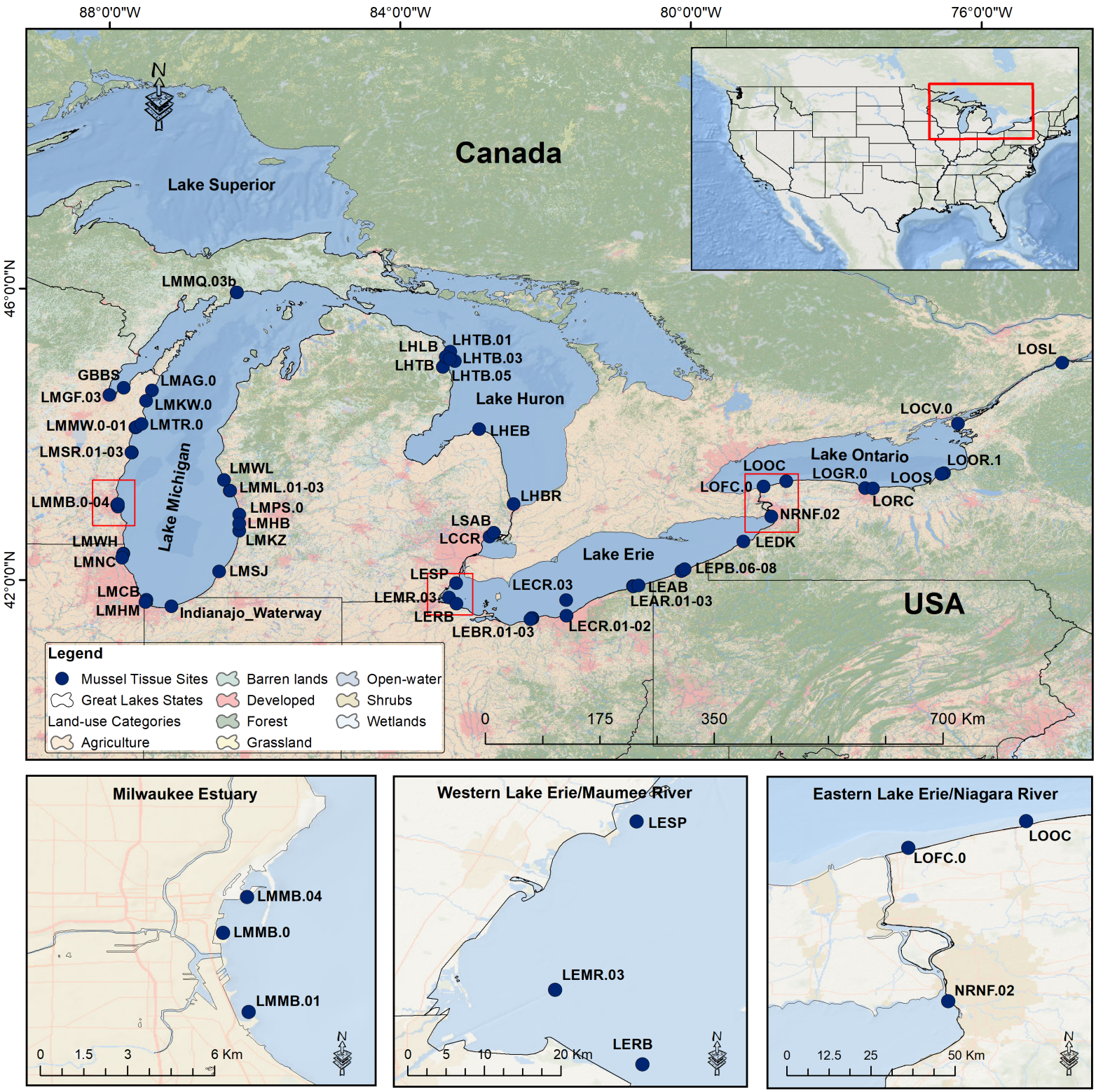


Figure 2. Map depicting Great Lakes Mussel Watch sites (●) sampled in 2022. Additional information on MWP site name, site code acronyms, and approximate nominal latitude and longitude for each site used in this study is provided in Table 1.

3.2 Analytical Methods

Chemical analyses in mussel tissue followed procedures established by the NOAA National Status and Trends (NS&T) Program. Detailed descriptions of analytical methods for legacy organic contaminants analyzed in this study can be found in Kimbrough et al, (2007). Laboratory results were subjected to regular NS&T performance-based quality assessment and quality control procedures for data accuracy and precision. Legacy organic contaminant data is presented in nanograms of contaminant per gram of dry tissue (ng/g dw) with the exception of butyltins which are reported as nanogram tin per gram dry weight (ng Sn/g dw), as they are organotin compounds. Chemical contaminant analyses have been conducted by multiple laboratories over time; however, inter-laboratory calibrations have been conducted to ensure data quality and consistency, with minimal concern for variability. In the MWP, legacy organic contaminants were analyzed by the Geochemical and Environmental Research Group or the Department of Oceanography at Texas A&M University in College Station, TX, from 1986 - 2000 and have been analyzed by TDI-Brooks International since 2000. The data in this report will be archived and publicly available at NOAA's National Centers for Environmental Information (NCEI) (https://www.ncei. noaa.gov/), and on the NCCOS Coastal Pollution Data Explorer (CPDE; https://argis.0SqSO9). For all contaminant classes, a background summary ("Chemical Description") and analysis summary ("Results Summary") can be found throughout this document.

3.3 Data Analysis

Data management and analysis were conducted using a combination of R version 4.0.2 (R Core Team, 2020), Microsoft Excel (Microsoft Corporation, 2018), and ArcGIS (ESRI, 2011). Concentrations of all compounds were blank corrected and any values below the method detection limit (MDL) were assigned a value of zero (although we acknowledge that trace amounts may still be present). The MDL is defined as the lowest concentration able to be detected with 99% confidence by the analytical instrument or method (as defined by the EPA in CFR Title 40 Appendix B to Part 136). Legacy organic compound concentrations are reported as the arithmetic sum of all the individual compound or congener concentrations within a contaminant group. For historical analysis in this report, these sums were the total of a subset of the compounds tested in 2022. Historic analysis dates back as early as possible while including the same subset of compounds over time.

Legacy contaminants were further aggregated into 11 compound groups: total butyltins, total PCBs, total PAHs, total DDTs, total chlordanes, total dieldrins, mirex, total chlorobenzenes, total HCHs, total endosulfans, and chlorpyrifos (Table 11). Data for each legacy contaminants detected in mussel tissue were statistically analyzed at site and regional scales using a series of analysis. Additionally, contamination levels detected in tissue samples at Great Lakes sampling locations in 2022 were compared to regional long-term NOAA NS&T monitoring data. The NS&T data used for comparison comprises mussel tissue sample concentrations collected by the national MWP in the Great Lakes since 1992. Comparison between legacy contamination levels detected in 2022 Great Lakes tissue samples and long-term NOAA NS&T Great Lakes monitoring data were performed using a series of unsupervised multivariate analysis and clustering algorithms [random forest (RF) unsupervised classification, and Fisher-Jenks natural breaks (NB) classification techniques; Anchang et al., 2016; Rey et al., 2016] on the sums of concentrations within each tissue legacy organic contaminant class assessed in this study. Random forest (RF) classification was used among other multivariate techniques to find patterns in legacy organic contaminants presence/absence data, as well as characterize the relationship between legacy contaminants concentration and study sites sampled in 2022. RF classification techniques were conducted using the R statistical software (R version 4.0.2; RCore Team, 2020), with the "randomForest" package for unsupervised RF classification, followed by the Fisher-Jenks algorithm cluster analysis technique (ArcMap 10.8 software; ESRI, 2011), which was used to group tissue legacy organic contaminants concentration sum results into their respective low, medium, and high classes (Anchang et al., 2016; Faridmarandi and Naja, 2014).

The Fisher-Jenks classification method clusters legacy contaminant concentrations into significantly different groups or ranges, by splitting the data into statistically homogenous groups/ranges by iteratively minimizing intra-group variation and maximizing the inter-group separation such that the values or concentration ranges contained within specific groups are more like each other than any other value or concentration range (high, medium and low) of a different group (Anchang et al., 2016; De Smith et al., 2007). It is important to note, the categories derived from the legacy contaminant clusters are not representative measurements that have exceeded any established regulatory thresholds; rather, these denoted concentrations that are significantly higher than the preceding category (Kimbrough et al., 2014; Swam et al., 2024). For each legacy contaminant class, sites are clustered into three groups to represent high contamination (value=3), medium contamination (value=2), and low contamination (value=1) levels, relative to all legacy contaminant data collected. Clustering results are reported in maps provided on the summary page of each legacy contaminant class. Any instances where legacy contaminants were "not detected" in mussel tissue at any Great Lakes sampling location, these contaminants were differentiated from low contamination groups (value=1), and are highlighted on the maps provided in the summary section for each legacy contaminant class.

Methods

Cluster analysis results were further summarized for all legacy contaminant classes per site assessed in this study in two heatmaps showing (1) cluster results of the 2022 study sites respective to concentrations found within the sites assessed in this study in mussel tissue, and (2) cluster results of the 2022 study sites based on national long-term NOAA NS&T monitoring data for the Great Lakes. The clustering process for comparing 2022 mussel tissue contaminant data only to sites analyzed within the 2022 Great Lakes study was similar to the process described for the historic MWP clustering. The contaminant sums were similarly clustered into respective high, medium, and low contamination groups. Additional statistical analyses were conducted using non-parametric Kruskal–Wallis tests, followed by pairwise comparisons, using Wilcoxon rank sum test and Dunn Kruskal–Wallis multiple comparison post hoc test ($p < 0.05$), with Benjamini-Hochberg (BH) method. Spearman's rank correlation was used to evaluate whether concentrations co-varied predictably as a function of time (Zar, 1999) and, therefore, imply a significant increasing or decreasing temporal trend.

Spearman's rank correlation procedure is a nonparametric technique that is free of assumptions about concentrations being normally distributed with a common variance about sites (Kimbrough et al., 2008). The variables used for the Spearman's test were year and legacy contaminants concentration sums. Regional temporal trends were also depicted using a three-point moving average of the yearly mean for all sites combined. Spearman's (non-parametric) rank correlation (ρ) analysis were also used to explore the relationship between each legacy organic contaminant class/groups and study sites. Spearman's (non-parametric) rank correlation (ρ) analyses were also conducted using the R statistical software (R version 4.0.2; R Core Team, 2020).

Legacy contaminant trends in mussel tissue were conducted using a moving average technique. Statistical moving average analysis is a smoothing operation that removes fluctuations from time series data and highlights the overall temporal trends. For each legacy contaminant group, the moving average technique was applied to aggregated annual data from all 18 long-term sites sampled in this study in the attempt to assess basin-wide mussel tissue legacy contaminant trends. Assessing legacy organic contaminants temporal trends across the Great Lakes provided a better picture of chemical contaminant concentration increases or decreases over time, which reflects the effectiveness of regulatory measures that banned some toxic legacy contaminants, and the benefits derived from restoration and clean-up efforts of these contaminants within the Great Lakes Basin.

To provide additional context for the magnitude of legacy contaminants concentrations detected in mussel tissue samples in 2022, legacy contaminant concentrations were further evaluated against the US FDA maximum permissible action levels for molluscan shellfish consumption for human health protection (FDA, 2011) and the US EPA Screening Values (SVs) (EPA, 2000) for recreational fishers. The US FDA Action and Tolerance Levels (Table A2) represent concentration limits at which the US FDA will take legal action to remove shellfish from the market to protect human health (FDA, 2011). The US EPA SVs (Table A2) were developed to provide guidance to state, local, regional, and tribal environmental health officials for their contaminant monitoring programs and for issuing fish and shellfish consumption advisories (EPA, 2000). The SVs represent a threshold concentration of concern for a chemical contaminant in fish and shellfish tissue for a critical toxic or carcinogenic effect in humans. In cases where both carcinogenic and non-carcinogenic SVs were available, the SV for the carcinogenic effects was used. Values higher than the SVs indicate where more intensive site-specific monitoring and/or evaluation of human health risks should be conducted (EPA, 2000). The US EPA and US FDA guideline values are reported on wet tissue weights basis, and where available for comparison, concentrations of chemical contaminants in mussels from the Great Lakes sampling locations in 2022 were converted to wet weights using the percent dry fraction (Table A1).

Methods

Table 1. MWP sites selected for the 2022 Great Lakes survey. Lakes sampled in 2022 are listed geographically from west to east across the Great Lakes Basin. Long-term/historic and recently established sites *(--) sampled in 2022 are depicted in the table below.*

Sampling Location	Site	State	Type	Historic/ Long-term MWP Sites	General Location	Specific Location	Latitude	Latitude
Lake Michigan	GBBS	WI	Nearshore	Long-term	Green Bay	Bayshore Park	44.6370	-87.8081
	LMMQ	WI	River	Long-term	Lake Michigan	Manistique River	45.9457	-86.2497
	LMMQ.03b	WI	River	--	Lake Michigan	Manistique River	45.9454	-86.2491
	Indianajo_Waterway	IN	Nearshore	--	Lake Michigan	Burns Harbor	41.6418	-87.1508
	LMAG.0	WI	River	--	Lake Michigan	Algoma	44.6011	-87.4188
	LMGF.03	WI	River	--	Lake Michigan	Green Bay Fox River	44.5413	-88.0016
	LMKW.0	WI	River	--	Lake Michigan	Kewaunee River	44.4591	-87.4991
	LMKZ	MI	River	--	Lake Michigan	Kalamazoo River	42.6766	-86.2141
	LMBA	WI	River	Long-term	Lake Michigan	Milwaukee Alternate	43.0461	-87.8793
	LMMB	WI	River	Long-term	Lake Michigan	Milwaukee Bay	43.0321	-87.8951
	LMMB.0	WI	River	--	Lake Michigan	Milwaukee Bay	43.0321	-87.8951
	LMMB.01	WI	River	--	Lake Michigan	Milwaukee Bay	43.0078	-87.8872
	LMMB.04	WI	River	--	Lake Michigan	Milwaukee Bay	43.0431	-87.8878
	LMMM	MI	River	Long-term	Lake Michigan	Menominee River	44.0961	-87.5912
	LMMU	MI	Nearshore	Long-term	Lake Michigan	Muskegon	44.2282	-86.3468
	LMML.01	MI	Nearshore	--	Lake Michigan	Muskegon Lake	43.2271	-86.3467
	LMML.02	MI	Nearshore	--	Lake Michigan	Muskegon Lake	43.2239	-86.3451
	LMML.03	MI	Nearshore	--	Lake Michigan	Muskegon Lake	43.2279	-86.3419
	LMMW.0	WI	River	--	Lake Michigan	Manitowoc	44.0933	-87.6455
	LMMW.01	WI	Nearshore	--	Lake Michigan	Manitowoc	44.1037	-87.6270
	LMPS.0	MI	River	--	Lake Michigan	Port Sheldon	42.9016	-86.2154
	LMSJ	MI	River	--	Lake Michigan	Saint Joseph River	42.1165	-86.4939
	LMSR	WI	River	Long-term	Lake Michigan	Sheboygan River	43.7516	-87.6973
	LMSR.01	WI	River	--	Lake Michigan	Sheboygan River	43.7507	-87.6949
	LMSR.02	WI	River	--	Lake Michigan	Sheboygan River	43.7526	-87.6975
	LMSR.03	WI	River	--	Lake Michigan	Sheboygan River	43.7507	-87.7004
	LMTR.0	WI	River	--	Lake Michigan	Two Rivers	44.1432	-87.5623
	LMWH	IL	River	--	Lake Michigan	Waukegan Harbor	42.3626	-87.8119
	LMWH.01	IL	River	--	Lake Michigan	Waukegan Harbor	42.3614	-87.8143
	LMWH.03	IL	River	--	Lake Michigan	Waukegan Harbor	42.3602	-87.8136
	LMWL	MI	River	--	Lake Michigan	White Lake	43.3750	-86.4267
	LMCB	IN	Nearshore	Long-term	Lake Michigan	Calumet Breakwater	41.7271	-87.4950
	LMHB	MI	Nearshore	Long-term	Lake Michigan	Holland Breakwater	42.7731	-86.2150
	LMHM	IN	Nearshore	Long-term	Lake Michigan	Hammond Marina	41.6986	-87.5082
	LMNC	IL	Nearshore	Long-term	Lake Michigan	North Chicago	42.3046	-87.8273
	TBLL	MI	Nearshore	Long-term	Lake Michigan	Leelanau State Park	45.2056	-85.5368

Methods

Table 1 (cont). MWP sites selected for the 2022 Great Lakes survey. Lakes sampled in 2022 are listed geographically from west to east across the Great Lakes Basin. Long-term/historic and recently established sites *(--) sampled in 2022 are depicted in the table below.*

Sampling Location	Site	State	Type	Historic/ Long-term MWP Sites	General Location	Specific Location	Latitude	Latitude
Lake Huron	LHEB	MI	Nearshore	--	Lake Huron	Eagle Bay	44.0705	-82.9157
	LHLB	MI	Nearshore	--	Lake Huron	Lincoln Bay	45.1339	-83.3111
	LHTB	MI	Nearshore	Long-term	Lake Huron	Thunder Bay	44.9221	-83.4135
	LHTB.01	MI	Nearshore	--	Lake Huron	Thunder Bay	45.0615	-83.3755
	LHTB.03	MI	Nearshore	--	Lake Huron	Thunder Bay	45.0060	-83.2498
	LHTB.05	MI	Nearshore	--	Lake Huron	Thunder Bay	45.0346	-83.3180
	LHBR	MI	Nearshore	Long-term	Lake Huron	Black River Canal	43.0443	-82.4386
	SBSR	MI	River	Long-term	Lake Huron	Saginaw River	43.6735	-83.8366
Lake St. Clair	LCCR	MI	Connecting Channel	--	Lake St. Clair	Clinton River	42.5940	-82.7703
	LSAB	MI	Connecting Channel	Long-term	Lake St. Clair	Anchor Bay	42.6491	-82.7110
Detroit River	DRMI	MI	Connecting Channel	Long-term	Detroit River	Mud Island	42.2377	-83.1402
	DRSE	MI	Connecting Channel	Long-term	Detroit River	South End	42.1069	-83.1355
Lake Erie	LEAR	OH	River	Long-term	Lake Erie	Ashtabula River	41.9112	-80.7876
	LEAR.01	OH	River	--	Lake Erie	Ashtabula River	41.9117	-80.7938
	LEAR.02	OH	River	--	Lake Erie	Ashtabula River	41.9179	-80.7956
	LEAR.03	OH	River	--	Lake Erie	Ashtabula River	41.9119	-80.7901
	LEBR	OH	River	--	Lake Erie	Black River	41.4743	-82.1810
	LEBR.01	OH	River	--	Lake Erie	Black River	41.4734	-82.1828
	LEBR.02	OH	River	--	Lake Erie	Black River	41.4773	-82.1834
	LEBR.03	OH	River	--	Lake Erie	Black River	41.4770	-82.1900
	LEBU	NY	River	Long-term	Lake Erie	Buffalo River	42.8800	-78.8916
	LECR	OH	River	Long-term	Lake Erie	Cuyahoga River	41.4994	-81.7188
	LECR.01	OH	River	--	Lake Erie	Cuyahoga River	41.5082	-81.7133
	LECR.02	OH	River	--	Lake Erie	Cuyahoga River	41.5095	-81.7136
	LECR.03	OH	River	--	Lake Erie	Cuyahoga River	41.7233	-81.7157
	LEMR	OH	River	Long-term	Lake Erie	Maumee River	41.7014	-83.4587
	LEMR.03	OH	Offshore	--	Lake Erie	Maumee River	41.7618	-83.3286
	LEPB	PA	River	Long-term	Lake Erie	Presque Isle Bay	42.1377	-80.0952
	LEPB.06	PA	River	--	Lake Erie	Presque Isle Bay	42.1297	-80.1132
	LEPB.07	PA	River	--	Lake Erie	Presque Isle Bay	42.1234	-80.1322
	LEPB.08	PA	River	--	Lake Erie	Presque Isle Bay	42.1475	-80.0857
	LERR	MI	River	Long-term	Lake Erie	River Raisin	41.8932	-83.3248

Methods

Table 1 (cont'd). MWP sites selected for the 2022 Great Lakes survey. Lakes sampled in 2022 are listed geographically from west to east across the Great Lakes Basin. Long-term/historic and recently established sites *(--) sampled in 2022 are depicted in the table below.*

Sampling Location	Site	State	Type	Historic/ Long-term MWP Sites	General Location	Specific Location	Latitude	Latitude
Lake Erie	LEAB	OH	River	Long-term	Lake Erie	Ashtabula	41.9246	-80.7183
	LEDK	NY	Nearshore	Long-term	Lake Erie	Dunkirk	42.5291	-79.2776
	LELR	OH	Nearshore	Long-term	Lake Erie	Lorain	41.4611	-82.2070
	LERB	OH	Offshore	Long-term	Lake Erie	Reno Beach	41.6745	-83.2261
	LESP	OH	Offshore	Long-term	Lake Erie	Stony Point	41.9586	-83.2330
	LEOW	OH	Offshore	Long-term	Lake Erie	Old Woman Creek	41.3850	-82.5187
	SBPP	OH	Offshore	Long-term	Lake Erie	Peach Orchard Pt.	41.6596	-82.8250
Niagara River	NRNF	NY	Nearshore	Long-term	Niagara River	Niagara Falls	43.0468	-78.8920
	NRNF.02	NY	Nearshore	--	Niagara River	Niagara Falls	42.8742	-78.8953
Lake Ontario	LOCV	NY	Connecting Channel	Long-term	Lake Ontario	Cape Vincent	44.1442	-76.3247
	LOCV.0	NY	Connecting Channel	--	Lake Ontario	Cape Vincent	44.1441	-76.3246
	LOEC	NY	Nearshore	Long-term	Lake Ontario	Eightmile Creek	43.3387	-78.7187
	LOFC.0	NY	Nearshore	--	Lake Ontario	Fourmile Creek	43.2840	-79.0019
	LOGR.0	NY	River	--	Lake Ontario	Genesee River	43.2599	-77.6024
	LOOR.1	NY	River	--	Lake Ontario	Oswego River	43.4652	-76.5150
	LOOC	NY	Nearshore	Long-term	Lake Ontario	Olcott	43.3553	-78.6866
	LOOS	NY	Nearshore	Long-term	Lake Ontario	Oswego	43.4528	-76.5508
	LORC	NY	Nearshore	Long-term	Lake Ontario	Rochester	43.2578	-77.4953
	LOSL	NY	River	Long-term	Lake Ontario	St.Lawrence River	44.9798	-74.8910
Hudson River	HRCI	NY	River	Long-term	Hudson River	Cruger Island	42.0338	-73.9293
	HRPK	NY	River	Long-term	Hudson River	Poughkeepsie	41.7088	-73.9405

4.0 RESULTS - TOTAL BUTYLTINS

4.1 Butyltins Chemical Description

For this document, Total Butyltins analyzed in 2022 is the sum of 4 organometallic compounds (monobutyltin, dibutyltin, tributyltin, and tetrabutyltin), and Total Butyltins analyzed historically is the sum of 3 organometallic compounds (Table 2). Tetrabutyltin is a precursor of tributyltin, which is the parent compound of diubtyltin and monobutyltin, its less toxic transformation products. Tributyltin is the most toxic of the four compounds. Tributyltin has had a variety of uses ranging from a biocide in antifouling paints to a catalyst and an ingredient in glass coatings (Bennett, 1996; Birchenough et al., 2002). In the late 1960s, tributyltin was incorporated into an antifouling polymer paint system, quickly becoming one of the most effective paints used on boat hulls (Birchenough et al., 2002). Beginning in 1989, the use of tributyltin as an antifouling agent was banned in the US for use on non-aluminum vessels smaller than 25 meters in length (Gibbs and Bryan, 1996). However, the continued use of tributyltin on ships and other antifouling paint applications increased the ubiquity of the compound in aquatic environments. Thus, tributyltin and its metabolites continue to be detected in many components of coastal and marine ecosystems in the U.S.

Butyltins can be highly toxic in multiple forms as they naturally degrade in the environment. Tributyltin was first shown to have biocidal properties in the 1950s (Bennett, 1996; Evans, 1970). The presence of tributyltin in the environment has been linked to endocrine disruption (Batley, 1996; Strand et al., 2009). In the mid-1970s, the use of tributyltin was linked to abnormal shell development and poor weight gain in oysters and, more recently, to an imposex condition (females developing male characteristics) in marine gastropod mollusks (Batley, 1996; Strand et al., 2009). There is no FDA-recommended safety level for butyltins in fish and fish products.

In the aquatic environment, tributyltin is degraded by microorganisms and sunlight into monobutyltin and dibutyltin (Bennett, 1996). Experiments have shown that the half-life of tributyltin is on the order of days, and degradation to monobutyltin takes approximately a month. However, in deeper anoxic sediments, the half-life of tributyltin appears to be on the order of 2-4 years or longer (Batley, 1996). Although banned decades ago, tributyltin and its metabolites continue to be detected in many components of the environment. While organotin compounds are readily bioaccumulated by aquatic organisms from water, there is no evidence for biomagnification up the food chain (WHO, 1980). Tributyltin is sparingly soluble in water and is readily associated with suspended particles in the water column. Butyltins are persistent in the aquatic environment and accumulate in sediment; therefore, they will continue to affect the aquatic environment (Gibbs and Bryan, 1996; EPA, 2003). The release of organotins into the atmosphere is insignificant due to their low vapor pressure and rapid photodegradation.

Table 2. Butyltin compounds composing the "Total Butyltins" summation for the 2022 Great Lakes survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2022	Analyzed in Historical Data
Monobutyltin	●	●
Dibutyltin	●	●
Tributyltin	●	●
Tetrabutyltin	●	

Results - Total Butyltins

4.2 Magnitude and Distribution of Butyltins in Mussel Tissue in 2022

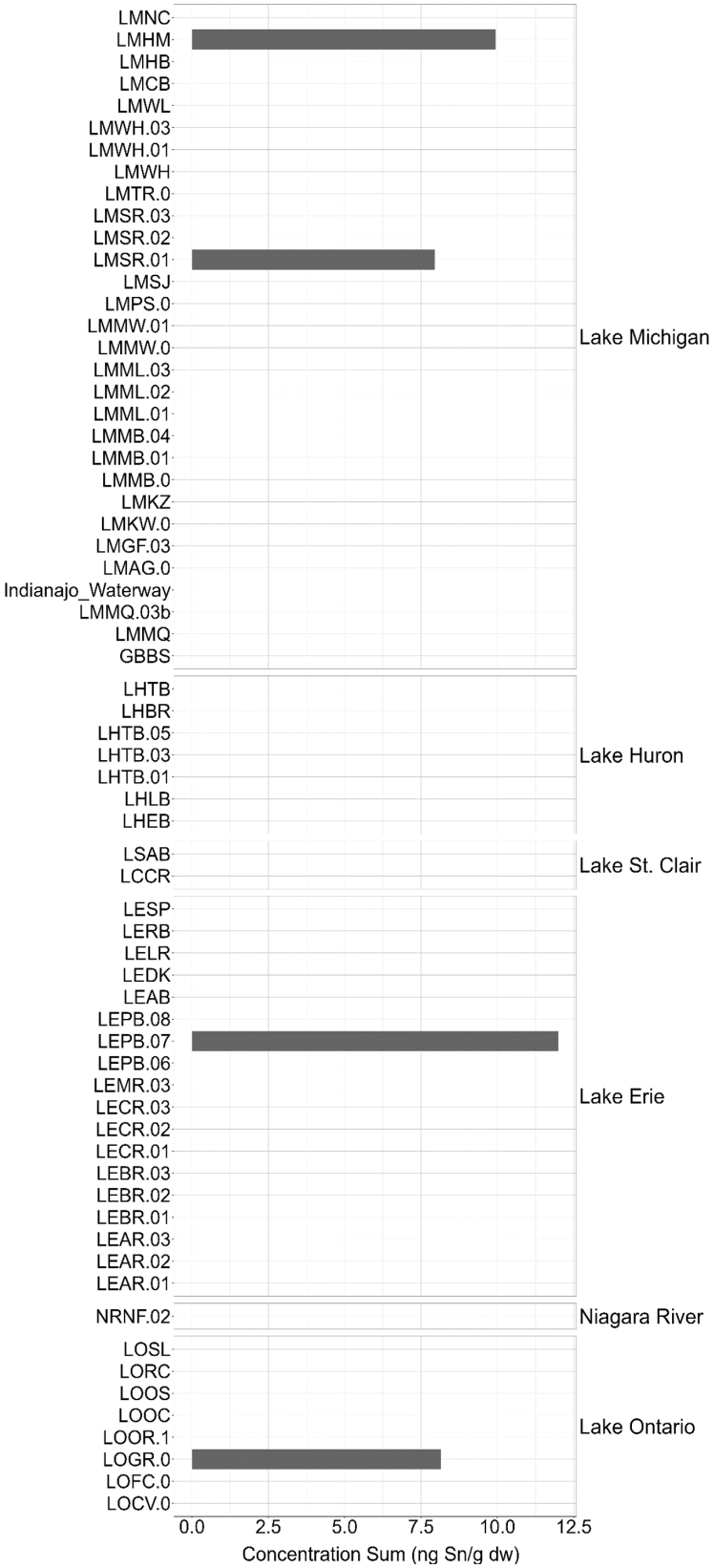


Figure 3. Bar graph showing the magnitude of Total Butyltin concentration sums detected in mussel tissue in the Great Lakes Basin during the 2022 sampling period. Lakes are listed geographically from west to east across the Great Lakes Basin.

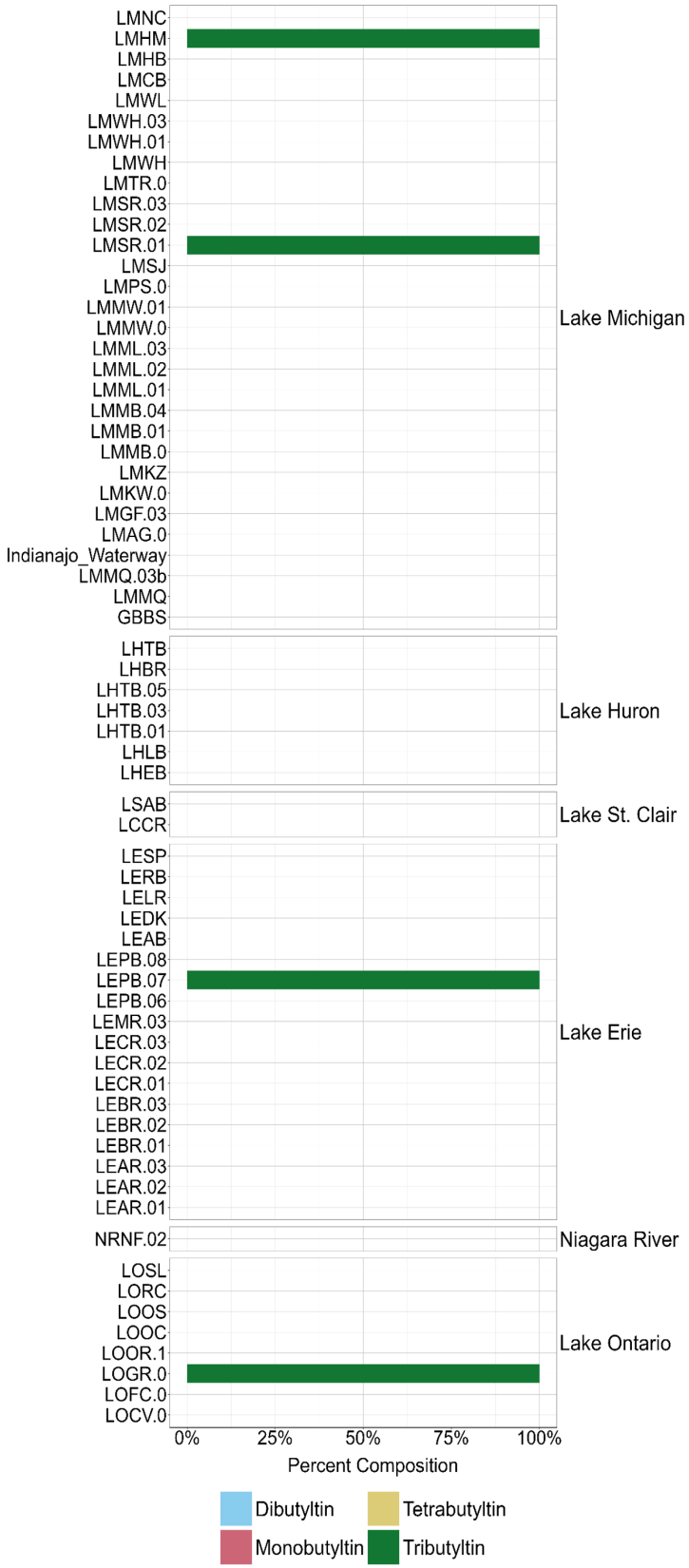


Figure 4. Percent composition of individual butyltin compounds in Total Butyltin concentration sums per site in the Great Lakes Basin during the 2022 sampling period. Lakes are listed geographically from west to east across the Great Lakes Basin.

Results - Total Butyltins

4.3 Historical Context of Butyltins Magnitude and Distribution in Mussel Tissue

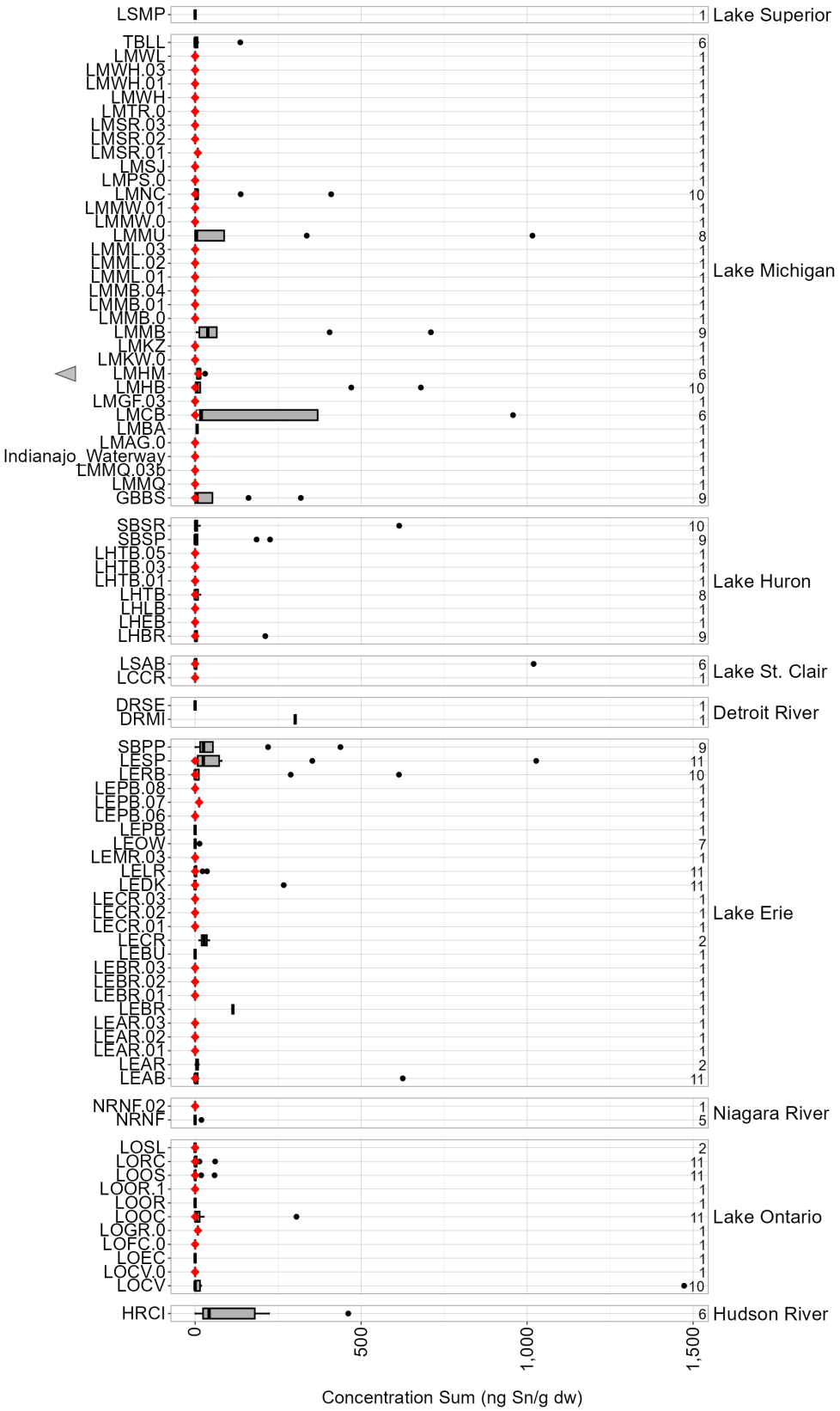


Figure 5. Total Butyltin concentration sums (ng Sn/g dw) in mussel tissue in 2022 (◆ - red diamonds), compared to boxplots of the historic Total Butyltin concentration (ng Sn/g dw) sums for each site (1992-2022) and results of the trend analysis showing significant increasing (▻) or decreasing (◅) contamination trends over time (gray triangles). Site SBPP (oullier) was removed for visualization purpose. Additional information is provided in Table 13 and Table A6. Lakes are listed geographically from west to east across the Great Lakes Basin. The number of years each site has been sampled since 1992 is noted at the side (right) of the plot for each site.

Results - Total Butyltins

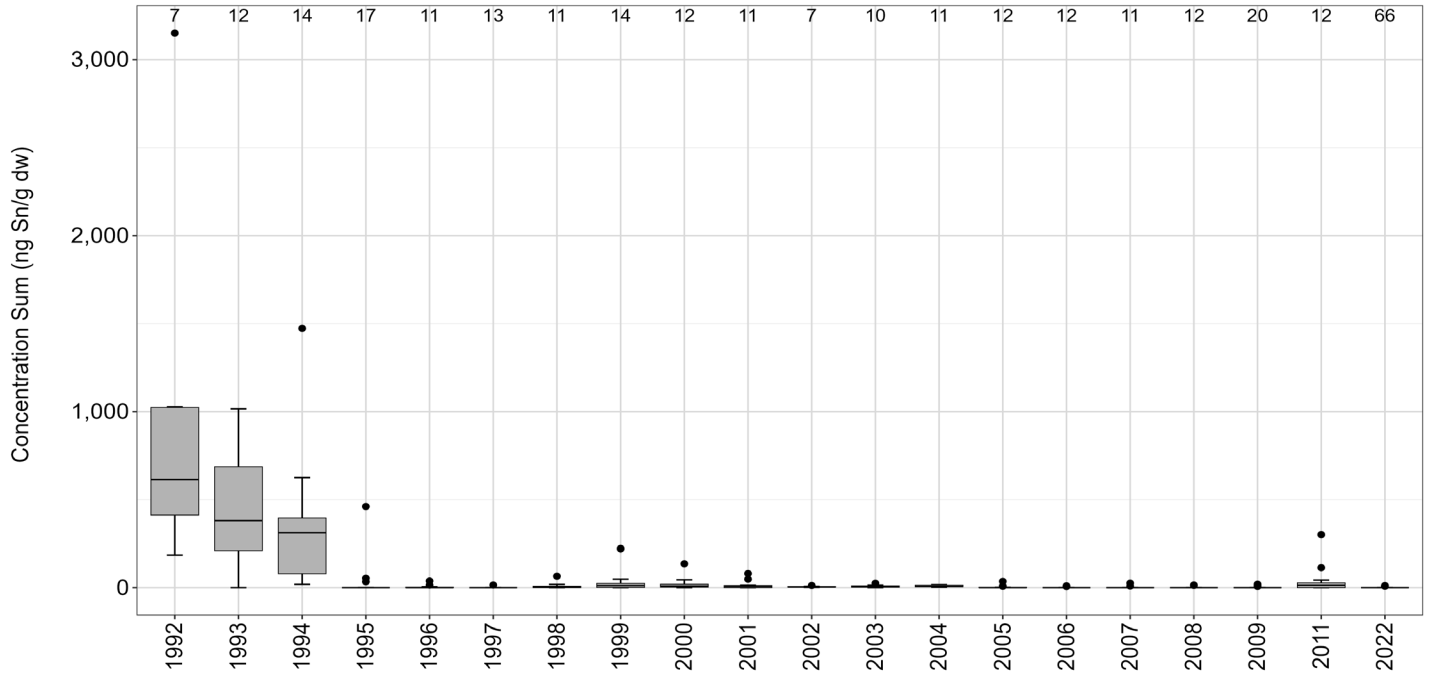


Figure 6. Boxplots representing the historic Total Butyltin concentration sums measured in mussel tissue of the sites analyzed in this study. The number of sites that were sampled each year is noted at the top of the plot.

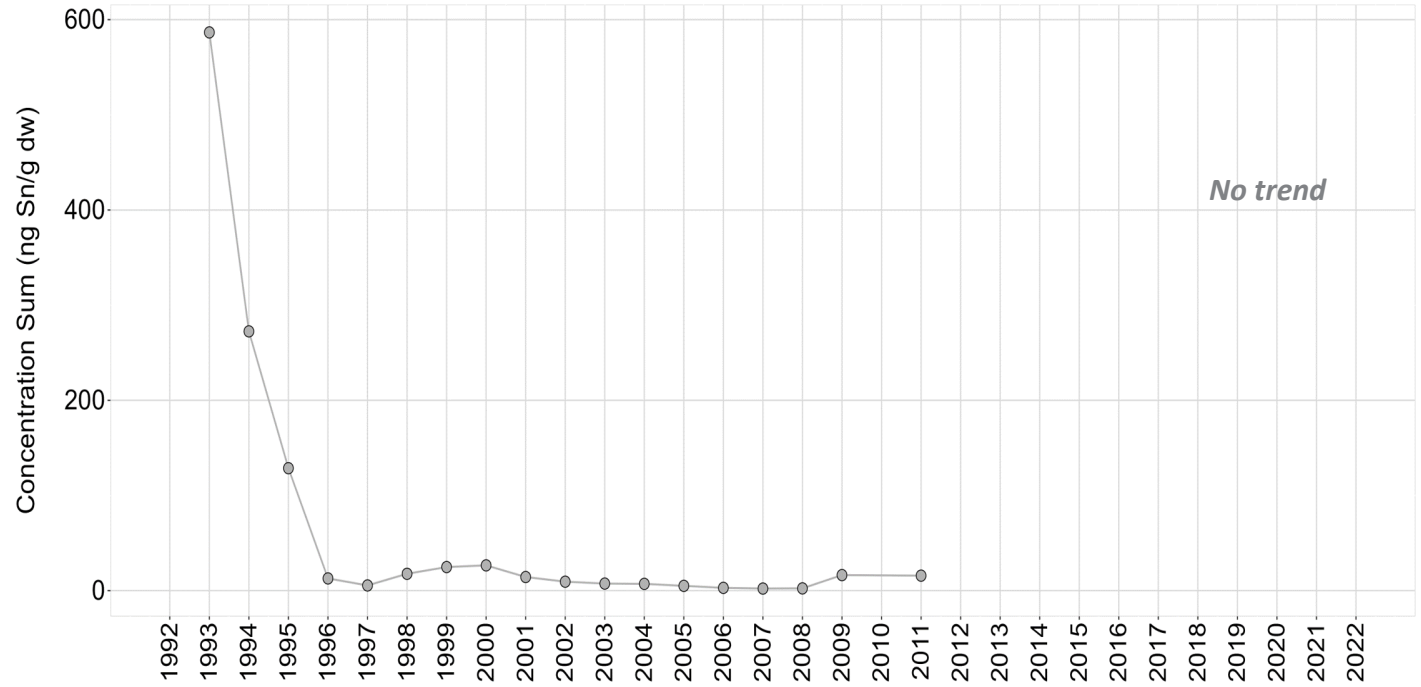


Figure 7. Three-point moving average of the yearly mean Total Butyltin concentration sums measured in mussel tissue at sites analyzed in this study.

Results - Total Butyltins

4.4 Butyltins Summary

2022 Dreissenid Mussel Tissue Results (sum of 4 compounds):

- Total Butyltins were detected in dreissenid mussels at 6% (4/66) of the sites surveyed in 2022 (Figure 3; Figure 4; Figure 8).
- Total Butyltins concentration summary statistics (Figure 3; Figure 4):
  - Range: 0.00 – 12.0 ng Sn/g dw
  - Minimum detectable concentration sum was detected at LMSR.01 (7.96 ng Sn/g dw)
  - Maximum concentration sum was detected at LEPB.07 (12.0 ng Sn/g dw)
  - Median: 9.05 ng Sn/g dw
  - Mean ± SD: 9.51 ± 1.88 ng Sn/g dw

Historical Context for Dreissenid Mussel Tissue (sum of 3 compounds):

- 94% (62/66) of sites were above their historic median concentrations in 2022 (Figure 5).
- No significant regional temporal trend of Total Butyltin concentration sums was observed across the Great Lakes Basin ($\alpha = 0.05$; Figure 6; Figure 7).
- 0 sites assessed across the Great Lakes Basin showed significant increasing temporal trends of Total Butyltin concentration sums ($\alpha = 0.05$; Figure 5).
- 1 site (LMHM) assessed across the Great Lakes Basin showed significant decreasing temporal trends of Total Butyltin concentration sums ($\alpha = 0.05$; Figure 5; Table 13; Table A6)
- The percentage of sites sampled in 2022 grouped into each cluster based on historic Great Lakes data (Figure 9):
  - 100% (66/66) of sites detected in the low cluster (1.80 - 369.2 ng Sn/g dw)
  - 0% of sites detected in the medium cluster (369.3 - 1,473.0 ng Sn/g dw)
  - 0% of sites detected in the high cluster (1,473.1 - 3,150.8 ng Sn/g dw)

General Observations:

- Total Butyltin concentration sums were generally low or not detected in dreissenid mussel tissue at sites surveyed in the Great Lakes in 2022 (Figure 3; Figure 5; Figure 8).
  - There was one instance of medium contamination (LMHM; 9.94 ng Sn/g dw) and one instance of high contamination (LEPB.07; 12.0 ng Sn/g dw) detected in dreissenid mussels in 2022.
- Total Butyltins was only detected at one (LMHM) of the eighteen long-term MWP sites sampled in 2022. Historically, highest Total Butyltin concentration sums were detected in mussels from sites sampled in Niagara River (19 ng Sn/g dw), Detroit River (301.3 ng Sn/g dw), Hudson River (461.3 ng Sn/g dw), Lake Huron (614.7 ng Sn/g dw), Lake Michigan (1,016.0 ng Sn/g dw), Lake St. Clair (1,019.6 ng Sn/g dw), Lake Ontario (1,473.0 ng Sn/g dw), and Lake Erie (3,150.8 ng Sn/g dw; Figure 5; Figure 10).
- Butyltin contamination levels across the Great Lakes Basin may reflect the historic use, current levels from urbanization, industrial activities, and the persistence of these compounds (Apeti and Lauenstein, 2006; Bartlett et al., 2005).
- The continued presence and detection of tributyltin (the butyltin parent compound) in mussel tissue suggests that there continues to be inputs into the Great Lakes aquatic environment, perhaps from sources such as resuspended sediments, the refurbishing of historic vessels, or from vessels originating outside the US, since tributyltin has been banned in the US for use on most vessels since 1989.
- Total Butyltin concentration sums were below the US EPA recommended screening values for both recreational and subsistence fishers (Table A2; Table A3).

Results - Total Butyltins

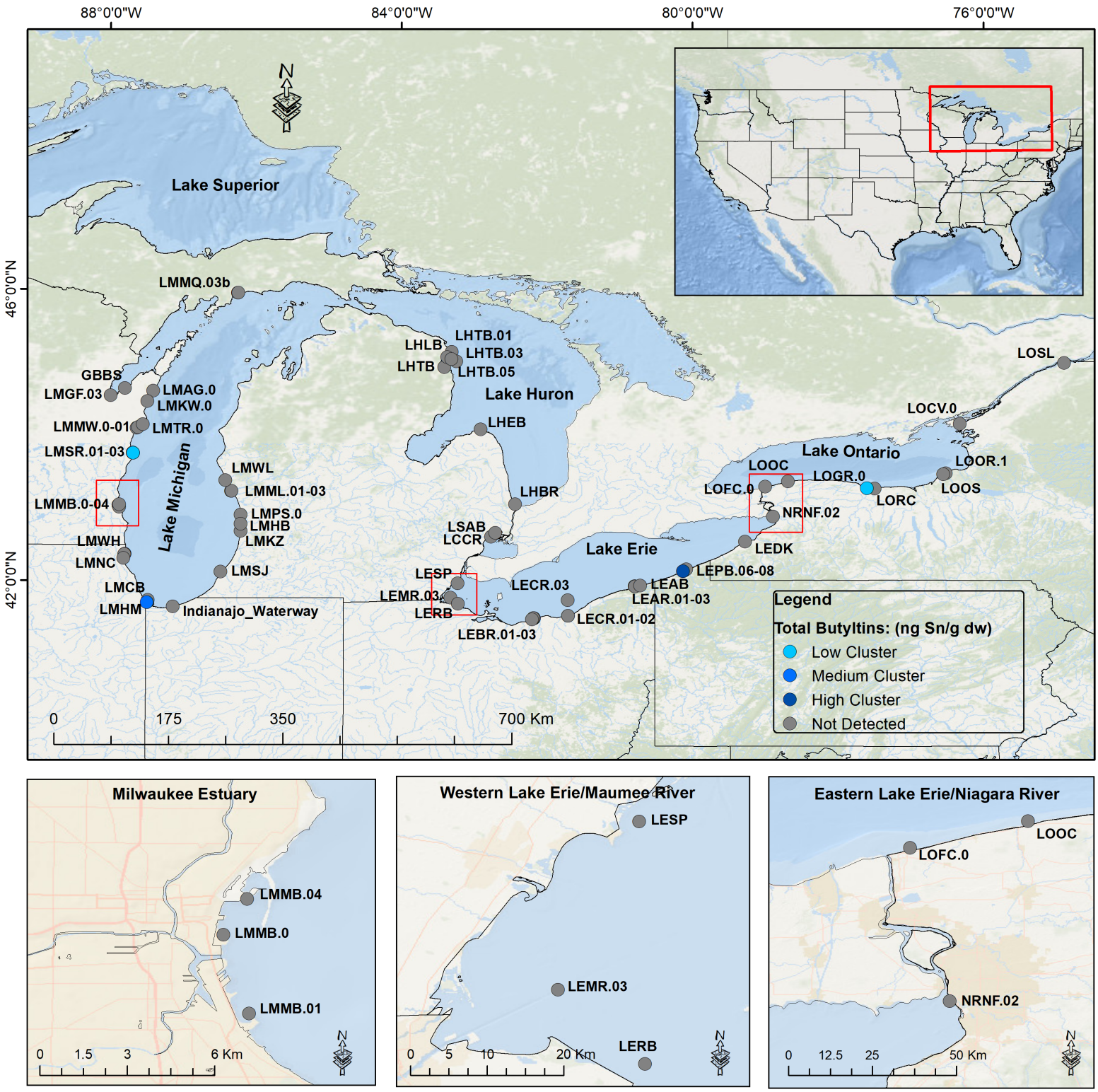


Figure 8. Total Butyltin concentration sums (ng Sn/g dw) measured in Great Lakes mussel tissue during 2022. Some mussel sampling locations are shown to overlap due to the size of the circles. Colors represent contamination magnitude and clusters as follows: low (●), medium (●), high (●), and not detected (●).

Results - Total Butyltins

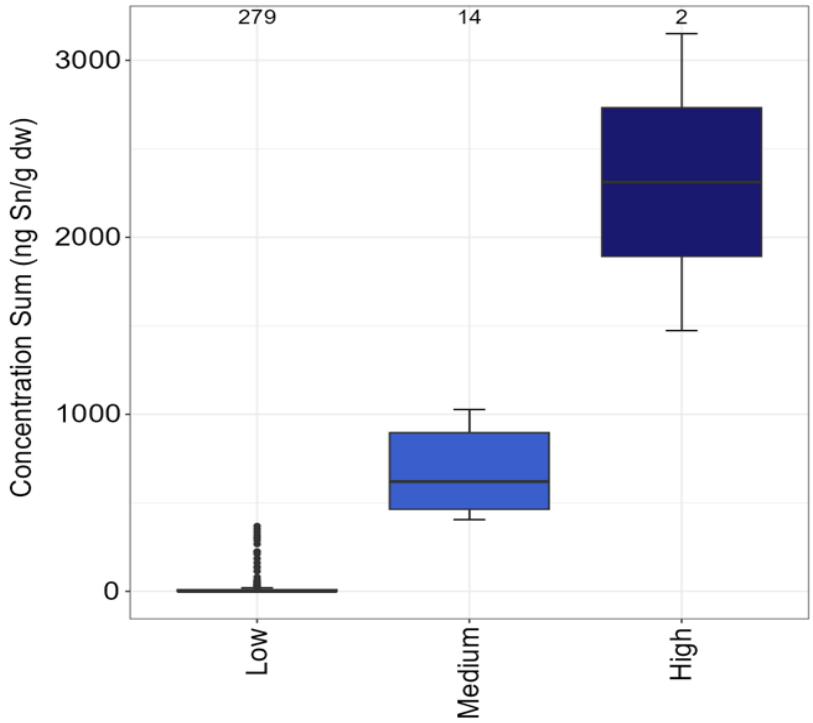


Figure 9. Total Butyltin concentration sums (ng Sn/g dw) measured in Great Lakes mussel tissue during 1992-2022. Colors represent contamination magnitude as follows: low ( ), medium ( ), and high ( ).

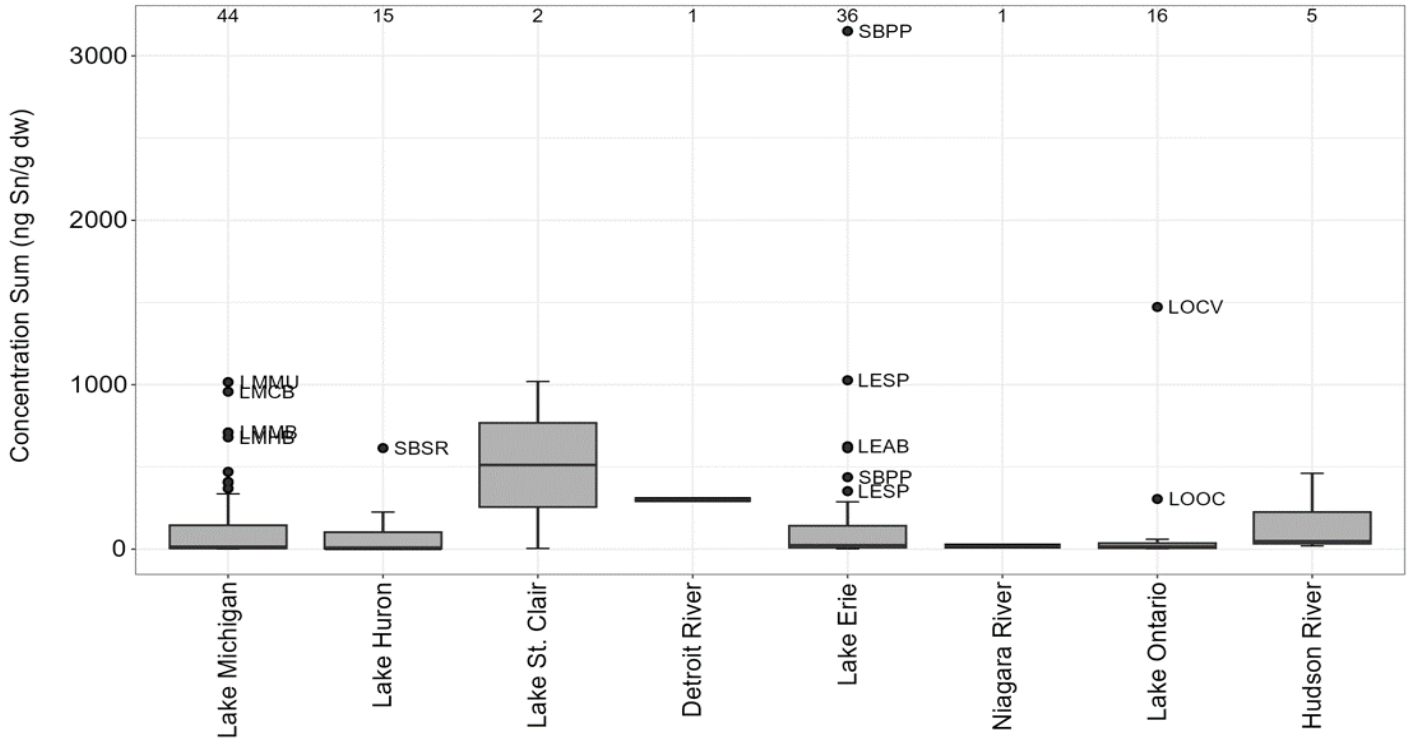


Figure 10. Total Butyltin concentration sums (ng Sn/g dw) measured in Great Lakes mussels between 1992-2022. Lakes and major rivers including connecting channels sampled during 1992-2022 are listed geographically from west to east across the Great Lakes Basin. The number of butyltin samples collected across each lake and major rivers including connecting channels sampled during 1992-2022 is noted at the top of the plot.

5.0 RESULTS - TOTAL CHLORDANES

5.1 Chlordanes Chemical Description

For this document, Total Chlordanes analyzed both in 2022 and historically in mussel tissue samples are the sum of 7 compounds (Table 3). Chlordane belongs to a group of organic pesticides called cyclodienes. Technical chlordane, an insecticide, was used in the US from 1948-1983 to control insect pests in agricultural and urban settings. It was also the predominant insecticide for the control of subterranean termites (Kamble et al., 1992). Chlordane restriction for food production was implemented in 1978, while agricultural uses were banned in 1983 and all uses were banned by 1988 (Pagano and Garner, 2020). These compounds are some of the most ubiquitous contaminants measured by the Mussel Watch Program.

The US FDA has established a 0.3 ppm wet weight safety level in all fish for both chlordane and heptachlor/ heptachlor epoxide (FDA, 2011). Human exposure to chlordane can occur through eating crops from contaminated soil, fish, and shellfish from contaminated waters or breathing contaminated air. Chlordane can enter the body by being absorbed through the skin, inhalation, and ingestion. At high levels, chlordane can affect the nervous system, digestive system, brain, and liver and is also carcinogenic (Trivedi et al., 2024). Chlordane is highly toxic to invertebrates and fish (Eisler, 1990).

Chlordane is removed from both soil and water sources primarily by volatilization and particle-bound runoff. In air, chlordane degrades through photolysis and oxidation. Chlordane exists in the atmosphere primarily in the vapor-phase, but the particle-bound fraction is important for long-range transport. Chlordane is prevalent in the Arctic due to the grasshopper effect, the geochemical process by which certain chemicals are vaporized and transported from warmer to colder regions of the Earth, and is distributed in the food web (Hargrave et al., 1992). Chlordane binds to dissolved organic matter, further facilitating its transport in natural waters.

Table 3. Chlordane compounds composing the "Total Chlordanes" summation for the 2022 Great Lakes survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2022	Analyzed in Historical Data
Alpha-Chlordane	●	●
Trans-Nonachlor	●	●
Gamma-Chlordane	●	●
Cis-Nonachlor	●	●
Heptachlor	●	●
Heptachlor-Epoxide	●	●
Oxychlordane	●	●

Results - Total Chlordanes

5.2 Magnitude and Distribution of Chlordanes in Mussel Tissue in 2022

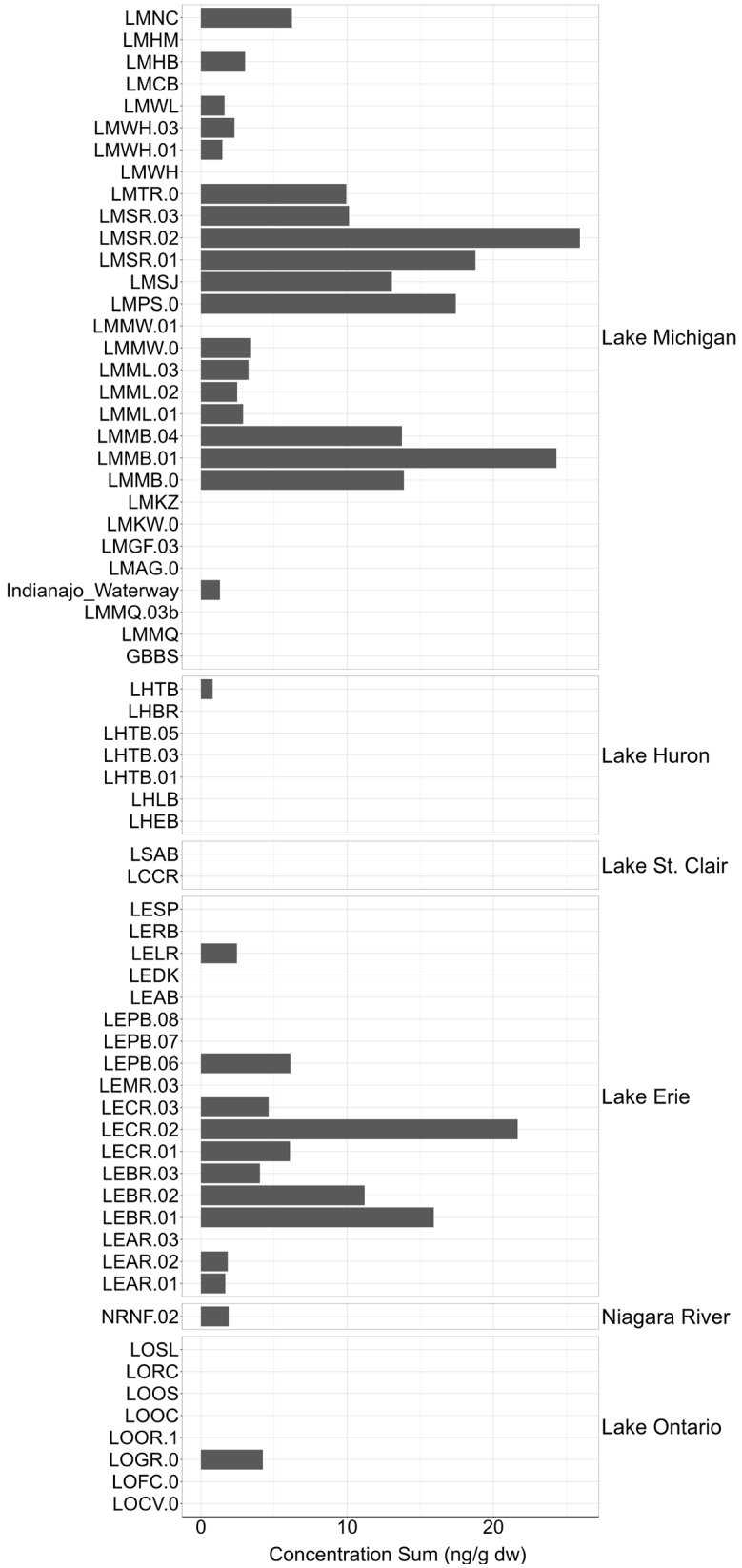


Figure 11. Bar graph showing the magnitude of Total Chlordane concentration sums detected in mussel tissue in the Great Lakes Basin during the 2022 sampling period. Lakes are listed geographically from west to east across the Great Lakes Basin.

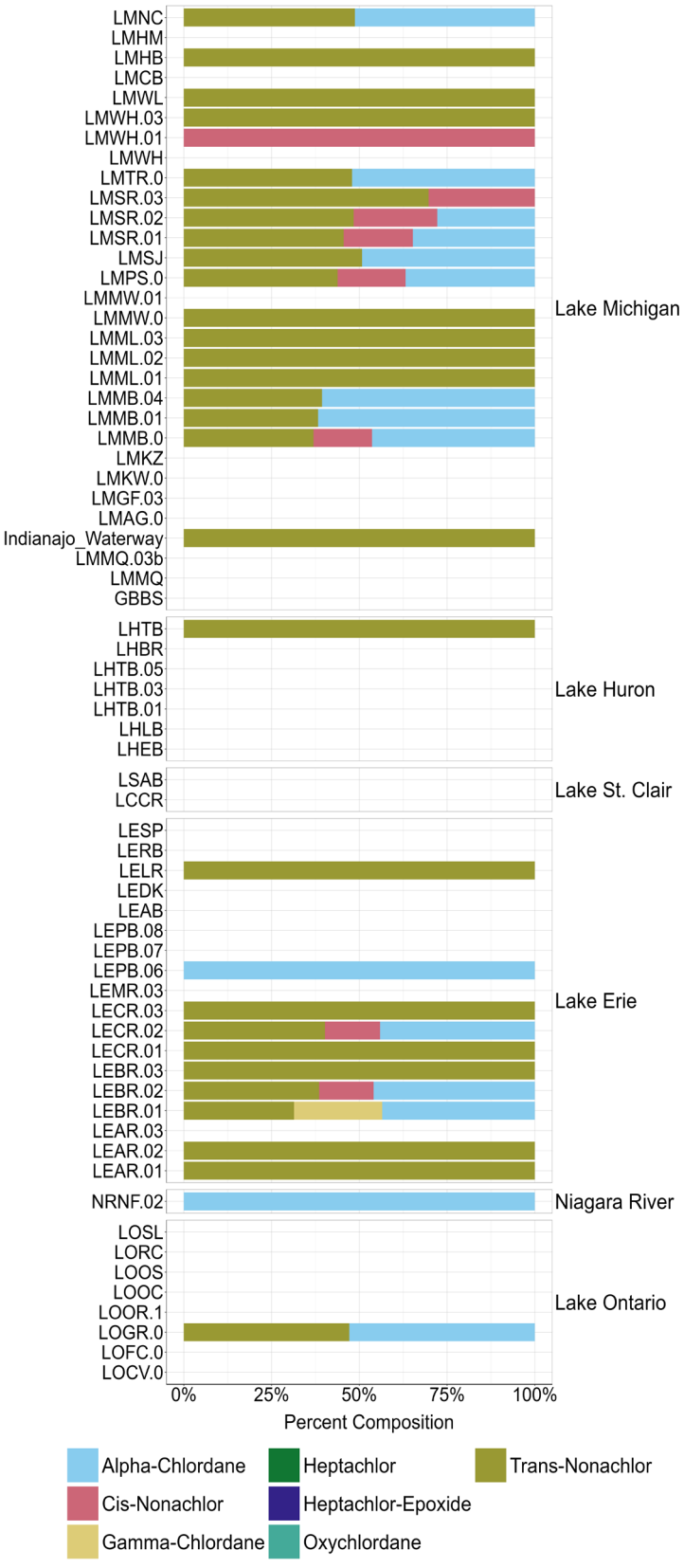


Figure 12. Percent composition of individual chlordane contaminants in Total Chlordane concentration sums per site in the Great Lakes Basin during the 2022 sampling period. Lakes are listed geographically from west to east across the Great Lakes Basin.

Results - Total Chlordanes

5.3 Historical Context of Chlordanes Magnitude and Distribution in Mussel Tissue

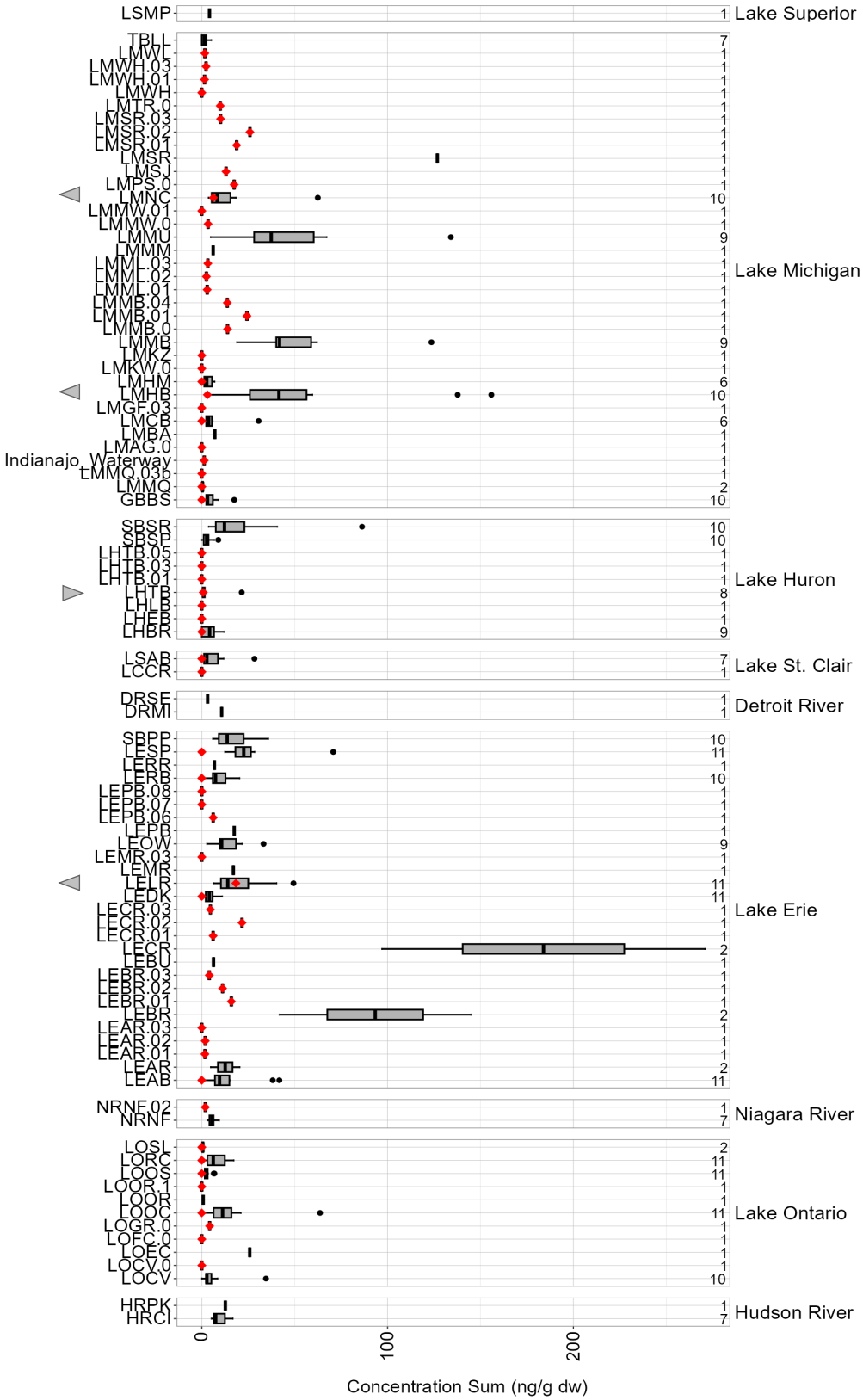


Figure 13. Total Chlordane concentration sums (ng/g dw) in mussel tissue in 2022 (◆ - red diamonds), compared to boxplots of the historic Total Chlordane concentration sums (ng/g dw) for each site (1992-2022) and results of the trend analysis showing significant increasing (▷) or decreasing (◁) contamination trends over time (gray triangles). Additional information is provided in Table 13 and Table A6. Lakes are listed geographically from west to east across the Great Lakes Basin. The number of years each site has been sampled since 1992 is noted at the side (right) of the plot for each site.

Results - Total Chlordanes

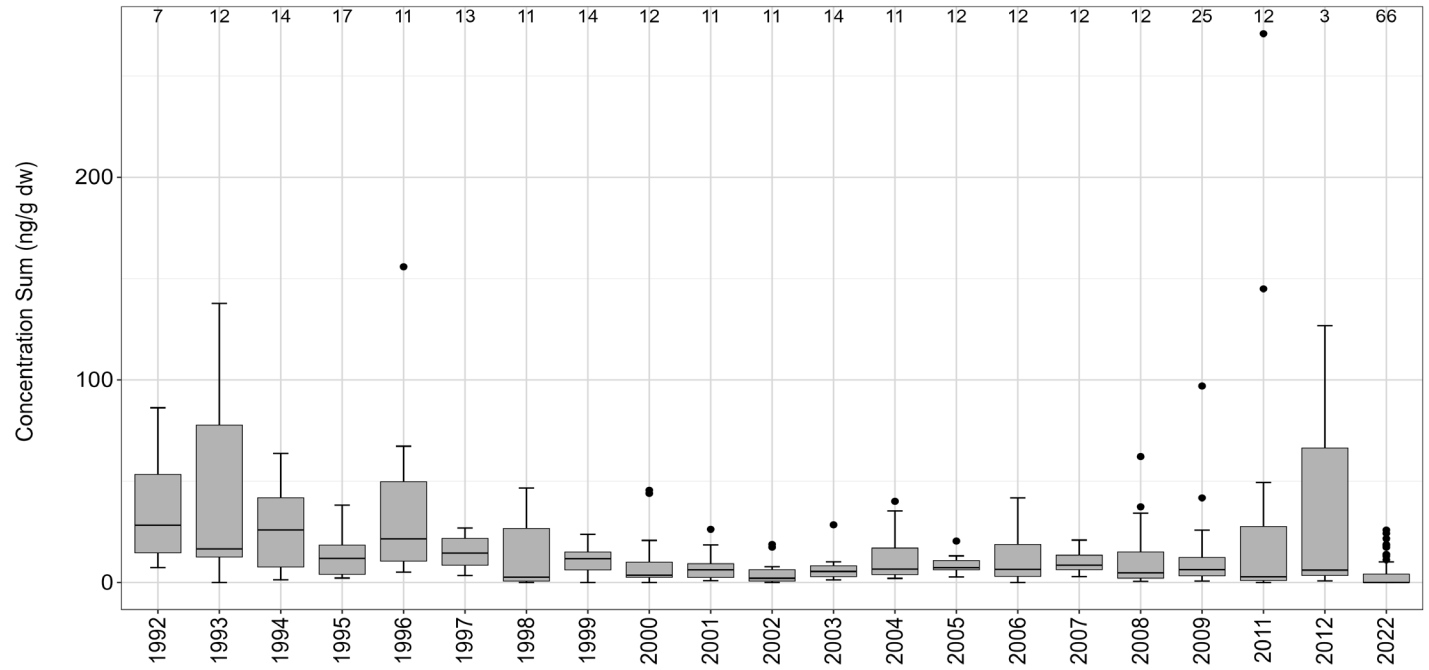


Figure 14. Boxplots representing the historic Total Chlordane concentration sums (ng/g dw) measured in mussel tissue of the sites analyzed in this study. The number of sites that were sampled each year is noted at the top of the plot.

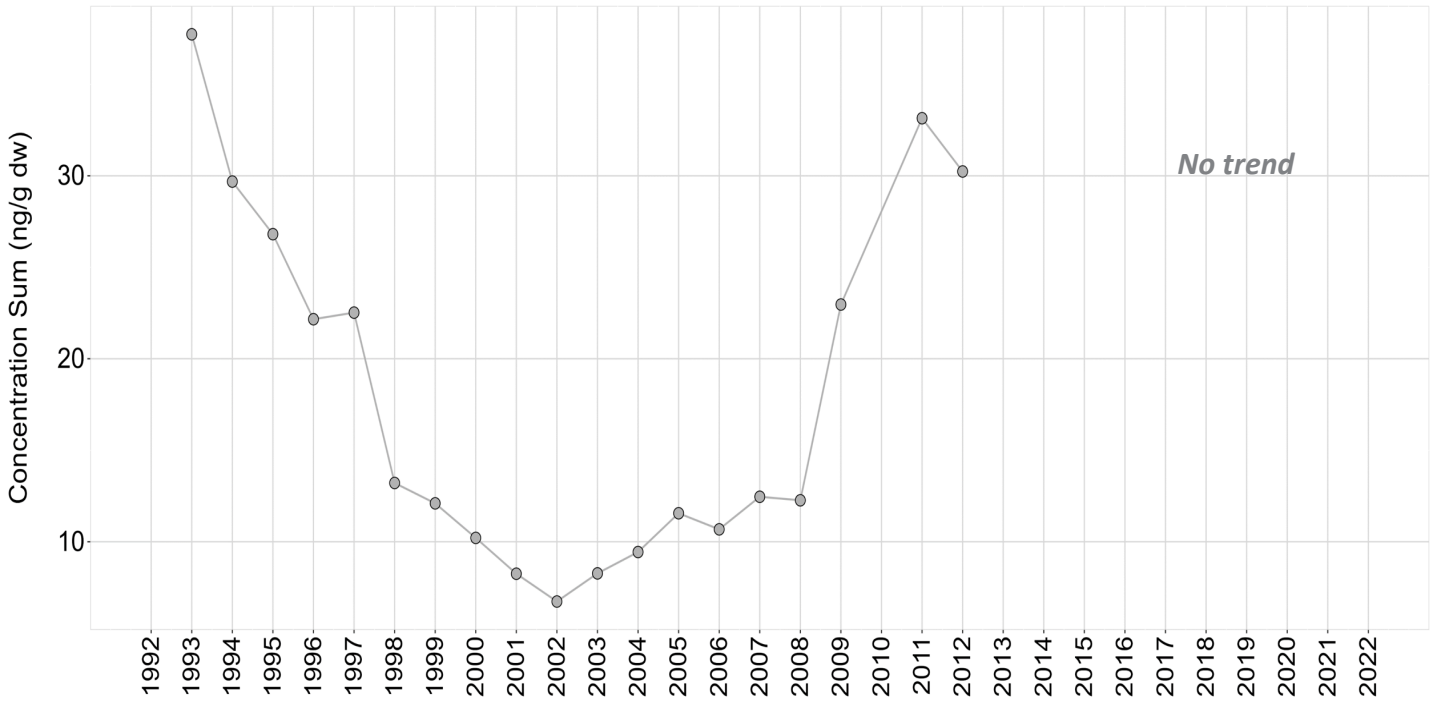


Figure 15. Three-point moving average of the yearly mean Total Chlordane concentration sums (ng/g dw) measured in mussel tissue at sites analyzed in this study.

Results - Total Chlordanes

5.4 Chlordanes Summary

2022 Dreissenid Mussel Tissue Results (sum of 7 compounds):

- Total Chlordanes were detected in dreissenid mussels at 48% (32/66) of the sites surveyed in 2022 (Figure 11; Figure 12; Figure 16).
- Total Chlordanes concentration summary statistics (Figure 11; Figure 12):
  - Range: 0.00 – 25.9 ng/g dw
  - Minimum detectable concentration sum was detected at LHTB (0.798 ng/g dw)
  - Maximum concentration sum was detected at LMSR.02 (25.9 ng/g dw)
  - Median: 4.43 ng/g dw
  - Mean ± SD: 7.97 ± 7.42 ng/g dw

Historical Context for Dreissenid Mussel Tissue (sum of 7 compounds):

- 52% (34/66) of sites were above their historic median concentrations in 2022 (Figure 13).
- No significant regional temporal trend of Total Chlordane concentration sums was observed across the Great Lakes Basin sampling locations between 1992-2022 ($\alpha = 0.05$; Figure 14; Figure 15).
- 1 site (LHTB) assessed across the Great Lakes Basin showed significant increasing temporal trends of Total Chlordane concentration sums ($\alpha = 0.05$; Figure 13; Table 13; Table A6).
- 3 sites (LMHB, LMNC, and LELR) assessed across the Great Lakes Basin showed significant decreasing temporal trends of Total Chlordane concentration sums ($\alpha = 0.05$; Figure 13; Table 13; Table A6).
- The percentage of sites sampled in 2022 grouped into each cluster based on historic Great Lakes data (Figure 17):
  - 100% (66/66) of sites in the low cluster (0.00 – 27.2 ng/g dw)
  - 0% of sites in the medium cluster (27.3 – 97.0 ng/g dw)
  - 0% of sites in the high cluster (97.1 – 270.8 ng/g dw)

General Observations:

- Total Chlordane concentration sums were generally low or not detected in dreissenid mussel tissue at sites surveyed in the Great Lakes in 2022 (Figure 11; Figure 13; Figure 16).
  - There were six instances of medium contamination (LMTR.0, LMSR.03, LEBR.02, LMSJ, LMMB.04, and LMMB.0) and five instances of high contamination (LMPS.0, LMSR.01, LECR.02, LMMB.01, and LMSR.02) detected in mussel tissue in 2022 (Figure 16).
- Overall, elevated chlordane concentrations detected in mussel tissue in 2022 were generally from sites sampled in Lake Michigan: Sheboygan River (LMSR.01, LMSR.02, LMSR.03; Concentration range: 10.1 - 25.9 ng/g dw), and Milwaukee Bay (LMMB.0, LMMB.01, LMMB.04; Concentration range: 13.7 - 24.3 ng/g dw; Figure 16).
- Total Chlordanes were only detected in dreissenid mussels at four (LMHB, LMNC, LHTB, and LELR) of the eighteen long-term MWP sites sampled in 2022.
- Historically, highest chlordane concentrations were detected in mussels from sites sampled in Lake Superior (4.14 ng/g dw), Niagara River (9.42 ng/g dw), Detroit River (10.7 ng/g dw), Hudson River (16.8 ng/g dw), Lake St. Clair (28.3 ng/g dw), Lake Ontario (63.7 ng/g dw), Lake Huron-Saginaw Bay (86.3 ng/g dw), Lake Michigan (155.8 ng/g dw), and Lake Erie (270.8 ng/g dw; Figure 13; Figure 18).
- The continued presence of chlordane residue in mussel tissue suggests continued input into the Great Lakes aquatic environment from likely sources such as resuspended sediments, and chlordane residue biotic degradation and abiotic weathering in soils (Ouyang et al., 2005).
- Total Chlordane concentration sums detected in mussel tissue in 2022, were below the US EPA recommended screening values for both recreational and subsistence fishers (Table A2; Table A3).

Results - Total Chlordanes

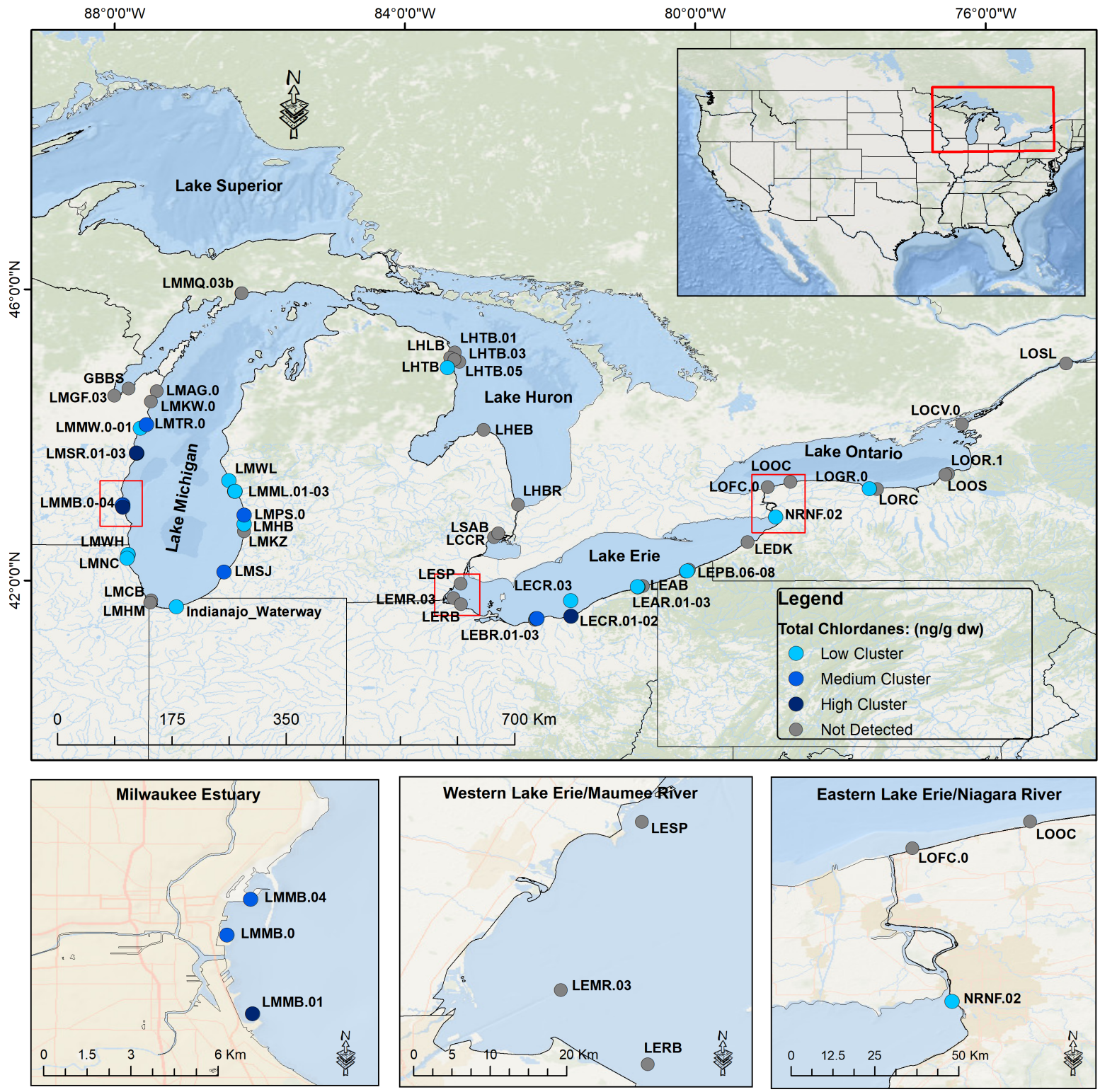


Figure 16. Total Chlordane concentration sums (ng/g dw) measured in Great Lakes mussel tissue during 2022. Some mussel sampling locations are shown to overlap due to the size of the circles. Colors represent contamination magnitude and clusters as follows: low ( ), medium ( ), high ( ), and not detected ( ).

Results - Total Chlordanes

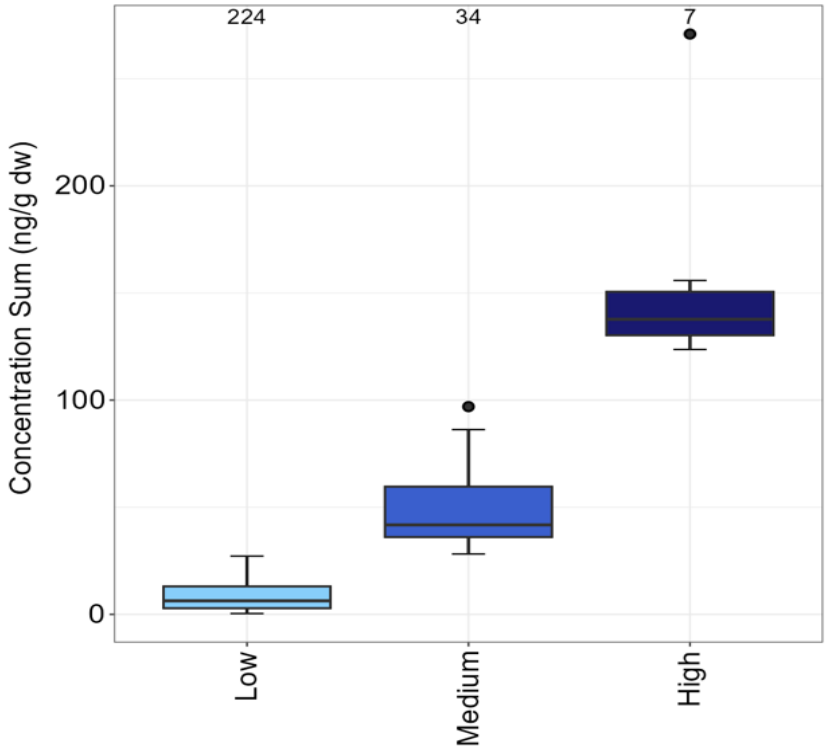


Figure 17. Total Chlordane concentration sums (ng/g dw) measured in Great Lakes mussel tissue during 1992-2022. Colors represent contamination magnitude as follows: low ( ), medium ( ), and high ( ).

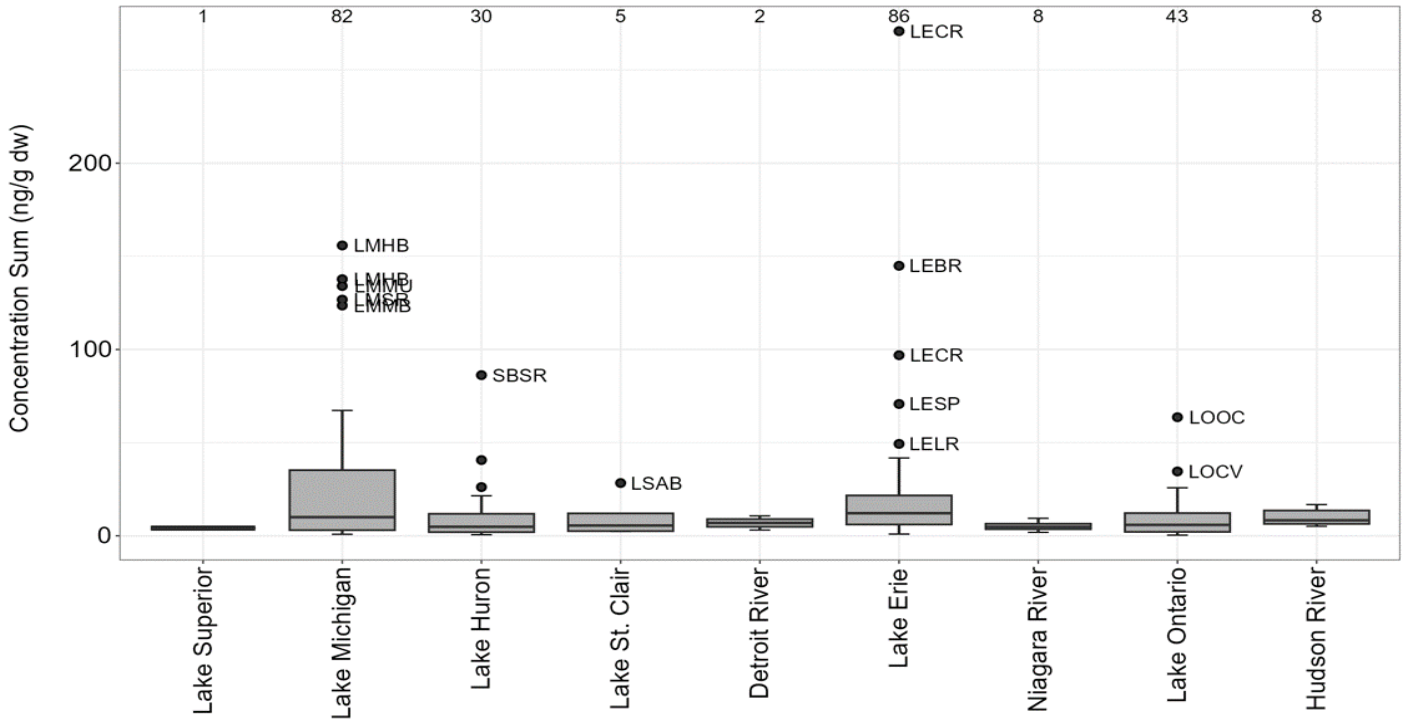


Figure 18. Total Chlordane concentration sums (ng/g dw) measured in Great Lakes mussel tissue during 1992-2022. Lakes and major rivers including connecting channels sampled during 1992-2022 are listed geographically from west to east across the Great Lakes Basin. The number of chlordane samples collected across each lake and major rivers including connecting channels sampled during 1992-2022 is noted at the top of the plot.

6.0 RESULTS - TOTAL CHLOROBENZENES

6.1 Chlorobenzenes Chemical Description

For this document, Total Chlorobenzenes analyzed both in 2022 and historically are the sum of 5 compounds (Table 4). Chlorobenzenes belong to the family of organic halogen compounds and are widely used as degreasers, chemical intermediates and solvents for pesticide formulations, adhesives, paints, polishes, dyes, and drugs (Pravasi, 2014). For example, pentachloroanisole comes from the biomethylation of pentachlorophenol, a chemical used as a general biocide, fungicide, bactericide, herbicide, molluscicide, algaecide and insecticide by agriculture and other industries, including textiles, paints, oil drilling and forestry (Canada, 2012). Although chlorobenzenes are not banned, their production has decreased by 60% since the peak in 1960 due primarily to regulations on DDT, in which it was used as part of the manufacturing process (ATSDR, 1990).

There is inadequate evidence to classify chlorobenzenes as carcinogens. However, animal studies indicate that livers, kidneys, and the central nervous system are affected by exposure to chlorobenzenes (ATSDR, 1990). Chlorobenzenes typically evaporate rapidly into the air or are broken down by bacteria and, as a result, are unlikely to build up in the food chain.

Table 4. Chlorobenzene compounds composing the "Total Chlorobenzenes" summation for the 2022 Great Lakes survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2022	Analyzed in Historical Data
1,2,3,4-Tetrachlorobenzene	●	●
1,2,4,5-Tetrachlorobenzene	●	●
Hexachlorobenzene	●	●
Pentachloroanisole	●	●
Pentachlorobenzene	●	●

Results - Total Chlorobenzenes

6.2 Magnitude and Distribution of Chlorobenzenes in Mussel Tissue in 2022

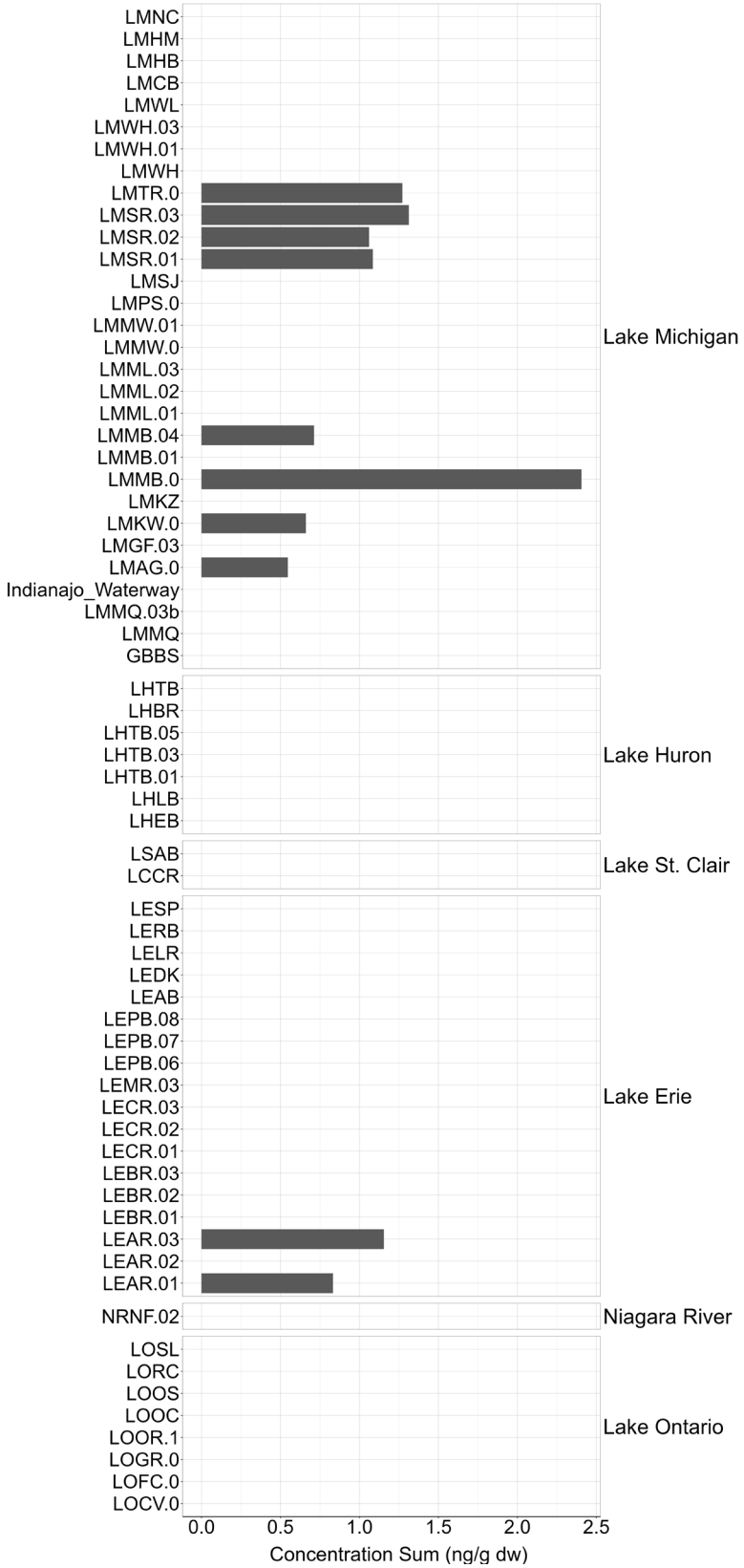


Figure 19. Bar graph showing the magnitude of Total Chlorobenzene concentration sums detected in mussel tissue in the Great Lakes Basin during the 2022 sampling period. Lakes are listed geographically from west to east across the Great Lakes Basin.

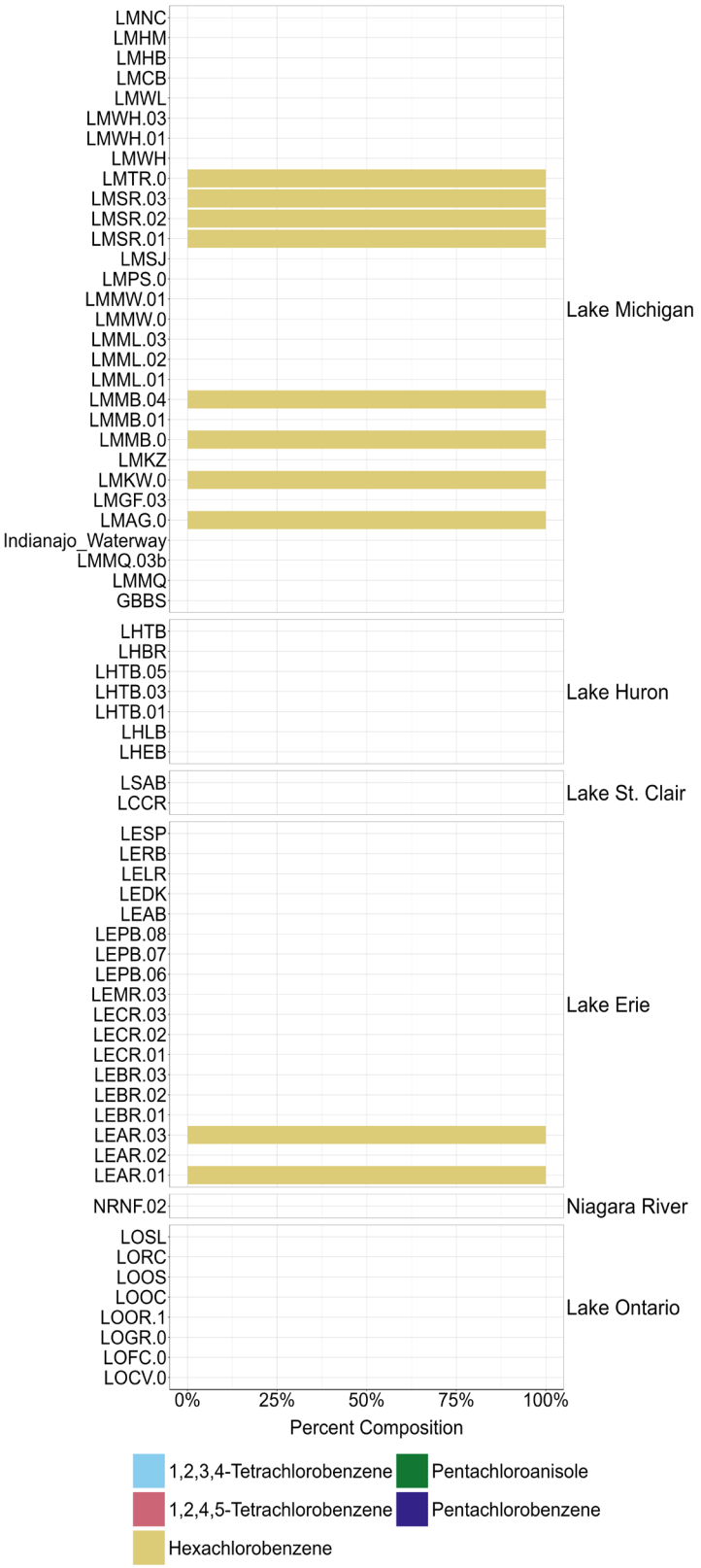


Figure 20. Percent composition of individual chlorobenzene contaminants in Total Chlorobenzene concentration sums per site in the Great Lakes Basin during the 2022 sampling period. Lakes are listed geographically from west to east across the Great Lakes Basin.

Results - Total Chlorobenzenes

6.3 Historical Context of Chlorobenzenes Magnitude and Distribution in Mussel Tissue

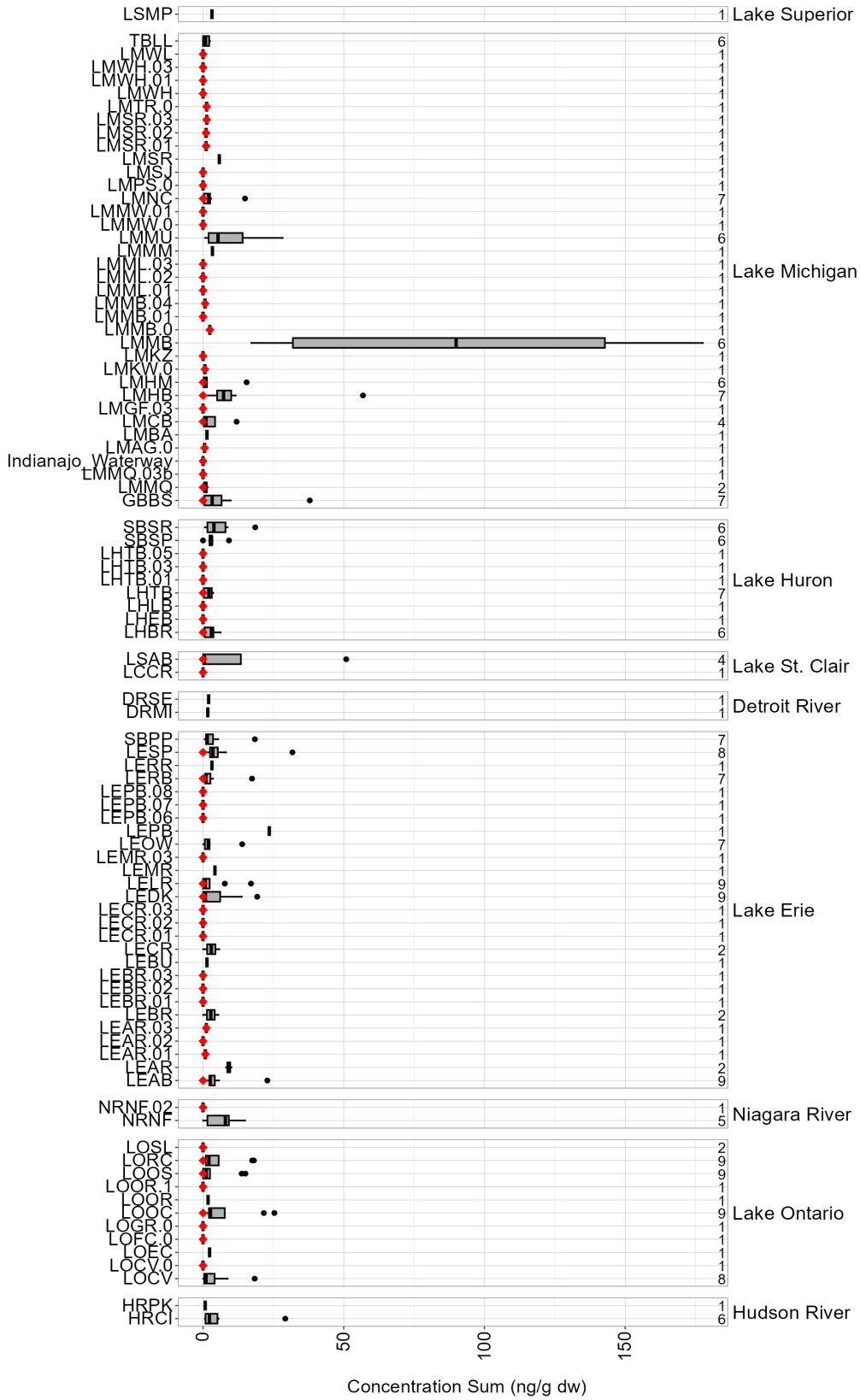


Figure 21. Total Chlorobenzene concentration sums (ng/g dw) in mussel tissue in 2022 (◆ - red diamonds), compared to boxplots of the historic Total Chlorobenzene concentration sums (ng/g dw) for each site (1992-2022) and results of the trend analysis showing significant increasing (▷) or decreasing (◁) contamination trends over time (gray triangles). Additional information is provided in Table 13 and Table A6. Lakes are listed geographically from west to east across the Great Lakes Basin. The number of years each site has been sampled since 1992 is noted at the side (right) of the plot for each site.

Results - Total Chlorobenzenes

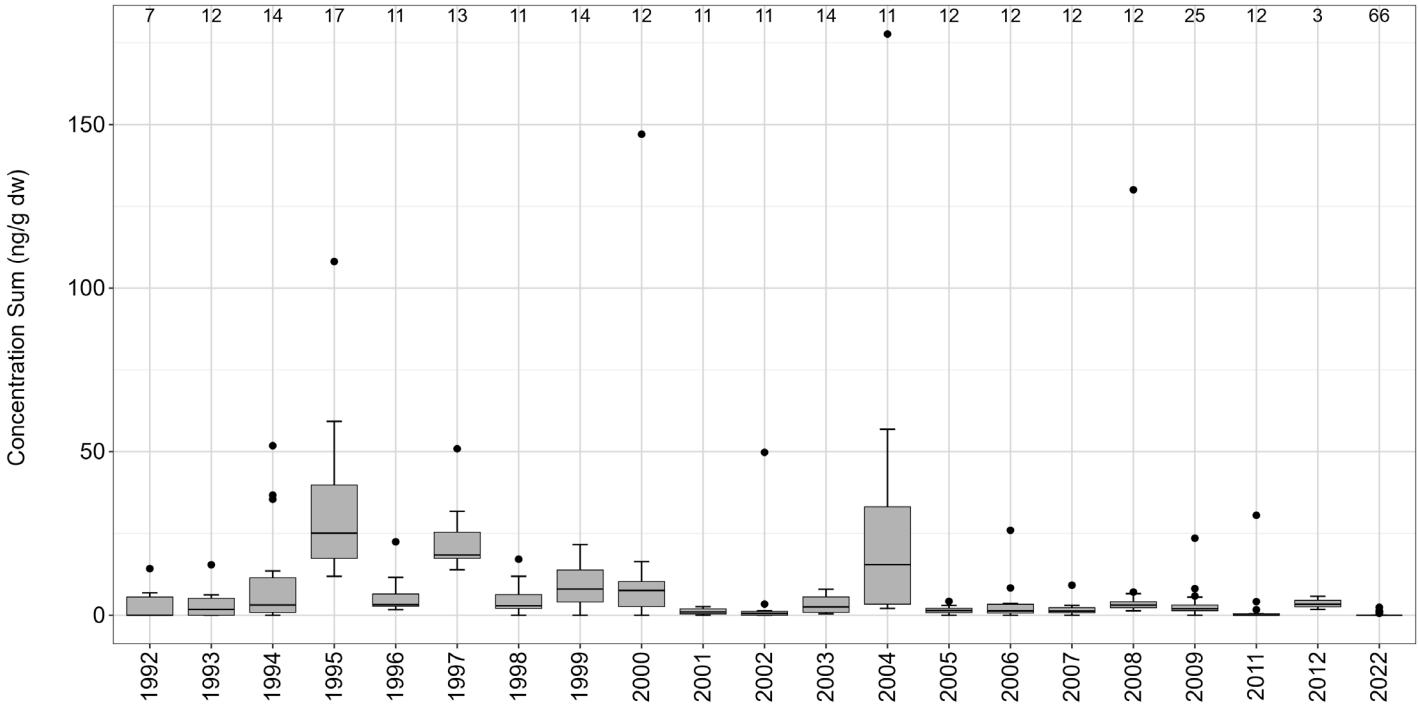


Figure 22. Boxplots representing the historic Total Chlorobenzene concentration sums (ng/g dw) measured in mussel tissue of the sites analyzed in this study. The number of sites that were sampled each year is noted at the top of the plot.

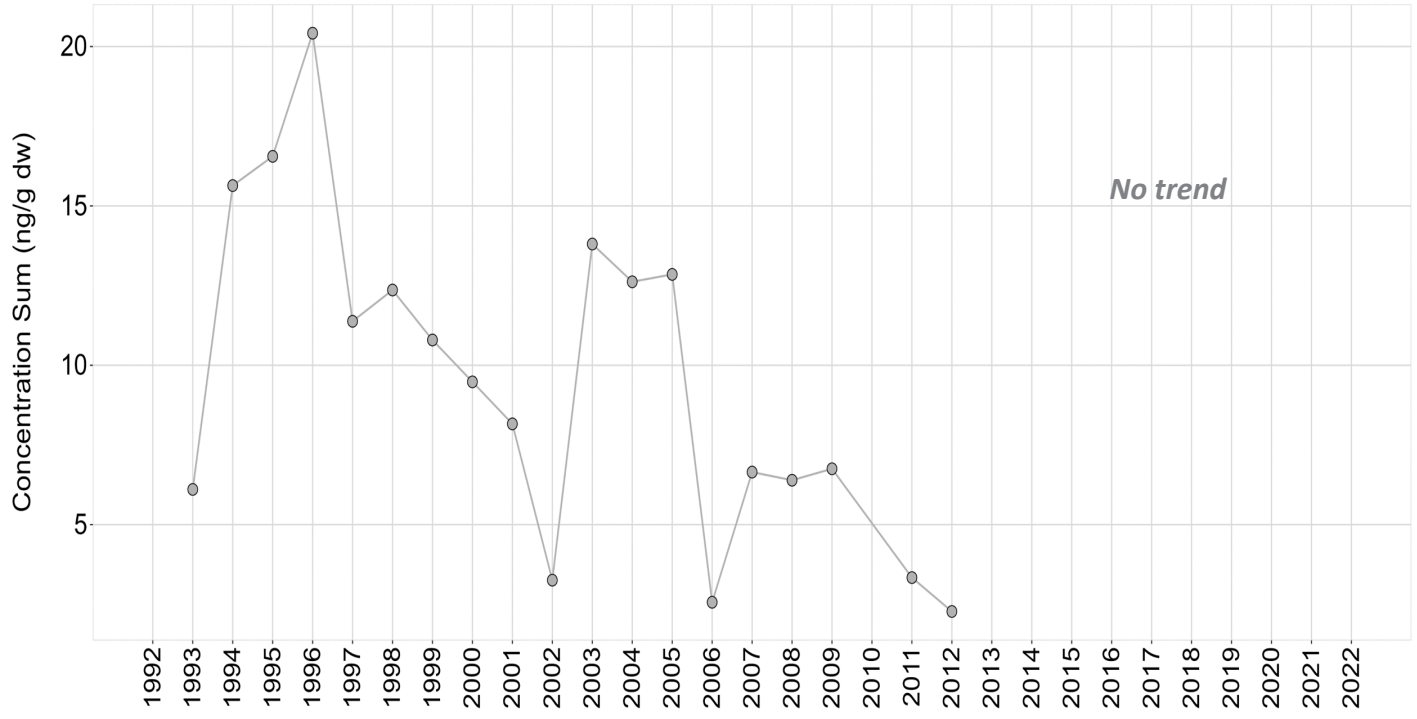


Figure 23. Three-point moving average of the yearly mean Total Chlorobenzene concentration sums (ng/g dw) measured in mussel tissue at sites analyzed in this study.

Results - Total Chlorobenzenes

6.4 Chlorobenzenes Summary

2022 Dreissenid Mussel Tissue Results (sum of 5 compounds):

- Total Chlorobenzenes were detected in dreissenid mussels at 15% (10/66) of the sites surveyed in 2022 (Figure 19; Figure 20; Figure 24).
- Total Chlorobenzenes concentration summary statistics (Figure 19; Figure 20):
  - Range: 0.00 – 2.41 ng/g dw
  - Minimum detectable concentration sum was detected at LMAG.0 (0.546 ng/g dw)
  - Maximum concentration sum was detected at LMMB.0 (2.41 ng/g dw)
  - Median: 1.07 ng/g dw
  - Mean ± SD: 1.10 ± 0.528 ng/g dw

Historical Context for Dreissenid Mussel Tissue (5 compounds):

- 85% (56/66) of sites were above their historic median concentrations in 2022 (Figure 21).
- No significant regional temporal trend of Total Chlorobenzene concentration sums were observed across the Great Lakes Basin sampling locations between 1992-2022 ($\alpha = 0.05$; Figure 22; Figure 23).
- 0 sites assessed across the Great Lakes Basin showed significant decreasing temporal trends of Total Chlorobenzene concentration sums ($\alpha = 0.05$; Figure 21).
- 0 sites assessed across the Great Lakes Basin showed significant increasing temporal trends of Total Chlorobenzene concentration sums ($\alpha = 0.05$; Figure 21).
- The percentage of sites sampled in 2022 grouped into each cluster based on historic Great Lakes data (Figure 25):
  - 100% (66/66) of sites in the low cluster (0.437 – 16.4 ng/g dw)
  - 0% of sites in the medium cluster (16.5 – 59.3 ng/g dw)
  - 0% of sites in the high cluster (59.4 – 177.7 ng/g dw)

General Observations:

- Total Chlorobenzenes concentration sums were generally low or not detected in dreissenid mussel tissue at sites surveyed in the Great Lakes in 2022 (Figure 19; Figure 21; Figure 24).
  - There were three instances of medium contamination (LMSR.02, LMSR.01, and LEAR.03) and three instances of high contamination (LMTR.0, LMSR.03, and LMMB.0) detected in dreissenid mussel tissue in 2022 (Figure 24).
- Historically, highest chlorobenzene concentrations were detected in mussels from sites sampled in Detroit River (1.98 ng/g dw), Lake Superior (3.15 ng/g dw), Lake Huron (18.6 ng/g dw), Lake Erie (41.9 ng/g dw), Niagara River (44.5 ng/g dw), Lake St. Clair (50.9 ng/g dw), Lake Ontario (59.3 ng/g dw), Hudson River (108.1 ng/g dw), and Lake Michigan (177.7 ng/g dw; Figure 21; Figure 26).
- The continued presence of chlorobenzenes residue in mussel tissue suggests that there continues to be inputs into the Great Lakes aquatic environment from likely sources such as resuspended sediments, and chlorobenzenes residue biotic degradation and abiotic weathering in soils (Brahushi et al., 2017).
- Total Chlorobenzene concentration sums were well below the US EPA recommended screening values for both recreational and subsistence fishers (Table A2; Table A3).

Results - Total Chlorobenzenes

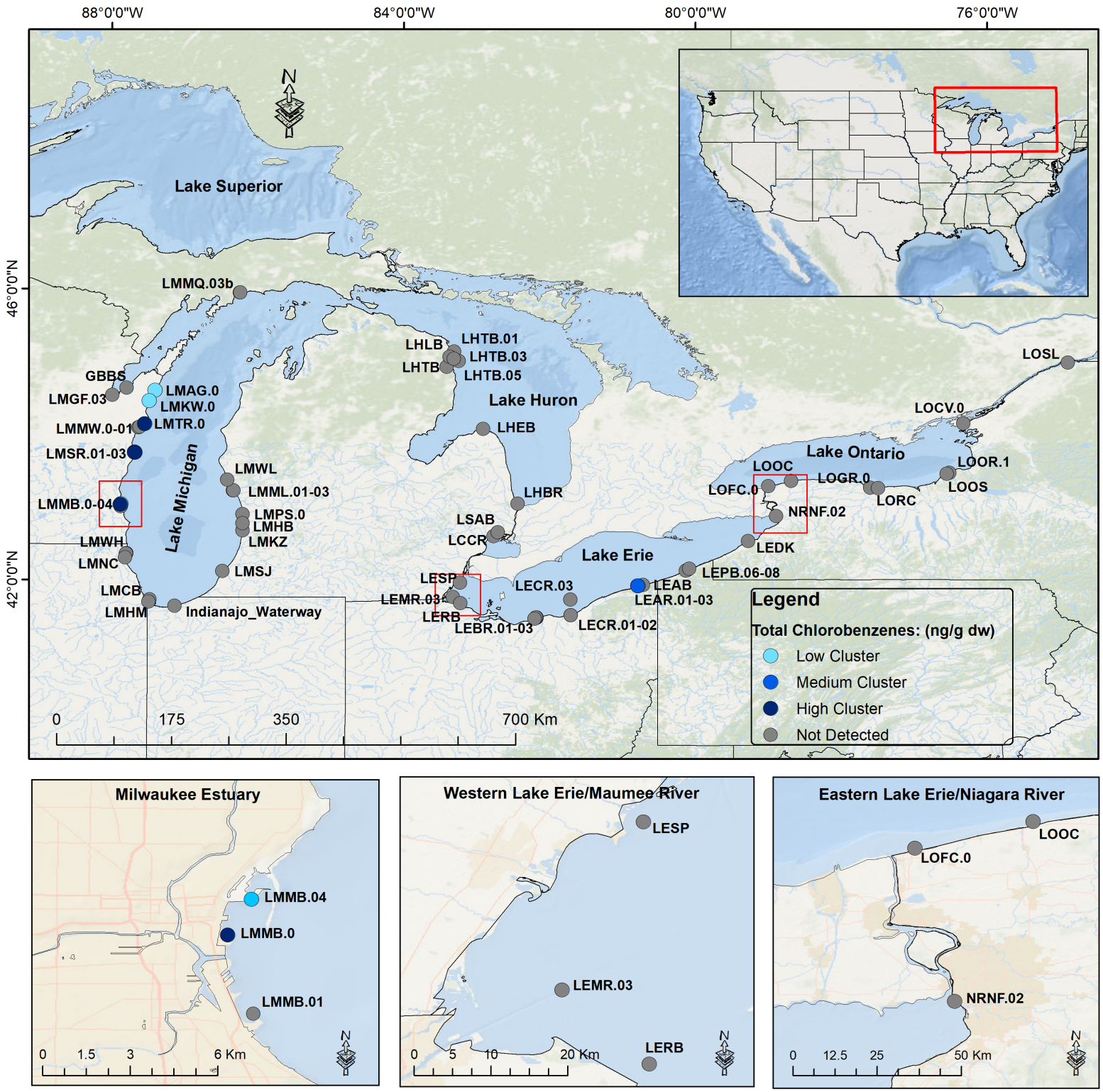


Figure 24. Total Chlorobenzene concentration sums (ng/g dw) measured in Great Lakes mussel tissue during 2022. Some mussel sampling locations are shown to overlap due to the size of the circles. Colors represent contamination magnitude and clusters as follows: low ( ), medium ( ), high ( ), and not detected ( ).

Results - Total Chlorobenzenes

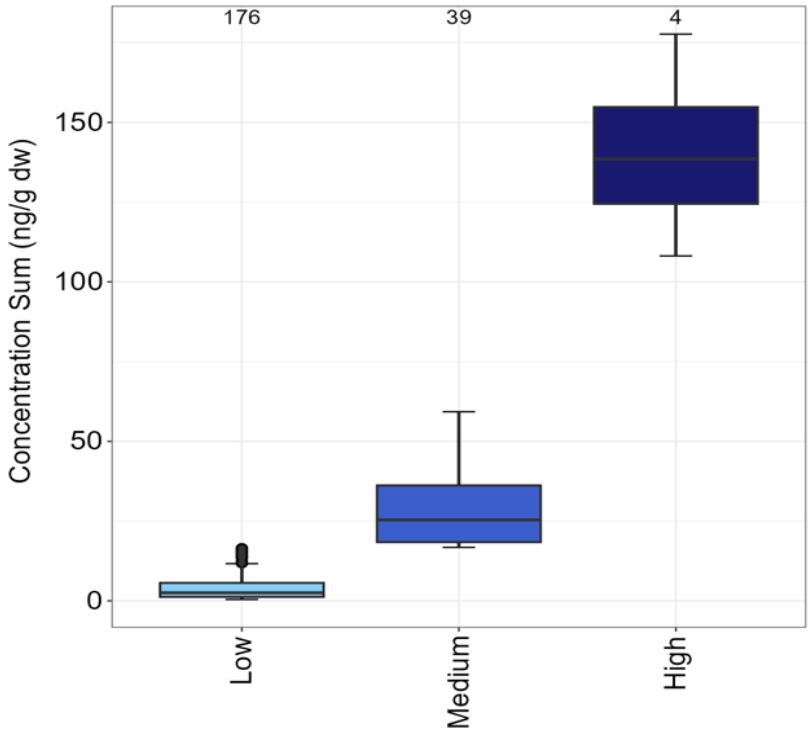


Figure 25. Total Chlorobenzene concentration sums (ng/g dw) measured in Great Lakes mussel tissue during 1992-2022. Colors represent contamination magnitude as follows: low ( ), medium ( ), and high ( ).

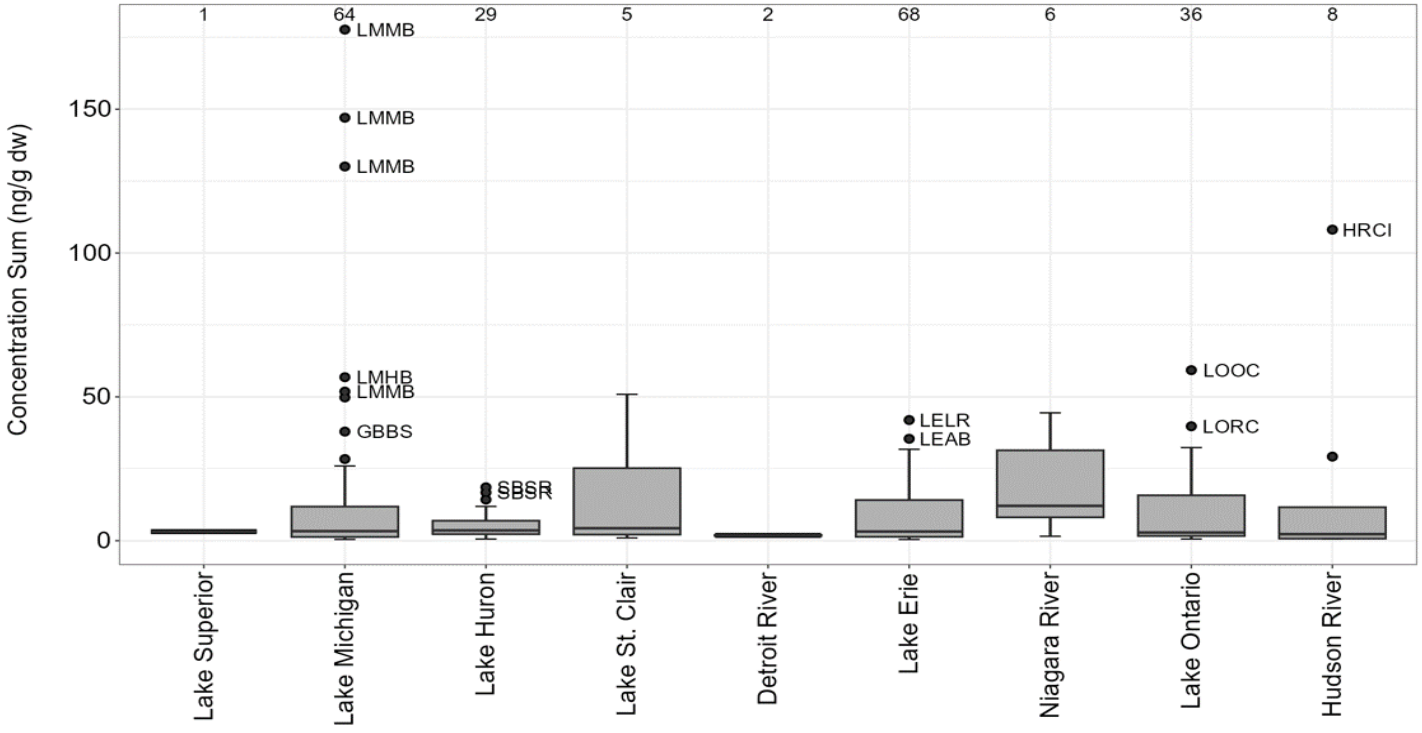


Figure 26. Total Chlorobenzene concentration sums (ng/g dw) measured in Great Lakes mussels between 1992-2022. Lakes and major rivers including connecting channels sampled during 1992-2022 are listed geographically from west to east across the Great Lakes Basin. The number of chlorobenzene samples collected across each lake and major rivers including connecting channels sampled during 1992-2022 is noted at the top of the plot.

7.0 RESULTS - CHLORPYRIFOS

7.1 Chlorpyrifos Chemical Description

For this study, chlorpyrifos analyzed both in 2022 and historically was a single compound. Chlorpyrifos is a white, crystal-like solid with a strong odor that has been used as an (organophosphate) pesticide in the United States since 1965 in both agricultural and non-agricultural settings (ATSDR, 1997; EPA, 2022). In 2000, there was a voluntary agreement to eliminate, phase out, and modify specific uses of chlorpyrifos, specifically private homeowner usage except for roaches, discontinuing uses on tomato plants and lowering the maximum residue level of grapes (EPA, 2022). As of 2022, chlorpyrifos is no longer used on food and animal feed crops, although it may still be used on non-fruit bearing trees (NPIC, 2023).

Exposure to chlorpyrifos may occur through exposure to pesticides by eating food contaminated with chlorpyrifos, breathing in fumes, or getting them on skin or eyes (NPIC, 2023). Exposure can also occur through contaminated groundwater, which can happen if products containing chlorpyrifos were used or mixed for application near a well for termite control (NPIC, 2023). Once it is inside the body, chlorpyrifos itself is not toxic, but when the body attempts to break it down, it creates a toxic form (NPIC, 2023). Typical symptoms may include sweating, headache, nausea, and dizziness, while more severe exposure can cause vomiting, abdominal muscle cramps, muscle twitching, tremors, weakness, and loss of coordination (ATSDR, 1997; NPIC, 2023). Generally, the body can excrete the contaminant within a few days (NPIC, 2023). Studies have not shown that chlorpyrifos is a carcinogen (ATSDR, 1997).

In the environment, chlorpyrifos can bind strongly to soil, where it can take weeks to years to break down (NPIC, 2023). Once in the soil, however, plant roots do not usually uptake chlorpyrifos, and it won't easily transfer to groundwater, although it may wash into rivers and streams (NPIC, 2023). Chlorpyrifos is very toxic to many bird species, causing thin eggshells and juvenile mortality (NPIC, 2023). It is also very toxic to fish and aquatic invertebrates, will bioaccumulate in larger animals, and is toxic to bees and earthworms (NPIC, 2023).

7.2 Magnitude and Distribution of Chlorpyrifos in Mussel Tissue in 2022

Chlorpyrifos was not detected at any sites in mussel tissue during the 2022 Great Lakes sampling period.

7.3 Historical Context of Chlorpyrifos Magnitude and Distribution in Mussel Tissue

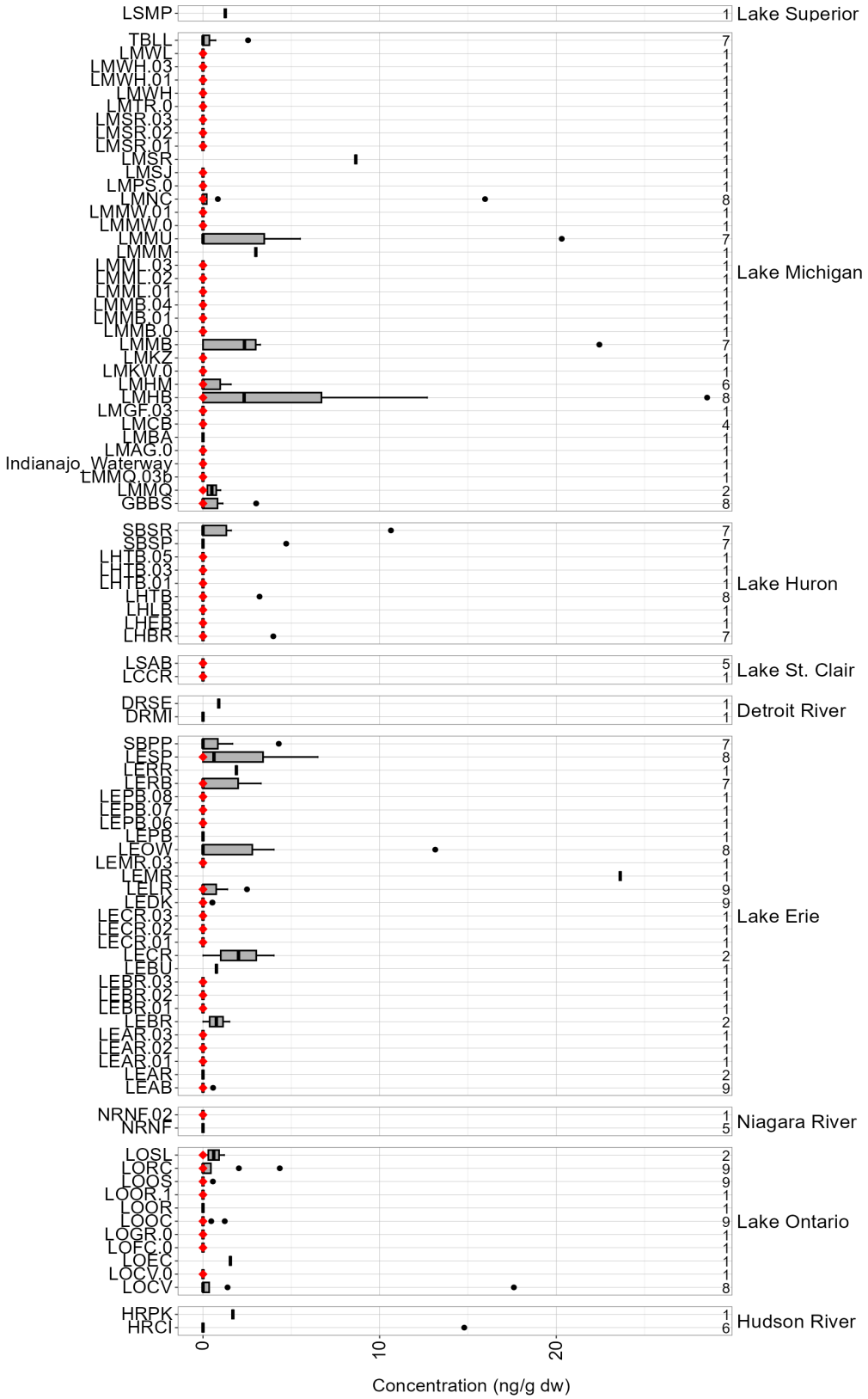


Figure 27. Chlorpyrifos concentrations (ng/g dw) in mussel tissue in 2022 (◆ - red diamonds), compared to boxplots of the historic chlorpyrifos concentration sums (ng/g dw) for each site (1992-2022) and results of the trend analysis showing significant increasing (▶) or decreasing (◀) contamination trends over time (gray triangles). Additional information is provided in Table 13 and Table A6. Lakes are listed geographically from west to east across the Great Lakes Basin. The number of years each site has been sampled since 1992 is noted at the side (right) of the plot for each site.

Results - Chlorpyrifos

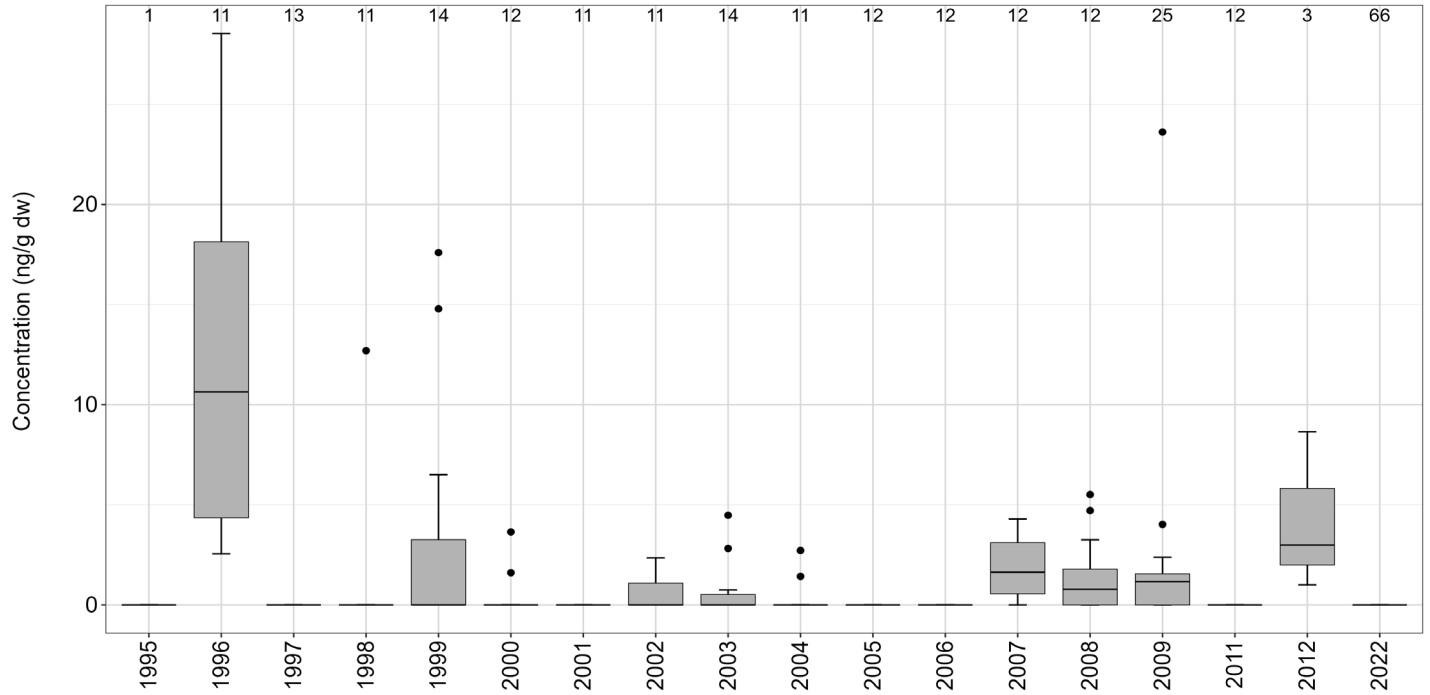


Figure 28. Boxplots representing the historic chlorpyrifos concentrations (ng/g dw) measured in mussel tissue of the sites analyzed in this study. The number of sites that were sampled each year is noted at the top of the plot.

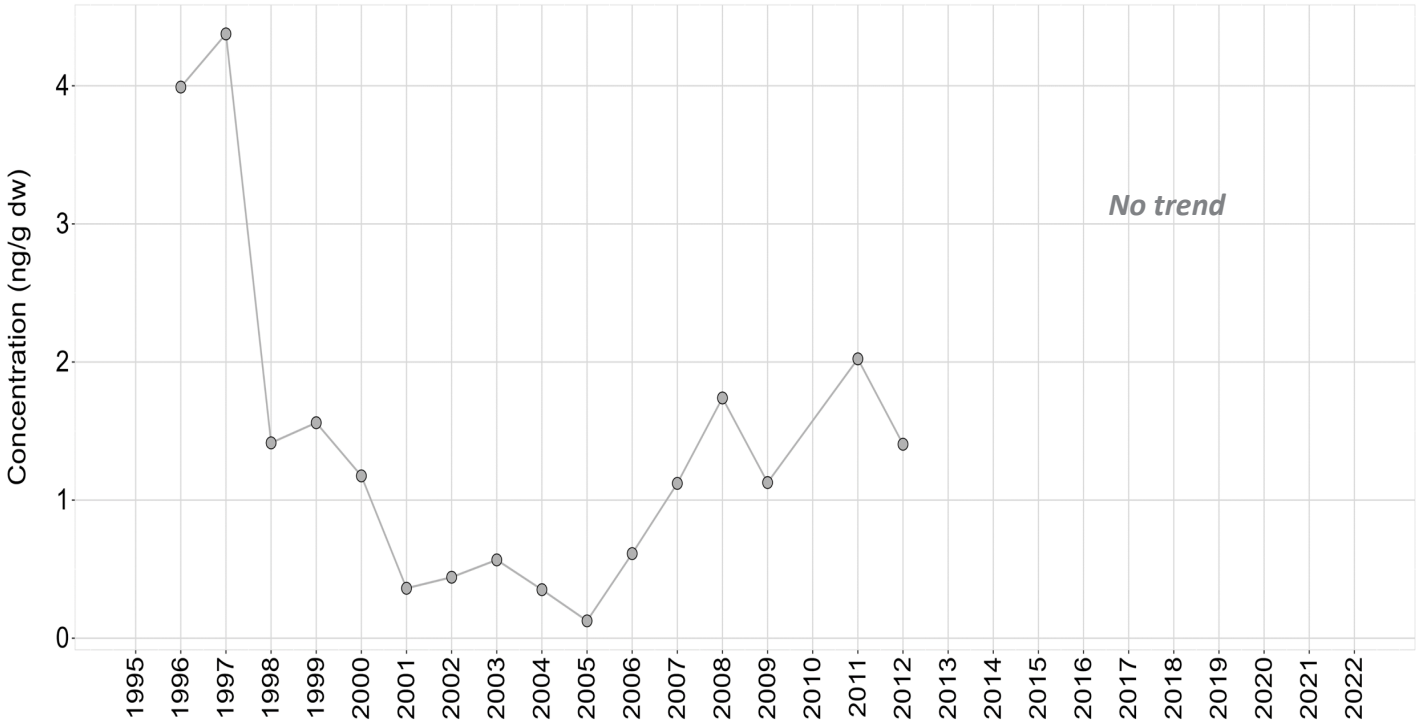


Figure 29. Three-point moving average of yearly mean chlorpyrifos concentrations (ng/g dw) measured in mussel tissue at sites analyzed in this study.

Results - Chlorpyrifos

7.4 Chlorpyrifos Summary

2022 Dreissenid Mussel Tissue Results (1 compound):

- Chlorpyrifos was not detected in dreissenid mussels at sites surveyed in the Great Lakes in 2022 (Figure 30).

Historical Context for Dreissenid Mussel Tissue (1 compound):

- 0% of sites were above their historic median concentrations in 2022 (Figure 27).
- There was no significant regional temporal trend of chlorpyrifos concentration sums observed across the Great Lakes Basin sampling locations between 1992-2022 ($\alpha = 0.05$; Figure 28; Figure 29).
- 0 sites assessed across the Great Lakes Basin showed significant decreasing temporal trends of chlorpyrifos concentration sums at ($\alpha = 0.05$; Figure 27).
- 0 sites assessed across the Great Lakes Basin showed significant increasing temporal trends of chlorpyrifos concentration sums at ($\alpha = 0.05$; Figure 27).
- The percentage of sites sampled in 2022 grouped into each cluster based on historic Great Lakes data (Figure 31):
  - 100% (66/66) of sites in the low cluster (0.00 – 3.29 ng/g dw)
  - 0% of sites in the medium cluster (3.30 – 10.6 ng/g dw)
  - 0% of sites in the high cluster (10.7 – 28.5 ng/g dw)

General Observations:

- Chlorpyrifos was not detected in dreissenid mussel tissue at any sites surveyed in the Great Lakes Basin in 2022 (Figure 27; Figure 30).
- Chlorpyrifos historical concentration sums (concentration range: 0.00 – 28.5 ng/g dw) measured in mussel tissue were generally low or not detected throughout the Great Lakes Basin between 1995-2022 (Figure 27; Figure 32).
- Historically, highest chlorpyrifos concentrations were detected in mussel tissue from sites sampled in Detroit River (0.8 ng/g dw), Lake Superior (1.26 ng/g dw), Lake Huron (10.6 ng/g dw), Hudson River (14.8 ng/g dw), Lake Ontario (17.6 ng/g dw), Lake Erie (23.6 ng/g dw), and Lake Michigan (28.5 ng/g dw; Figure 32).

Results - Chlorpyrifos

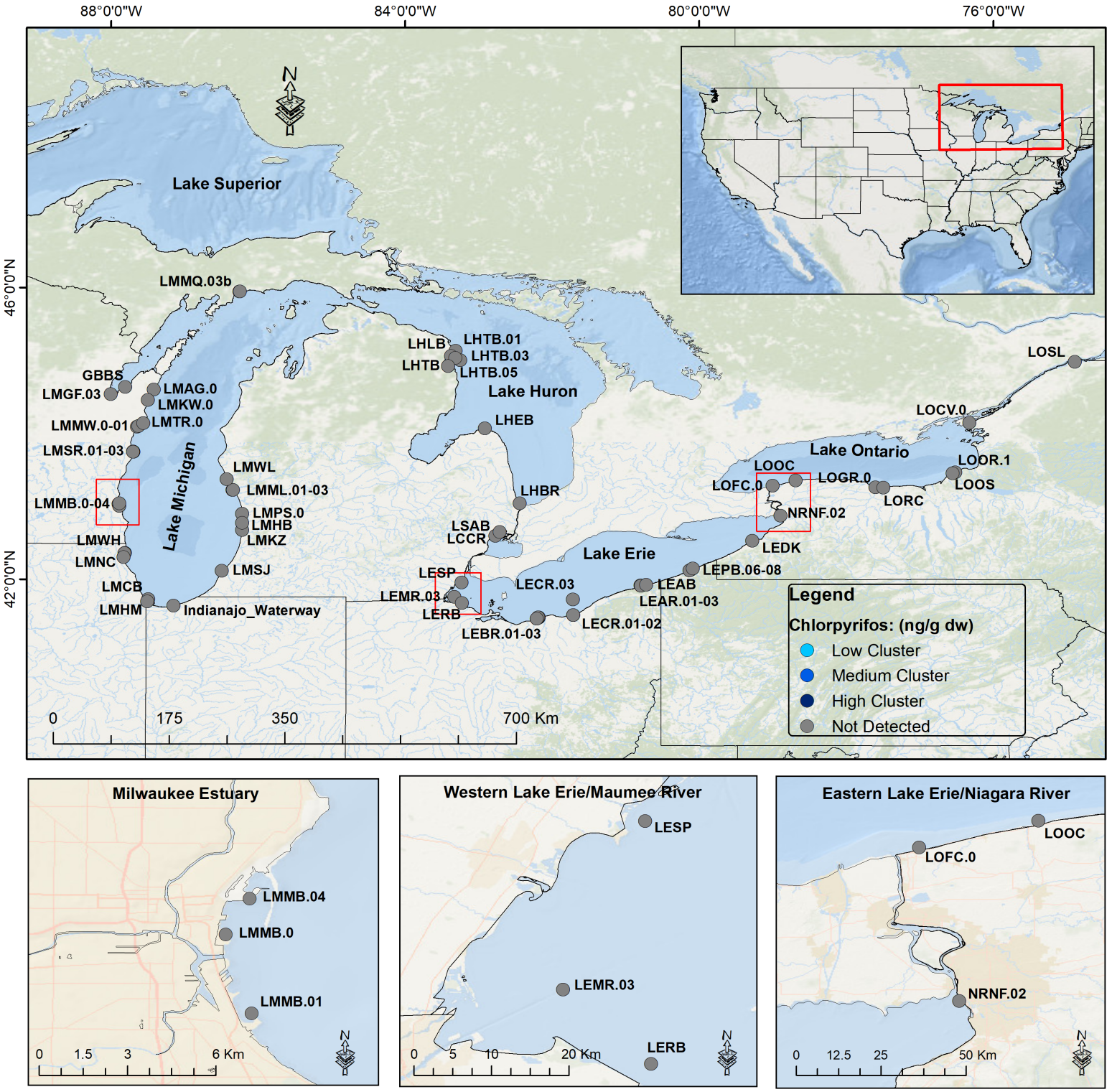


Figure 30. Chlorpyrifos were not detected in Great Lakes mussel tissue analyzed during 2022. Some mussel sampling locations are shown to overlap due to the size of the circles. Colors represent contamination magnitude and clusters as follows: low ( ), medium ( ), high ( ), and not detected ( ).

Results - Chlorpyrifos

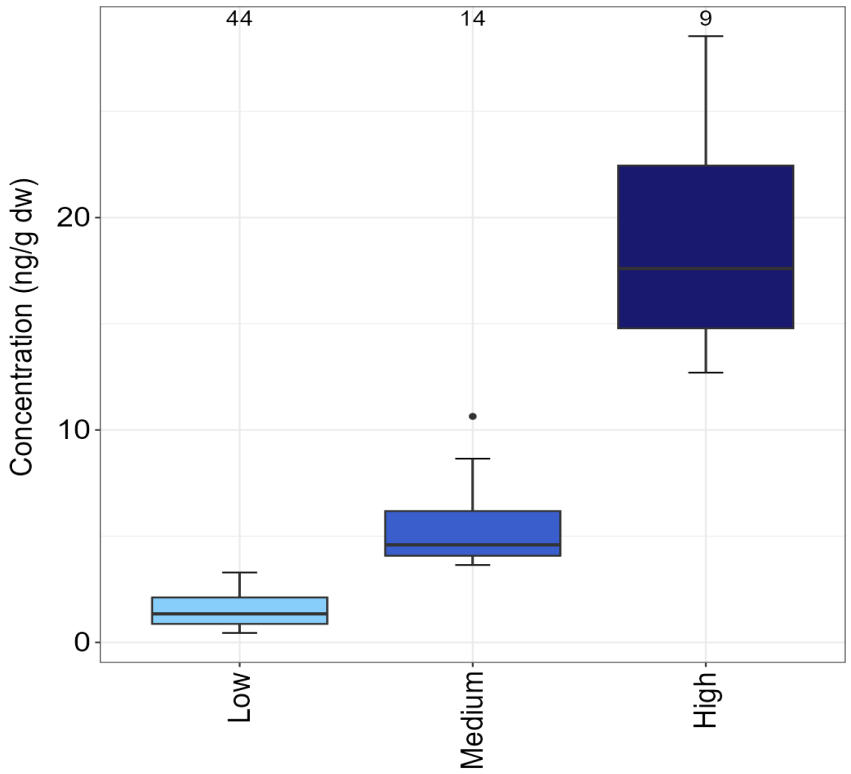


Figure 31. Chlorpyrifos concentrations (ng/g dw) measured in Great Lakes mussel tissue during 1995-2022. Colors represent contamination magnitude as follows: low ( ), medium ( ), and high ( ).

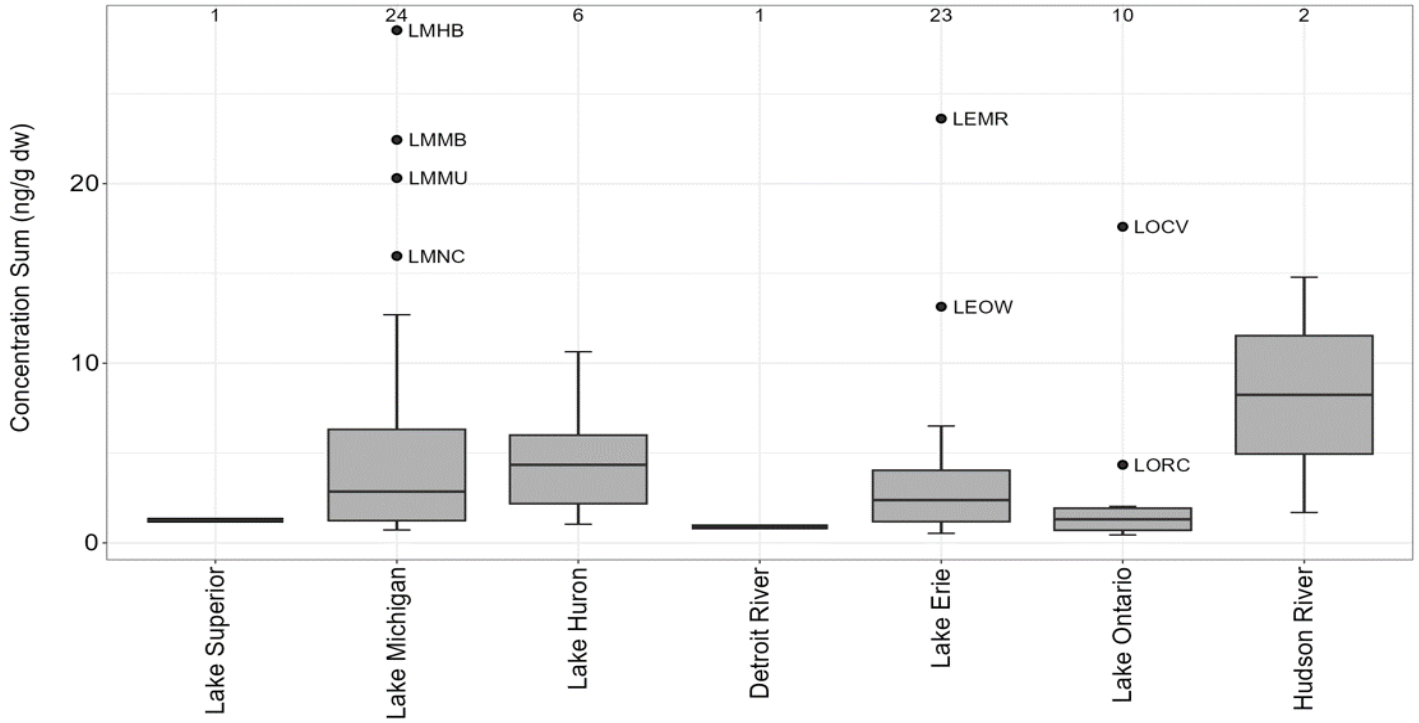


Figure 32. Chlorpyrifos concentrations (ng/g dw) measured in Great Lakes mussels between 1995-2022. Lakes and major rivers including connecting channels sampled during 1995-2022 are listed geographically from west to east across the Great Lakes Basin. The number of chlorpyrifos samples collected across each lake and major rivers including connecting channels sampled during 1995-2022 is noted at the top of the plot.

8.0 RESULTS - TOTAL DDTs

8.1 DDTs Chemical Description

For this document, Total DDTs analyzed in 2022 is the sum of 7 compounds and Total DDTs analyzed historically is the sum of 6 compounds (Table 5). With the exception of dichlorodiphenylchloroethylene (DDMU), the six compounds within Total DDTs (dichlorodiphenyltrichloroethane) are comprised of ortho and para forms of DDT and its transformation products DDE and DDD, the latter being the most predominant form found in the environment (ATSDR, 2022). Technical DDT, the insecticide, was composed of up to 14 compounds, of which 65-80% was the active ingredient, *p,p'*-DDT (4,4'-DDT). The next major component, *o,p'*-DDT (2,4'-DDT) (15-21%), is nearly inactive as an insecticide. DDT was used worldwide as an insecticide for agricultural pests and mosquito control. Its use in the United States was banned in 1972 (Zhou et al., 2018), but it is still used in some countries today. DDT was banned due to its environmental persistence, bioaccumulation, and toxicity to non-target organisms (ATSDR, 2022).

Due to its persistence and hydrophobic nature, DDT bioaccumulates in organisms. Organochlorine pesticides are typically neurotoxins, and DDT has been shown to interfere with the endocrine system (Rogan & Chen, 2005). DDT and its metabolite DDE were explicitly linked to eggshell thinning in birds (Lincer, 1975). The US FDA has established a 5-ppm wet weight safety level for DDT and DDE in all fish (FDA, 2011).

As the result of their persistence and heavy/extended use in the past, residues of DDT and other organochlorine pesticides (OCPs) can still be found in every environmental compartment, including biota, in which they can bioaccumulate in the fatty tissue of animals, and their toxicity continues to be a concern (ATSDR, 2022). In air, DDT and its metabolites can be rapidly broken down by sunlight; however, in soil they are only slowly broken down by microorganisms. Evaporation of DDT from soil, followed by long-distance transport, results in its widespread global distribution due to the grasshopper effect (Wania and Mackay, 1996). DDT that enters surface waters is subject to volatilization, adsorption to suspended particulates and sediment, and bioaccumulation. About half of the atmospheric DDT is adsorbed to particulates (Bidleman, 1988).

Table 5. DDT compounds composing the "Total DDTs" summation for the 2022 Great Lakes survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2022	Analyzed in Historical Data
2,4'-DDD	●	●
4,4'-DDD	●	●
2,4'-DDE	●	●
4,4'-DDE	●	●
2,4'-DDT	●	●
4,4'-DDT	●	●
DDMU	●	

8.2 Magnitude and Distribution of DDTs in Mussel Tissue in 2022

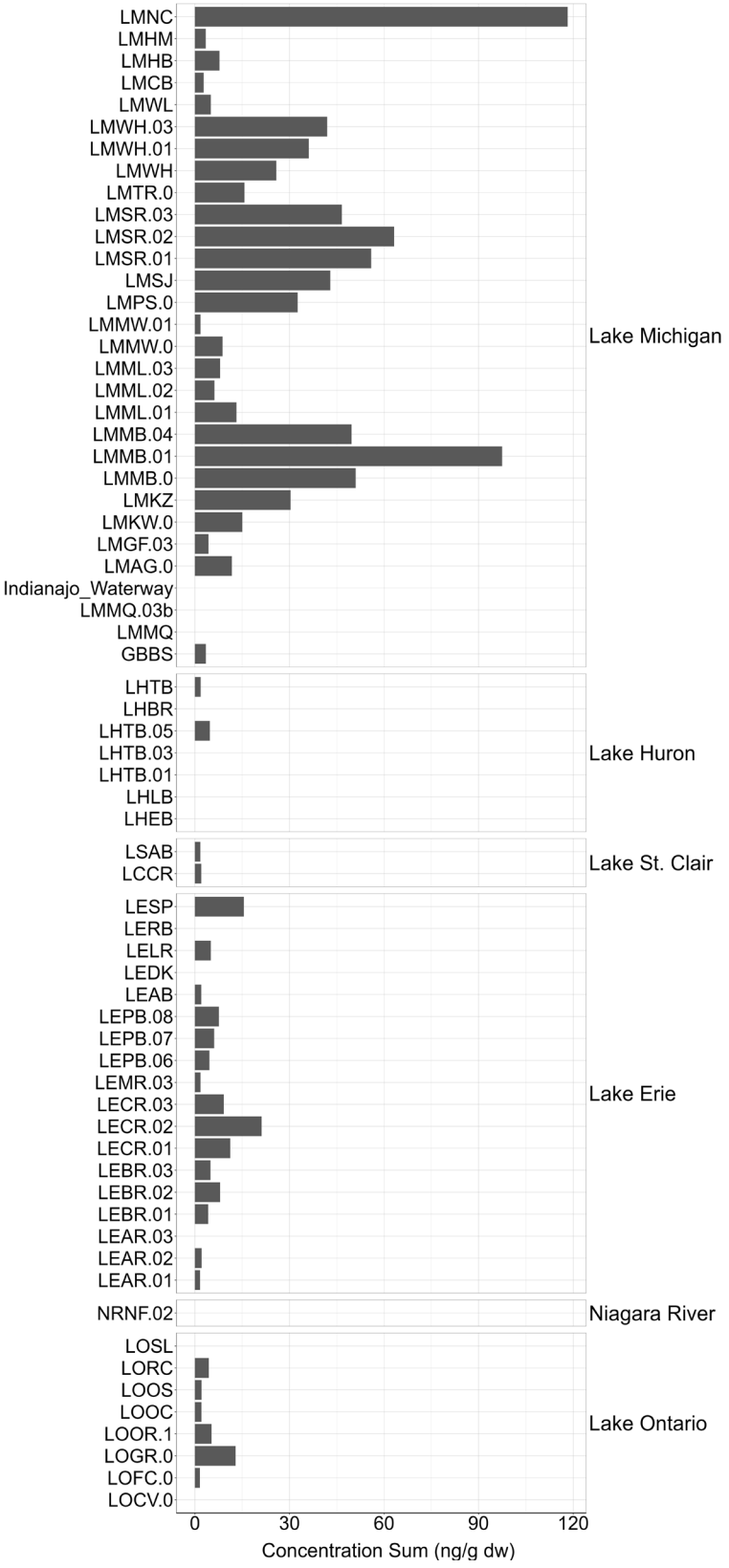


Figure 33. Bar graph showing the magnitude of Total DDT concentration sums detected in mussel tissue in the Great Lakes Basin during the 2022 sampling period. Lakes are listed geographically from west to east across the Great Lakes Basin.

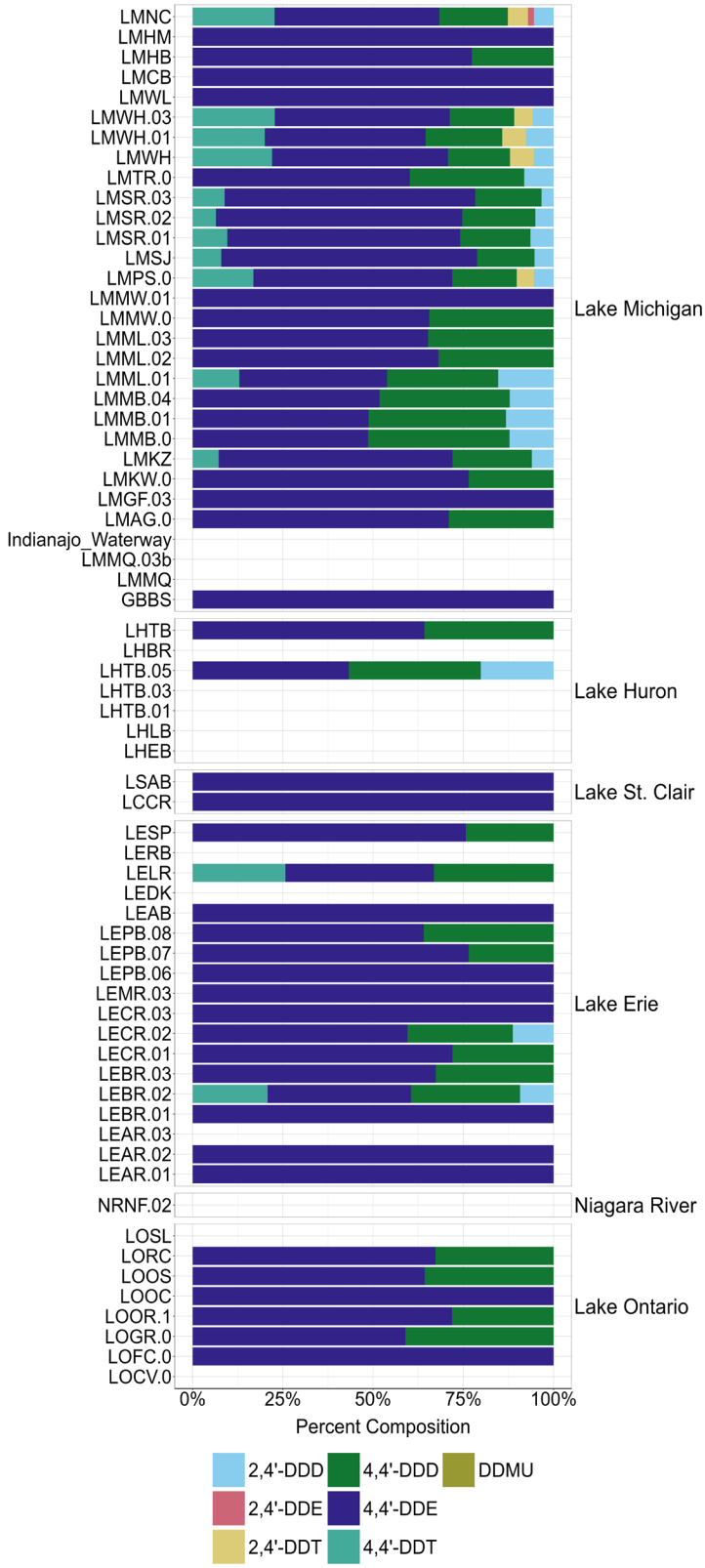


Figure 34. Percent composition of individual DDT contaminants in Total DDT concentration sums per site in the Great Lakes Basin during the 2022 sampling period. Lakes are listed geographically from west to east across the Great Lakes Basin.

8.3 Historical Context of DDTs Magnitude and Distribution in Mussel Tissue

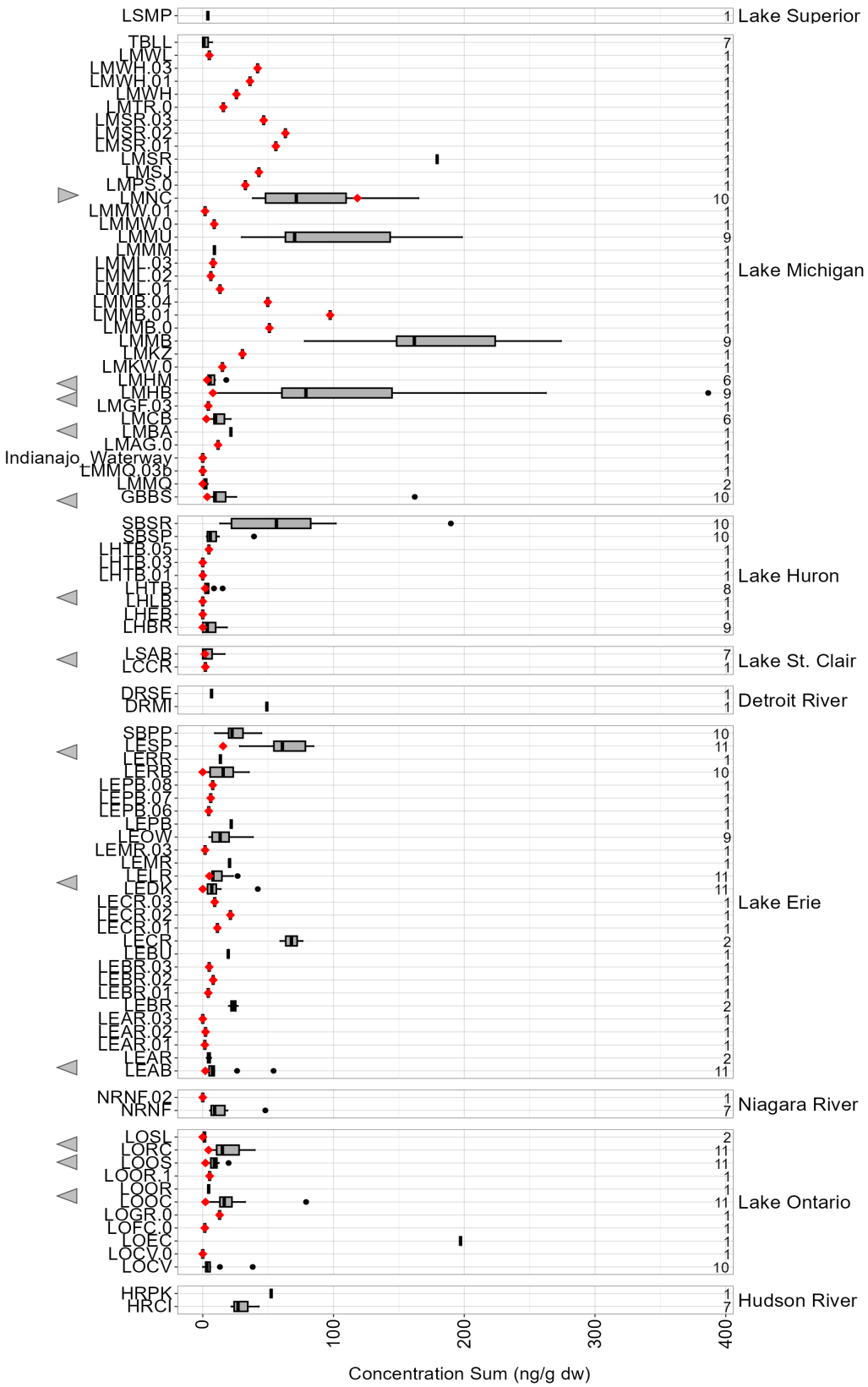


Figure 35. Total DDT concentration sums (ng/g dw) in mussel tissue in 2022 (◆ - red diamonds), compared to boxplots of the historic Total DDTs concentration sums (ng/g dw) for each site (1992-2022) and results of the trend analysis showing significant increasing (▻) or decreasing (◅) contamination trends over time (gray triangles). Additional information is provided in Table 13 and Table A6. Lakes are listed geographically from west to east across the Great Lakes Basin. The number of years each site has been sampled since 1992 is noted at the side (right) of the plot for each site.

Results - Total DDTs

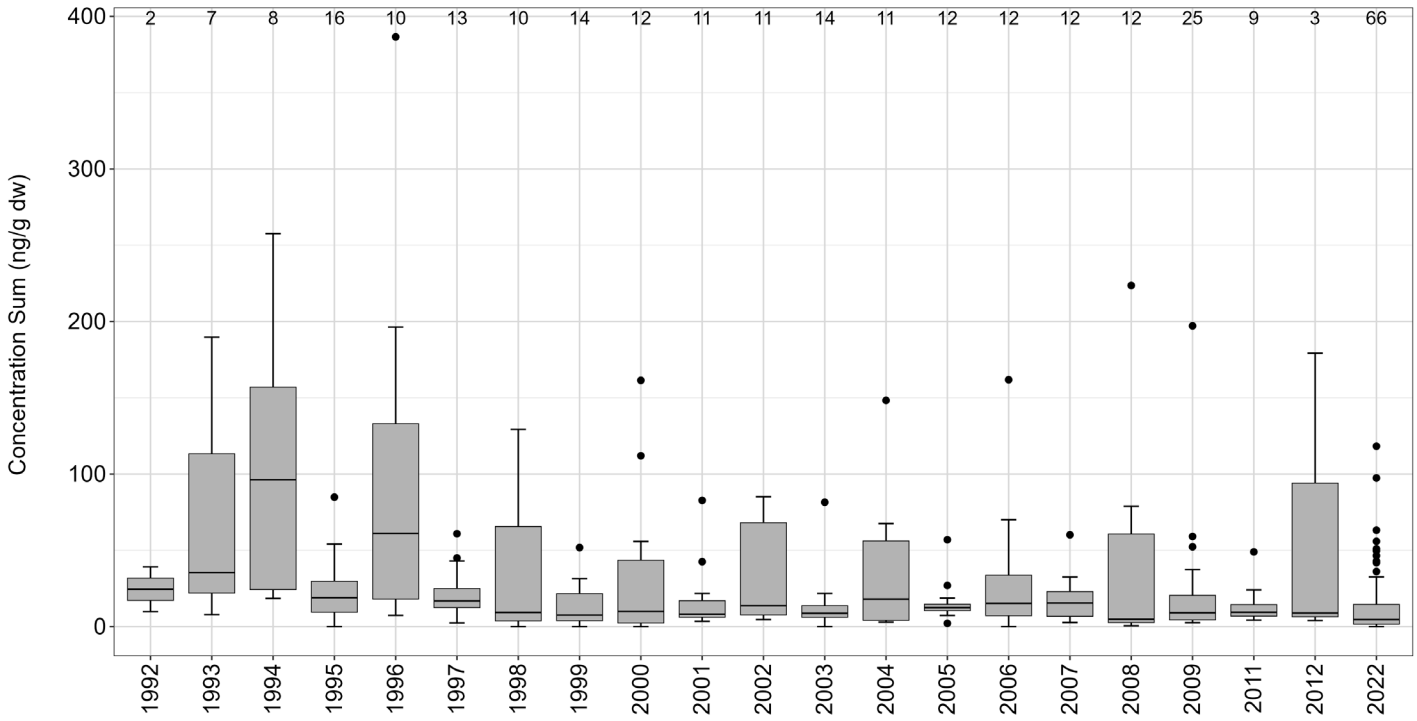


Figure 36. Boxplots representing the historic Total DDT concentration sums (ng/g dw) in mussel tissue of the sites analyzed in this study. The number of sites that were sampled each year is noted at the top of the plot.

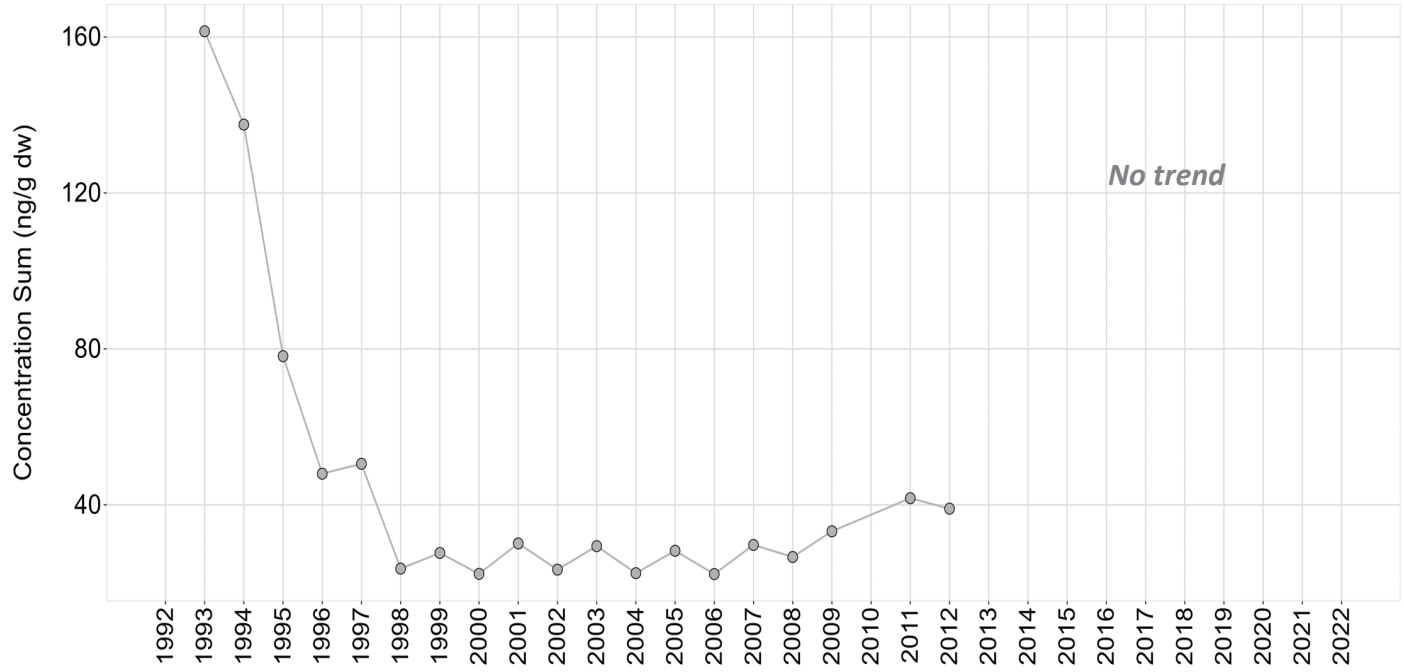


Figure 37. Three-point moving average of the yearly mean Total DDT concentration sums (ng/g dw) measured in mussel tissue at sites analyzed in this study.

Results - Total DDTs

8.4 DDTs Summary

2022 Dreissenid Mussel Tissue Results (sum of 7 compounds):

- Total DDTs were detected in dreissenid mussels at 79% (52/66) of the sites surveyed in 2022 (Figure 33; Figure 38).
- Total DDTs concentration summary statistics (Figure 33; Figure 34):
  - Range: 0.00 – 118.3 ng/g dw
  - Minimum concentration sum was detected at LOFC.0 (1.59 ng/g dw)
  - Maximum concentration sum was detected at LMNC (118.3 ng/g dw)
  - Median: 7.85 (ng/g dw)
  - Mean ± SD: 18.7 ± 24.9 (ng/g dw)

Historical Context for Dreissenid Mussel Tissue (6 compounds):.

- 23% (15/66) of sites were above their historic median concentrations in 2022 (Figure 35).
- No significant decreasing regional temporal trend of Total DDT concentration sums were observed across the Great Lakes Basin sampling locations between 1992-2022 ($\alpha = 0.05$; Figure 36; Figure 37).
- 1 site (LMNC) assessed across the Great Lakes Basin showed increasing temporal trends of Total DDT concentration sums ($\alpha = 0.05$; Figure 35; Table 13; Table A6).
- 12 sites (GBBS, LMCB, LMHB, LMHM, LHTB, LSAB, LEAB, LELR, LESP, LOOC, LOOS, and LORC) assessed across the Great Lakes Basin showed decreasing temporal trends of Total DDT concentration sums ($\alpha = 0.05$; Figure 35; Table 13; Table A6).
- The percentage of sites sampled in 2022 grouped into each cluster based on historic Great Lakes data (Figure 39):
  - 86% (57/66) of sites in the low cluster (0.00 – 36.1 ng/g dw)
  - 14% (9/66) of sites in the medium cluster (36.2 – 118.3 ng/g dw)
  - 0% of sites in the high cluster (118.4 – 386.6 ng/g dw)

General Observations:

- Total DDT concentration sums (concentration range: 0.00 – 118.3 ng/g dw) were generally low to medium, or not detected in dreissenid mussel tissue at sites surveyed in the Great Lakes in 2022 (Figure 33; Figure 35).
  - There were 11 instances of Total DDTs medium contamination (LMWH, LMKZ, LMPS.0, LMWH.01, LMWH.03, LMSJ, LMSR.03, LMMB.04, LMMB.0, LMSR.01, and LMSR.02) and two instance of high contamination (LMMB.01, and LMNC) detected in dreissenid mussel tissue in 2022 (Figure 38).
  - Mussel sampling locations sampled in Lake Michigan including LMMB.04, LMMB.0, LMMB.01, and LMNC were sampled near high-density urban areas such as Chicago and Milwaukee, suggesting ongoing contamination from localized point and diffuse sources.
- Total DDTs were only detected at twelve (GBBS, LEAB, LESP, LHTB, LMCB, LMHB, LMHM, LMNC, LOOC, LOOS, LORC, and LSAB) of the eighteen long-term sites sampled in 2022 (Concentration range: 1.71 - 118.3 ng/g dw; Table 11).
- Historically, highest DDT concentrations were detected in mussels from Lake Superior (3.89 ng/g dw), Lake St. Clair 16.9 ng/g dw), Niagara River (47.9 ng/g dw), Detroit River (49.0 ng/g dw), Hudson River (52.3 ng/g dw), Lake Erie (84.9 ng/g dw), Lake Huron (189.8 ng/g dw), Lake Ontario (197.2 ng/g dw), and Lake Michigan (386.6 ng/g dw; Figure 35; Figure 40).
- The continued presence of DDT residue in mussel tissue suggest continued input into the Great Lakes aquatic environment from likely sources such as resuspended sediments, input from urban and agricultural runoff (Zhou et al., 2018).
- Total DDT concentration sums were below the US EPA recommended screening values for both recreational and subsistence fishers (Table A2; Table A3).

Results - Total DDTs

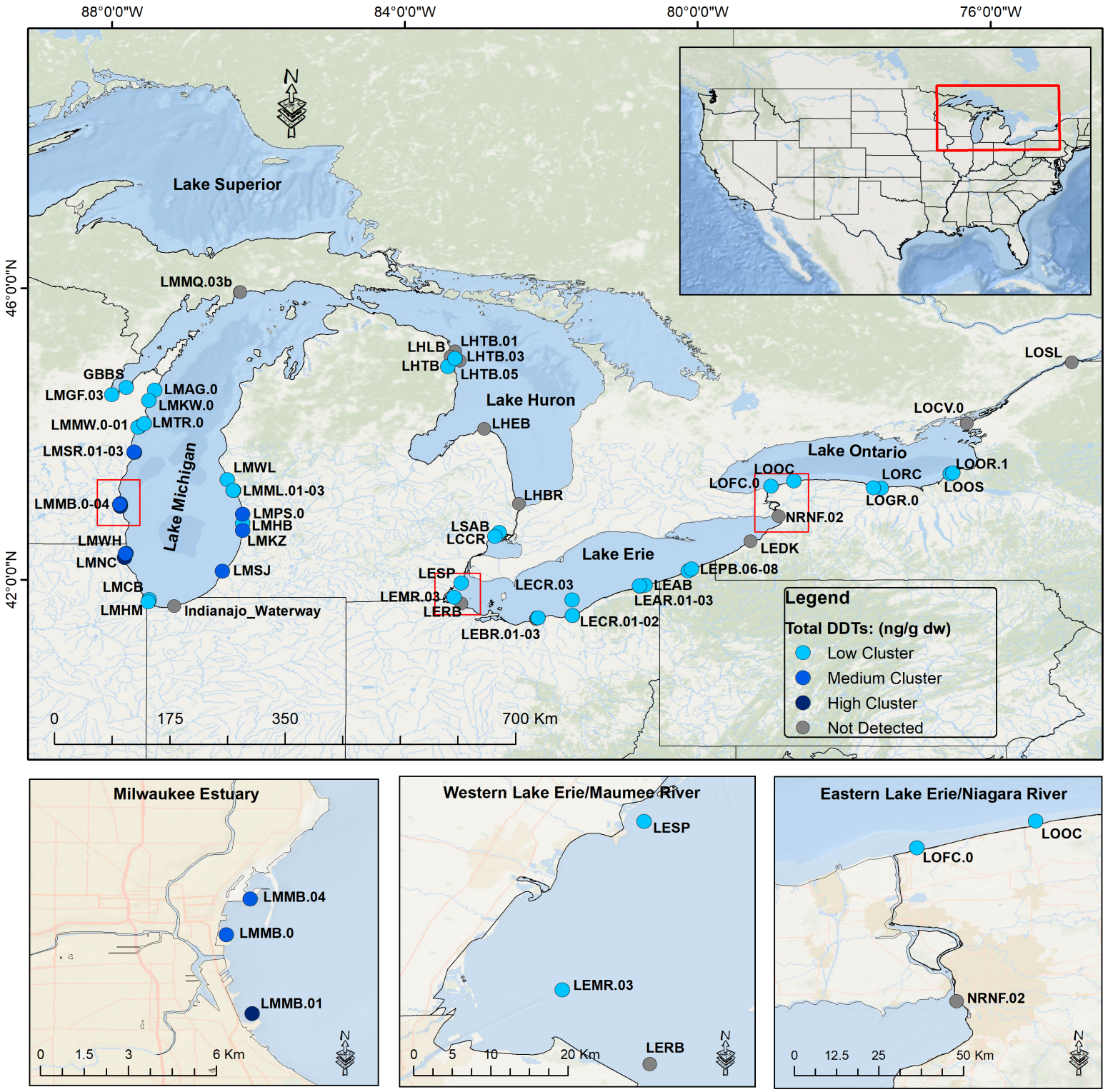


Figure 38. Total DDT concentration sums (ng/g dw) measured in Great Lakes mussel tissue during 2022. Some mussel sampling locations are shown to overlap due to the size of the circles. Colors represent contamination magnitude and clusters as follows: low ( ), medium ( ), high ( ), and not detected ( ).

Results - Total DDTs

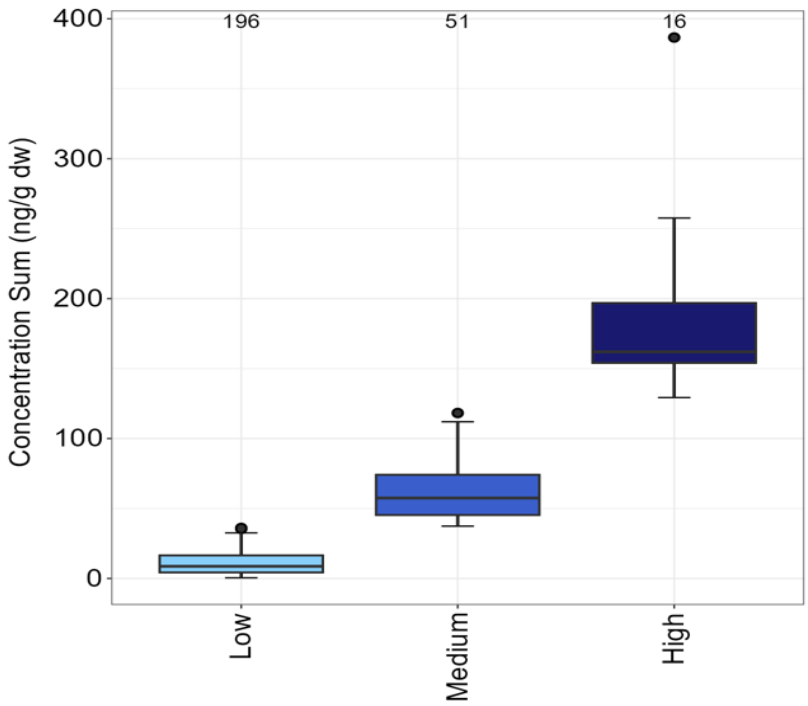


Figure 39. Total DDT concentration sums (ng/g dw) measured in Great Lakes mussel tissue during 1992-2022. Colors represent contamination magnitude as follows: low ( ), medium ( ), and high ( ).

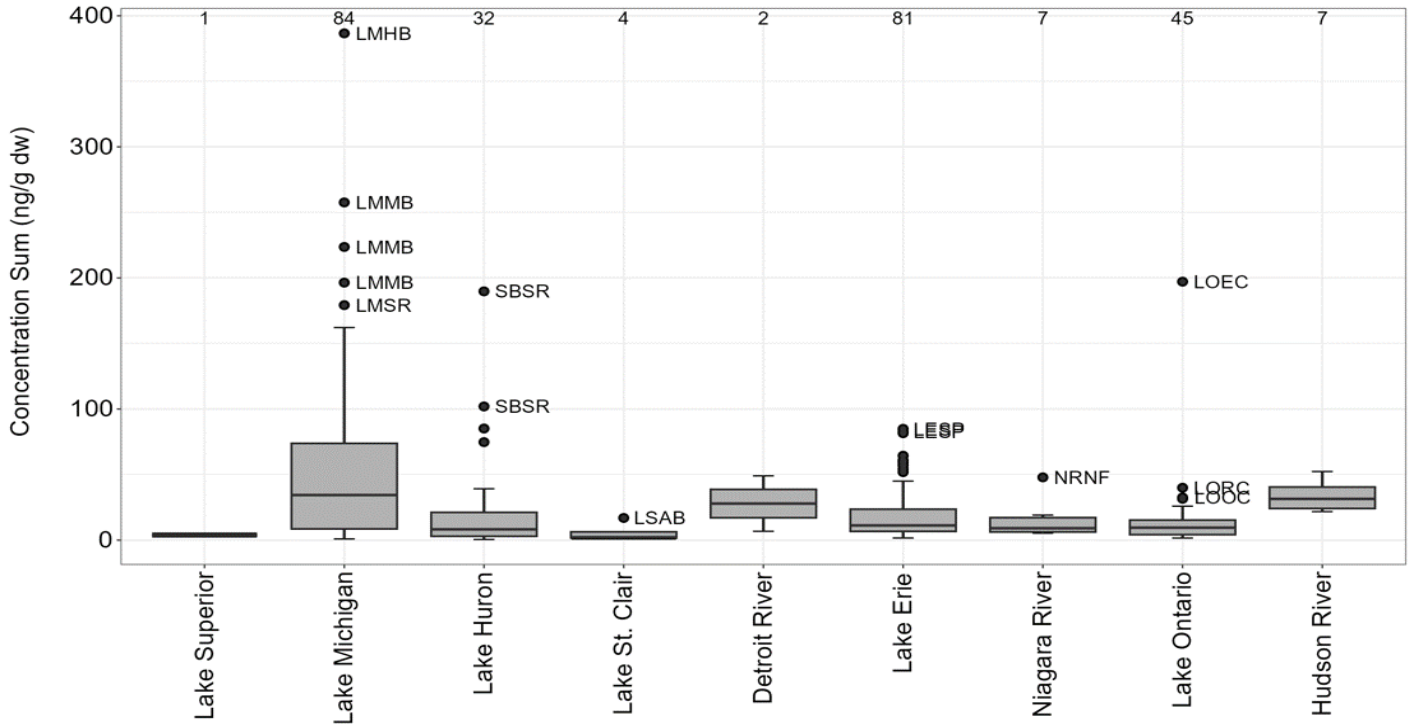


Figure 40. Total DDT concentration sums (ng/g dw) measured in Great Lakes mussels between 1992-2022. Lakes and major rivers including connecting channels sampled during 1992-2022 are listed geographically from west to east across the Great Lakes Basin. The number of DDT samples collected across each lake and major rivers including connecting channels sampled during 1992-2022 is noted at the top of the plot.

9.0 RESULTS - TOTAL DIELDRINS

9.1 Dieldrins Chemical Description

For this document, Total Dieldrins analyzed in 2022 is the sum of 5 compounds, and Total Dieldrins analyzed historically is the sum of 3 compounds (Table 6). Dieldrins were widely used as insecticides in the 1960s to control termites around buildings and protect crops from insects. In 1970, all uses of dieldrins were banned due to concern that they could cause severe aquatic environmental change and potential carcinogenicity (EPA, 1980). The ban was lifted in 1972 to allow limited use of dieldrins, primarily for termite control. All uses of dieldrins were finally banned in 1989 (EPA, 1990).

Exposure to dieldrins, whether through ingesting contaminated water and food products, including fish and shellfish, or inhaling indoor air in buildings treated with these insecticides, poses a significant health risk. Acute and long-term human exposures are associated with central nervous system intoxication (ATSDR, 2002). Because dieldrins can build up in the body and are slow to degrade, health effects can occur from long-term low dose exposure. Aldrin and dieldrin are carcinogenic to animals and classified as likely human carcinogens. The US FDA has established a 0.3 ppm wet weight safety level for aldrin and dieldrin in all fish (FDA, 2011).

The predominance of dieldrin in the environment can be explained by dieldrin being resistant to further transformation and aldrin being readily converted to dieldrin in the environment by sunlight and bacteria (ATSDR, 2002). Dieldrin bioaccumulates and biomagnifies through aquatic food chains and has been detected in tissues of freshwater and saltwater fish, and marine mammals. Aldrin and dieldrin applied to soil are tightly bound but may be transported to streams and rivers by soil erosion. Volatilization is the primary loss mechanism from soil. Dieldrin undergoes very slow and minor degradation to photodieldrin in marine environments (ATSDR, 2002).

Table 6. Dieldrin compounds composing the "Total Dieldrins" summation for the 2022 Great Lakes survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2022	Analyzed in Historical Data
Aldrin	●	●
Dieldrin	●	●
Endrin	●	●
Endrin Ketone	●	
Endrin Aldehyde	●	

9.2 Magnitude and Distribution of Dieldrins in Mussel Tissue in 2022

Dieldrins were not detected in mussel tissue at any site during the 2022 Great Lakes sampling period.

9.3 Historical Context of Dieldrins Magnitude and Distribution in Mussel Tissue

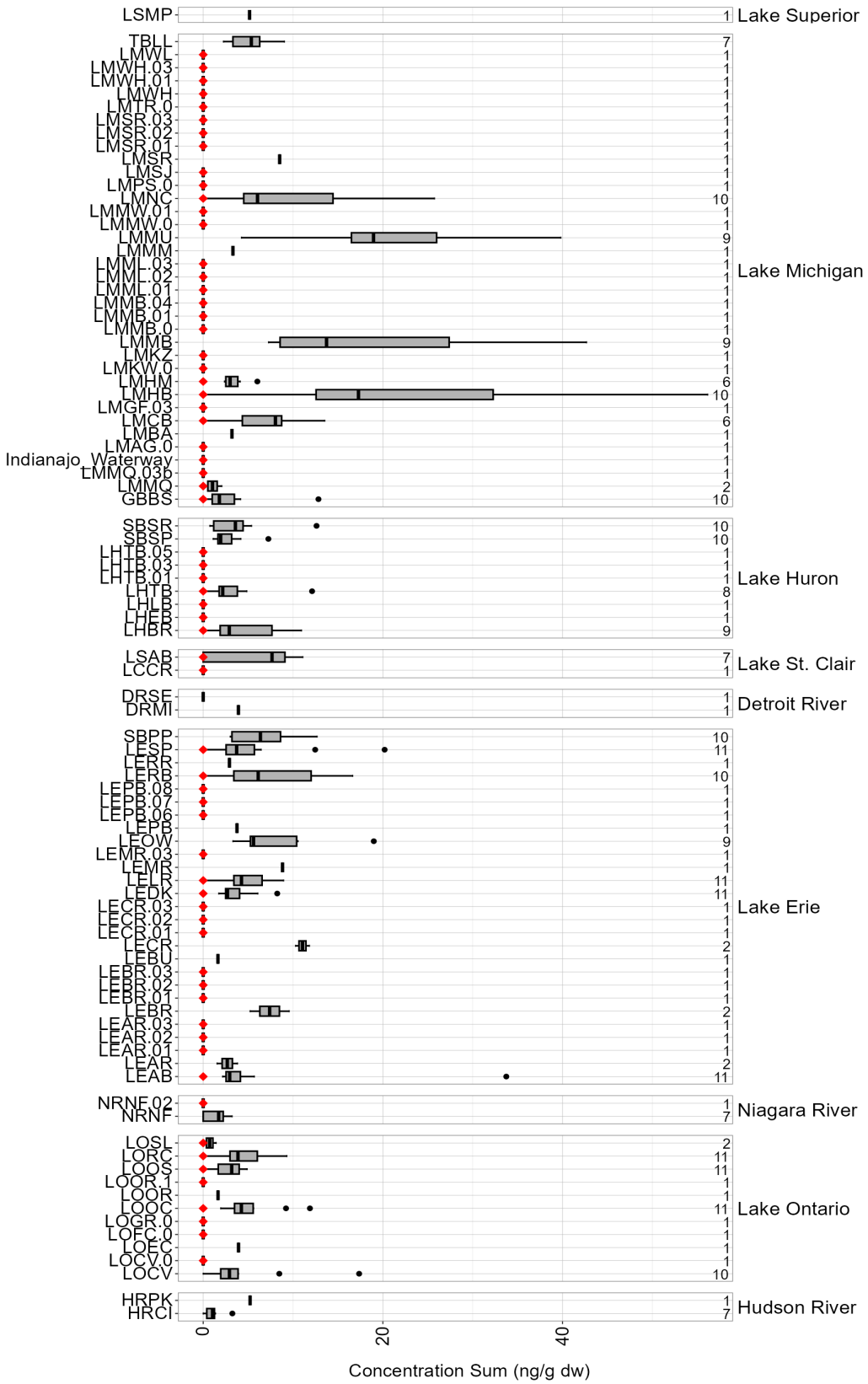


Figure 41. Total Dieldrin concentration sums (ng/g dw) measured in mussel tissue in 2022 (◆ - red diamonds), compared to boxplots of the historic Total Dieldrin concentration sums (ng/g dw) for each site (1992-2022) and results of the trend analysis showing significant increasing (▷) or decreasing (◁) contamination trends over time (gray triangles). Additional information is provided in Table 13 and Table A6. Lakes are listed geographically from west to east across the Great Lakes Basin. The number of years each site has been sampled since 1992 is noted at the side (right) of the plot for each site.

Results - Total Dieldrins

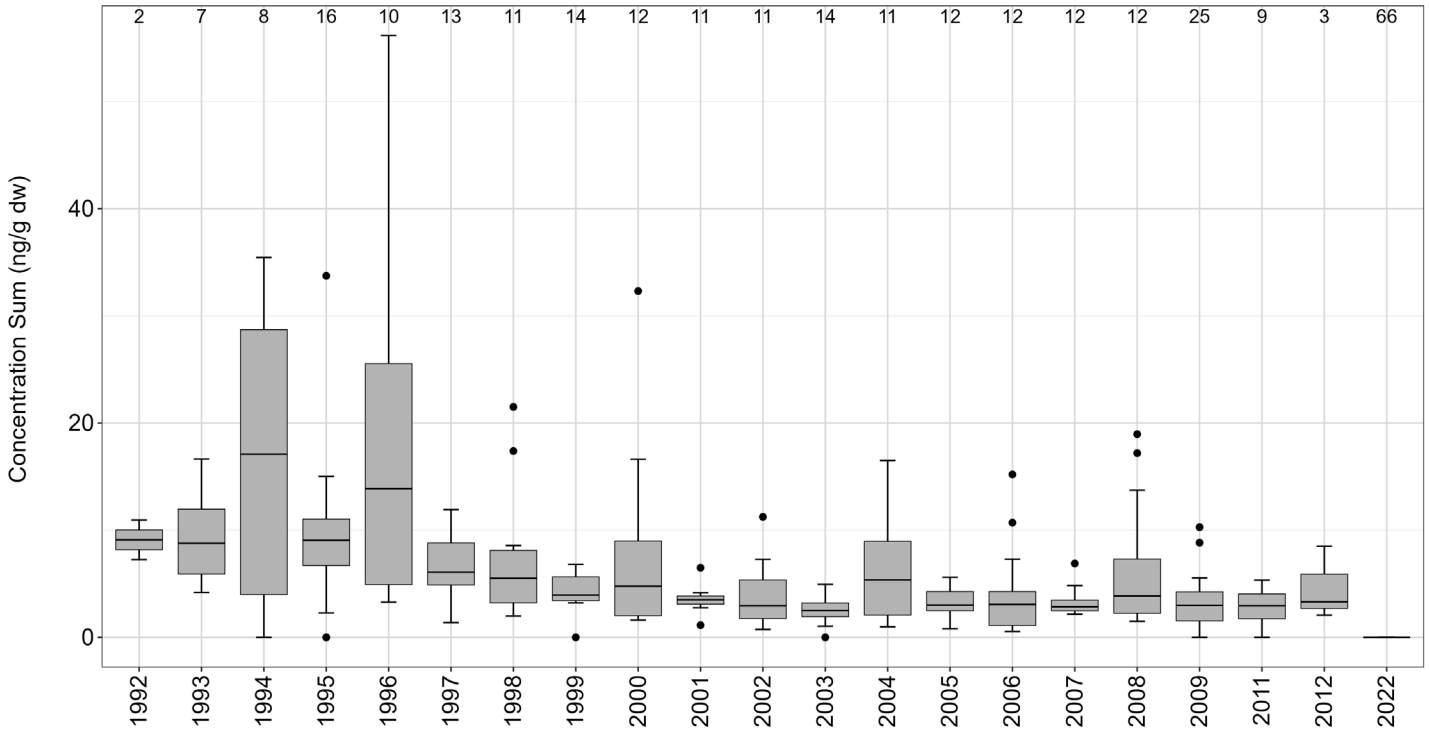


Figure 42. Boxplots representing the historic Total Dieldrin concentration sums (ng/g dw) measured in mussel tissue of the sites analyzed in this study. The number of sites that were sampled each year is noted at the top of the plot.

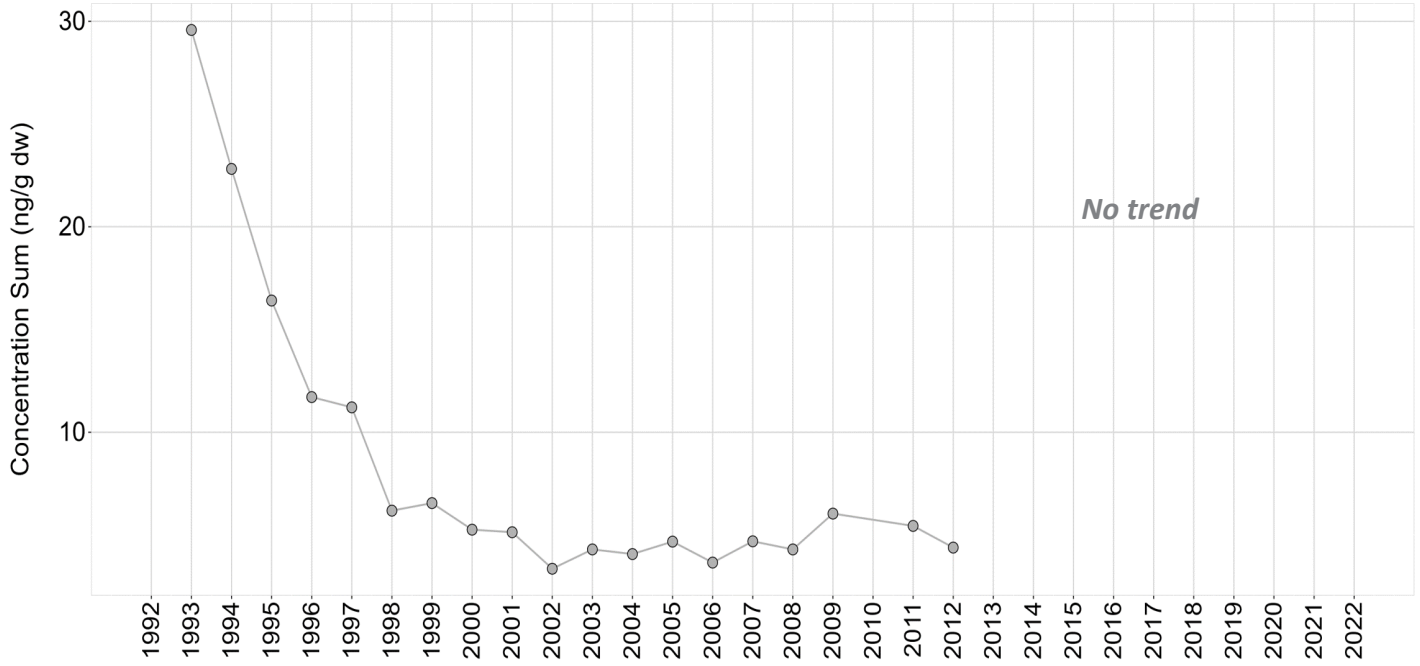


Figure 43. Three-point moving average of the yearly mean Total Dieldrin concentration sums (ng/g dw) measured in mussel tissue at sites analyzed in this study.

Results - Total Dieldrins

9.4 Dieldrins Summary

2022 Dreissenid Mussel Tissue Results (sum of 5 compounds):

- Total Dieldrins were not detected in dreissenid mussels at sites surveyed in the Great Lakes in 2022 (Figure 44).

Historical Context for Dreissenid Mussel Tissue (sum of 3 compounds):

- No significant regional temporal trend of Total Dieldrin concentration sums were observed across the Great Lakes Basin sampling locations between 1992-2022 ($\alpha = 0.05$; Figure 42; Figure 43).
- 0 sites assessed across the Great Lakes Basin showed significant decreasing temporal trends of Total Dieldrins concentration sums ($\alpha = 0.05$; Figure 41).
- 0 sites assessed across the Great Lakes Basin showed significant increasing temporal trends of Total Dieldrins concentration sums ($\alpha = 0.05$; Figure 41).
- The percentage of sites sampled in 2022 grouped into each cluster based on historic Great Lakes data (Figure 45):
  - 100% (66/66) of sites in the low cluster (0.00 – 7.66 ng/g dw)
  - 0% of sites in the medium cluster (7.67 – 21.5 ng/g dw)
  - 0% of sites in the high cluster (21.6 – 56.2 ng/g dw)

General Observations:

- Dieldrins were not detected in dreissenid mussel tissue at sites sampled throughout the Great Lakes in 2022 (Figure 41; Figure 44).
- Historically, highest Total Dieldrin concentrations were detected in mussels from sites sampled in Niagara River (3.22 ng/g dw), Detroit River (3.92 ng/g dw), Lake Superior (5.16 ng/g dw), Hudson River (5.22 ng/g dw), Lake Ontario (9.31 ng/g dw), Lake Huron (10.9 ng/g dw), Lake St. Clair (11.1 ng/g dw), Lake Erie (33.7 ng/g dw), and Lake Michigan (56.2 ng/g dw; Figure 41; Figure 46).

Results - Total Dieldrins

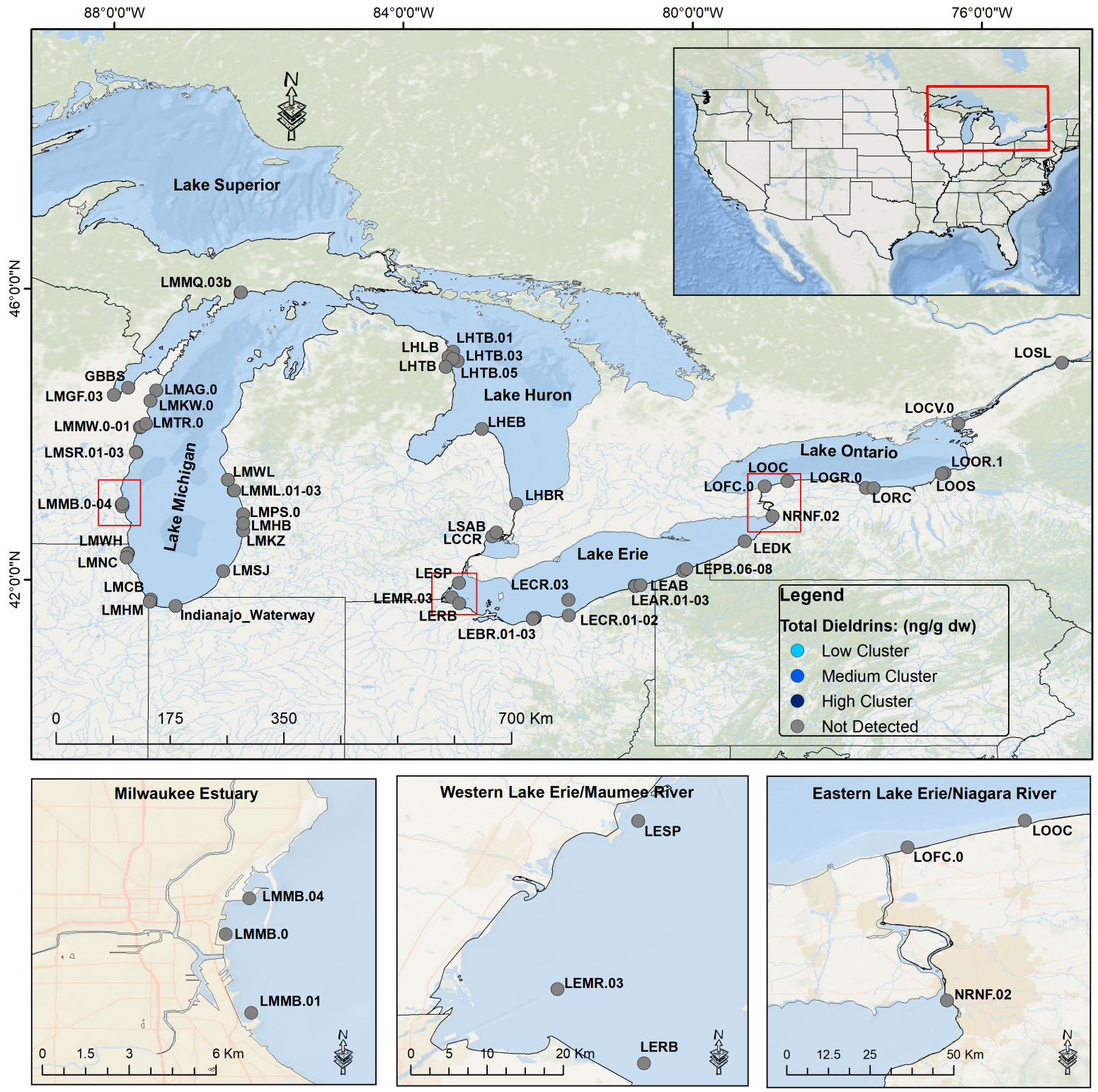


Figure 44. Total Dieldrin were not detected in Great Lakes mussel tissue analyzed during 2022. Some mussel sampling locations are shown to overlap due to the size of the circles. Colors represent contamination magnitude and clusters as follows: low ( ), medium ( ), high ( ), and not detected ( ).

Results - Total Dieldrins

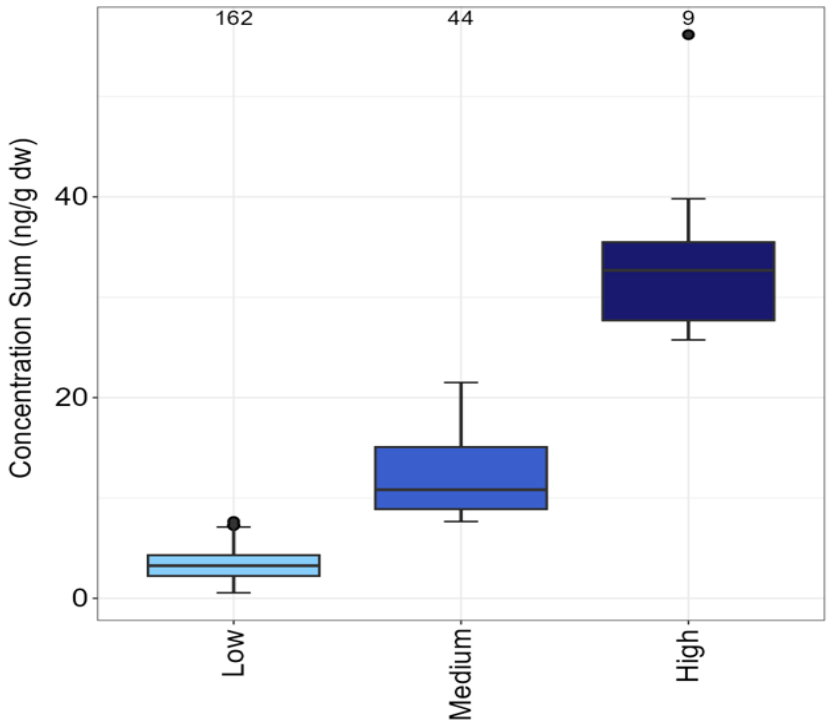


Figure 45. Total Dieldrin concentration sums (ng/g dw) measured in Great Lakes mussel tissue during 1992-2012. Colors represent contamination magnitude as follows: low ( ), medium ( ), and high ( ).

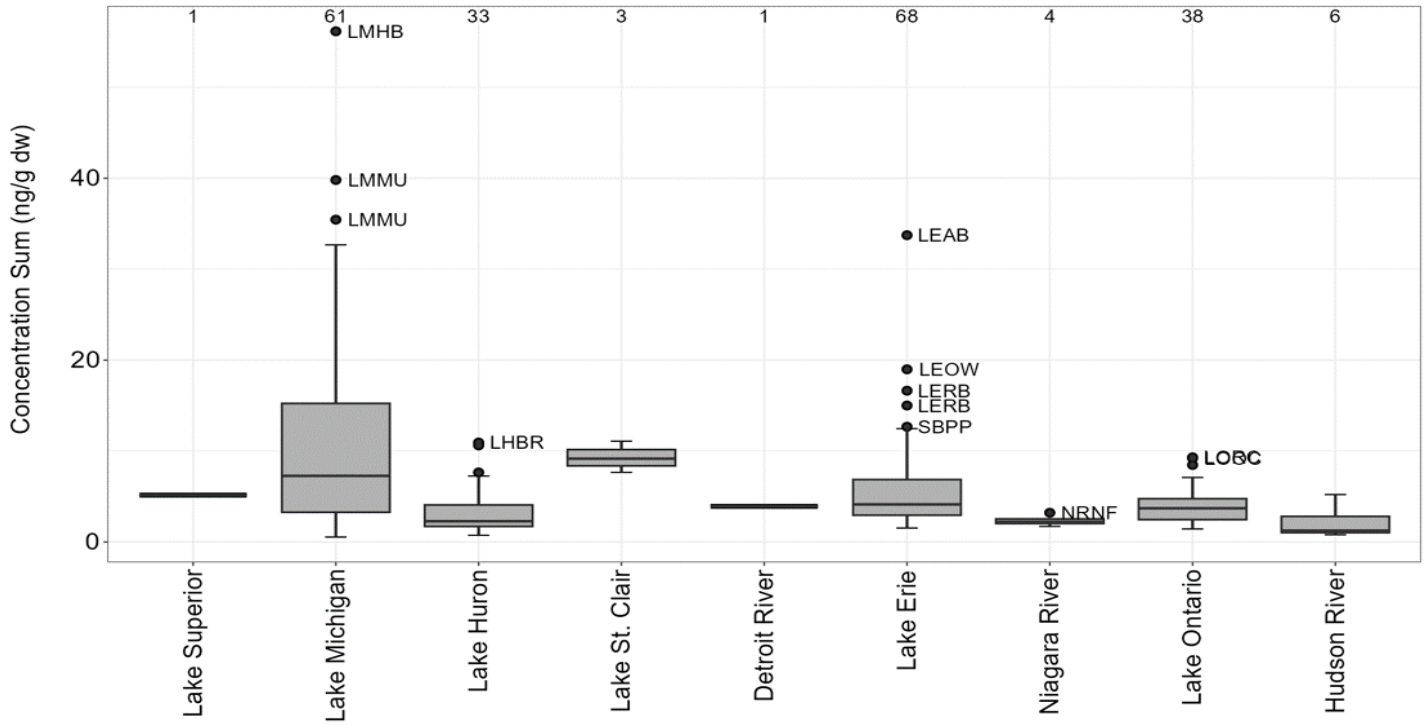


Figure 46. Total Dieldrin concentration sums (ng/g dw) measured in Great Lakes mussels between 1992-2022. Lakes and major rivers including connecting channels sampled during 1992-2022 are listed geographically from west to east across the Great Lakes Basin. The number of dieldrin samples collected across each lake and major rivers including connecting channels sampled during 1992-2022 is noted at the top of the plot.

10.0 RESULTS - TOTAL ENDOSULFANS

10.1 Endosulfans Chemical Description

For this document, Total Endosulfans analyzed both in 2022 and historically are the sum of 3 compounds (Table 7). Technical grade endosulfan is a mixture of two isomers (Endosulfan I and II), and Endosulfan sulfate is a product of oxidation that can be found in technical grade endosulfan. Endosulfan was a restricted-application pesticide used to treat certain crops against aphids, beetles, leafhoppers, whiteflies, etc. (ATSDR, 2015). Endosulfans were restricted to certain crops before their phase-out in 2016 (ATSDR, 2015).

The general population is exposed to endosulfans through diet, breathing contaminated air, ingesting contaminated water, or being in contact with contaminated soil or plants (ATSDR, 2015). In humans, endosulfan appears to accumulate in the liver, kidneys, and brain and is typically excreted within a few days or weeks. Endosulfan accumulation has been shown to primarily effect the nervous system. Exposure to high levels of endosulfans can induce hyperactivity and convulsion. No evidence suggests that endosulfan can cause cancer or any other disease in humans (ATSDR, 2015).

Endosulfan can be found in the atmosphere, soil, and water (ATSDR, 2015). Endosulfans can travel long distances by air and may be broken down by sunlight. Endosulfans attach to soil particles, which limits their movement and transport across environmental compartments. Endosulfans change into the less toxic endosulfan diol in water, but endosulfan sulfate is more resistant to breaking down. Endosulfans can bioaccumulate in organisms living in contaminated water (ATSDR, 2015).

Table 7. Endosulfan compounds composing the "Total Endosulfans" summation for the 2022 Great Lakes survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2022	Analyzed in Historical Data
Endosulfan I	●	●
Endosulfan II	●	●
Endosulfan Sulfate	●	●

10.2 Magnitude and Distribution of Endosulfans in Mussel Tissue in 2022

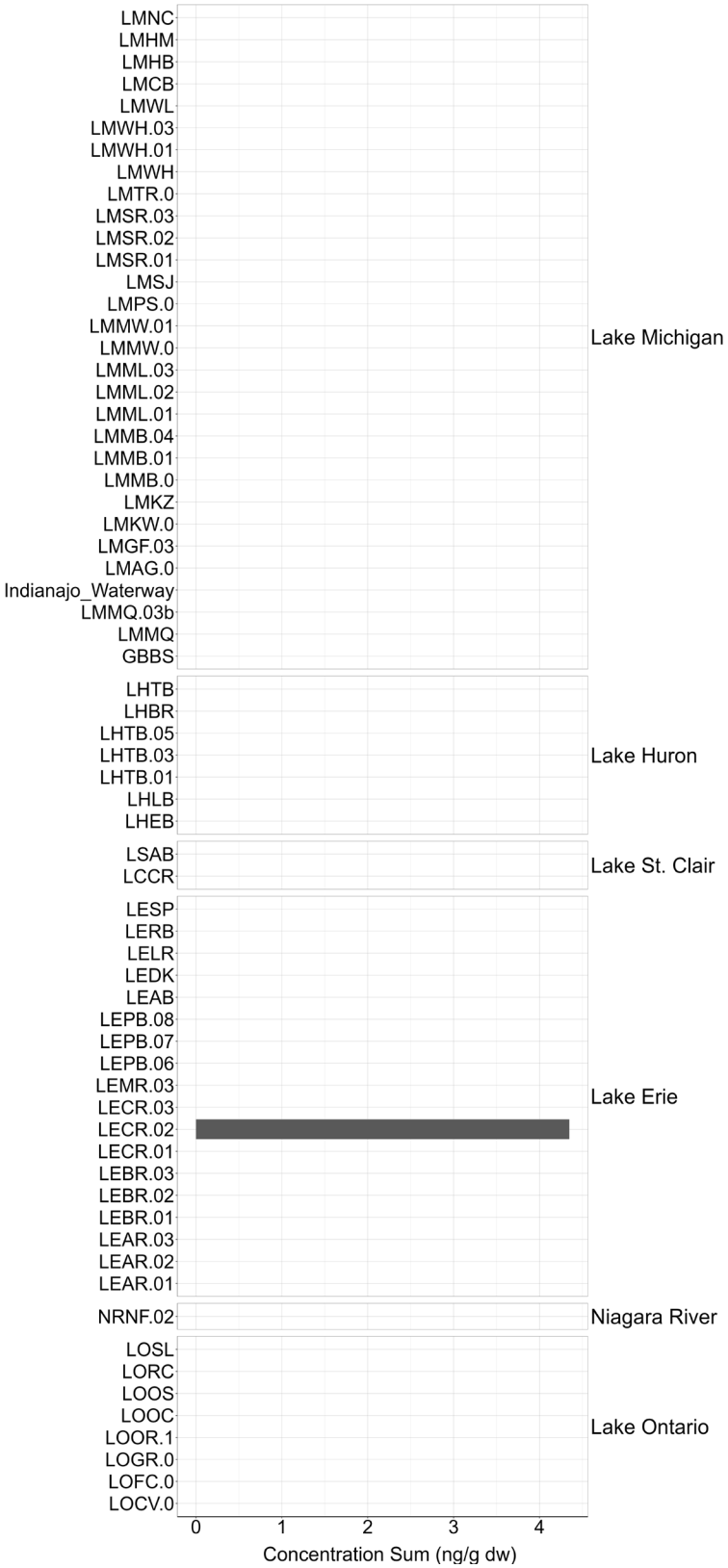


Figure 47. Bar graph showing the magnitude of Total Endosulfan concentration sums detected in mussel tissue in the Great Lakes Basin during the 2022 sampling period. Lakes are listed geographically from west to east across the Great Lakes Basin.

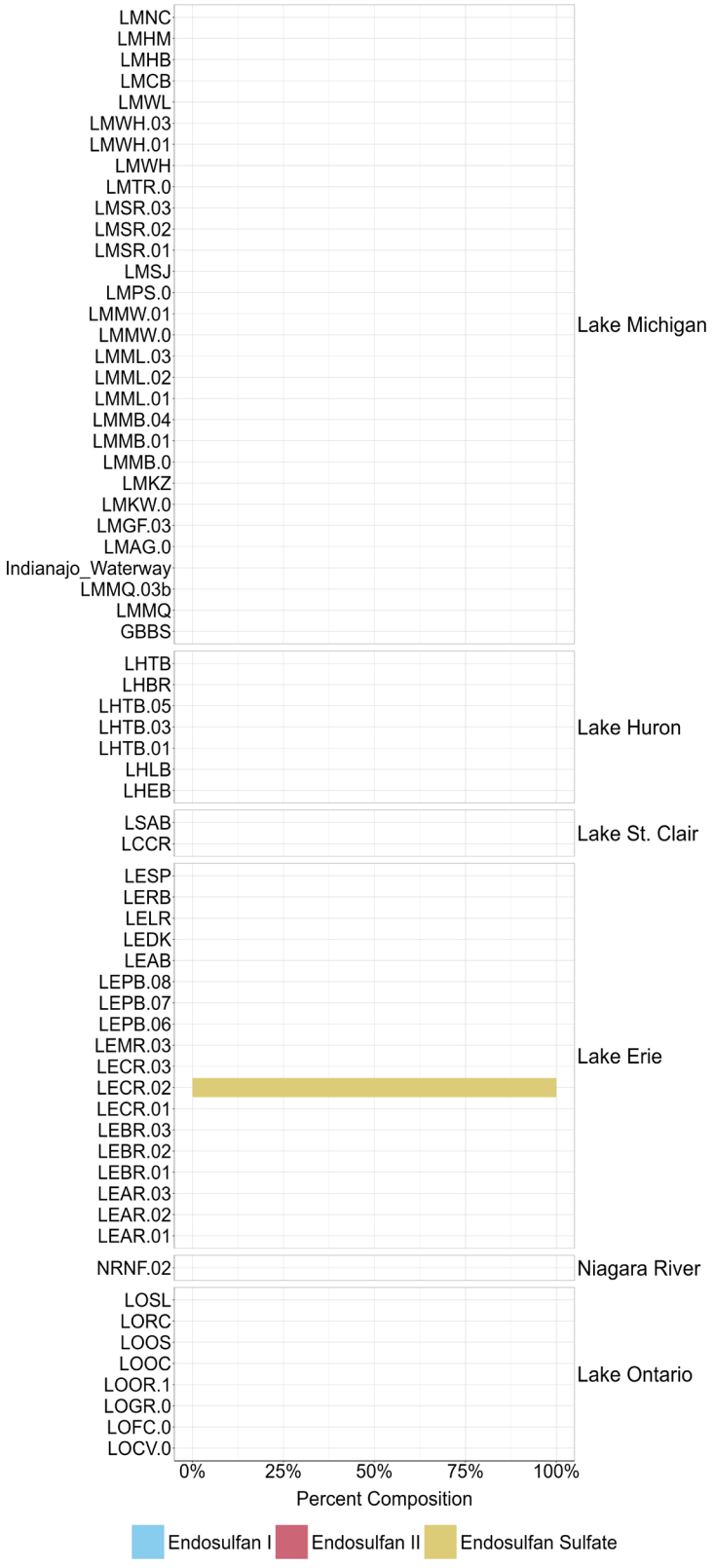


Figure 48. Percent composition of individual endosulfans contaminants in Total Endosulfan concentration sums per site in the Great Lakes Basin during the 2022 sampling period. Lakes are listed geographically from west to east across the Great Lakes Basin.

10.3 Historical Context of Endosulfans Magnitude and Distribution in Mussel Tissue

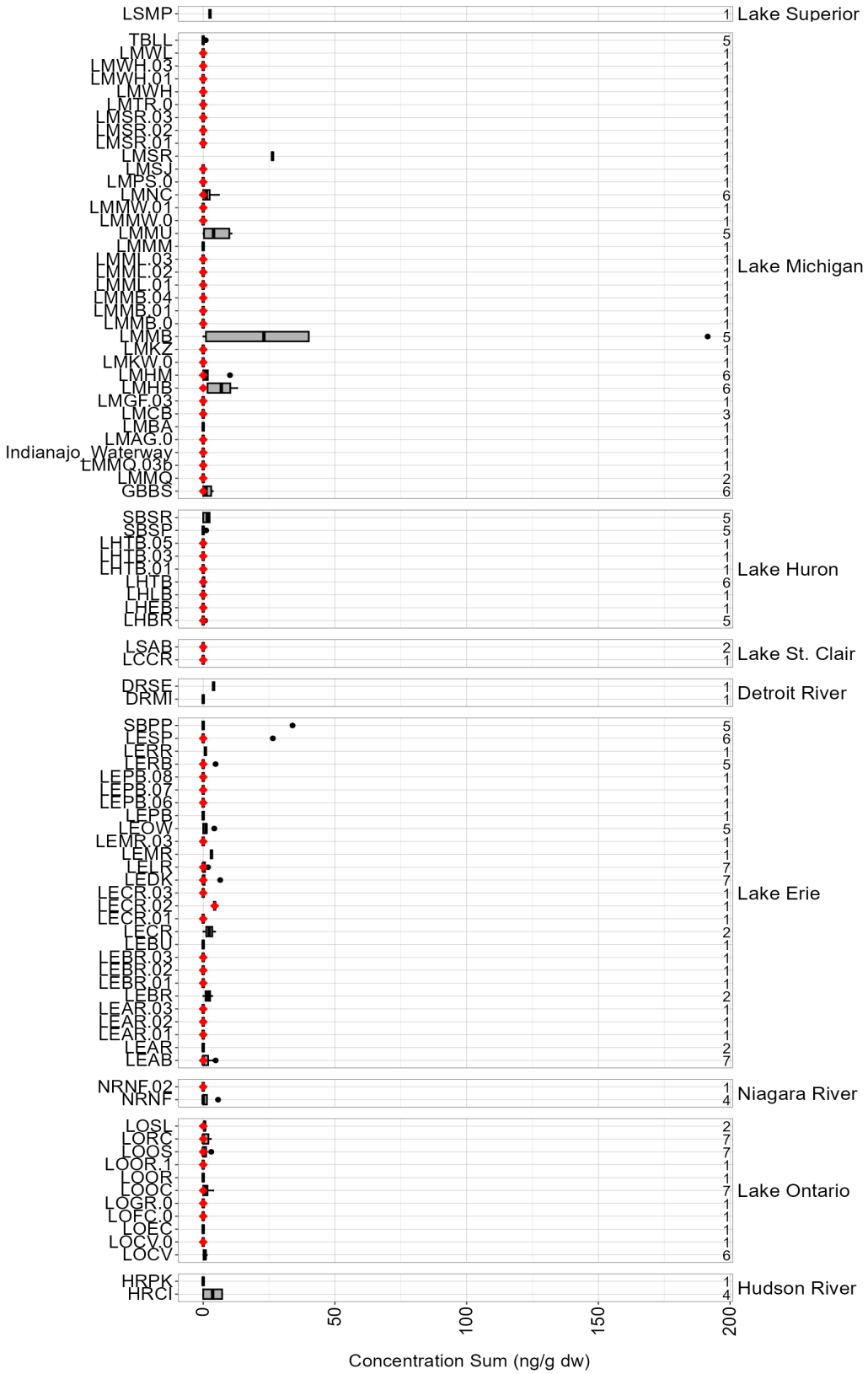


Figure 49. Total Endosulfan concentration sums (ng/g dw) measured in mussel tissue in 2022 (◆ - red diamonds), compared to boxplots of the historic Total Endosulfan concentration sums (ng/g dw) for each site (1995-2022) and results of the trend analysis showing significant increasing (▶) or decreasing (◀) contamination trends over time (gray triangles). Additional information is provided in Table 13 and Table A6. Lakes are listed geographically from west to east across the Great Lakes Basin. The number of years each site has been sampled since 1992 is noted at the side (right) of the plot for each site.

Results - Total Endosulfans

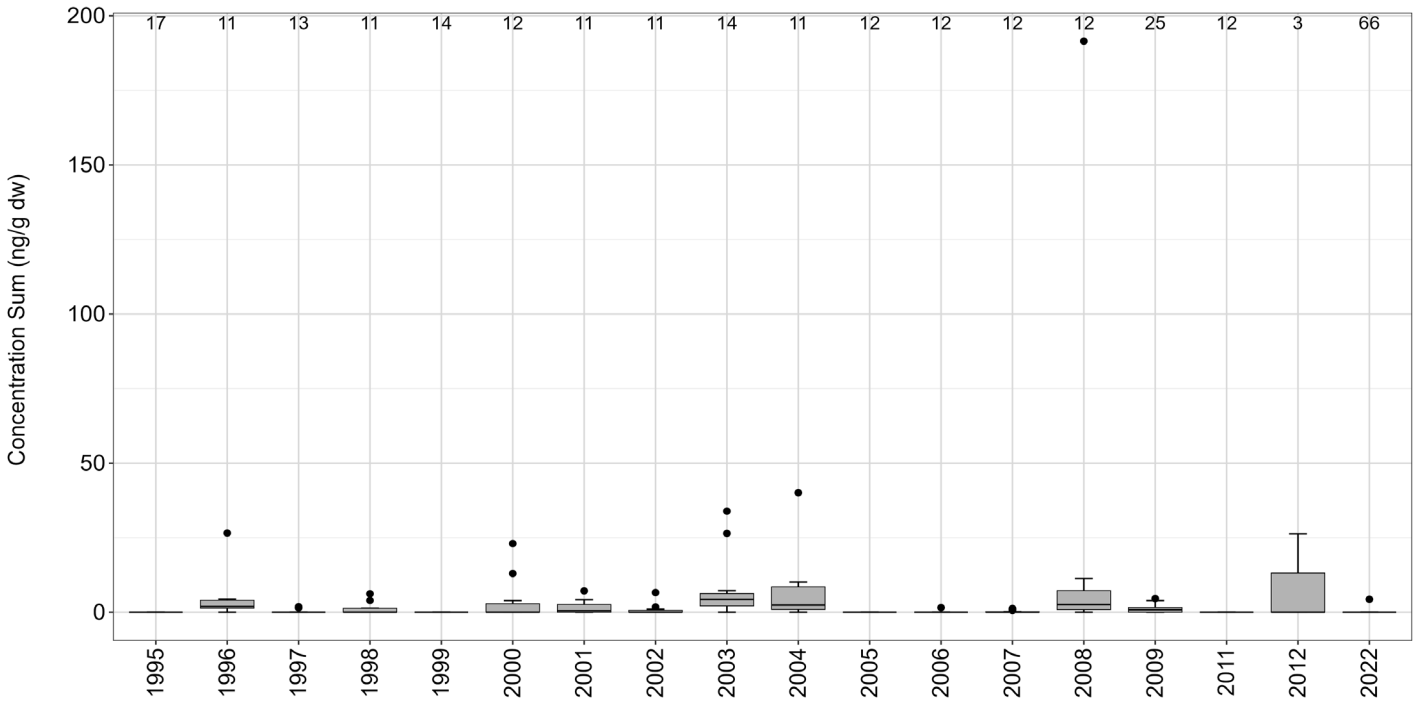


Figure 50. Boxplots representing the historic Total Endosulfan concentration sums (ng/g dw) measured in mussel tissue of the sites analyzed in this study. The number of sites that were sampled each year is noted at the top of the plot.

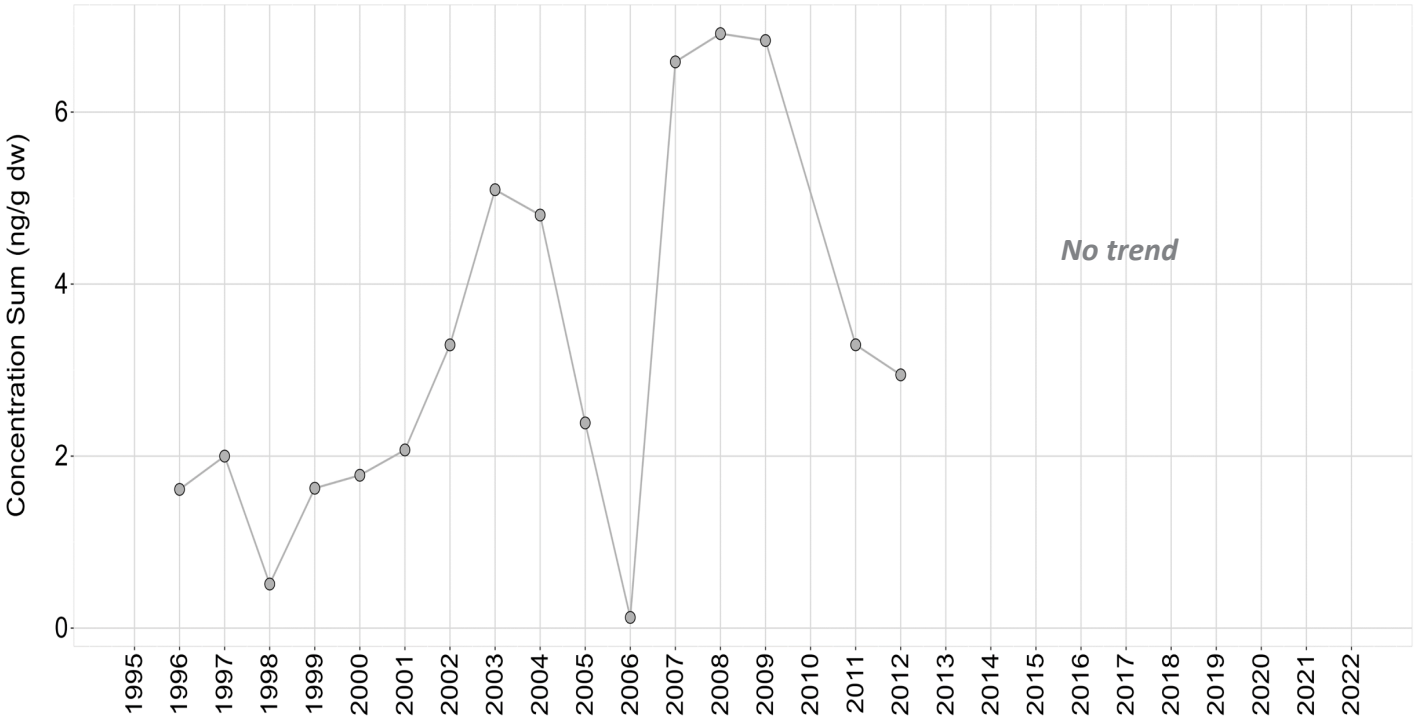


Figure 51. Three-point moving average of the yearly mean Total Endosulfan concentration sums (ng/g dw) measured in mussel tissue at sites analyzed in this study.

Results - Total Endosulfans

10.4 Endosulfans Summary

2022 Dreissenid Mussel Tissue Results (sum of 3 compounds):

- Endosulfans were detected in dreissenid mussels at one site (LECR.02; 4.35 ng/g dw) surveyed in the Great Lakes in 2022 (Figure 47; Figure 48; Figure 52).

Historical Context for Dreissenid Mussel Tissue (sum of 3 compounds):

- No significant decreasing regional temporal trend of Total Endosulfan concentration sums were observed across the Great Lakes Basin sampling locations between 1995-2022 ($\alpha = 0.05$; Figure 50; Figure 51).
- 0 sites assessed across the Great Lakes Basin showed significant decreasing temporal trends of Total Endosulfan concentration sums ($\alpha = 0.05$; Figure 49).
- 0 sites assessed across the Great Lakes Basin showed significant increasing temporal trends of Total Endosulfan concentration sums ($\alpha = 0.05$; Figure 49).
- The percentage of sites sampled in 2022 grouped into each cluster based on historic Great Lakes data (Figure 53):
  - 100% (66/66) of sites in the low cluster (0.00 – 13.0 ng/g dw)
  - 0% of sites in the medium cluster (13.1 – 40.1 ng/g dw)
  - 0% of sites in the high cluster (40.2 – 191.5 ng/g dw)

General Observations:

- Endosulfan was detected in dreissenid mussel tissue at one site (LECR.02) surveyed in the Great Lakes in 2022 (Figure 47; Figure 52).
- Endosulfans (organochlorine insecticides) residue can adsorb to particulates and persist in soil and/or sediment (Pait et al., 1992; Rand et al., 2010). As such, their continued presence and detection in mussel tissue is likely attributable to release/input from these medium.
- Historically, highest endosulfan concentrations were detected in mussels from sites sampled in Lake Superior (2.56 ng/g dw), Lake Ontario (3.89 ng/g dw), Detroit River (3.93 ng/g dw), Lake Huron (3.97 ng/g dw), Niagara River (5.64 ng/g dw), Hudson River (7.24 ng/g dw), Lake Erie (33.9 ng/g dw), and Lake Michigan (191.5 ng/g dw; Figure 49; Figure 54).
- Endosulfan concentration was below the US EPA recommended screening values for both recreational and subsistence fishers (Table A2; Table A3).

Results - Total Endosulfans



Figure 52. Total Endosulfan concentration sums (ng/g dw) measured in Great Lakes mussel tissue during 2022. Some mussel sampling locations are shown to overlap due to the size of the circles. Colors represent contamination magnitude and clusters as follows: low ( ), medium ( ), high ( ), and not detected ( ).

Results - Total Endosulfans

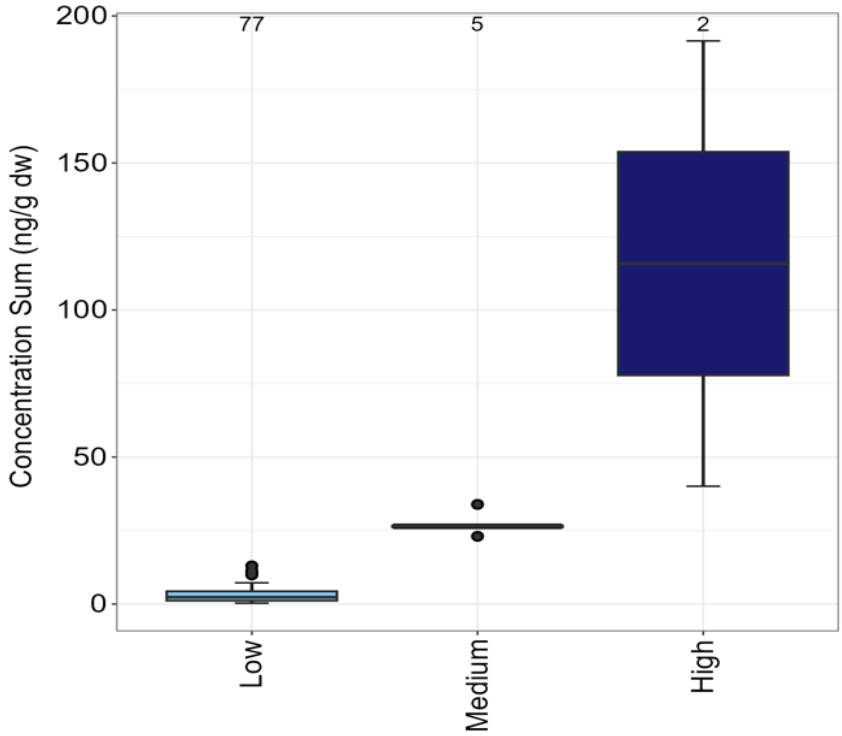


Figure 53. Total Endosulfan concentration sums (ng/g dw) measured in Great Lakes mussel tissue during 1995-2022. Colors represent contamination magnitude as follows: low ( ), medium ( ), and high ( ).

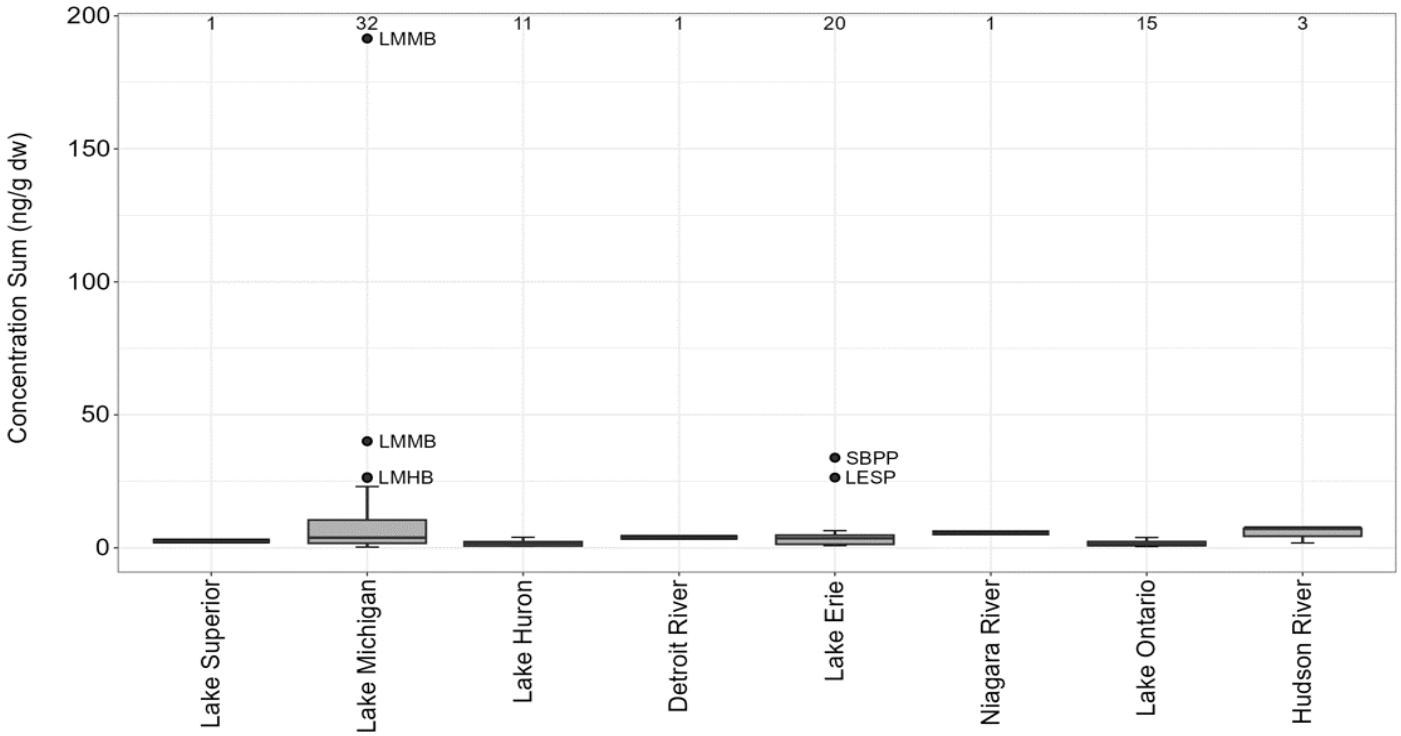


Figure 54. Total Endosulfan concentration sums (ng/g dw) measured in Great Lakes mussels between 1995-2022. Lakes and major rivers including connecting channels sampled during 1996-2022 are listed geographically from west to east across the Great Lakes Basin. The number of dieldrin samples collected across each lake and major rivers including connecting channels sampled during 1992-2022 is noted at the top of the plot.

11.0 RESULTS - TOTAL HCHs

11.1 HCHs Chemical Description

For this document, Total HCHs analyzed both in 2022 and historically are the sum of 4 compounds (Table 8). Technical grade HCH (hexachlorocyclohexane) contains the alpha, beta, gamma, delta, and epsilon forms of HCH. Almost all insecticidal properties are found in gamma-HCH (lindane), which is used as an insecticide on fruit, vegetables, and forest crops. It is also found in lotion, cream, or shampoo as a prescription to treat head and body lice and scabies (ATSDR, 2005). In 2009, the Stockholm Convention on Persistent Organic Pollutants implemented an international ban on the use of lindane in agriculture. However, it allowed a 5-year extension for its use in the treatment of head lice and scabies (UNEP, 2009). The US did not ratify the convention; however, the EPA requested the voluntary cancellation of the last agricultural use of lindane in 2006 (EPA, 2006).

All of the HCH isomers are toxic to animals to varying degrees and are persistent in the environment. The Department of Health and Human Services (DHHS), International Agency for Research on Cancer (IARC), and the EPA vary in their classification of HCH as a human carcinogen. However, technical HCH, alpha-HCH, and beta-HCH are listed by all three as at least possible human carcinogens (ATSDR, 2005). In 2015, based on a review of the most recent data on lindane, the IARC modified its classification from “probably carcinogenic to humans” to “known to cause human cancer” (IARC, 2015).

HCH can be broken down into less toxic substances by algae, bacteria, and fungi in sediments and water, but it is a slow process. HCH has been shown to accumulate in the fatty tissue of fish (Ortega-Retuerta et al., 2008).

Table 8. HCH compounds composing the “Total HCHs” summation for the 2022 Great Lakes survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2022	Analyzed in Historical Data
Alpha-Hexachlorocyclohexane	●	●
Beta-Hexachlorocyclohexane	●	●
Gamma-Hexachlorocyclohexane	●	●
Delta-Hexachlorocyclohexane	●	●

11.2 Magnitude and Distribution of HCHs in Mussel Tissue in 2022

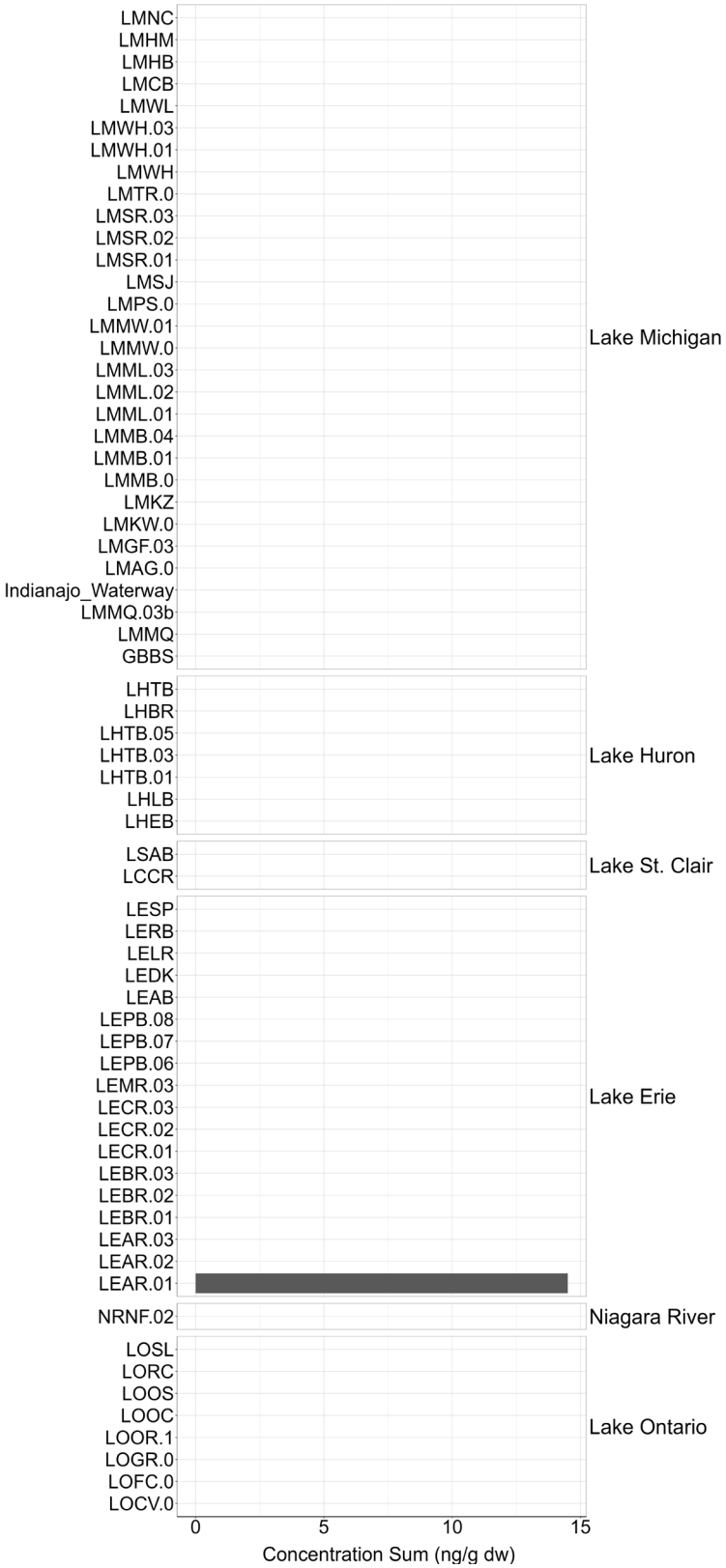


Figure 55. Bar graph showing the magnitude of Total HCH concentration sums detected in mussel tissue in the Great Lakes Basin during the 2022 sampling period. Lakes are listed geographically from west to east across the Great Lakes Basin.

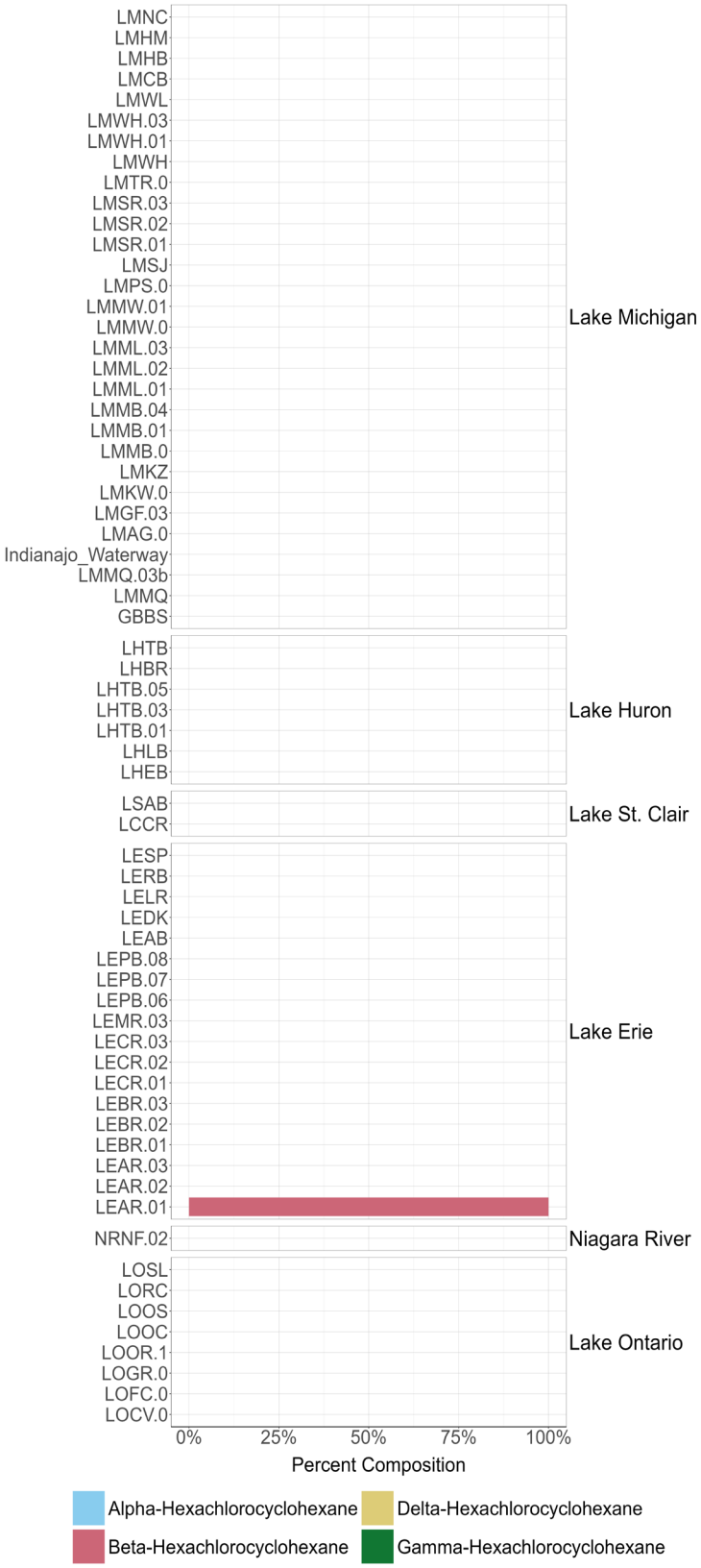


Figure 56. Percent composition of individual HCH contaminants measured in Total HCH concentration sums per site in the Great Lakes Basin during the 2022 sampling period. Lakes are listed geographically from west to east across the Great Lakes Basin.

11.3 Historical Context of HCHs Magnitude and Distribution in Mussel Tissue

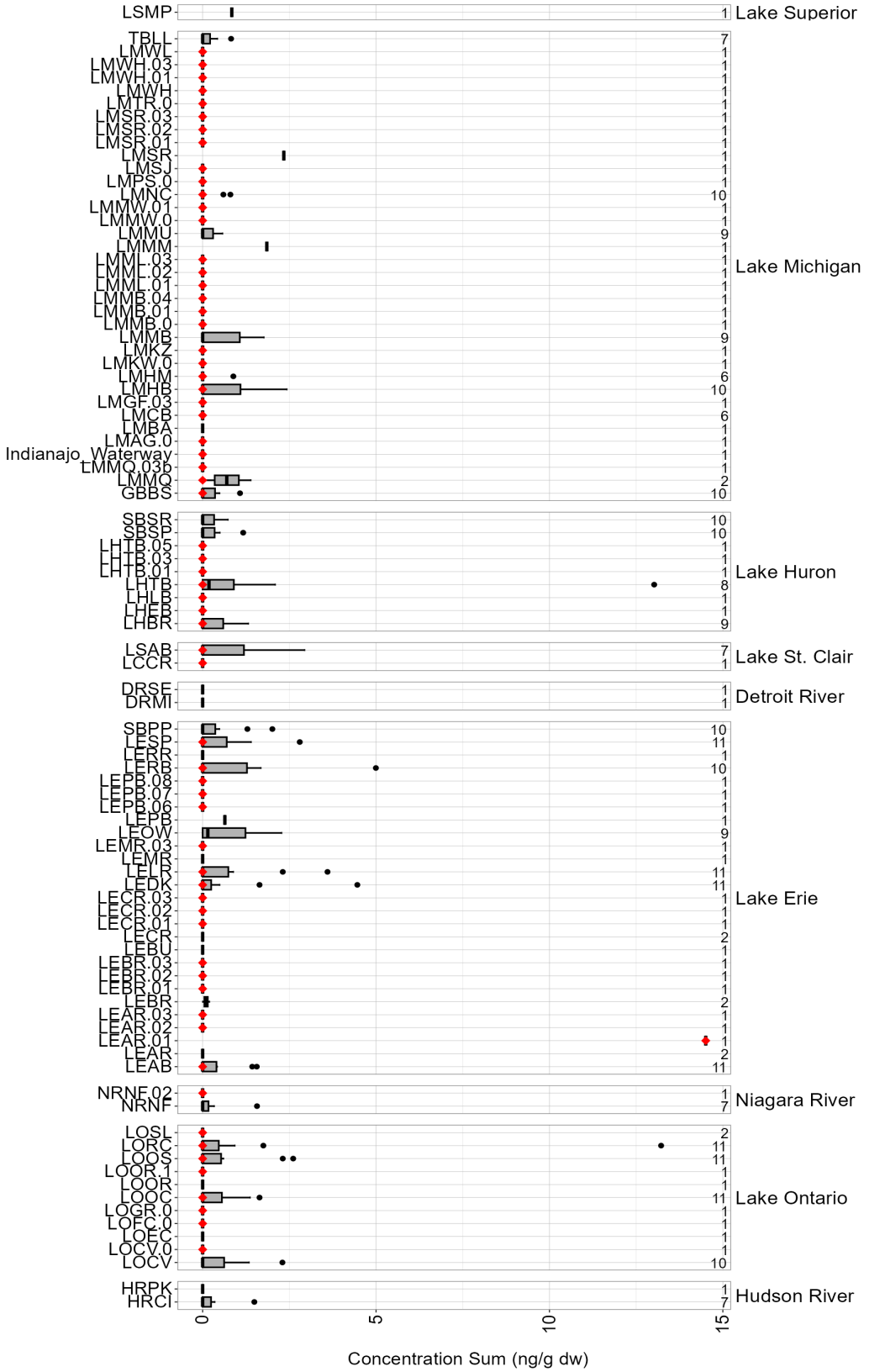


Figure 57. Total HCH concentration sums (ng/g dw) measured in mussel tissue in 2022 (◆ - red diamonds), compared to boxplots of the historic Total HCH concentration sums for each site (1992-2022) and results of the trend analysis showing significant increasing (►) or decreasing (◄) contamination trends over time (gray triangles). Additional information is provided in Table 13 and Table A6. Lakes are listed geographically from west to east across the Great Lakes Basin. The number of years each site has been sampled since 1992 is noted at the side (right) of the plot for each site.

Results - Total HCHs

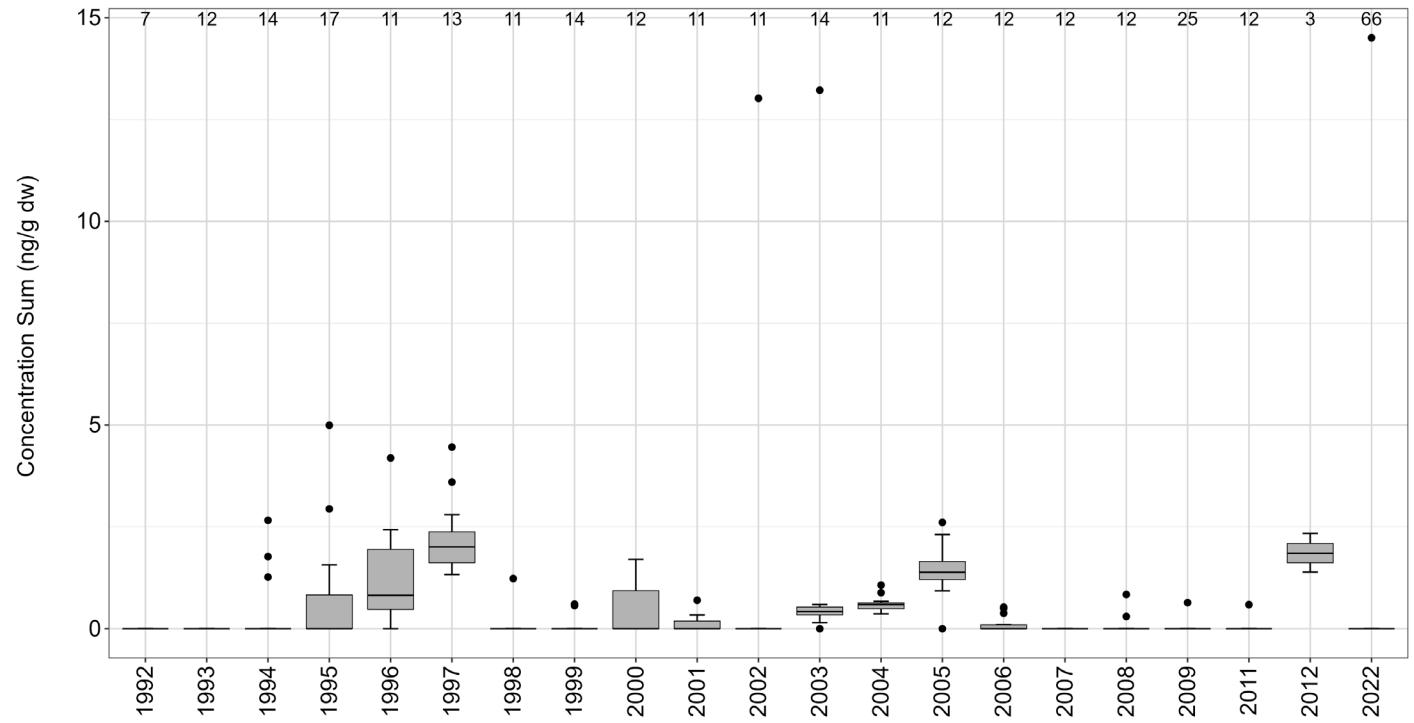


Figure 58. Boxplots representing the historic Total HCH concentration sums (ng/g dw) measured in mussel tissue of the sites analyzed in this study. The number of sites that were sampled each year is noted at the top of the plot.

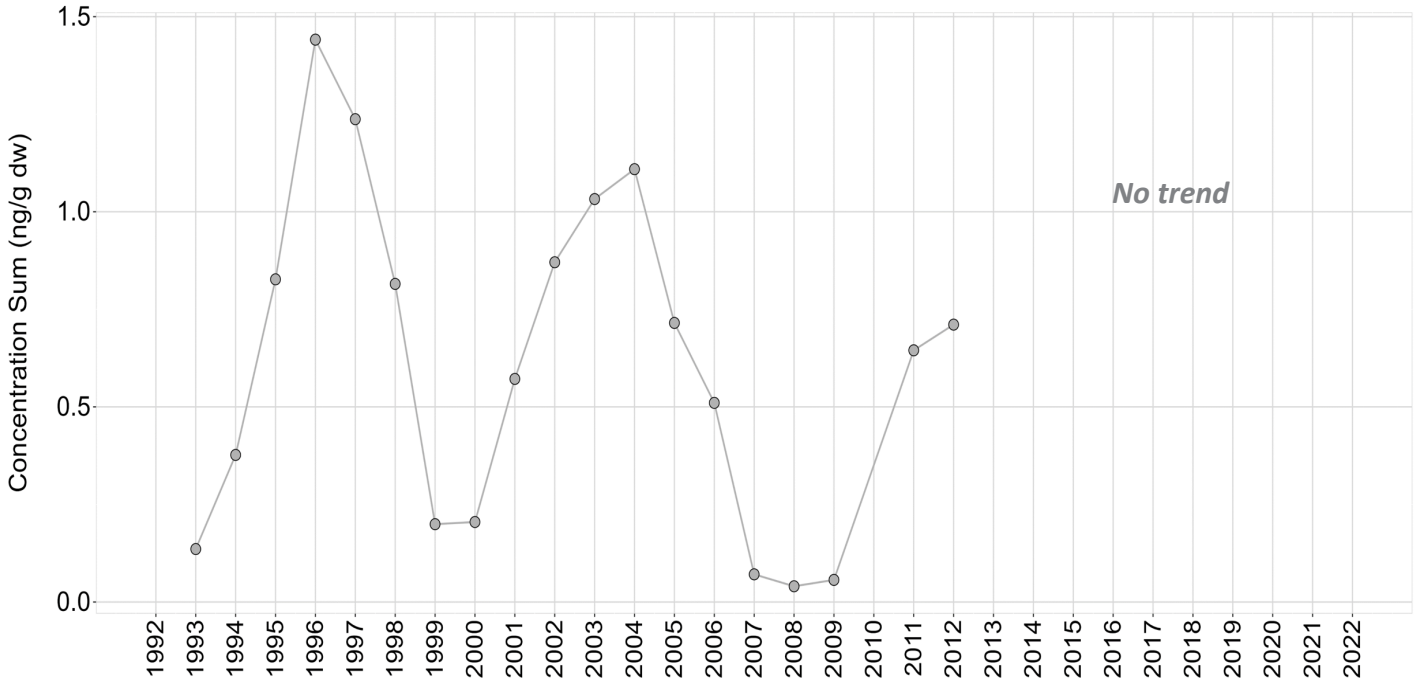


Figure 59. Three-point moving average of the yearly mean Total HCH concentration sums (ng/g dw) measured in mussel tissue at sites analyzed in this study.

Results - Total HCHs

11.4 HCHs Summary

2022 Dreissenid Mussel Tissue Results (sum of 4 compounds):

- HCHs were detected in dreissenid mussels at one site (LEAR.01; 14.6 ng/g dw) surveyed in the Great Lakes in 2022 (Figure 55; Figure 56; Figure 60).

Historical Context for Dreissenid Mussel Tissue (4 compounds):

- No significant decreasing regional temporal trend of Total HCHs concentration sums were observed across the Great Lakes Basin sampling locations between 1992-2022 ($\alpha = 0.05$; Figure 58; Figure 59).
- 0 sites assessed across the Great Lakes Basin showed significant decreasing temporal trends of Total HCHs concentration sums ($\alpha = 0.05$; Figure 57).
- 0 sites assessed across the Great Lakes Basin showed significant increasing temporal trends of Total HCHs concentration sums ($\alpha = 0.05$; Figure 57).
- The percentage of sites sampled in 2022 grouped into each cluster based on historic Great Lakes data (Figure 61):
  - 98.5% (65/66) of sites in the low cluster (0.00 – 1.85 ng/g dw)
  - 0% of sites in the medium cluster (1.86 – 5.00 ng/g dw)
  - 1.5% (1/66) of sites in the high cluster (5.01 – 14.6 ng/g dw)

General Observations:

- HCHs was detected in dreissenid mussel tissue at one site (LEAR.01) sampled in the Great Lakes in 2022 (Figure 55; Figure 60).
  - It is noteworthy that this is the highest HCH concentration ever detected in the Great Lakes Basin.
- Historically, highest HCH concentrations were detected in mussels Lake Superior (0.84 ng/g dw), Hudson River (1.49 ng/g dw), Niagara River (1.57 ng/g dw), Lake Michigan (2.43 ng/g dw), Lake St. Clair (2.94 ng/g dw), Lake Huron (13.0 ng/g dw), Lake Ontario (13.2 ng/g dw), and Lake Erie (14.6 ng/g dw; Figure 57; Figure 62).
- Total HCH concentration sums were below the US EPA recommended screening values for both recreational and subsistence fishers (Table A2; Table A3).

Results - Total HCHs

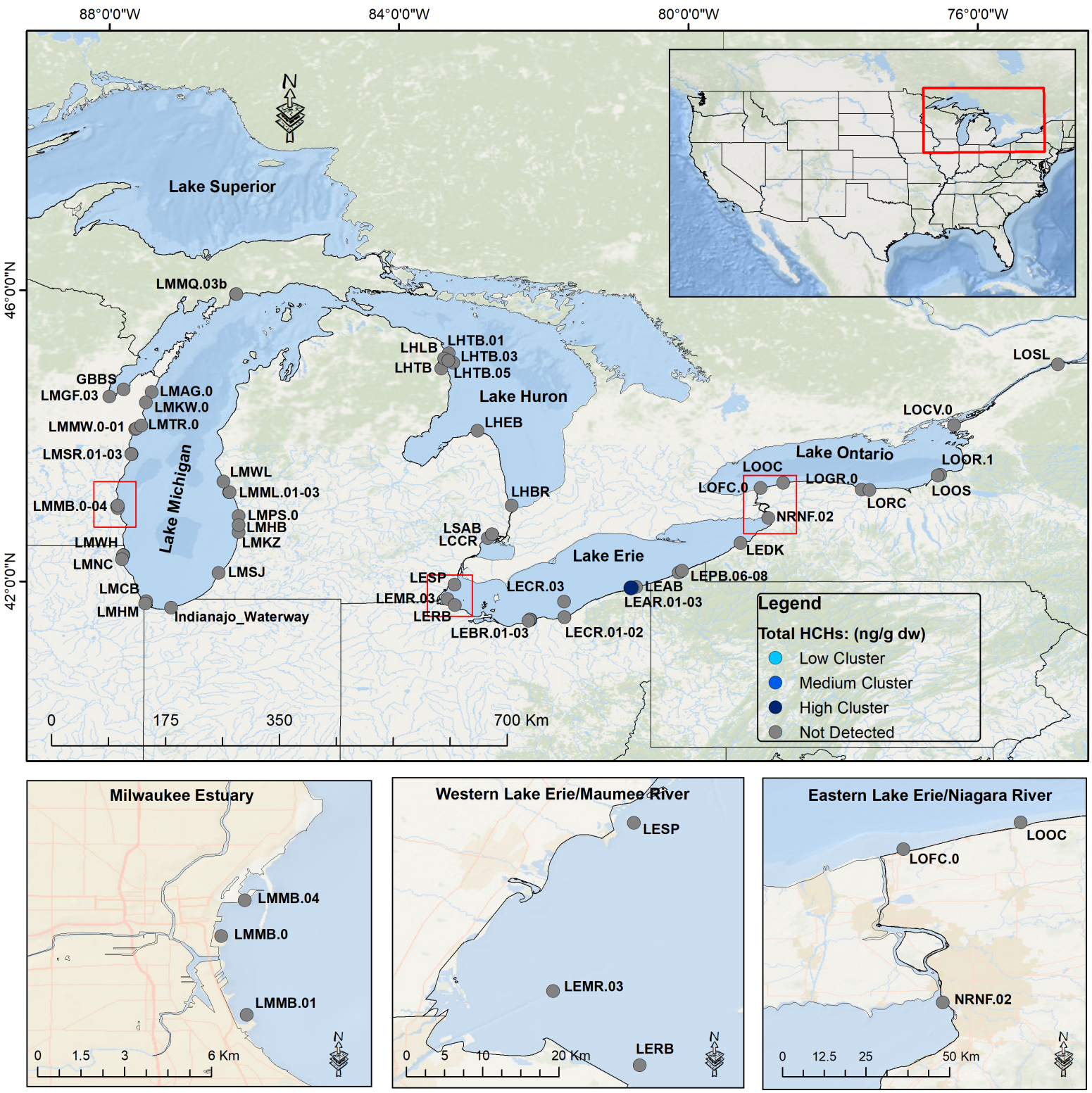


Figure 60. Total HCH concentration sums (ng/g dw) measured in Great Lakes mussel tissue during 2022. Some mussel sampling locations are shown to overlap due to the size of the circles. Colors represent contamination magnitude and clusters as follows: low ( ), medium ( ), high ( ), and not detected ( ).

Results - Total HCHs

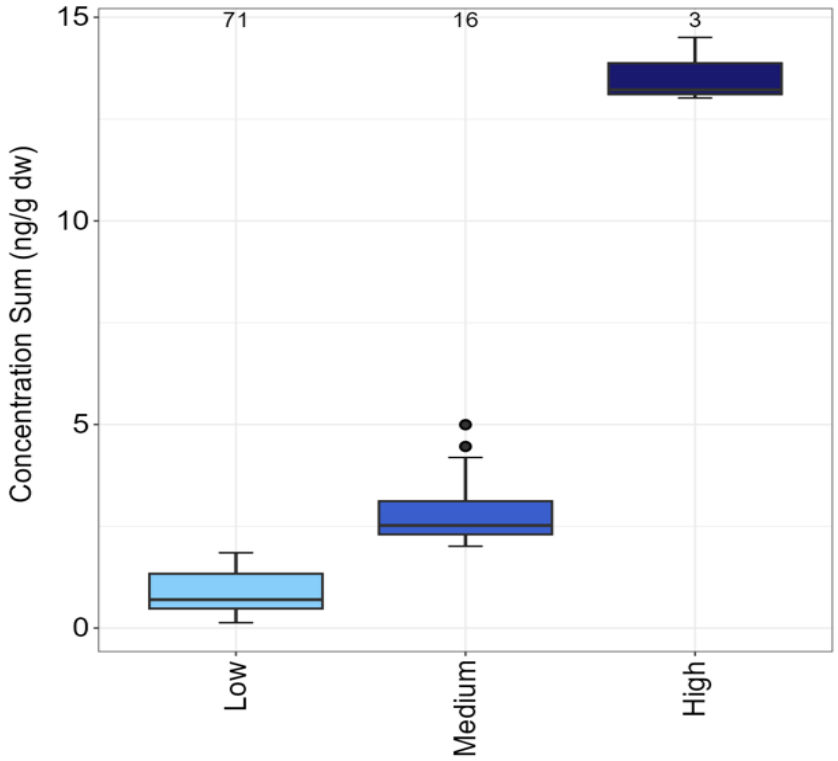


Figure 61. Total HCH concentration sums (ng/g dw) measured in Great Lakes mussel tissue during 1992-2022. Colors represent contamination magnitude as follows: low ( ), medium ( ), and high ( ).

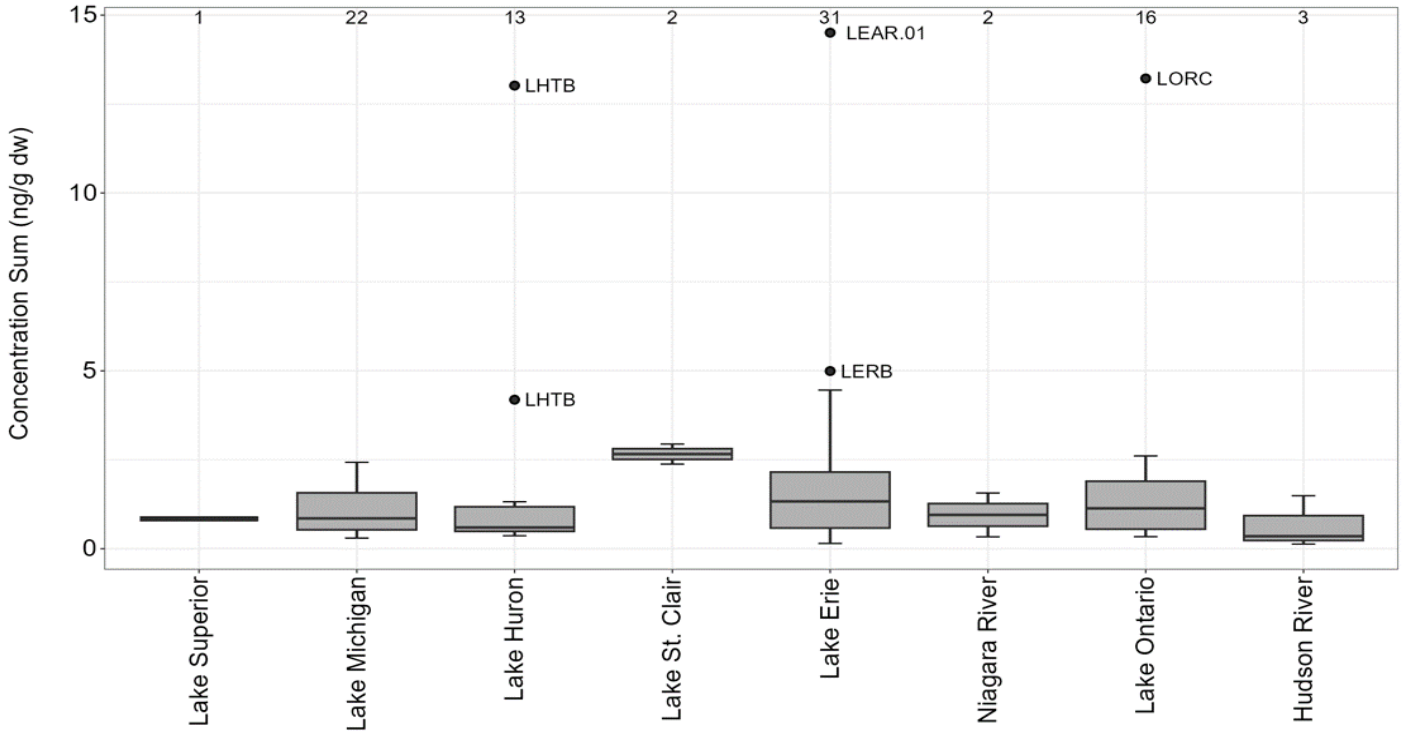


Figure 62. Total HCH concentration sums (ng/g dw) measured in Great Lakes mussels between 1992-2022. Lakes and major rivers including connecting channels sampled during 1992-2022 are listed geographically from west to east across the Great Lakes Basin. The number of HCH samples collected across each lake and major rivers including connecting channels sampled during 1994-2022 is noted at the top of the plot.

12.0 RESULTS - MIREX

12.1 Mirex Chemical Description

For this document, mirex analyzed both in 2022 and historically is a single compound. Mirex was commercially introduced in the US in 1959 for use in pesticide formulations and as an industrial fire retardant (ATSDR, 2020; Kumari and Marathe, 2024). Mirex was used in rubber, plastic, paints, paper, and electrical goods. In the 1960s, mirex (a chlorinated insecticide) was commonly used to control imported fire ants in southern States. Mirex was banned in the US in 1978, except for use on pineapples, until stock on hand were exhausted (ATSDR, 2020; Zhou et al., 2018).

Exposure to mirex typically occurs through dermal contact with contaminated soil and ingestion of local wildlife or contaminated food, as mirex bioaccumulates in organisms and adheres to soil particles, which can contaminate crops (ATSDR, 2020; Kumari and Marathe, 2024). Due to their limited solubility in water and nonvolatile nature, exposure through drinking water and inhalation is unlikely. The primary consequences of exposure to mirex in animals can be seen in the liver, kidneys, selected developmental endpoints, and the thyroid. Specific negative consequences include decreased glycogen storage in the liver, increased glomerulosclerosis and proteinuria in the kidneys, ocular lesions in newborns, and increases in cystic follicles in the thyroid. Decreased fertility, testicular atrophy, reproductive failure, and marked developmental toxicity have been observed following exposure to mirex. Mirex has also been classified as a carcinogen to animals (ATSDR, 2020).

Mirex atmospheric transport is unlikely due to its hydrophobic nature and low vapor pressure (ATSDR, 2020). Conversely, mirex will bind tightly to soil, so will be highly immobile (half-life of up to 10 years; Kumari and Marathe, 2024). Therefore, mirex residue mostly enters surface waters as a result of soil runoff. In water, mirex will then likely bind to dissolved organic matter. Mirex has been shown to bioaccumulate in animals and terrestrial plants. Mirex breaks down extremely slowly in the environment, with the primary process of degradation being photolysis in water and photolysis or anaerobic biodegradation in soil. Any detected concentrations are most likely due to residual chemicals rather than new sources (ATSDR, 2020).

12.2 Magnitude and Distribution of Mirex in Mussel Tissue in 2022

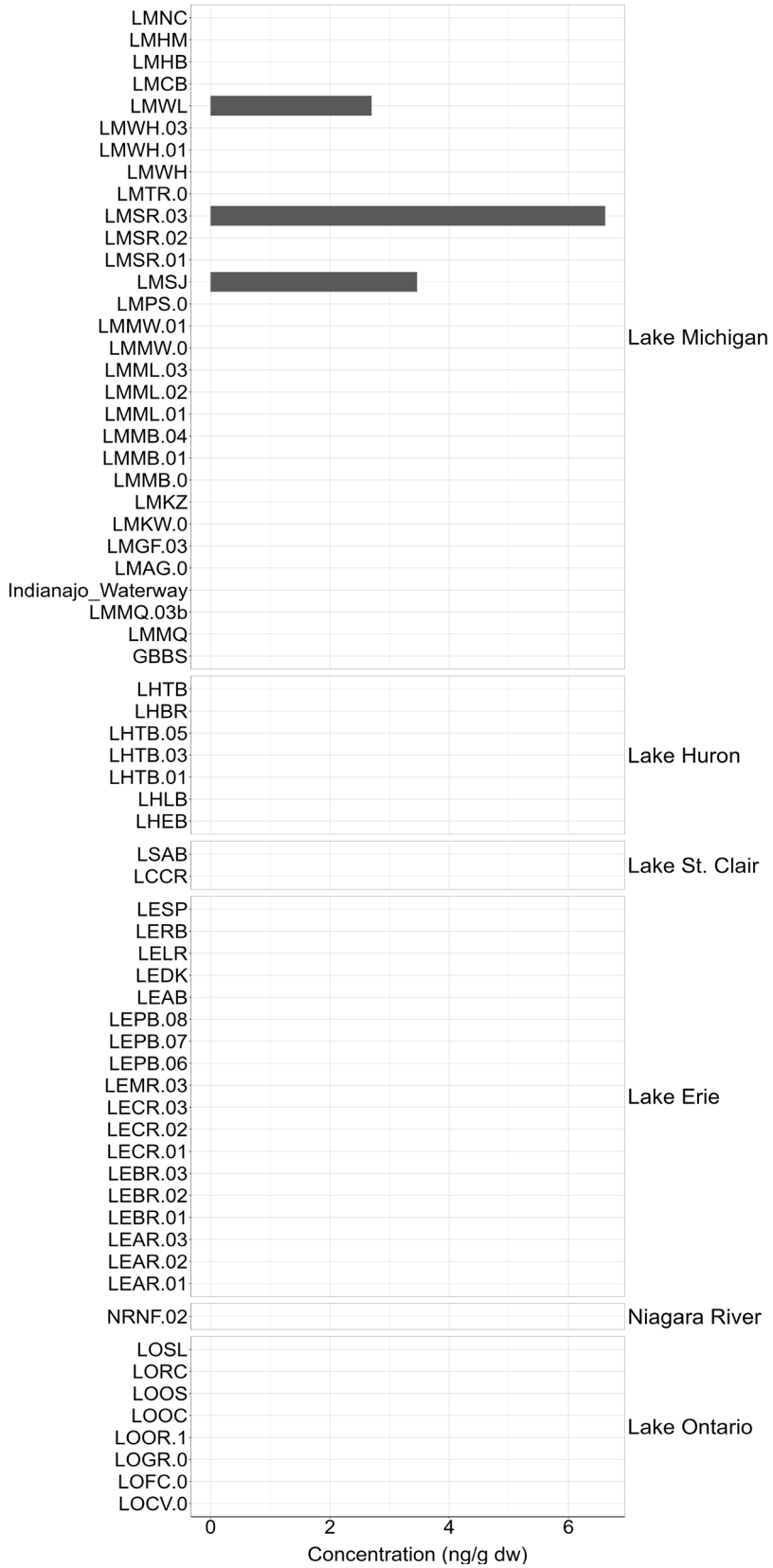


Figure 63. Bar graph showing the magnitude of mirex concentrations measured in mussel tissue in the Great Lakes Basin during the 2022 sampling period. Lakes are listed geographically from west to east across the Great Lakes Basin.

12.3 Historical Context of Mirex Magnitude and Distribution in Mussel Tissue

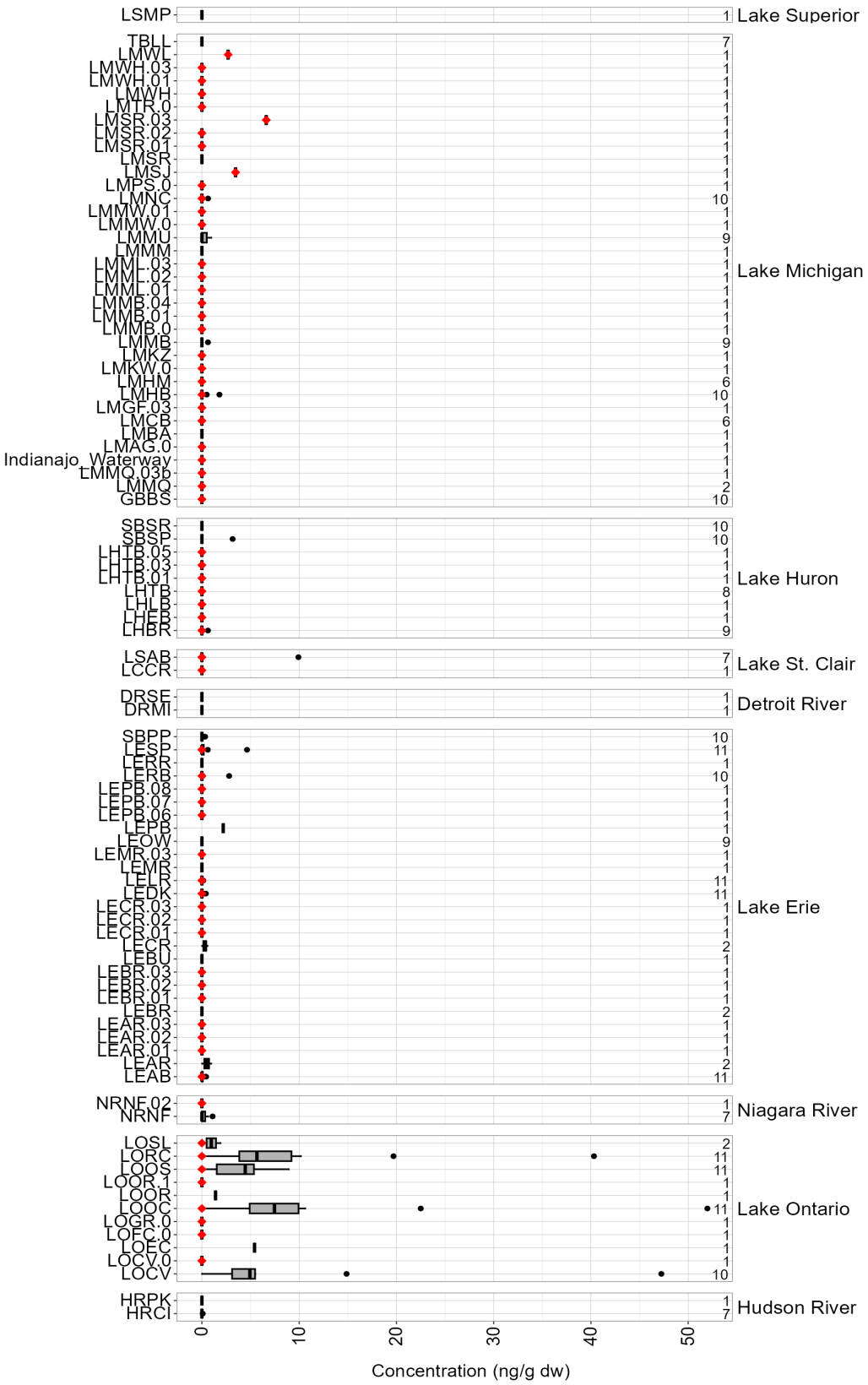


Figure 64. Mirex concentrations (ng/g dw) measured in mussel tissue in 2022 (◆ - red diamonds), compared to boxplots of the historic Mirex concentrations (ng/g dw) for each site (1992-2022) and results of the trend analysis showing significant increasing (▷) or decreasing (◁) contamination trends over time (gray triangles). Additional information is provided in Table 13 and Table A6. Lakes are listed geographically from west to east across the Great Lakes Basin. The number of years each site has been sampled since 1992 is noted at the side (right) of the plot for each site.

Results - Mirex

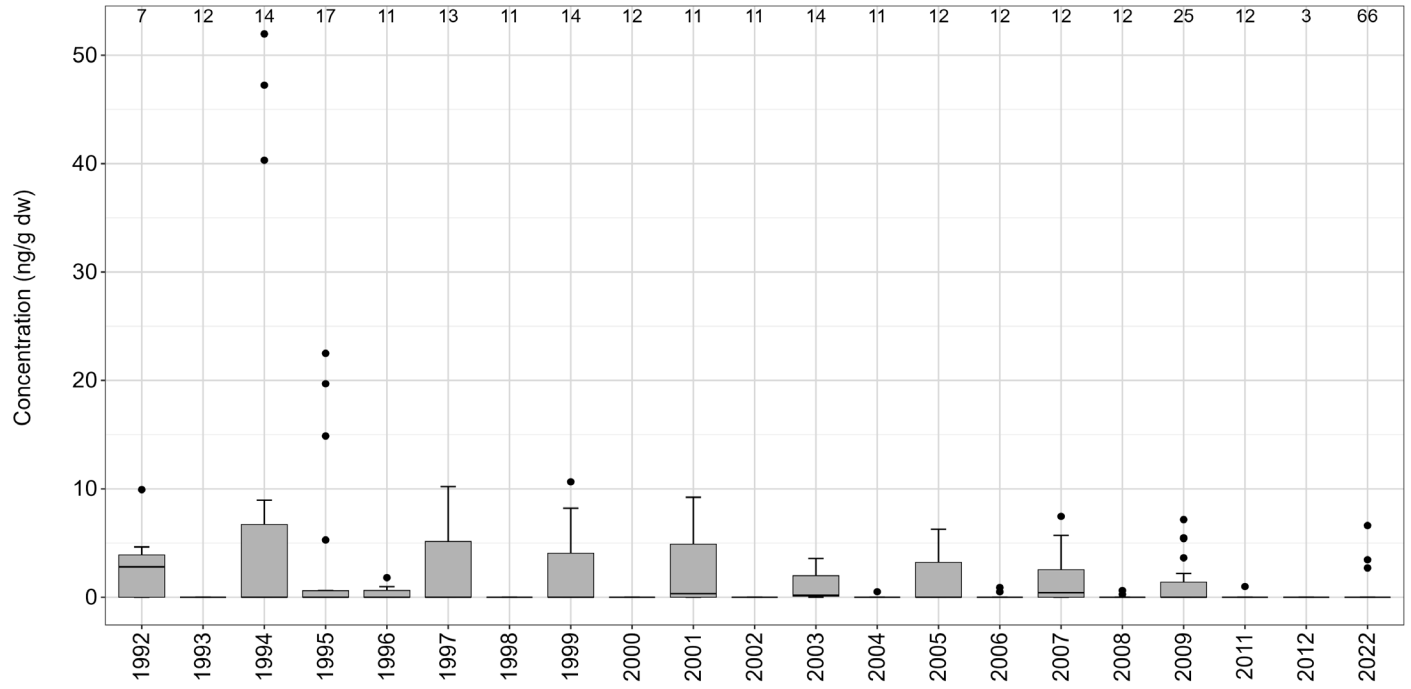


Figure 65. Boxplots representing the historic mirex concentrations (ng/g dw) measured in mussel tissue of the sites analyzed in this study. The number of sites that were sampled each year is noted at the top of the plot.

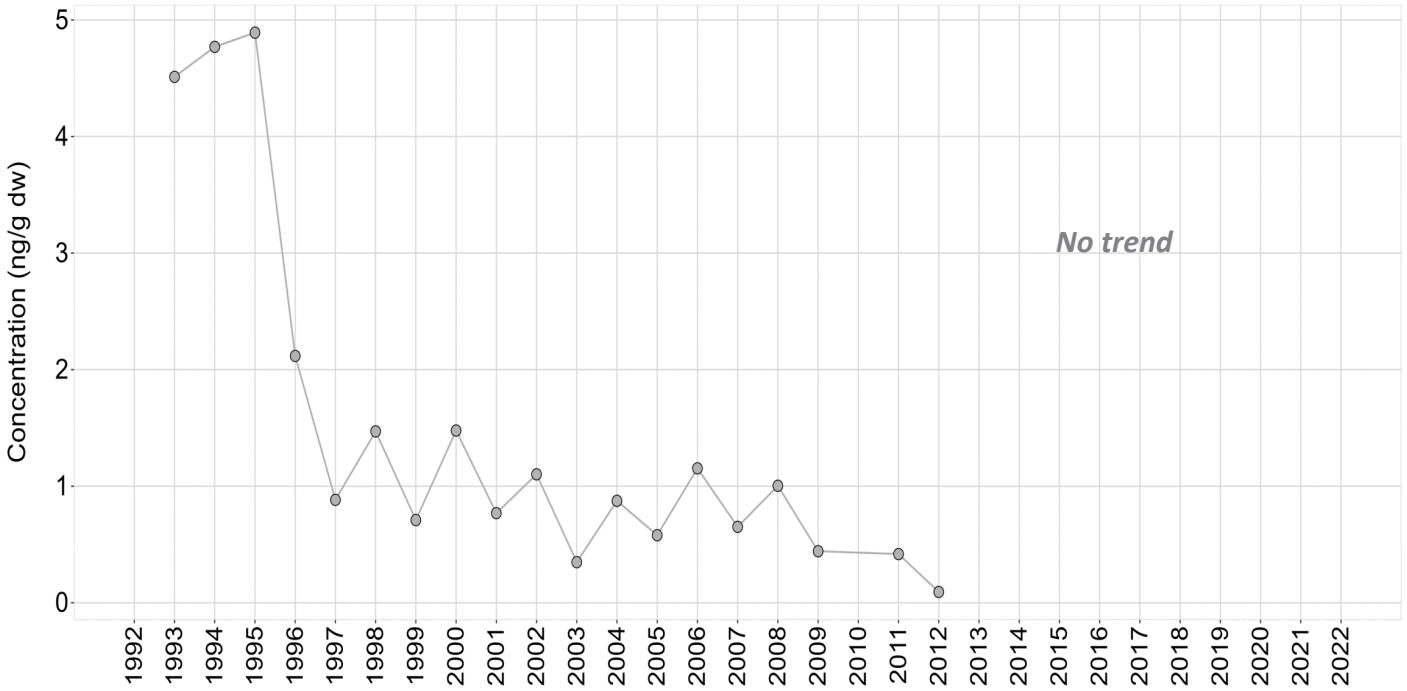


Figure 66. Three-point moving average of the yearly mean mirex concentrations (ng/g dw) measured in mussel tissue at sites analyzed in this study.

12.4 Mirex Summary

2022 Dreissenid Mussel Tissue Results (1 compound):

- Mirex was detected in dreissenid mussels at three sites (LMSJ, LMSR.03, and LMWL) surveyed in the Great Lakes in 2022 (Figure 63; Figure 67).
- Mirex concentration results (Figure 63):
  - Range: 0.00 – 6.61 (ng/g dw)
  - Minimum concentration sum was detected at LMWL (2.69 ng/g dw)
  - Maximum concentration sum was detected at LMSR.03 (6.61 ng/g dw)
  - Median: 3.46 (ng/g dw)
  - Mean ± SD: 4.26 ± 2.08 (ng/g dw)

Historical Context for Dreissenid Mussel Tissue (1 compound):

- 5% (3/66) of sites were above their historic median concentrations in 2022 (Figure 64).
- No significant regional temporal trend of mirex concentrations were observed across the Great Lakes Basin sampling locations between 1992-2022 ($\alpha = 0.05$; Figure 65; Figure 66).
- 0 sites assessed across the Great Lakes Basin showed significant decreasing temporal trends of mirex concentrations ($\alpha = 0.05$; Figure 64).
- 0 sites assessed across the Great Lakes Basin showed significant increasing temporal trends of mirex concentrations ($\alpha = 0.05$; Figure 64).
- The percentage of sites sampled in 2022 grouped into each cluster based on historic Great Lakes data (Figure 68):
  - 98.5% (65/66) of sites in the low cluster (0.00 – 4.72 ng/g dw)
  - 1.5% (1/66) of sites in the medium cluster (4.73 – 22.5 ng/g dw)
  - 0% of sites in the high cluster (22.6 – 51.9 ng/g d

General Observations:

- Mirex was only detected in dreissenid mussel tissue at three sites (LMSJ, LMSR.03, and LMWL) in the Great Lakes in 2022.
  - Mirex was only detected in dreissenid mussels from sites sampled in Lake Michigan.
  - Highest mirex contamination detected in dreissenid mussels were from sites sampled in Sheboygan River (LMSR.03).
  - Overall, mirex concentration (concentration range: 0.00 – 6.61 ng/g dw) was generally low, or not detected in dreissenid mussel tissue at sites surveyed in the Great Lakes in 2022 (Figure 64; Figure 65; Figure 67).
- Historically, highest mirex concentrations were detected in mussels from sites sampled in Hudson River (0.081 ng/g dw), Niagara River (1.09 ng/g dw), Lake Huron (3.16 ng/g dw), Lake Erie (4.64 ng/g dw), Lake Michigan (6.61 ng/g dw), Lake St. Clair (9.92 ng/g dw), and Lake Ontario (51.9 ng/g dw; Figure 64; Figure 69).
- The continued presence of mirex in mussel tissue suggests continued input into he Great Lakes aquatic environment from likely sources such as resuspended sediments, their historical use and links to non-point sources associated with mirex use as pesticides (Apeti and Lauenstein, 2006; Kumari and Marathe, 2024).
- Mirex concentrations were below the US EPA recommended screening values for both recreational and subsistence fishers (Table A2; Table A3).

Results - Mirex

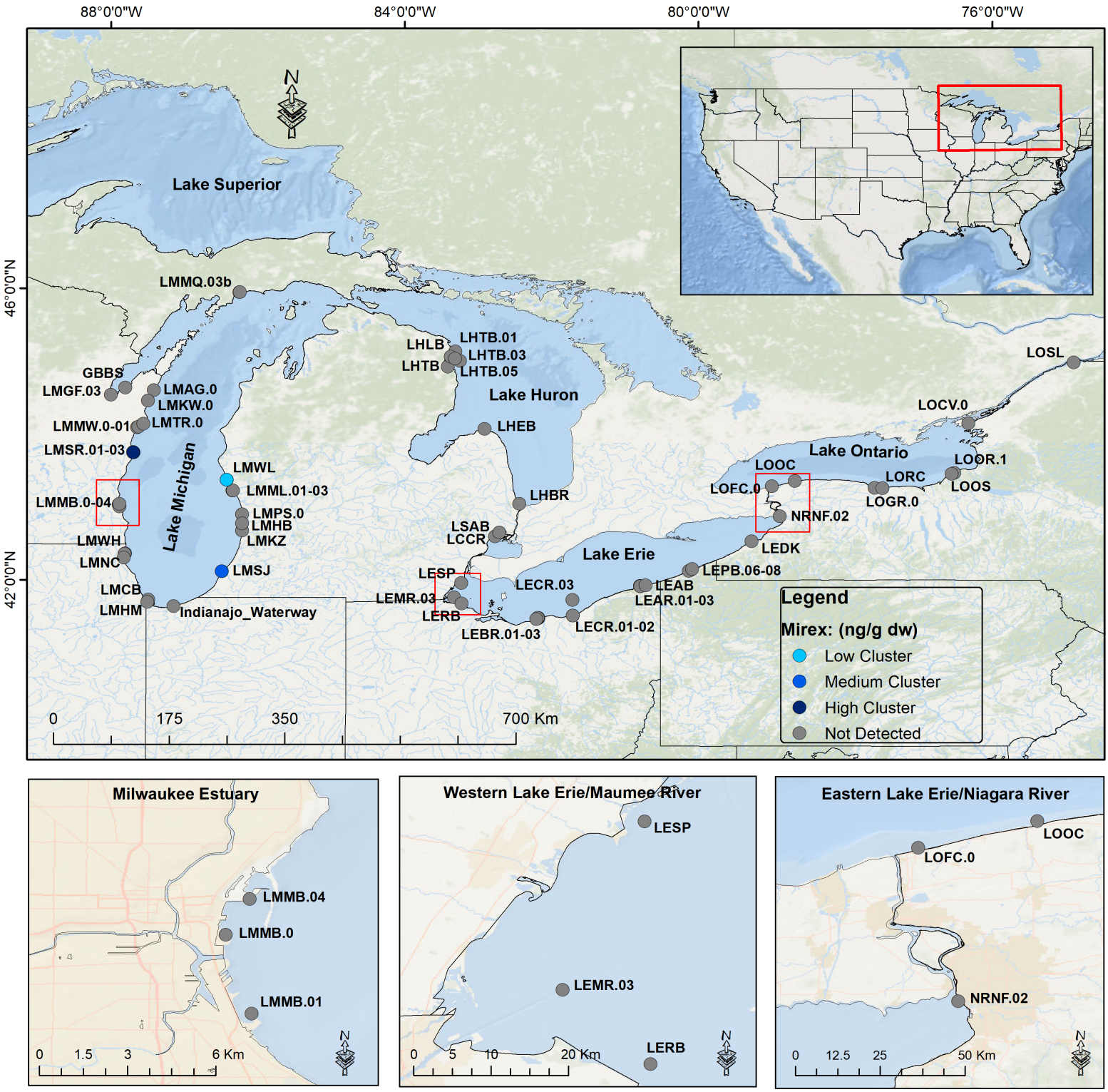


Figure 67. Mirex concentrations (ng/g dw) measured in Great Lakes mussel tissue during 2022. Some mussel sampling locations are shown to overlap due to the size of the circles. Colors represent contamination magnitude and clusters as follows: low ( ), medium ( ), high ( ), and not detected ( ).

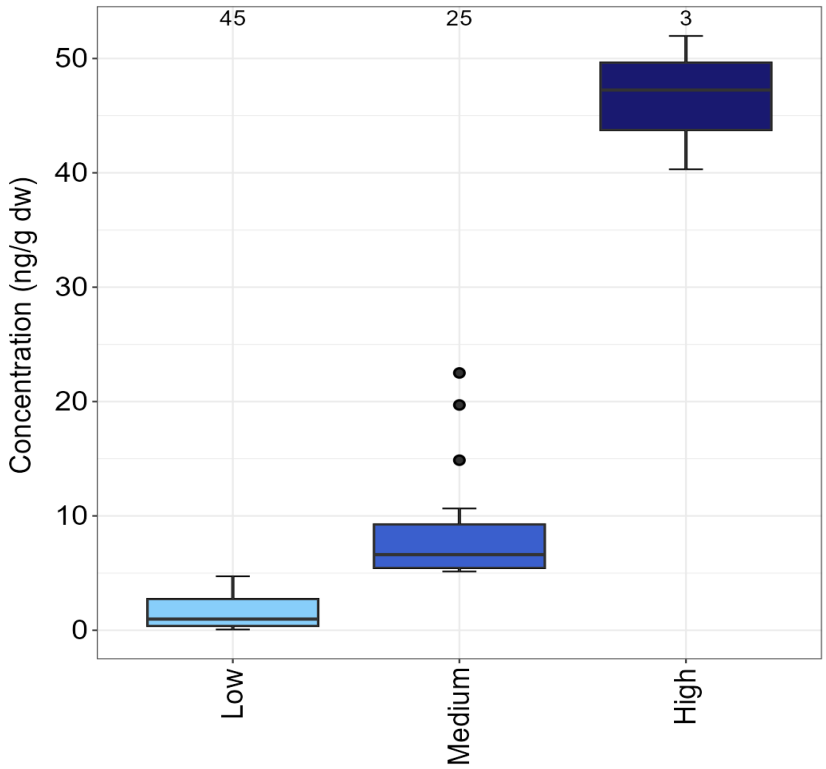


Figure 68. Mirex concentrations (ng/g dw) measured in Great Lakes mussel tissue during 1992-2022. Colors represent contamination magnitude as follows: low ( ), medium ( ), and high ( ).

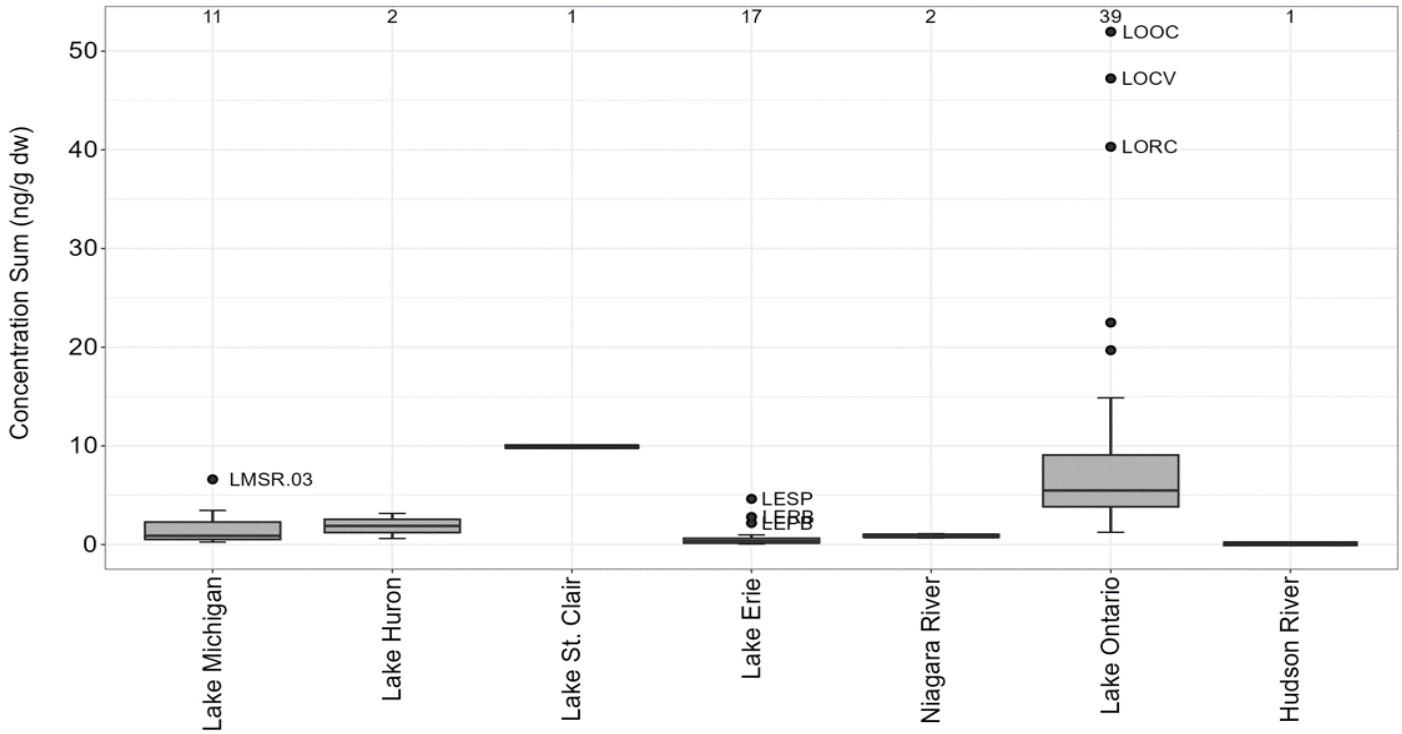


Figure 69. Mirex concentrations (ng/g dw) measured in Great Lakes mussels between 1992-2022. Lakes and major rivers including connecting channels sampled during 1992-2022 are listed geographically from west to east across the Great Lakes Basin. The number of mirex samples collected across each lake and major rivers including connecting channels sampled during 1992-2022 is noted at the top of the plot.

13.0 RESULTS - TOTAL PAHs

13.1 PAHs Chemical Description

For this document, Total PAHs analyzed in 2022 is the sum of 64 compounds, and Total PAHs analyzed historically is the sum of 39 compounds (Table 9). PAHs (polycyclic aromatic hydrocarbons) are formed from the fusion of benzene rings during the incomplete combustion of organic materials (Richter and Howard, 2000). PAHs are found in creosote, soot, petroleum, coal, and tar. They are the only organic contaminants measured by the Mussel Watch Program that have natural sources (forest fires and volcanoes) in addition to anthropogenic sources (automobiles emissions, home heating, coal fired power plants). PAHs can also enter the aquatic environment by discharge from industrial and wastewater treatments plants (ATSDR, 1995), or can volatilize from an oil spill if they have a small molecular weight.

Made up of hundreds of compounds, PAHs exhibit a wide range of toxicities. While many aquatic organisms, like fish, can metabolize PAHs, marine invertebrates, such as oysters and mussels, are less able to metabolize them efficiently and, as such, can be better indicators of overall environmental exposure (Neff, 1985). The PAH contents of plants and animals may be much higher than PAH contents of soil or water in which they live (ATSDR, 1995). This might be attributed to the lipophilic structure of PAHs which dictates the rate of uptake and transport of these compounds in plants and animals (Şahin, T., Dalğa, S., and Ölmez, M. 2022). Many of the PAHs that bioaccumulate in aquatic and terrestrial organisms are toxic and some, including benzo(a)pyrene, benz(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, dibenzo(a,h)anthracene, and indeno(1,2,3-c,d)pyrene, are likely carcinogens (ATSDR, 1995). Toxic responses to PAHs in aquatic organisms include reproduction inhibition, mutations, liver abnormalities, and mortality. Exposure to aquatic organisms results from oil spills, boat exhaust, and urban runoff. Human exposure to PAHs can result from exposure to smoke from forest fires, automobile exhaust, home heating using wood, grilling, and cigarettes. There is no US FDA-recommended safety level for PAHs in fish and fish products; however, the US EPA Recreational and Subsistence Fishery Screening Values are 5.47 ng/g ww and 0.673 ng/g ww, respectively (EPA, 2000).

The fate and transport of PAHs are variable and dependent on the physical properties of each individual compound. Most PAHs strongly associate with particles; larger PAH compounds (high molecular weight) associate to a higher degree with particles relative to smaller PAH compounds (low molecular weight). Smaller compounds predominate in petroleum products whereas larger compounds are associated with combustion (ATSDR, 1995).

Table 9. PAH compounds composing the "Total PAHs" summation for the 2022 Great Lakes survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2022	Analyzed in Historical Data
Acenaphthene	●	●
Acenaphthylene	●	●
Anthracene	●	●
Benz[a]anthracene	●	●
Benzo[a]fluoranthene	●	
Benzo[a]pyrene	●	●
Benzo[b]fluoranthene	●	●
Benzo[b]fluorene		
Benzo[e]pyrene	●	●
Benzo[g,h,i]perylene	●	●
Benzo[k]fluoranthene	●	●
Benzothiophene	●	
Biphenyl	●	●
C1-Benzothiophene	●	
C1-Chrysenes	●	●

Results - Total PAHs

Table 9 (cont). PAH compounds composing the "Total PAHs" summation for the 2022 Great Lakes survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2022	Analyzed in Historical Data
C1-Decalin	●	
C1-Dibenzo[a,h]anthracene	●	
C1-Dibenzothiophenes	●	●
C1-Fluoranthenes_Pyrenes	●	●
C1-Fluorenes	●	●
C1-Naphthalenes	●	●
C1-Naphthobenzothiophene	●	
C1-Phenanthrenes_Anthracenes	●	●
C2-Benzothiophene	●	
C2-Chrysenes	●	●
C2-Decalin	●	
C2-Dibenzo[a,h]anthracene	●	
C2-Dibenzothiophenes	●	●
C2-Fluoranthenes_Pyrenes	●	
C2-Fluorenes	●	●
C2-Naphthalenes	●	●
C2-Naphthobenzothiophene	●	
C2-Phenanthrenes_Anthracenes	●	●
C3-Benzothiophene	●	
C3-Chrysenes	●	●
C3-Decalin	●	
C3-Dibenzo[a,h]anthracene	●	
C3-Dibenzothiophenes	●	●
C3-Fluoranthenes_Pyrenes	●	
C3-Fluorenes	●	●

Table 9 (cont). PAH compounds composing the "Total PAHs" summation for the 2022 Great Lakes survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2022	Analyzed in Historical Data
C3-Naphthalenes	●	●
C3-Naphthobenzothiophene	●	
C3-Phenanthrenes_Anthracenes	●	●
C4-Benzothiophenes	●	
C4-Chrysenes	●	●
C4-Decalin	●	
C4-Dibenzothiophenes	●	
C4-Fluoranthenes_Pyrenes	●	
C4-Naphthalenes	●	●
C4-Naphthobenzothiophenes	●	
C4-Phenanthrenes_Anthracenes	●	●
Carbazole	●	
Chrysene	●	●
Decalin	●	
Dibenzo[a,h]anthracene	●	●
Dibenzofuran	●	
Dibenzothiophene	●	●
Fluoranthene	●	●
Fluorene	●	●
Indeno[1,2,3-c,d]pyrene	●	●
Naphthalene	●	●
Naphthobenzothiophene	●	
Perylene	●	●
Phenanthrene	●	●
Pyrene	●	●

13.2 Magnitude and Distribution of PAHs in Mussel Tissue in 2022

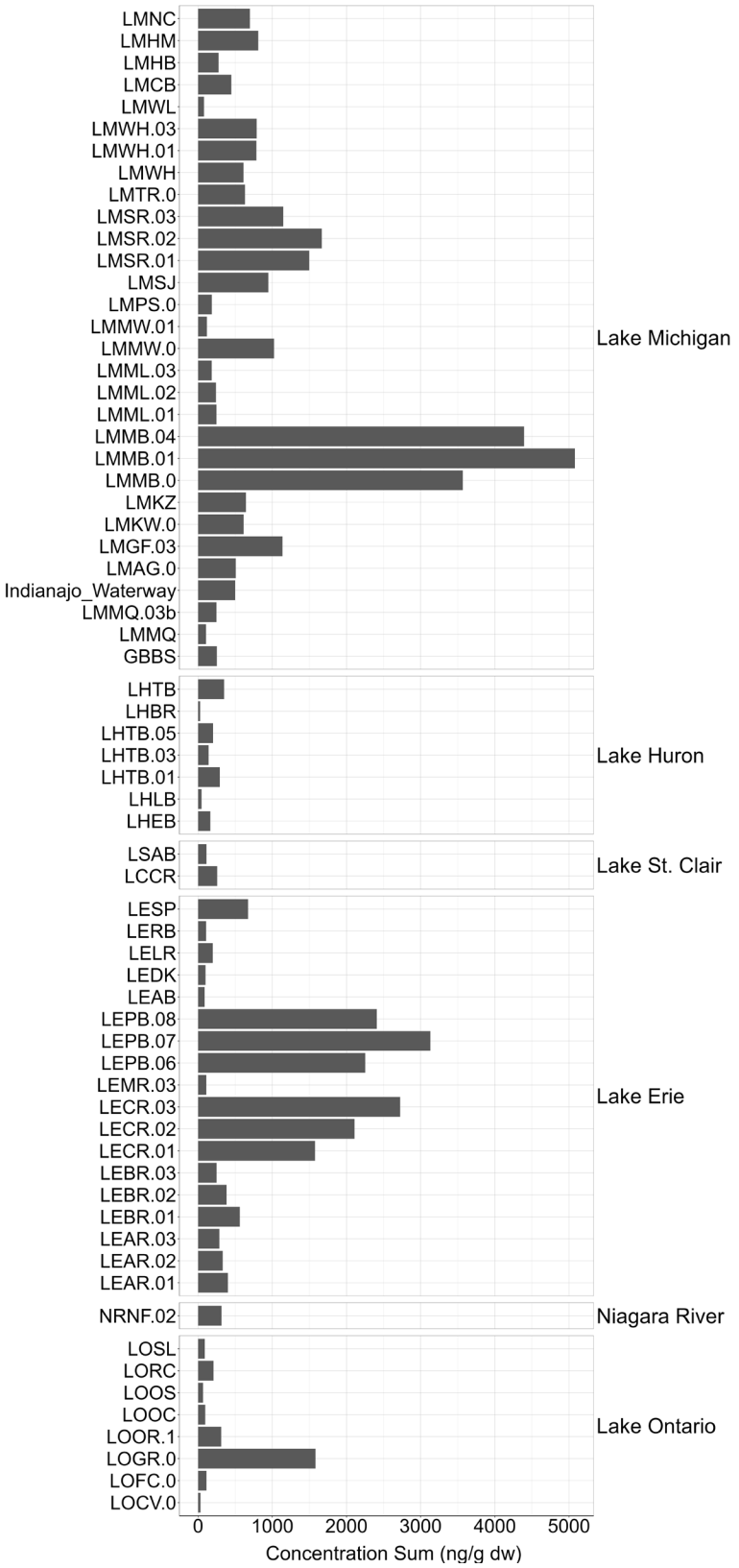


Figure 70. Bar graph showing the magnitude of Total PAH concentration sums detected in mussel tissue in the Great Lakes Basin during the 2022 sampling period. Lakes are listed geographically from west to east across the Great Lakes Basin.

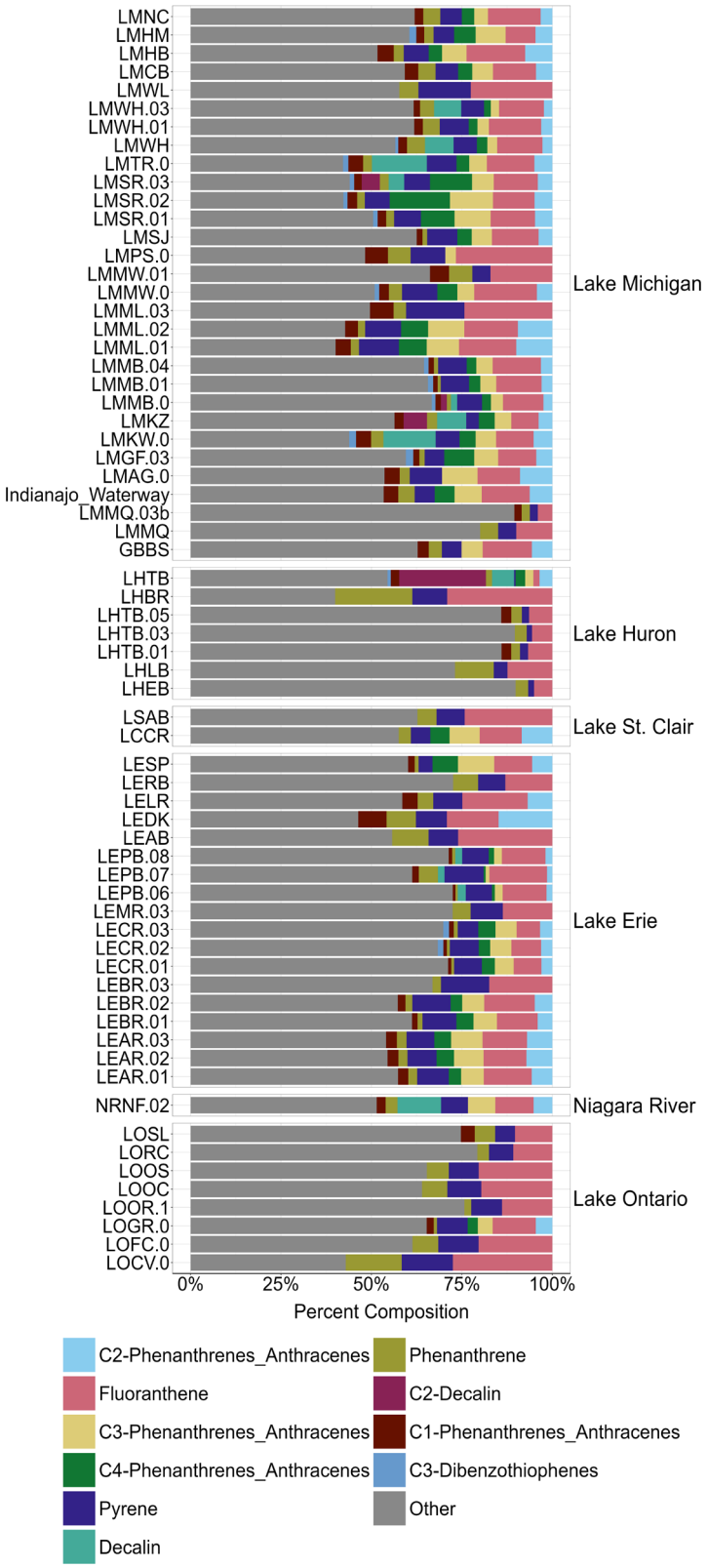


Figure 71. Percent composition of individual PAH contaminants in Total PAH per site in the Great Lakes Basin during the 2022 sampling period. Lakes are listed geographically from west to east across the Great Lakes Basin. The 10 most prevalent PAH compounds were chosen to be included in this stacked boxplot, the remaining compounds were grouped together as "Other".

13.3 Historical Context of PAHs Magnitude and Distribution in Mussel Tissue

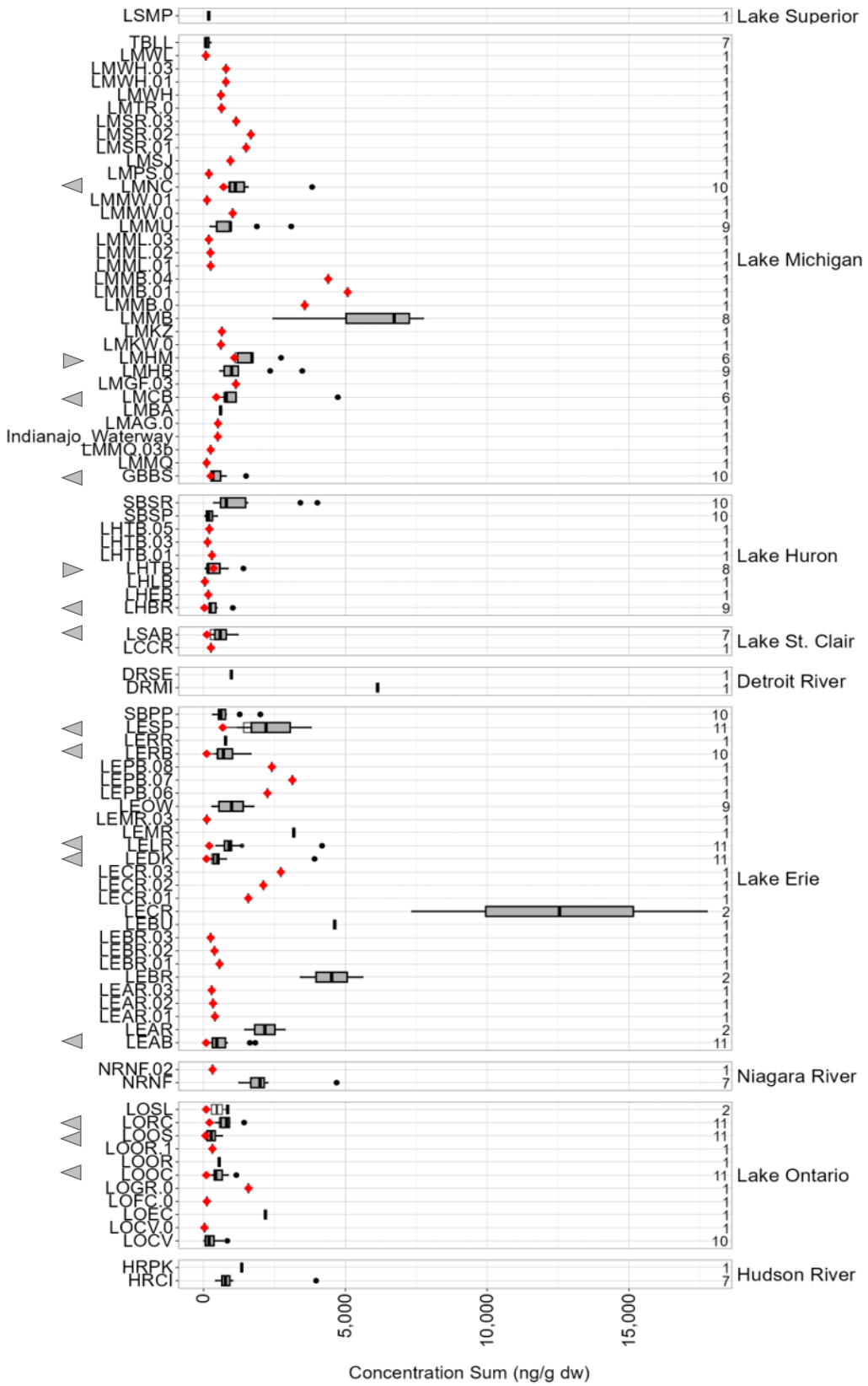


Figure 72. Total PAHs concentration sums (ng/g dw) measured in mussel tissue in 2022 (◆ - red diamonds), compared to boxplots of the historic Total PAH concentration sums for each site (1992-2022) and results of the trend analysis showing significant increasing (▻) or decreasing (◅) contamination trends over time (gray triangles). Sites LEPB and LMMB (outliers) were removed for visualization purposes. Additional information is provided in Table 13 and Table A6. Lakes are listed geographically from west to east across the Great Lakes Basin. The number of years each site has been sampled since 1992 is noted at the side (right) of the plot for each site.

Results - Total PAHs

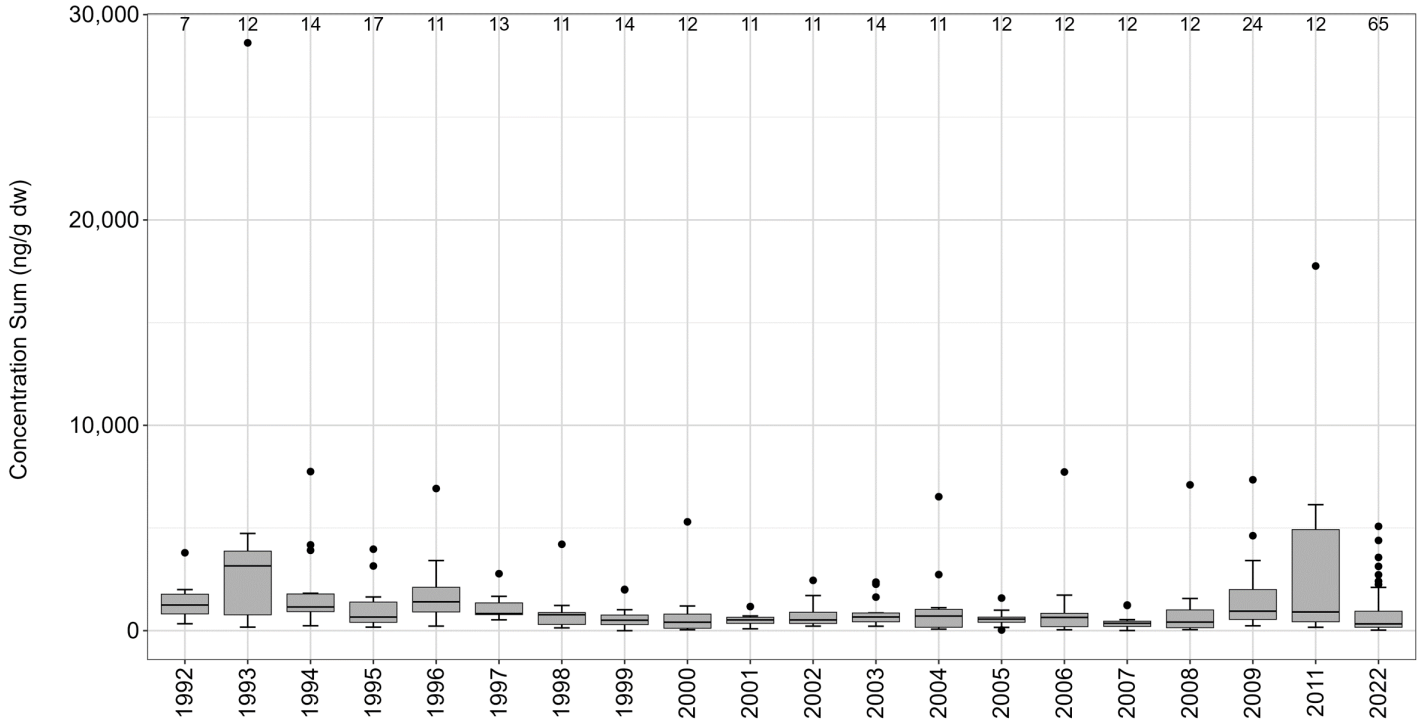


Figure 73. Boxplots representing the historic Total PAH concentration sums (ng/g dw) in mussel tissue of the sites analyzed in this study. The number of sites that were sampled each year is noted at the top of the plot. Site LEPB in 2009 was a high outlier (103,905 ng/g dw), so it was removed to aid in visualization.

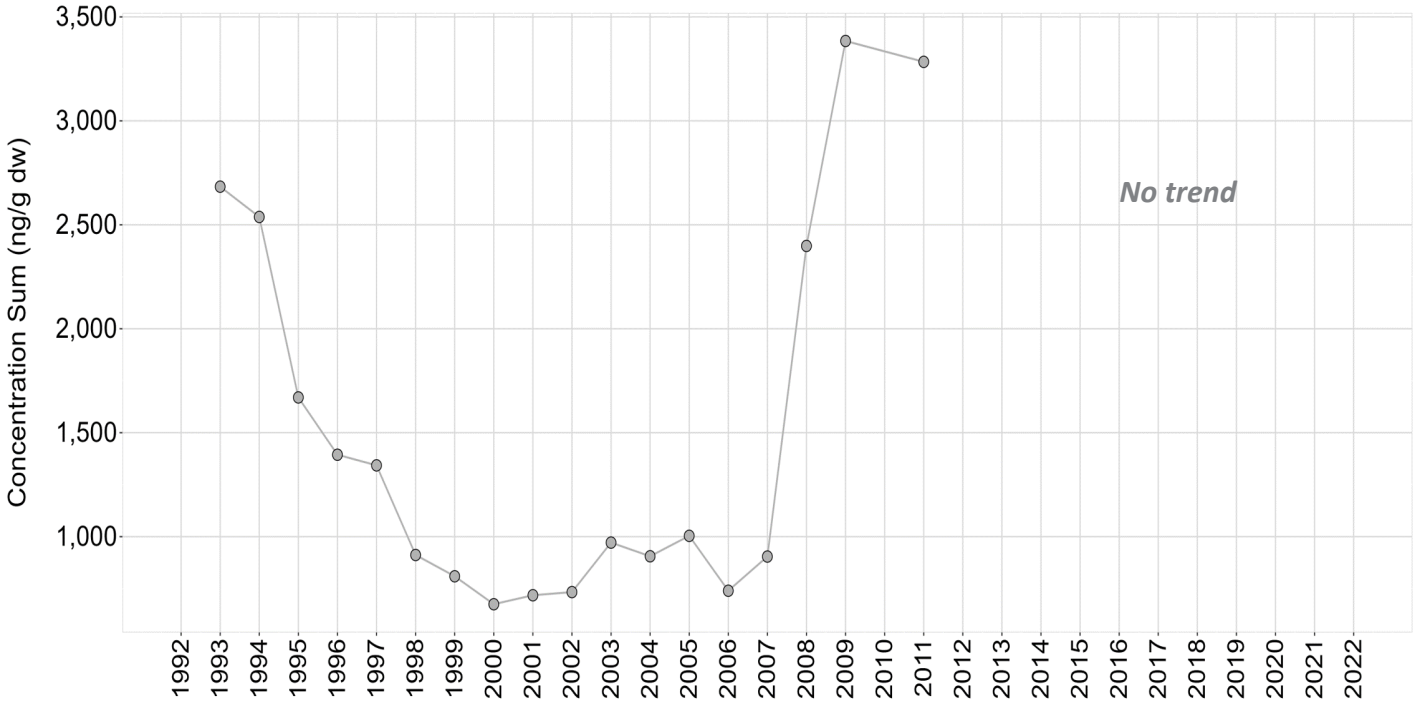


Figure 74. Three-point moving average of the yearly mean Total PAH concentration sums (ng/g dw) in mussel tissue in the sites analyzed in this study.

13.4 PAHs Summary

2022 Dreissenid Mussel Tissue Results (sum of 64 compounds):

- Total PAHs were detected in dreissenid mussels at 100% (66/66) of the sites surveyed in the Great Lakes in 2022 (Figure 70; Figure 75).
- Total PAHs concentration summary statistics (Figure 70; Figure 71):
  - Range: 0.00 – 5,080.0 ng/g dw
  - Minimum concentration sum was detected at site LHBR (27.1 ng/g dw)
  - Maximum concentration sum was detected at site LMMB.01 (5,080.0 ng/g dw)
  - Median: 322.9 ng/g dw
  - Mean ± SD: 787.9 ± 1,066.0 ng/g dw

Historical Context for Dreissenid Mussel Tissue (sum of 39 compounds):

- 0% of sites were above their historic median concentrations in 2022 (Figure 72).
- No significant regional temporal trend of Total PAHs concentration sums was observed across the Great Lakes Basin sampling locations between 1992-2022 ($\alpha = 0.05$; Figure 73; Figure 74).
- 2 sites (LMHM and LHTB) assessed across the Great Lakes Basin showed increasing temporal trends of Total PAH concentration sums ($\alpha = 0.05$; Figure 72; Table 13; Table A6).
- 13 sites (GBBS, LMCB, LMNC, LHBR, LSAB, LEAB, LEDK, LELR, LERB, LESP, LOOC, LOOS, and LORC) assessed across the Great Lakes Basin showed decreasing temporal trends of Total PAH concentration sums ($\alpha = 0.05$; Figure 72; Table 13; Table A6).
- The percentage of sites sampled in 2022 grouped into each cluster based on historic Great Lakes data (Figure 76):
  - 100% of sites in the low cluster (0.00 – 5,616.0 ng/g dw)
  - 0% of sites in the medium cluster (5,616.1 – 17,757.4 ng/g dw)
  - 0% of sites in the high cluster (17,757.5 – 103,905.0 ng/g dw)

General Observations:

- Total PAHs concentration sums (concentration range: 0.00 – 5,080.0 ng/g dw) were generally low in dreissenid mussel tissue at sites surveyed in the Great Lakes in 2022 (Figure 72; Figure 76).
  - There were twelve instances of medium contamination (LMHM, LMMW.0, LMGF.03, LMSR.03, LMSR.01, LECR.01, LOGR.0, LMSR.02, LECR.02, LEPB.06, LEPB.08, and LECR.03) and four instance of high contamination (LEPB.07, LMMB.0, LMMB.04, and LMMB.01) detected in dreissenid mussel tissue at sites surveyed in 2022 (Figure 75).
- Total PAHs were only detected at 15 (GBBS, LMCB, LMHM, LMNC, LHBR, LHTB, LSAB, LEAB, LEDK, LELR, LERB, LESP, LOOC, LOOS, and LORC) of the eighteen long-term MWP sites surveyed in 2022.
- Historically, highest Total PAH concentrations were detected in mussels from sites sampled in Lake Superior (178.5 ng/g dw), Lake St. Clair (1213.9 ng/g dw), Lake Ontario (2180.2 ng/g dw), Hudson River (3967.9 ng/g dw), Lake Huron (4,013.5 ng/g dw), Niagara River (4,691.8 ng/g dw), Detroit River (6,136.2 ng/g dw), Lake Michigan (2,8625.6 ng/g dw), and Lake Erie (103,905 ng/g dw; Figure 72; Figure 77).
- The continued presence of PAH compounds in mussel tissue suggests continued inputs into the Great Lakes aquatic environment from resuspended sediments, and likely anthropogenic emission sources such as industrial activities, mobile (e.g., vehicular exhaust), domestic, and agricultural sources, and active runoff from impervious surfaces (Baldwin et al., 2016; Kimbrough et al., 2021; Patel et al., 2020).
- For sampling locations surveyed in 2022, Total PAH concentration sums at 30 sites exceeded the US EPA screening values for subsistence fishers; while 33 sites exceeded the US EPA recommended screening values for recreational fishers (Table A2; Table A3).

Results - Total PAHs

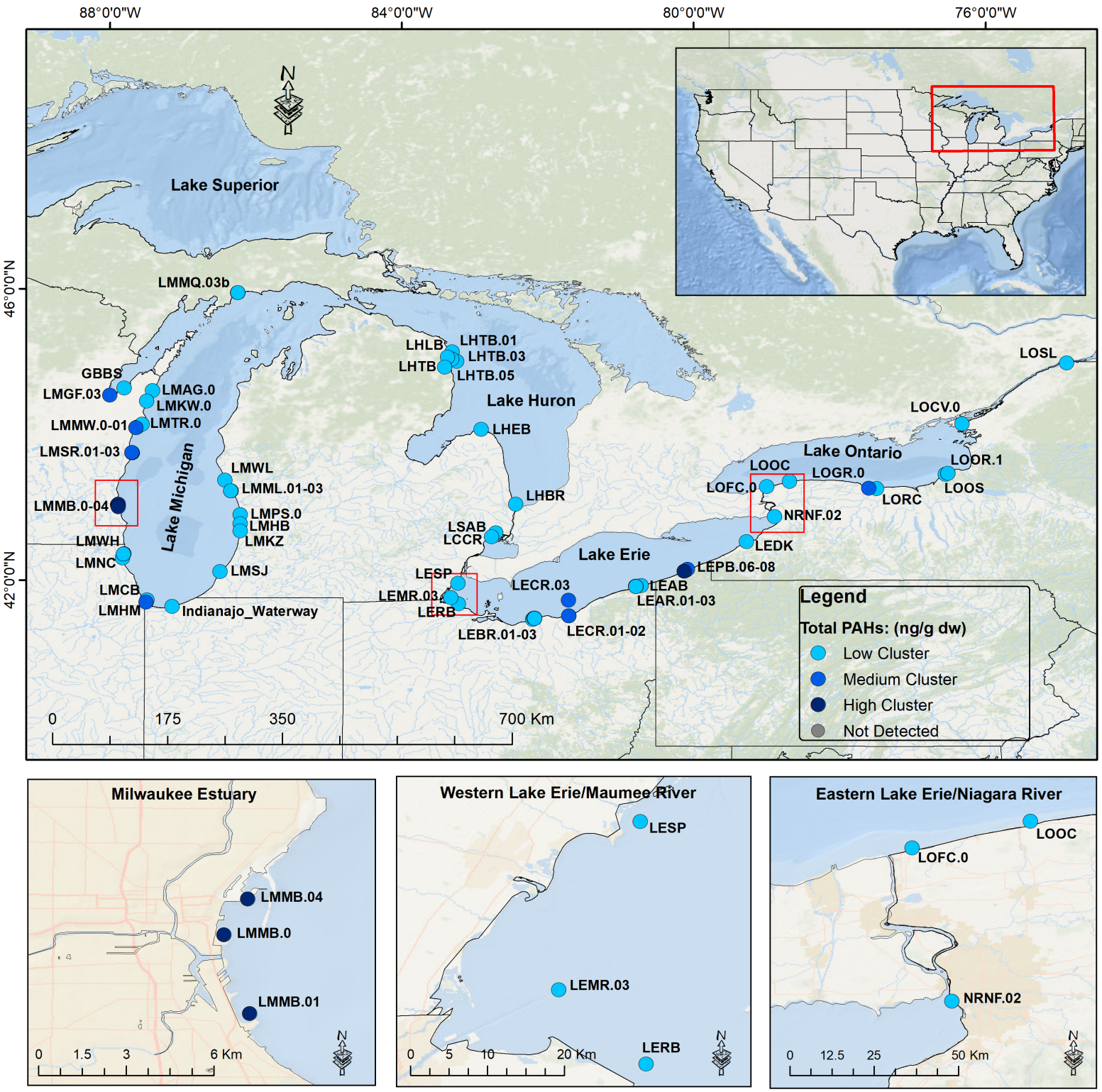


Figure 75. Total PAH concentration sums (ng/g dw) measured in Great Lakes mussel tissue during 2022. Some mussel sampling locations are shown to overlap due to the size of the circles. Colors represent contamination magnitude and clusters as follows: low ( ), medium ( ), high ( ), and not detected ( ).

Results - Total PAHs

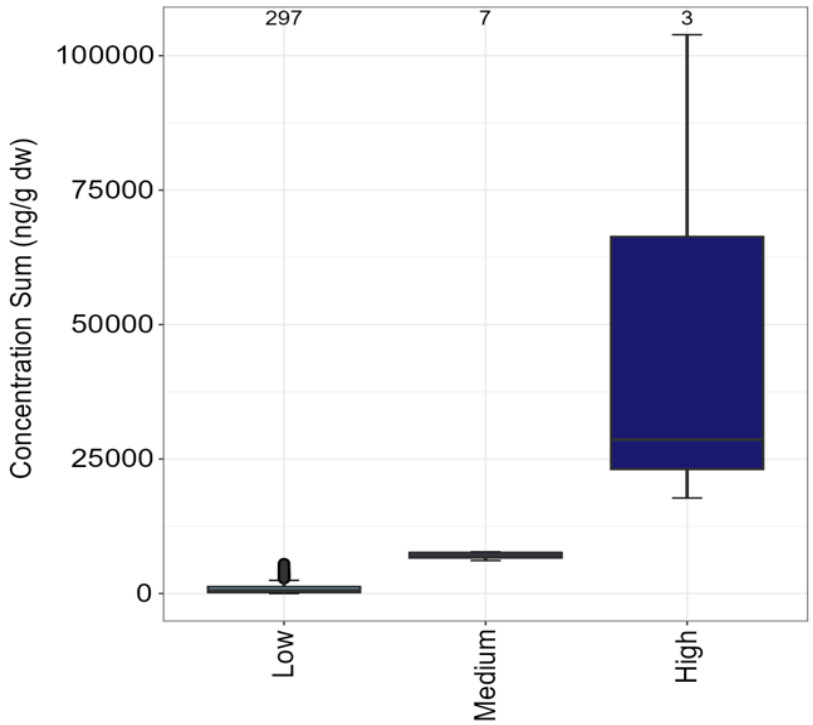


Figure 76. Total PAH concentration sums (ng/g dw) measured in Great Lakes mussel tissue during 1992-2022. Colors represent contamination magnitude as follows: low ( ), medium ( ), and high ( ).

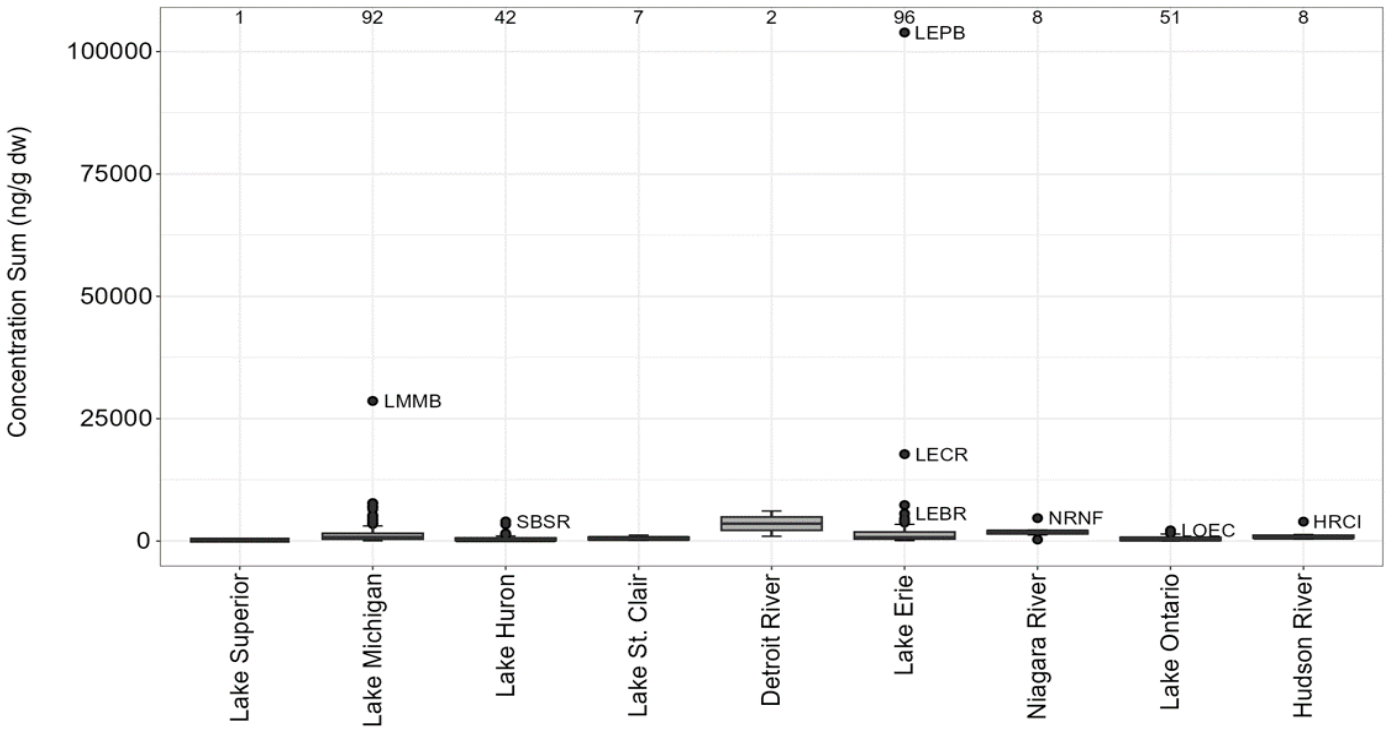


Figure 77. Total PAH concentration sums (ng/g dw) measured in Great Lakes mussels between 1992-2022. Lakes and major rivers including connecting channels sampled during 1992-2022 are listed geographically from west to east across the Great Lakes Basin. The number of PAH samples collected across each lake and major rivers including connecting channels sampled during 1992-2022 is noted at the top of the plot.

14.0 RESULTS - TOTAL PCBs

14.1 PCBs Chemical Description

For this document, Total PCBs analyzed in 2022 is the sum of 156 compounds and Total PCBs analyzed historically is the sum of 18 compounds typically used to assess PCBs (Table 10). Polychlorinated Biphenyls (PCBs) are synthetic organic compounds used in numerous applications, including insulators in electrical transformers and capacitors, hydraulic and heat transfer fluids/coolants, pesticides, adhesives, and plasticizers in paints (ATSDR, 2000; Cornwell et al., 2014). PCBs have a biphenyl ring structure (two benzene rings with a carbon-to-carbon bond) and a varying number (1-10) of chlorine atoms. There are 209 individual PCB compounds or congeners possible. PCBs were manufactured in the US between 1929 and 1977. In the US, a single manufacturer produced all PCBs, and the commercial products were referred to as Aroclors, which are mixtures of PCB congeners. Approximately 65% of PCBs manufactured in the US were used in electrical applications (Eisler and Belisle, 1996). Other applications were lubricants, hydraulic fluids, paints, adhesives, plasticizers, and flame retardants (Kimbrough et al., 2008). Their original environmental introduction was due to improper disposal and leakage. Current pollution sources include volatilization and runoff from landfills, leaks from old electrical equipment, and dredging of contaminated sediments (WHO and IPCS, 1993). PCBS were banned in the US in the 1970s (Zhou et al., 2018).

PCBs have been linked to many health issues, adversely affecting reproduction, growth, metabolism, and survival in animals (Eisler and Belisle, 1996). PCBs are associated with skin ailments, neurological, and immunological responses and at high doses can decrease motor skills and cause memory loss. Other effects can include hepatotoxicity, immunotoxicity, neurotoxicity, low birth weight, and teratogenicity (Eisler and Belisle, 1996). Exposure to PCBs in fish has been linked to reduced growth, reproductive impairment, and vertebral abnormalities (Eisler and Belisle, 1996). PCBs have also been shown to cause cancer in laboratory animals and are likely carcinogens in humans (ATSDR, 2000). The main human exposure route for PCBs is through eating contaminated seafood and meats, which is the reason for many consumption advisories. The US FDA safety level for PCBs in all fish (edible portion) is 2 ppm wet weight, irrespective of which mixture of PCBs is present in the residue (FDA, 2011). The US EPA Recreational and Subsistence Fishery Screening Values are 20 ng/g ww and 2.45 ng/g ww, respectively (EPA, 2000).

PCBs readily accumulate in the tissues of organisms, including filter feeders, fish, and marine mammals. Although no longer manufactured in the US, ecosystem contamination by PCBs is widespread due to their environmental persistence, slow degradation, and tendency to bioaccumulate. Small amounts of PCBs may remain dissolved in water, but the majority adhere to fine sediment and organic particles and can take years to degrade (Gao et al., 2013).

As a note, the combination of PCB compounds comprising individual groups has changed over time. The core 18 compounds that are used for most PCB comparisons were the basis for inclusion in historical analysis, and so any compound in the data that included one of those 18 compounds was included in the summary, even if the grouping changed over time to include other compounds as well (e.g., PCB105 was analyzed until 2016 when the analyte grouping changed to PCB105_127).

Results - Total PCBs

Table 10. PCB compounds composing the "Total PCBs" summation for the 2022 Great Lakes survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2022	Analyzed in Historical Data
PCB1	●	
PCB2	●	
PCB3	●	
PCB4_10	●	
PCB6	●	
PCB7_9	●	
PCB8_5	●	●
PCB11	●	
PCB12	●	
PCB13	●	
PCB14	●	
PCB15	●	
PCB16_32	●	
PCB17	●	
PCB18	●	●
PCB19	●	
PCB22_51	●	
PCB23	●	
PCB24_27	●	
PCB25	●	
PCB26	●	
PCB28_31	●	●
PCB29	●	
PCB30	●	
PCB33_53_20_21	●	
PCB34	●	
PCB35	●	
PCB36	●	
PCB38	●	
PCB39	●	
PCB40_57	●	
PCB42_59_37	●	
PCB43	●	
PCB44	●	●
PCB45	●	

Table 10 (cont). PCB compounds composing the "Total PCBs" summation for the 2022 Great Lakes survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2022	Analyzed in Historical Data
PCB46_69_73	●	
PCB47_48_75	●	
PCB49	●	
PCB50	●	
PCB52	●	●
PCB54	●	
PCB55	●	
PCB56_60	●	
PCB58	●	
PCB61_74	●	
PCB62	●	
PCB63	●	
PCB65	●	
PCB66_80	●	●
PCB67	●	
PCB68_41_64	●	
PCB71	●	
PCB72	●	
PCB76_70	●	
PCB78	●	
PCB79	●	
PCB81	●	
PCB82	●	
PCB85	●	
PCB88	●	
PCB89_113	●	
PCB91	●	
PCB92	●	
PCB94	●	
PCB96_103	●	
PCB97_125_86	●	
PCB99	●	
PCB100	●	
PCB101_84_90	●	●
PCB102_98	●	

Table 10 (cont). PCB compounds composing the "Total PCBs" summation for the 2022 Great Lakes survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2022	Analyzed in Historical Data
PCB104	●	
PCB105_127	●	●
PCB106_107	●	
PCB109	●	
PCB110_77	●	
PCB111_115_87	●	
PCB112	●	
PCB114_131_122	●	
PCB116_117	●	
PCB118_108	●	●
PCB119	●	
PCB120_83	●	
PCB121_93_95	●	
PCB124	●	
PCB128_167	●	●
PCB129_126	●	
PCB130	●	
PCB134_133	●	
PCB135	●	
PCB136_154	●	
PCB138_164_163	●	●
PCB140	●	
PCB141_179	●	
PCB142_146_161	●	
PCB143	●	
PCB144	●	
PCB147	●	
PCB148_145	●	
PCB149_123_139	●	
PCB150	●	
PCB151	●	
PCB152	●	
PCB153_132_168	●	●
PCB155	●	
PCB156_171_202	●	

Table 10 (cont). PCB compounds composing the "Total PCBs" summation for the 2022 Great Lakes survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2022	Analyzed in Historical Data
PCB159	●	
PCB160_158	●	
PCB162	●	
PCB165_131	●	
PCB166	●	
PCB169	●	
PCB170_190	●	●
PCB174	●	
PCB175	●	
PCB176_137	●	
PCB177	●	
PCB180_193	●	●
PCB181	●	
PCB183	●	
PCB184	●	
PCB185	●	
PCB186_178	●	
PCB187_182	●	●
PCB188	●	
PCB189	●	
PCB191	●	
PCB192_172	●	
PCB194	●	
PCB195_208	●	●
PCB196_203	●	
PCB197	●	
PCB199	●	
PCB200	●	
PCB201_157_173	●	
PCB204	●	
PCB205	●	
PCB206	●	●
PCB207	●	
PCB209	●	●

14.2 Magnitude and Distribution of PCBs in Mussel Tissue in 2022

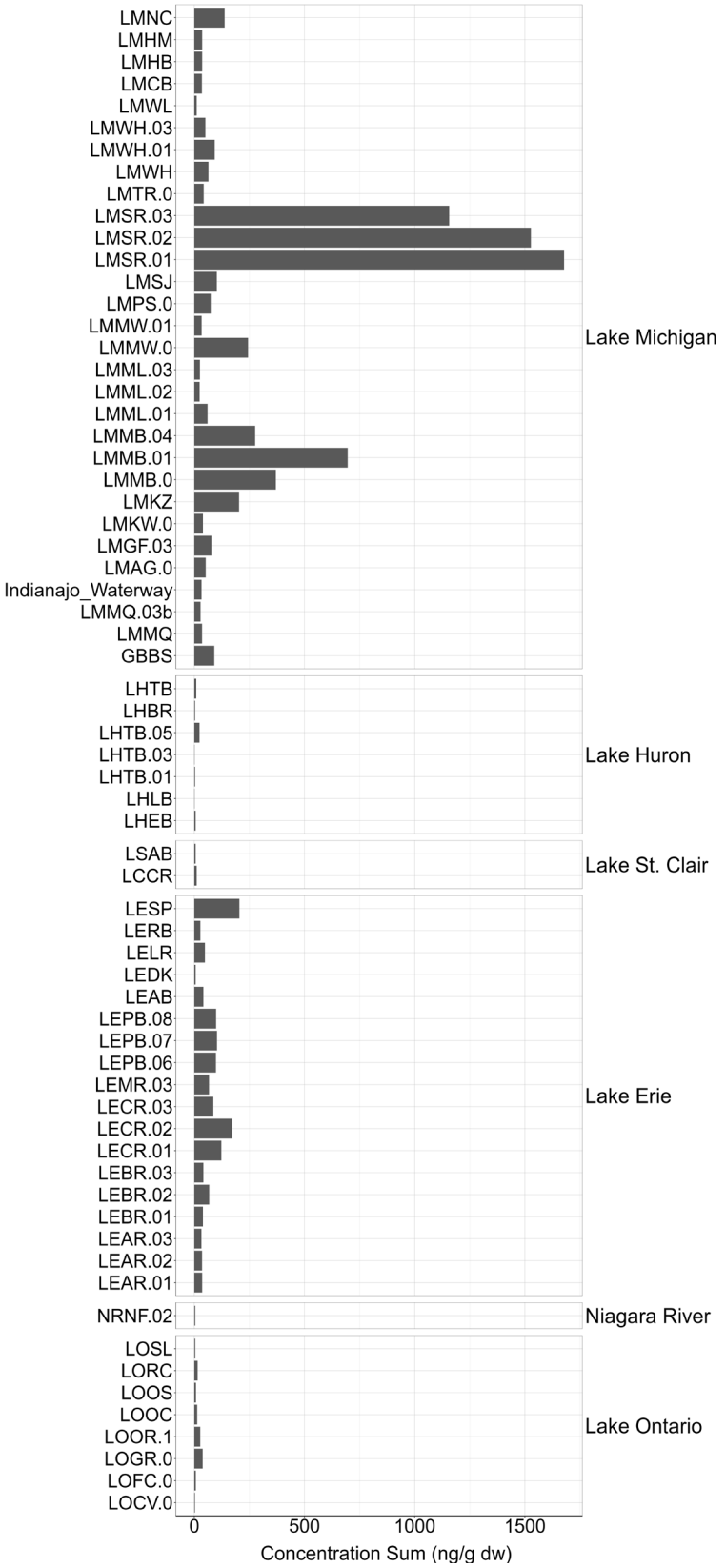


Figure 78. Bar graph showing the magnitude of Total PCB concentration sums detected in mussel tissue in the Great Lakes Basin during the 2022 sampling period. Lakes are listed geographically from west to east across the Great Lakes Basin.

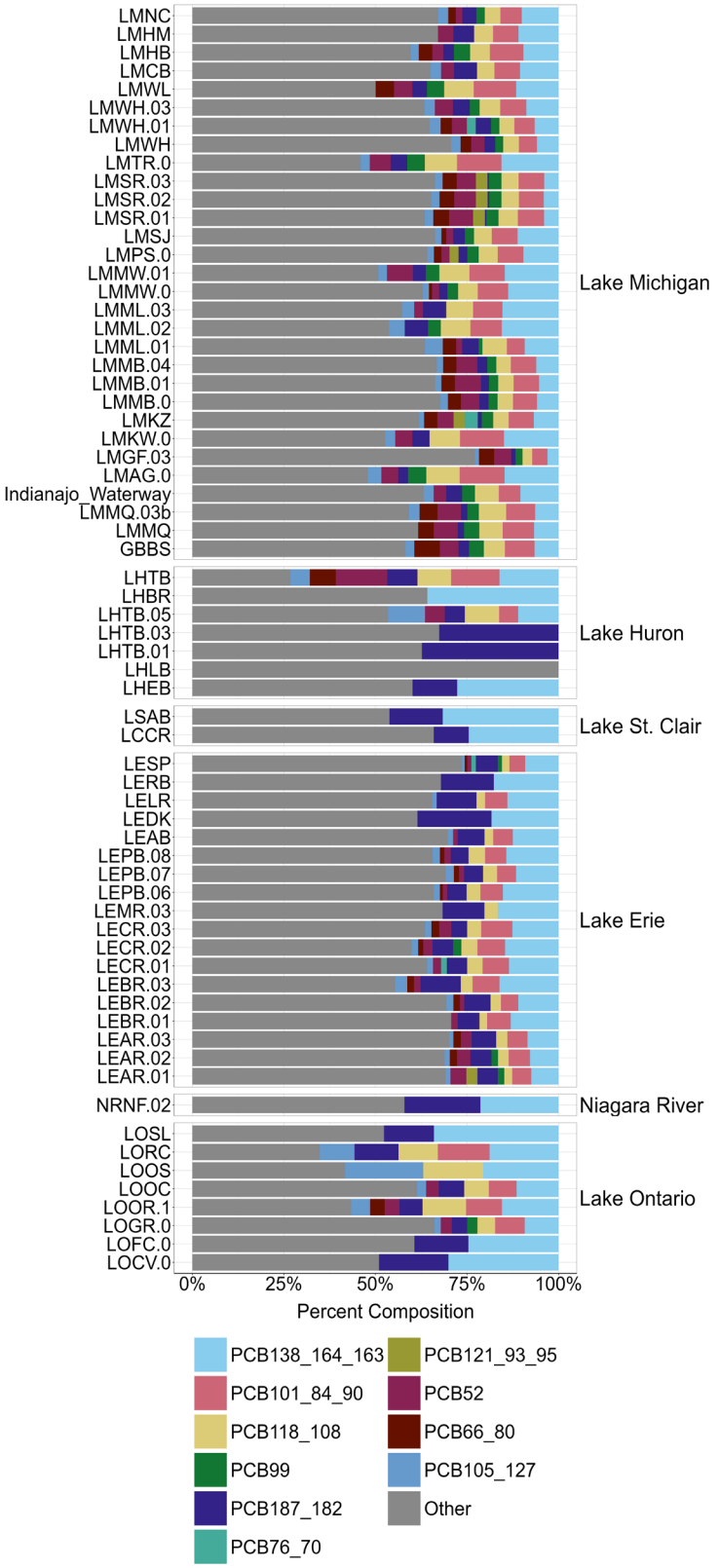


Figure79. Percent composition of individual PCB contaminants in Total PCB concentration sums per site in the Great Lakes Basin during the 2022 sampling period. Lakes are listed geographically from west to east across the Great Lakes Basin. The 10 most prevalent PCB compounds were chosen to be included in this stacked boxplot, the remaining compounds were grouped together as "Other".

14.3 Historical Context of PCBs Magnitude and Distribution in Mussel Tissue

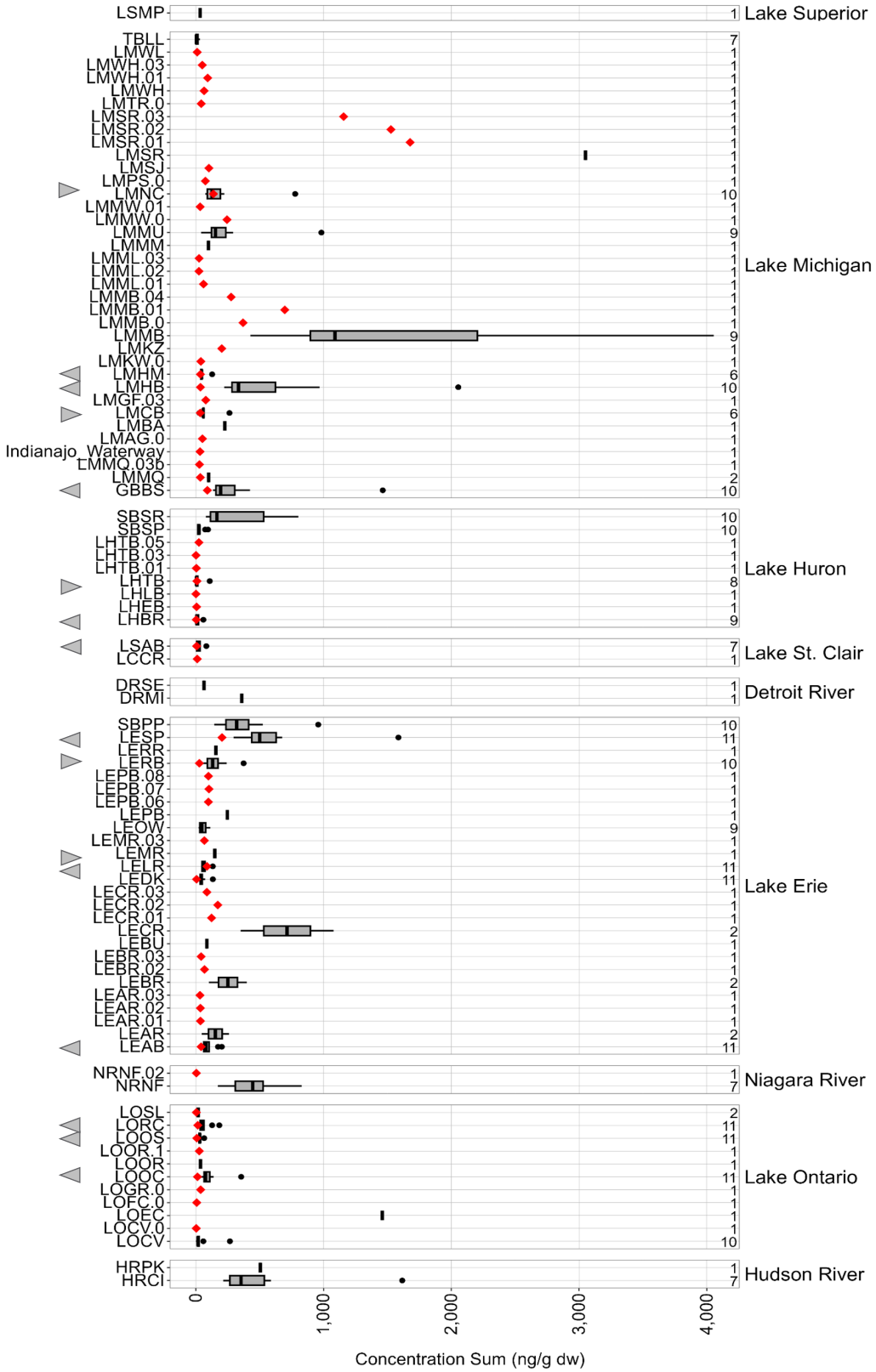


Figure 80. Total PCB concentration sums (ng/g dw) measured in mussel tissue in 2022 (◆ - red diamonds), compared to boxplots of the historic Total PCB concentration sums (ng/g dw) for each site (1992-2022) and results of the trend analysis showing significant increasing (▶) or decreasing (◀) contamination trends over time (gray triangles). Additional information is provided in Table 13 and Table A6. Lakes are listed geographically from west to east across the Great Lakes Basin. The number of years each site has been sampled since 1992 is noted at the side (right) of the plot for each site.

Results - Total PCBs

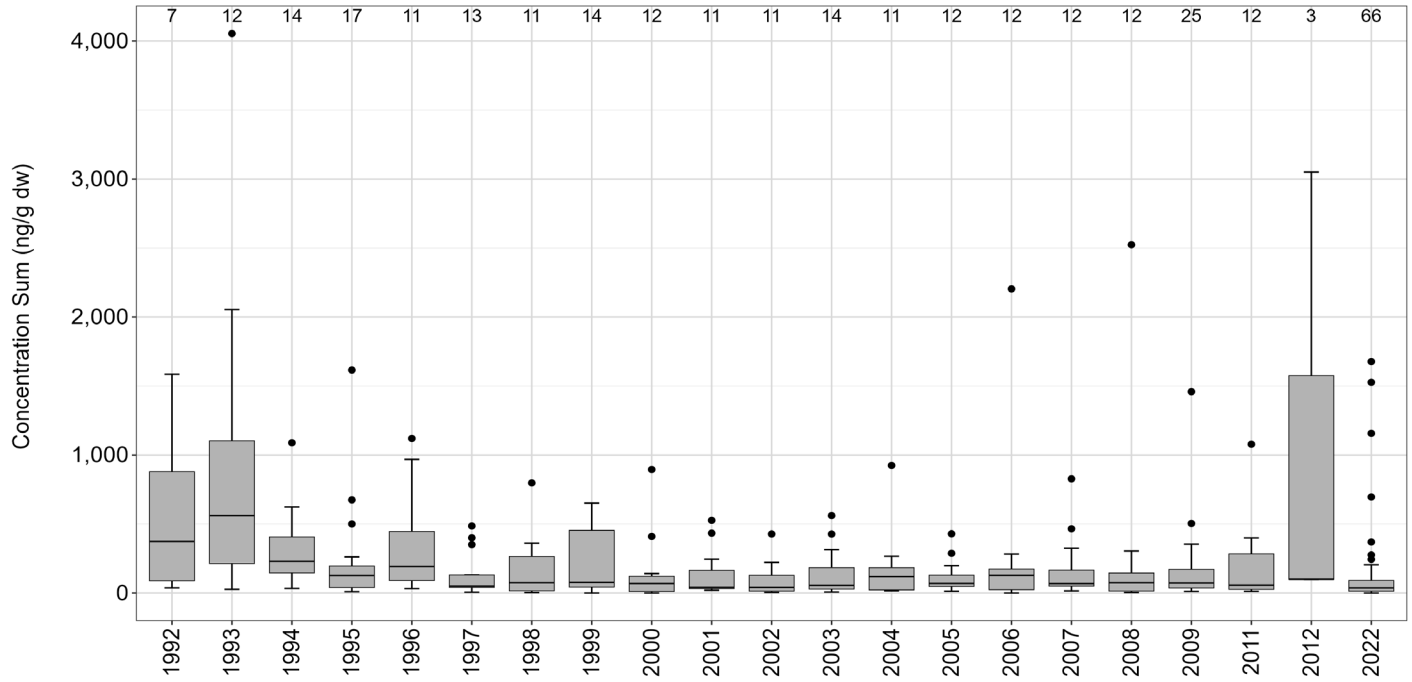


Figure 81. Boxplots representing the historic Total PCB concentration sums (ng/g dw) measured in mussel tissue of the sites analyzed in this study. The number of sites that were sampled each year is noted at the top of the plot.

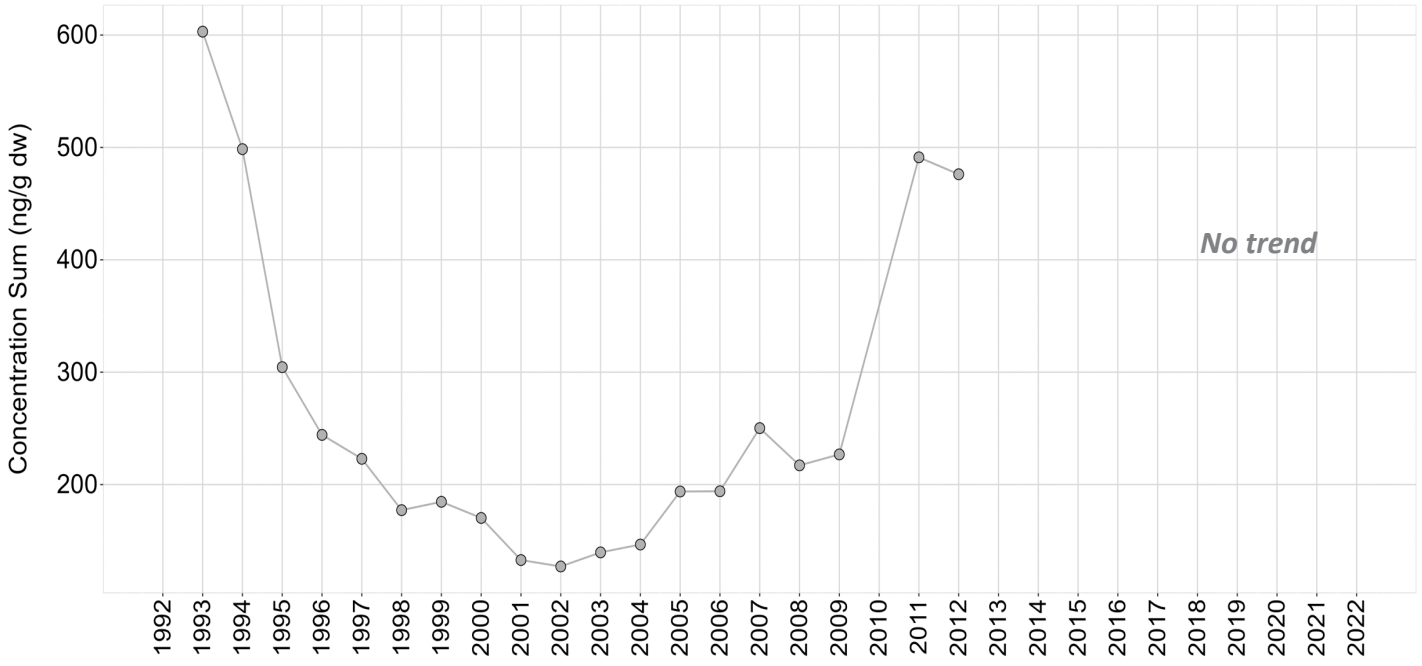


Figure 82. Three-point moving average of the yearly mean Total PCB concentration sums (ng/g dw) measured in mussel tissue in the sites analyzed in this study.

14.4 PCBs Summary

2022 Dreissenid Mussel Tissue Results (sum of 156 compounds):

- Total PCBs were detected in dreissenid mussels at 100% (66/66) of the sites surveyed in the Great Lakes in 2022 (Figure 78; Figure 83)
- Total PCBs concentration summary statistics (Figure 78; Figure 79):
  - Range: 0 – 1,677.5 ng/g dw
  - Minimum detectable concentration sum was detected LHLB (0.982 ng/g dw)
  - Maximum concentration sum was detected at LMSR.01 (1,677.5 ng/g dw)
  - Median: 37.0 ng/g dw
  - Mean ± SD: 136.8 ± 316.6 ng/g dw

Historical Context for Dreissenid Mussel Tissue (sum of 18 compounds):

- 0% of sites were above their historic median concentrations in 2022 (Figure 80).
- No significant regional temporal trend of Total PCBs concentration sums was observed across the Great Lakes Basin sampling locations between 1992-2022 ($\alpha = 0.05$; Figure 81; Figure 82).
- 5 sites (LMCB, LMNC, LHTB, LELR, and LERB) assessed across the Great Lakes Basin showed increasing temporal trends of Total PCB concentration sums ($\alpha = 0.05$; Figure 80; Table 13; Table A6).
- 11 sites (GBBS, LMHB, LMHM, LHBR, LSAB, LEAB, LEDK, LESP, LOOC, LOOS, and LORC) assessed across the Great Lakes Basin showed decreasing temporal trends of Total PCB concentration sums ($\alpha = 0.05$; Figure 80; Table 13; Table A6).
- The percentage of sites sampled in 2022 grouped into each cluster based on historic Great Lakes data (Figure 84):
  - 92% (61/66) of sites in the low cluster (0.00 – 334.6 ng/g dw)
  - 5% (3/66) of sites in the medium cluster (334.7 – 1,156.9 ng/g dw)
  - 3% (2/66) of sites in the high cluster (1,157.0 – 4,054.3 ng/g dw)

General Observations:

- Total PCBs concentration sums were generally low in dreissenid mussel tissue at sites surveyed in the Great Lakes in 2022 (Figure 80; Figure 81).
  - There were six instances of medium contamination (LMKZ, LESP, LMMW.0, LMMB.04, LMMB.0, and LMMB.01) and three instance of high contamination (LMSR.03, LMSR.02, and LMSR.01) detected in dreissenid mussel tissue in 2022 (Figure 83).
- Historically, highest Total PCB concentrations were detected in mussels from sites sampled in Lake Superior (34.1 ng/g dw), Lake St. Clair (82.2 ng/g dw), Detroit River (359.3 ng/g dw), Lake Huron (802.8 ng/g dw), Niagara River (827.7 ng/g dw), Lake Ontario (1,459.0 ng/g dw), Lake Erie (1,586.1 ng/g dw), Hudson River (1,615.9 ng/g dw), and Lake Michigan (4,054.3 ng/g dw; Figure 80; Figure 85).
- The continued presence of PCB compounds in mussel tissue suggests continued inputs into the Great Lakes aquatic environment from likely urban sources (due to their strong urban signature; Hafner and Hites, 2003), as well as input from uncontrolled sources, and resuspended sediment (Hornbuckle et al., 2006).
- For sampling locations analyzed in 2022, Total PCBs concentration sums at 10 sites exceeded the US EPA screening values for subsistence fishers, and 3 sites exceeded US EPA recommended screening values for recreational fishers (Table A2; Table A3).

Results - Total PCBs

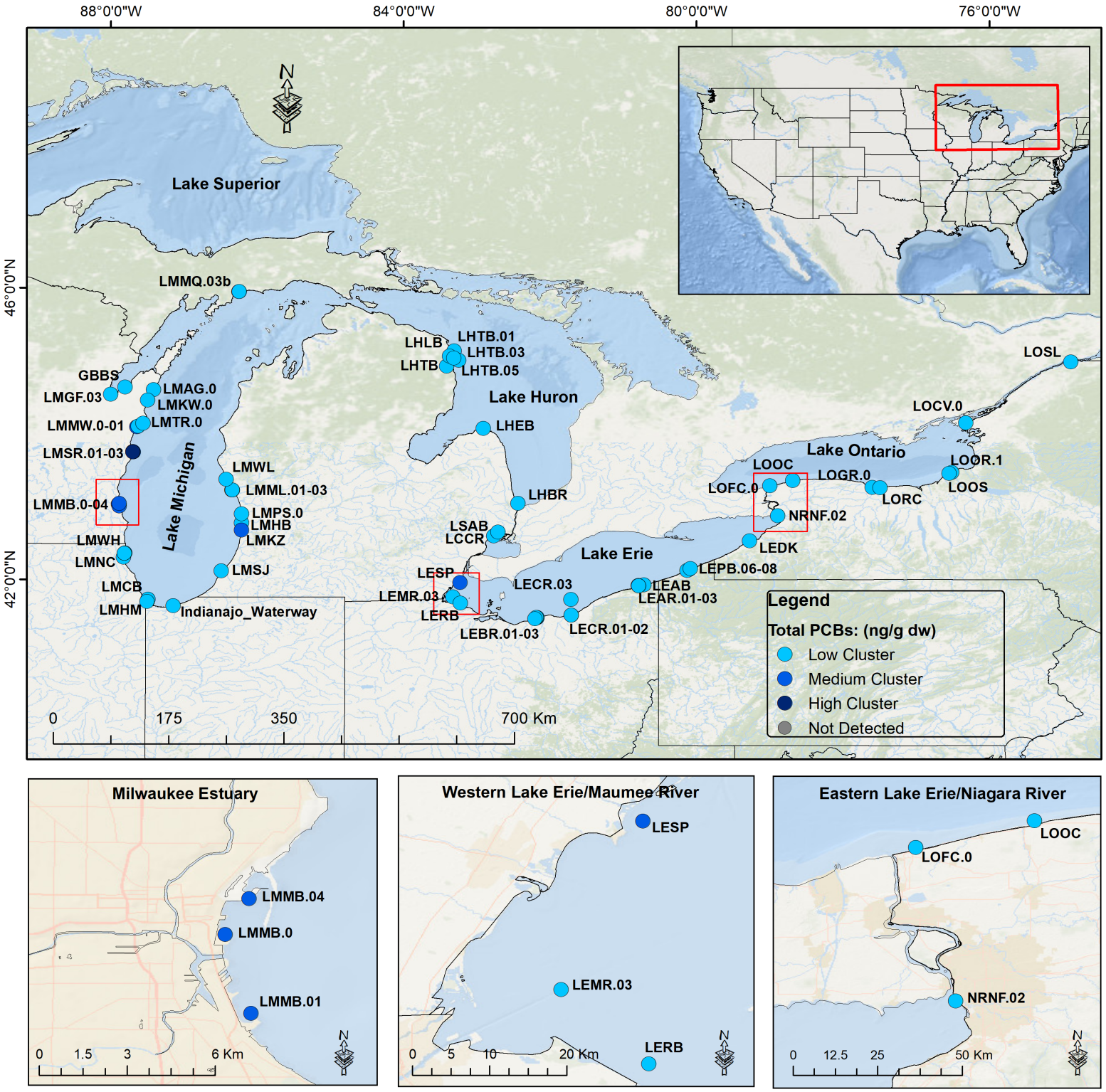


Figure 83. Total PCB concentration sums (ng/g dw) measured in Great Lakes mussel tissue during 2022. Some mussel sampling locations are shown to overlap due to the size of the circles. Colors represent contamination magnitude and clusters as follows: low ( ), medium ( ), high ( ), and not detected ( ).

Results - Total PCBs

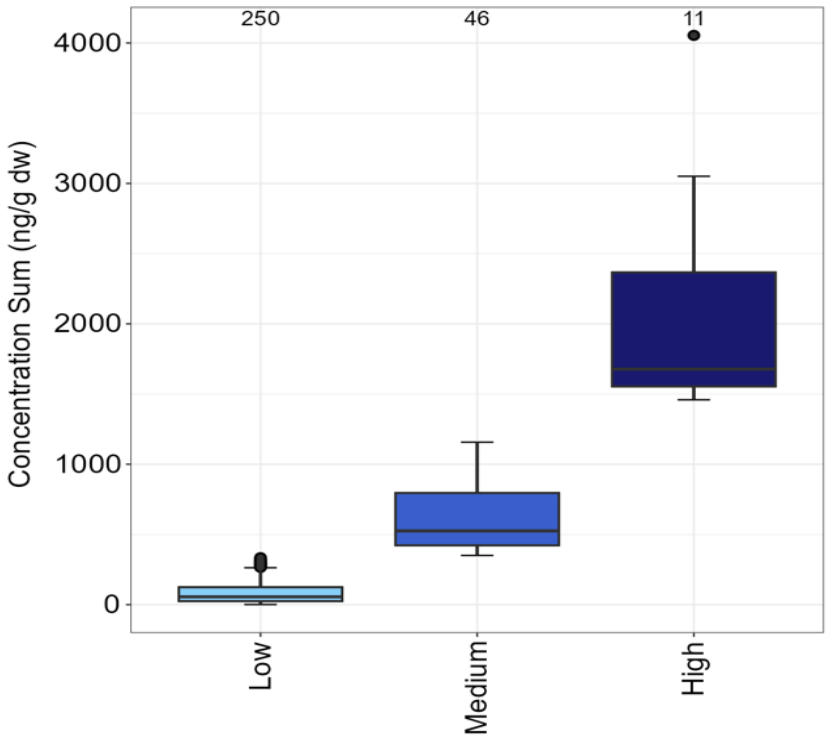


Figure 84. Total PCB concentration sums (ng/g dw) measured in Great Lakes mussel tissue during 1992-2022. Colors represent contamination magnitude as follows: low ( ), medium ( ), and high ( ).

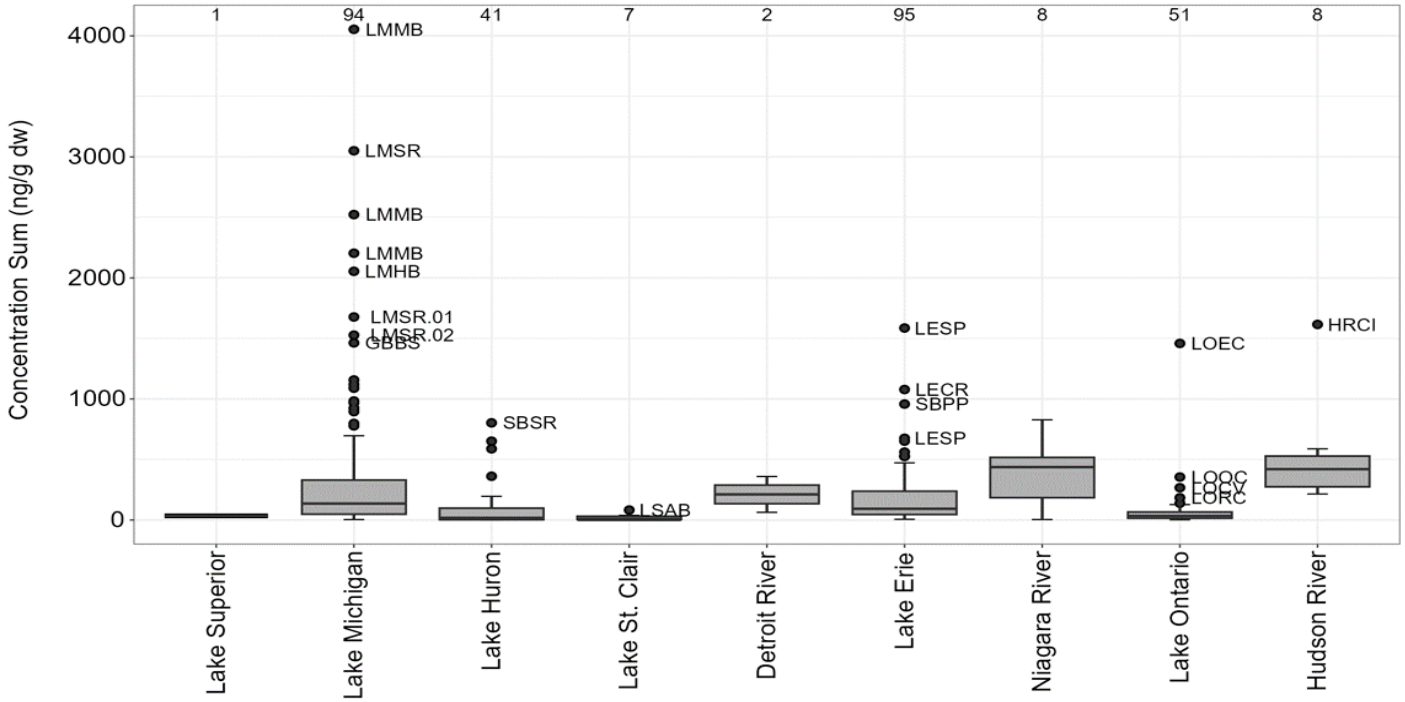


Figure 85. Total PCB concentration sums (ng/g dw) measured in Great Lakes mussels between 1992-2022. Lakes and major rivers including connecting channels sampled during 1992-2022 are listed geographically from west to east across the Great Lakes Basin. The number of PCB samples collected across each lake and major rivers including connecting channels sampled during 1992-2022 is noted at the top of the plot.

Summary

15.0 SUMMARY

This study addresses the magnitude and environmental occurrence of legacy organic contaminants measured in dreissenid mussels in 2022 across four Great Lakes (Lake Michigan, Lake Huron, Lake Erie, and Lake Ontario), with the aim of highlighting the complex nature of these contaminants in the Great Lakes Basin, even after being banned or restricted for decades. We further discuss the spatial distribution and composition profile of these compounds as they are detected in dreissenid mussel tissue basin-wide. Separate result summaries for each legacy organic contaminant group can be found in the summary subsection of each legacy organic contaminant section within this document. This summary furher integrates all contamination results into one analysis to assess overall contamination of MWP sites sampled in the Great Lakes Basin, both with respect to the other sites analyzed within this 2022 study, and to the larger historical MWP dataset for the region. Long-term MWP monitoring tissue data dating back to 1992, provide evidence of the presence of legacy organic contaminants in dreissenid mussels at elevated concentrations in the Great Lakes, pointing towards the need for long-term monitoring of some legacy contaminants in the region. This report aimed to address the objectives to: 1) provide information on legacy organic contaminants magnitude, geographical distribution, and temporal trends measured in lower-trophic level organisms (i.e., dreissenid mussels) across the Great Lakes Basin; 2) compare and contextualize legacy contaminants in the Great Lakes in 2022 to previous studies conducted in the region; 3) provide information on legacy contaminants to inform and support future NOAA/NCCOS MWP and Great Lakes management and policy decisions; and 4) make data on legacy organic contaminants electronically available to state, local, and coastal resource managers in the Great Lakes region; to further reduce and abate the continued occurrence of these contaminants in the Great Lakes aquatic environments.

15.1 Legacy Contaminants Occurrence and Concentration

Of the 11 legacy contaminant groups analyzed in this study, 9 legacy groups were detected at least once in dreissenid mussels basin-wide. With the exception of chlorpyrifos and Total Dieldrins (aldrin, dieldrin, endrin, endrin ketone, and endrin aldehyde), all Great Lakes sampling locations assessed for legacy organic contaminants during the 2022 sampling period had at least one contaminant group present in mussel tissue (Table 11). The absence of detections of chlorpyrifos and of dieldrins in mussel tissue in the Great Lakes in 2022 suggest that these legacy contaminants may no longer be widely present in the region at detectable levels. However, further sampling may be needed to confirm (Figure 30; Figure 44; Figure 86; Table 11). Total DDTs (range: 0 - 118.3 ng/g dw), Total PCBs (range: 0 - 1,677.5 ng/g dw), and Total PAH contaminant groups (range: 0 - 5,080.1 ng/g dw) were detected at the highest concentrations in mussel tissue, with the overall concentration for some compounds within these groups varying by several orders of magnitude (Table 11). The concentrations observed in our study for these contaminant groups were similar or higher compared to previous studies that measured legacy contaminants in dreissenid mussels (Fuller et al., 2023; Kimbrough et al., 2014; Kimbrough et al., 2021; Legrand et al., 2023; Sericano et al., 2014), tree swallows (*Tachycineta bicolor*; Custer et al., 2016; Custer et al., 2021; Custer et al., 2024), and fish tissue (Visha et al., 2018; Zhou et al., 2018).

In regards to environmental occurrence, Total Chlordanes (48%) > Total DDTs (79%) > Total PAHs (100%) > Total PCBs (100%) were detected frequently in mussel tissue during the 2022 survey (Table 12). This emphasizes the ubiquity of these legacy contaminants, even with environmental legislation and restrictions, often imposed many years prior. The frequent detection of these contaminants in mussel tissue might also reflect their historical use, as well as the high uptake and bioaccumulation of these organic compounds in mussels (Edwards et al., 2014; Fuller et al., 2023; Legrand et al., 2023). This also showcases how long-lasting these contaminants can be in the environment, and why they still merit continuous monitoring and assessment. Equally important, the historical use, coupled with anthropogenic activities (e.g., industrial, domestic, and urban wastewater uses), including the discharge of these legacy organic compounds across the Great Lakes Basin are well-documented (Baldwin et al., 2016; Cromwell et al., 2014; Kimbrough et al., 2014). The above conditions have resulted in the impairment of various areas of the Great Lakes (Cromwell et al., 2014; Hartig et al., 2020).

Comparatively, several contaminant groups including Total Endosulfans , Total HCHs, Mirex , Total Butyltins, and Total Chlorobenzenes were detected at relatively low frequencies in mussel tissue, with overall occurrence rates < 16% (Table 12). However, although these legacy contaminants were infrequently detected and measured at low concentrations in mussel tissue, apical and biological effects resulting from low-dose effects may still pose a risk (e.g., additive and synergistic mixture effects, transgenerational effects, etc.,) to both lower and upper trophic-level aquatic organisms (Fuller et al., 2023; Fuller et al., 2025; Legrand et al., 2023).

Summary

*Table 11. Legacy organic contaminant groups total concentrations measured in mussel tissue samples in the Great Lakes in 2022. Individual sites are listed by their general location (associated lake/river) sampled. Long-term/historic MWP sites (****bold italicized***) sampled in 2022 are highlighted below. Lakes are listed geographically from west to east across the Great Lakes Basin.*

Sampling Location	Site	Total Butyltins (n=3)	Total Chlordanes (n=7)	Total Chloro-benzenes (n=5)	Chlor-pyrifos (n=1)	Total DDTs (n=6)	Total Dieldrins (n=3)	Total Endosulfans (n=3)	Total HCHs (n=4)	Mirex (n=1)	Total PAHs (n=39)	Total PCBs (n=18)
Lake Michigan	***GBBS**	0.00	0.00	0.00	0.00	3.45	0.00	0.00	0.00	0.00	251.5	90.0
	Indianajo_Waterway	0.00	1.29	0.00	0.00	0.00	0.00	0.00	0.00	0.00	499.4	32.9
	LMAG.0	0.00	0.00	0.546	0.00	11.7	0.00	0.00	0.00	0.00	506.8	51.8
	LMGF.03	0.00	0.00	0.00	0.00	4.25	0.00	0.00	0.00	0.00	1136.9	77.7
	LMKW.0	0.00	0.00	0.661	0.00	15.0	0.00	0.00	0.00	0.00	612.3	39.2
	LMKZ	0.00	0.00	0.00	0.00	30.4	0.00	0.00	0.00	0.00	643.3	203.1
	LMMB.0	0.00	13.9	2.41	0.00	50.9	0.00	0.00	0.00	0.00	3567.7	369.8
	LMMB.01	0.00	24.3	0.00	0.00	97.4	0.00	0.00	0.00	0.00	5080.1	696.2
	LMMB.04	0.00	13.7	0.712	0.00	49.7	0.00	0.00	0.00	0.00	4394.8	276.2
	LMML.01	0.00	2.89	0.00	0.00	13.2	0.00	0.00	0.00	0.00	246.8	60.1
	LMML.02	0.00	2.48	0.00	0.00	6.18	0.00	0.00	0.00	0.00	239.7	24.5
	LMML.03	0.00	3.25	0.00	0.00	7.94	0.00	0.00	0.00	0.00	183.6	25.4
	***LMMQ**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	105.3	35.1
	LMMQ.03b	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	246.6	28.3
	LMMW.0	0.00	3.37	0.00	0.00	8.76	0.00	0.00	0.00	0.00	1024.7	244.1
	LMMW.01	0.00	0.00	0.00	0.00	1.77	0.00	0.00	0.00	0.00	118.8	33.6
	LMPS.0	0.00	17.4	0.00	0.00	32.6	0.00	0.00	0.00	0.00	185.9	74.6
	LMSJ	0.00	13.1	0.00	0.00	42.9	0.00	0.00	0.00	3.46	946.1	102.2
	LMSR.01	7.96	18.8	1.09	0.00	55.9	0.00	0.00	0.00	0.00	1497.9	1677.5
	LMSR.02	0.00	25.9	1.06	0.00	63.2	0.00	0.00	0.00	0.00	1665.7	1527.3
	LMSR.03	0.00	10.1	1.31	0.00	46.6	0.00	0.00	0.00	6.61	1147.3	1156.9
	LMTR.0	0.00	9.93	1.27	0.00	15.7	0.00	0.00	0.00	0.00	631.3	42.3
	LMWH	0.00	0.00	0.00	0.00	25.8	0.00	0.00	0.00	0.00	611.1	64.7
	LMWH.01	0.00	1.47	0.00	0.00	36.1	0.00	0.00	0.00	0.00	785.1	92.5
	LMWH.03	0.00	2.28	0.00	0.00	41.9	0.00	0.00	0.00	0.00	789.2	50.4
	LMWL	0.00	1.61	0.00	0.00	5.03	0.00	0.00	0.00	2.70	79.4	10.5
	***LMCB**	0.00	0.00	0.00	0.00	2.76	0.00	0.00	0.00	0.00	447.6	34.6
	***LMHB**	0.00	3.01	0.00	0.00	7.76	0.00	0.00	0.00	0.00	274.6	36.1
	***LMHM**	9.94	0.00	0.00	0.00	3.40	0.00	0.00	0.00	0.00	1084.4	35.9
	***LMNC**	0.00	6.23	0.00	0.00	118.3	0.00	0.00	0.00	0.00	699.3	137.2
Lake Huron	***LHBR**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	27.1	3.44
	LHEB	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	164.1	6.08
	LHLB	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	46.2	0.982
	***LHTB**	0.00	0.798	0.00	0.00	1.81	0.00	0.00	0.00	0.00	349.5	8.50
	LHTB.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	293.5	3.82
	LHTB.03	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	140.9	1.33
	LHTB.05	0.00	0.00	0.00	0.00	4.71	0.00	0.00	0.00	0.00	200.8	23.9

*Table 11 (cont). Legacy organic contaminant groups total concentrations measured in mussel tissue samples in the Great Lakes in 2022. Individual sites are listed by their general location (associated lake/river) sampled. Long-term/historic MWP sites (***bold italicized**) sampled in 2022 are highlighted below. Lakes are listed geographically from west to east across the Great Lakes Basin.*

Sampling Location	Site	Total Butyltins (n=3)	Total Chlordanes (n=7)	Total Chloro-benzenes (n=5)	Chlor-pyrifos (n=1)	Total DDTs (n=6)	Total Dieldrins (n=3)	Total Endosulfans (n=3)	Total HCHs (n=4)	Mirex (n=1)	Total PAHs (n=39)	Total PCBs (n=18)
Lake St. Clair	LCCR	0.00	0.00	0.00	0.00	2.01	0.00	0.00	0.00	0.00	258.2	10.8
	***LSAB**	0.00	0.00	0.00	0.00	1.71	0.00	0.00	0.00	0.00	113.2	6.28
Lake Erie	***LEAB**	0.00	0.00	0.00	0.00	1.98	0.00	0.00	0.00	0.00	88.2	41.1
	LEAR.01	0.00	1.67	0.832	0.00	1.61	0.00	0.00	14.5	0.00	400.4	36.1
	LEAR.02	0.00	1.82	0.00	0.00	2.12	0.00	0.00	0.00	0.00	331.3	35.6
	LEAR.03	0.00	0.00	1.15	0.00	0.00	0.00	0.00	0.00	0.00	284.5	32.4
	LEBR.01	0.00	15.9	0.00	0.00	4.20	0.00	0.00	0.00	0.00	561.2	38.9
	LEBR.02	0.00	11.2	0.00	0.00	7.95	0.00	0.00	0.00	0.00	384.0	67.9
	LEBR.03	0.00	4.03	0.00	0.00	4.92	0.00	0.00	0.00	0.00	247.7	41.3
	LECR.01	0.00	6.08	0.00	0.00	11.2	0.00	0.00	0.00	0.00	1576.9	122.9
	LECR.02	0.00	21.7	0.00	0.00	21.2	0.00	4.35	0.00	0.00	2107.4	171.8
	LECR.03	0.00	4.63	0.00	0.00	9.13	0.00	0.00	0.00	0.00	2722.5	86.2
	***LEDK**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	98.8	5.89
	***LELR**	0.00	18.7	0.00	0.00	9.24	0.00	0.00	0.00	0.00	197.9	86.9
	LEMR.03	0.00	0.00	0.00	0.00	1.77	0.00	0.00	0.00	0.00	110.6	66.7
	LEPB.06	0.00	6.11	0.00	0.00	4.59	0.00	0.00	0.00	0.00	2251.6	98.0
	LEPB.07	11.9	0.00	0.00	0.00	6.08	0.00	0.00	0.00	0.00	3131.0	102.6
	LEPB.08	0.00	0.00	0.00	0.00	7.61	0.00	0.00	0.00	0.00	2407.2	98.5
	***LERB**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	106.6	27.6
	***LESP**	0.00	0.00	0.00	0.00	15.5	0.00	0.00	0.00	0.00	673.8	204.9
Niagara River	NRNF.02	0.00	1.89	0.00	0.00	0.00	0.00	0.00	0.00	0.00	314.5	4.03
Lake Ontario	LOCV.0	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	30.2	3.19
	LOFC.0	0.00	0.00	0.00	0.00	1.59	0.00	0.00	0.00	0.00	113.7	6.86
	LOGR.0	8.15	4.22	0.00	0.00	12.9	0.00	0.00	0.00	0.00	1581.7	37.9
	LOOR.1	0.00	0.00	0.00	0.00	5.26	0.00	0.00	0.00	0.00	310.9	26.9
	***LOOC**	0.00	0.00	0.00	0.00	2.05	0.00	0.00	0.00	0.00	94.5	12.8
	***LOOS**	0.00	0.00	0.00	0.00	2.09	0.00	0.00	0.00	0.00	66.1	7.71
	***LORC**	0.00	0.00	0.00	0.00	4.43	0.00	0.00	0.00	0.00	206.6	15.4
	***LOSL**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	90.1	4.08

Table 12. Frequency of detection (% of total) of legacy organic contaminant groups in mussel tissue in 2022 at sites within the Great Lakes Basin.

Contaminant Group	Sites with Detects	Sites Analyzed	Detection Frequency
Total Butyltins	4	66	6%
Total Chlordanes	32	66	48%
Total Chlorobenzenes	10	66	15%
Chlorpyrifos	0	66	0%
Total DDTs	52	66	79%
Total Dieldrins	0	66	0%
Total Endosulfans	1	66	2%
Total HCHs	1	66	2%
Mirex	3	66	5%
Total PAHs	66	66	100%
Total PCBs	66	66	100%

15.2 Mussel Tissue Legacy Contaminants Cluster Analysis Summary

To identify the magnitude and extent of legacy organic contamination in the Great Lakes, mussel site contamination analysis was conducted in two ways. Initially, a multivariate cluster analysis was conducted for each legacy contaminant group at each mussel sampling location, of which the results are represented in subsequent heatmaps (Figure 87; Figure 89). A second multivariate cluster analysis was conducted using the total cluster value for each site to group sites into five categories (e.g., not detected, low, medium, high, very high; Figure 87 and Figure 89), with statistically different degrees of overall contamination. This two-pronged analysis was conducted, (1) respective to sites sampled within this study, and (2) respective to all historic MWP legacy contaminants analyzed in Great Lakes mussel tissue samples between 1992-2022. A breakdown of legacy contaminants cluster analysis respective to contaminants quantified in mussel tissue samples in 2022, and in all historic MWP legacy contaminants analyzed in dreissenid mussels between 1992-2022 is presented in the appendix section of this report (Table A4; Table A5).

On average, for analyses conducted respective only to sites analyzed in this 2022 study, 59% (39/66) of sites were clustered into the low contamination category, 29% (19/66) in the medium category, 10% (7/66) in the high category, and 2% (1/66) in the very high category (Figure 87). Overall, sites with high or very high legacy contamination tended to be closer to more densely populated areas such as sites sampled in Lake Michigan and Lake Erie (Figure 86; Figure 87). In contrast, sites categorized as having low or medium/moderate contamination tended to be in less populated areas. Additional analysis revealed differences in mussels sampled at inshore (river-tributary-harbor complexes) and offshore sampling locations in 2022. Overall legacy contaminant composition and concentrations varied across mussel sampling locations, with higher sum concentrations observed in mussels from riverine sites (concentration range: 0 - 5,898.1 ng/g dw), compared to nearshore (concentration range: 0 - 961.0 ng/g dw), offshore (concentration range: 0 - 894.3 ng/g dw), and connecting channel sampling locations (concentration range: 0 - 271.0 ng/g dw; Figure 86; Figure 87; Table 11). However, differences in riverine, nearshore, offshore, and connecting channel sampling locations legacy contaminant concentrations were not statistically significant (Kruskal-Wallis; $p > 0.05$).

For analyses conducted respective to mussel sampling locations analyzed between 1992-2022, 44% (48/110) of sites surveyed in 2022 were clustered into the low contamination category, while 16% (18/110) fell within the medium category (Figure 89). No sites surveyed in 2022 for legacy organic contaminants fell within the high or very high category, respective to overall sites assessed between 1992-2022. A general assessment of legacy contaminants examined across individual mussel sampling locations from 1992-2022 further revealed similar spatial patterns and chemical signatures for some legacy compound groups examined in mussel tissue. Elevated legacy contaminants composition and concentration observed in mussel tissue were mainly driven by detections at highly industrial sub-watersheds and sampling locations with elevated developed/urban clusters in Lakes Michigan, and Erie (Figure 89). Furthermore, sites

sampled from specific riverine systems which are influenced by domestic and urban waste streams (e.g., municipal solid waste, urban wastewater system, and stormwater runoff) depicted distinct chemical signatures.

Additional analysis revealed, inshore and nearshore sites sampled in Lake Michigan, Lake Huron, Lake Erie, and Lake Ontario which are influenced by domestic and urban sources, displayed distinct chemical signatures for several legacy contaminants (e.g., Total Chlordanes, Total Chlorobenzenes, Total DDTs, Total PAHs, and Total PCBs; Figure 13; Figure 23; Figure 26; Figure 35; Figure 40; Figure 72; Figure 77; Figure 80; Figure 85). Prior studies have also highlighted similar legacy contamination patterns and chemical signatures across the upper and lower Great Lakes (Kimbrough et al., 2014; McGoldrick and Murphy, 2015). Interestingly, the magnitude for several of these legacy contaminant groups detected in mussels from Great Lakes inshore and nearshore sampling locations, were similar or higher compared to results and findings for mussels, fish, and bird tissue in other reported studies (Apeti and Lauenstein, 2006; Custer et al., 2020; Fuller et al., 2025; Kimbrough et al., 2014; Macksasitorn et al., 2015; McGoldrick and Murphy, 2015).

Due to their high organic carbon, high water partition coefficient (Log K_{OC}), and long half-life, legacy contaminants detected in this study will strongly sorb to the organic fraction of bottom sediment and soil, thus reside in the Great Lakes terrestrial and aquatic systems for extended period of time (Al-Omran, 2019; Apeti and Lauenstein, 2006). Moreover, as demonstrated in previous studies, legacy organic compounds are non-biodegradable and tend to persist in bottom sediment and soil for a long time (Edwards et al., 2014; Kimbrough et al., 2008; Kour et al., 2021). As such, the above results would suggest elevated legacy contaminant loading and tissue measurements would likely be attributed to active sediment resuspension due to dynamic lake-seiche events (inshore and nearshore contaminant dynamics and cycling; Harrow-Lyle et al., 2023), and fresh inputs of legacy contaminants [resulting from contaminants being slowly leached from soils and transported into water bodies via atmospheric deposition (wet or dry deposition), and diffuse or point source discharge; Baldwin et al., 2016; Macksasitorn et al., 2015] occurring at these sampling locations. Equally important, elevated legacy contaminant tissue measurements may be associated with the presence of active emission sources, as well as elevated agricultural and urban runoff occurring at these sampling locations (Apeti and Lauenstein, 2006; Burlakova et al., 2018).

Overall, these results indicate legacy contaminants environmental occurrence, concentration, and composition generally differed among mussel sampling locations basin-wide. The above results have also shown legacy contaminants quantified in mussel tissue are also occurring as complex mixtures across the Great Lakes region, with legacy contamination more extensive at some upper and lower Great Lakes mussel sampling locations (Figure 87). As demonstrated in previous studies, the detection of these organic contaminants as complex mixtures in the Great Lakes is of concern, due to their synergistic, additive or antagonistic effects on each other, as well as their constituents, which have been shown to elicit adverse health effects on humans and aquatic organisms (Fuller et al., 2025; Hammond et al., 2023; Legrand et al., 2023; Maloney et al., 2023).

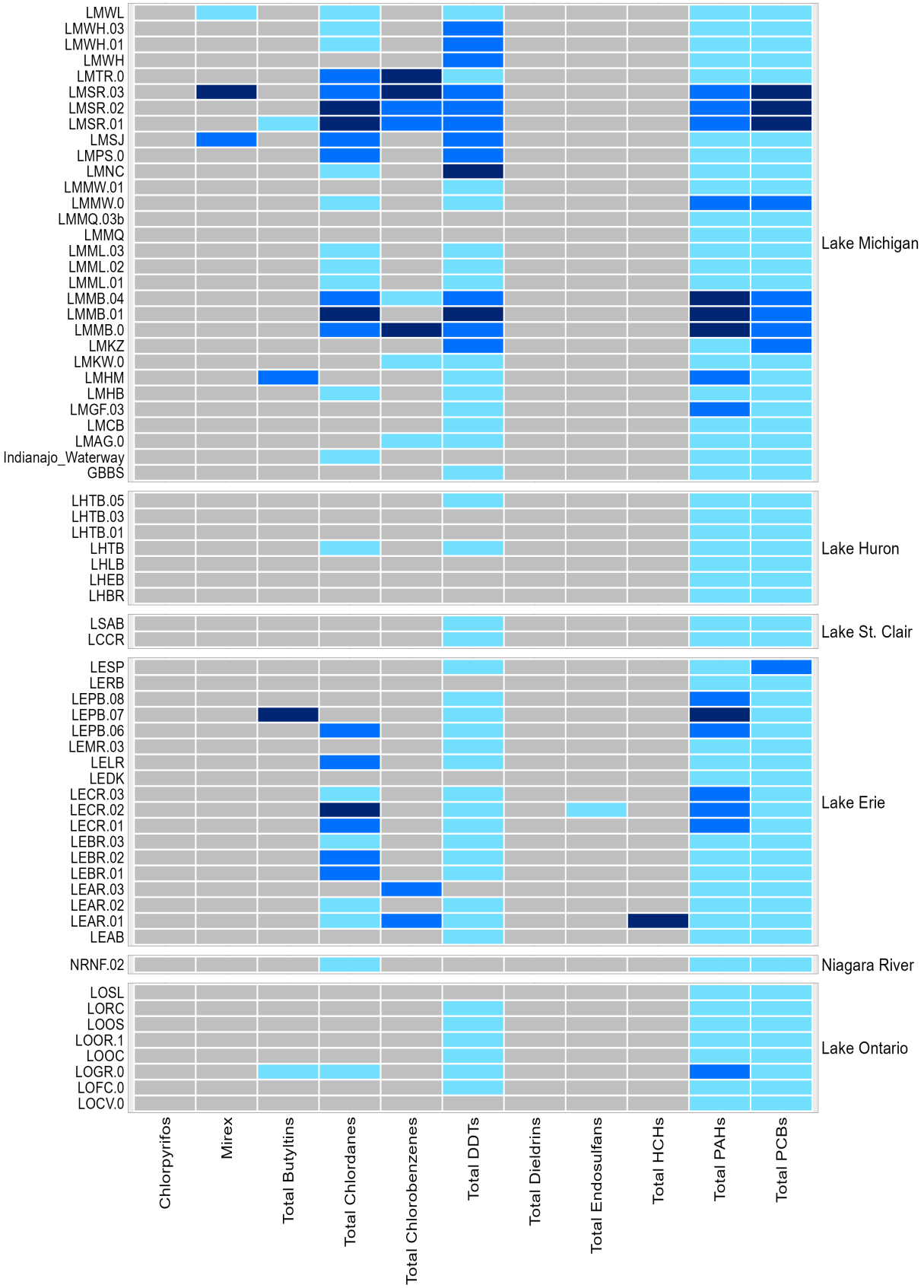


Figure 86. Heatmap depicting the distribution and magnitude of 2022 legacy compound group concentrations detected in Great Lakes dreissenid mussel tissue respective to sites assessed in 2022. Colors represent contamination magnitude as follows: low ( ), medium ( ), high ( ), and not detected ( ). Heatmap tiles with white represents missing or non-quantifiable data. Individual sampling locations are listed across the Great Lakes from west to east by their general location (associated lake/river) sampled. Additional information is provided in Table A4.

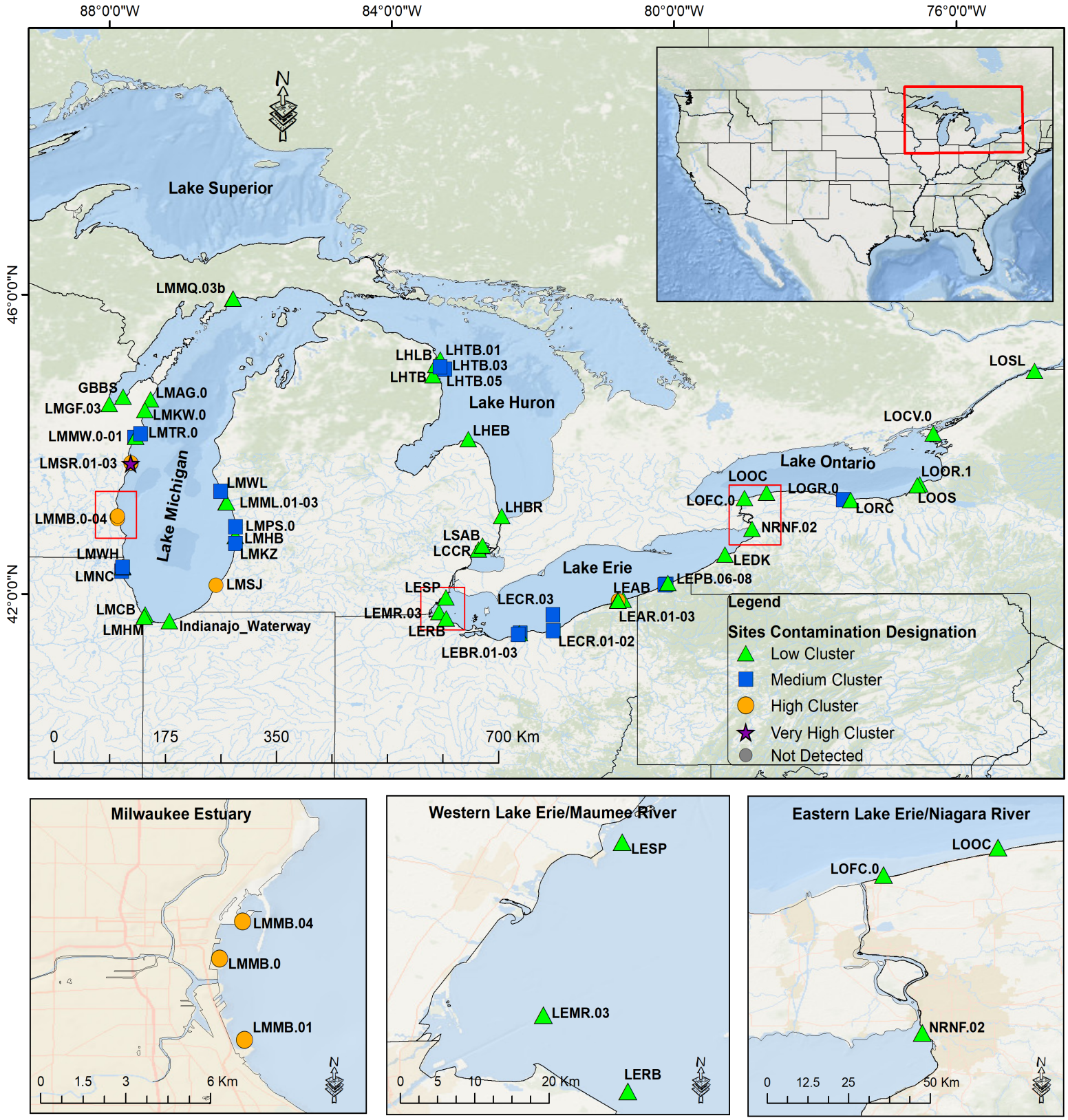


Figure 87. Map of MWP sites in the Great Lakes Basin highlighting sampling locations with Low (▲), Medium (■), High (●), and Very High (★) degrees of contamination in mussel tissue in 2022, respective to one another in this study. Some mussel sampling locations are shown to overlap due to the size of the symbols.

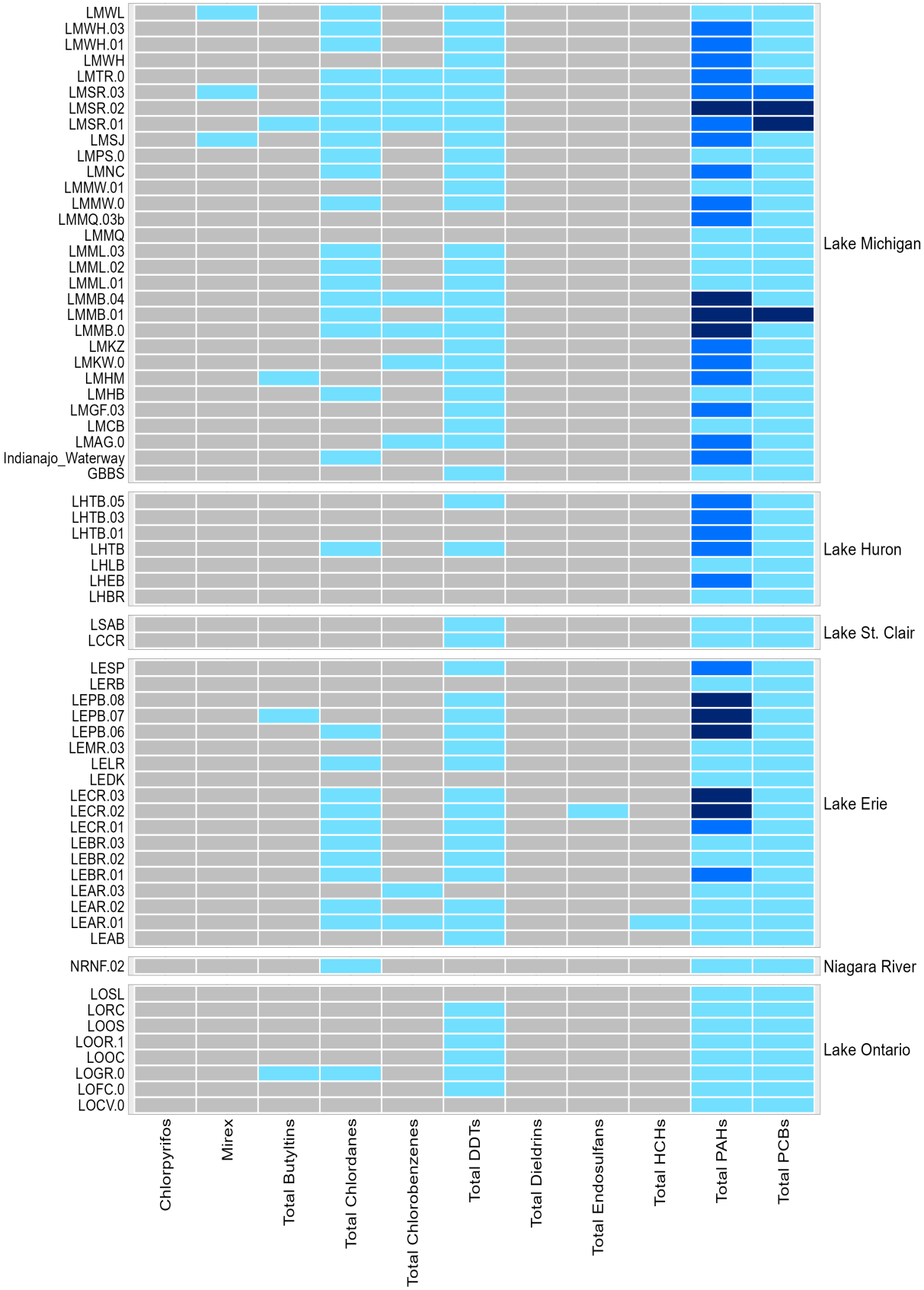


Figure 88. Heatmap showing the distribution and magnitude of legacy compounds concentration sums detected in 2022 Great Lakes mussel tissue samples, respective to all historic Great Lakes MWP legacy contaminants analyzed in dreissenid mussel tissue. Colors represent contamination magnitude as follows: low ( ), medium ( ), high ( ), and not detected ( ). Heatmap tiles with white represents missing or non-quantifiable data. Individual sampling locations are listed across the Great Lakes Basin from west to east by their general location (associated lake/river) sampled.Additional information is provided in Table A5.

Summary

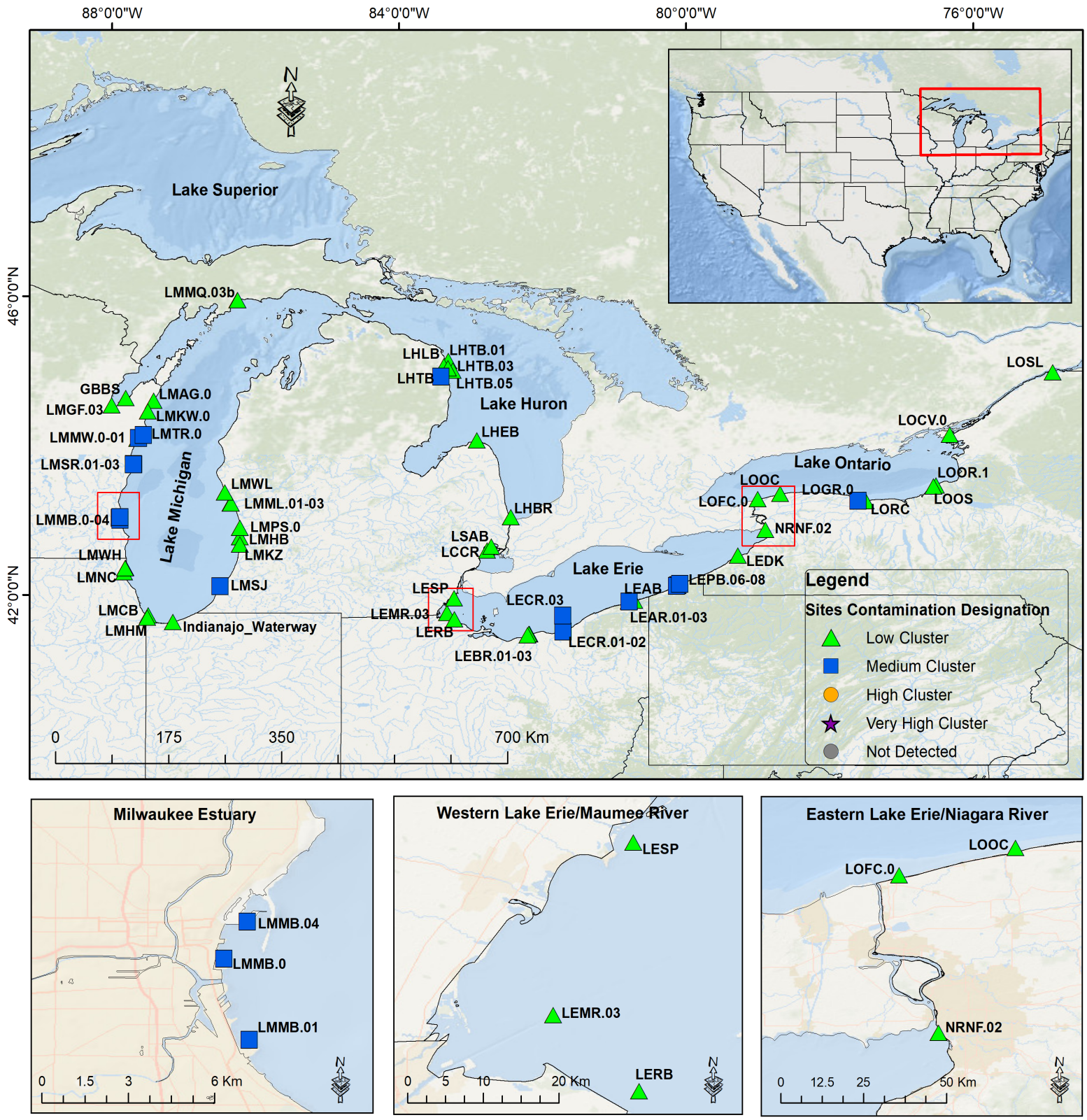


Figure 89. Map of MWP sites in the Great Lakes Basin highlighting sampling locations with Low (▲), Medium (■), High (●), and Very High (★) degrees of legacy contamination detected in dreissenid mussel tissue in 2022, respective to all historic Great Lakes MWP contamination analyzed in dreissenid mussel tissue. Some mussel sampling locations are shown to overlap due to the size of the symbols.

15.3 Legacy Contaminants Historical Analyses and Temporal Trends

Despite the restriction and application ban placed on legacy contaminants use and production in the U.S. since the early 1970s, these compounds are still being detected in the Great Lakes and their connecting channels aquatic systems. The detection of these organic contaminants in Great Lakes aquatic biota (i.e., lower and upper-trophic level organisms) is a significant environmental issue (Fuller et al., 2025; Hartig et al., 2020; Hull et al., 2014; Legrand et al., 2023; Li et al., 2018; McGoldrick and Murphy, 2015; Travis et al., 2020; Zhou et al., 2018). While it is useful to know which mussel sampling locations within the Great Lakes are most contaminated relative to other sites in the region, it is equally important to put the contamination of a region into perspective with other available data historically (Swam-Jaramillo et al., 2025). As such, historical analysis can allow us to observe contaminant trends, as well as identify patterns of legacy contaminant release, persistence, and exposure pathways for these organic compounds to the Great Lakes aquatic food-web. In addition, through an understanding of both the temporal and spatial variations, long-term monitoring programs such as the NOAA/NCCOS Mussel Watch Program (https://coastalscience.noaa.gov/science-areas/pollution/mussel-watch/) can serve its purpose of assessing potential contaminant exposure across regional and national scales. Contrasting temporal trends observed in mussel tissue samples, is one of the lines-of-evidence that allow us to observe the effects of regulations and restrictions on the concentrations of these organic compounds across the Great Lakes, with the emphasis of placing into perspective any recently detected concentrations recorded in mussel tissue samples.

The results from this 2022 study showed, the concentrations for a majority of the legacy contaminants assessed in mussel tissue samples are either declining or showing no directional change over time. For example, the concentration for organic groups such as Total Chlordanes, Total DDTs, Total PAHs, and Total PCBs were shown to decline at some mussel sampling locations in 2022 (Kruskal-Wallis; $p < 0.05$; Table 13; Table A6). The 2022 study results further confirm some legacy contaminant groups have persisted at background levels, suggesting that achieving further significant reductions in some contaminant concentrations may be challenging (Kimbrough et al., 2014). Overall, the decreasing trends observed for some legacy contaminants in this study would also suggest that these contaminants might have reached asymptotic levels across Great Lakes mussel sampling locations, and further reductions in contaminant levels by natural means alone will likely take some time, even decades (Apeti and Lauenstein, 2006; Carlson et al., 2010; Kimbrough et al., 2014). Furthermore, the absence of detections of chlorpyrifos and dieldrins in mussel tissue in the Great Lakes in 2022 suggest, these contaminants may no longer be widely present in the region at detectable levels, however further sampling may be needed to confirm these results (Figure 30; Figure 44; Figure 86; Table 11; Table 13).

Interestingly, an increase in concentrations was also observed for several legacy contaminant groups including Total Chlordanes (LHTB), Total DDTs (LMNC), Total PAHs (LMHM, and LHTB), and Total PCBs (LMCB, LMNC, LHTB, LELR, and LERB) at some long-term mussel sampling locations in 2022 (Figure 11, Figure 33, Figure 70; Figure 78; Table 13; Table A6), suggesting continued influence and input from environmental routes and dispersive pathways such as release from contaminated sediment, urban and agricultural runoff, release of municipal and industrial wastewater effluent, runoff from landfills, and inputs from atmospheric transport and deposition (Apeti et al., 2018; Edwards et al., 2024b; Guo et al., 2018; Hartig et al. 2020; McGoldrick and Murphy, 2015; Venier et al., 2016). In general, legacy compounds with increasing concentrations were higher in mussels that were sampled around developed/urban clusters, compared to non-urban sampling locations. Further analysis and sampling may be warranted to determine the current sources and pathways of contamination for these elevated levels of legacy organic contaminants identified at these mussel sampling locations.

Additionally, when compared to historic Great Lakes mussel tissue data (1992-2022), only two legacy organic contaminant groups (Total PAHs, and Total PCBs) were present at either medium or high level of contamination in mussel tissue at several sampling locations in the Great Lakes Basin in 2022 (Figure 88). Further monitoring should aim to continue analyzing the temporal trends of these contaminants on a regional scale (i.e., through periodic sampling and biennial sampling at select sites), with focus on areas and environmental variables of interest that could give further insight on range, variation, and potential localized point sources of contamination. For example, further sampling around upper Great Lakes areas such as the Waukegan Harbor, Sheboygan River, and Milwaukee Estuary areas could help clarify the higher overall contamination in these areas, relative to other areas sampled in the Great Lakes.

Overall, our 2022 study results confirm some legacy contaminants detected in mussels from lake inshore (tributaries, rivers, bays, and harbors), nearshore and offshore sampling locations are similar in complexity and occurrence to other comparative studies [e.g., The USEPA's Great Lakes Fish Monitoring and Surveillance Program (GLFMSP)], that have targeted and examined legacy contaminants in nearshore and offshore upper trophic-level fish populations throughout the Great Lakes Basin (Carlson et al., 2006; McGoldrick and Murphy, 2015; Pagano and Garner, 2019). Our results further highlight the importance of biomonitoring programs, such as the MWP, in sampling and detecting both legacy

contaminants on a larger spatial scale at nearshore and offshore lake complexes. Despite some legacy contaminants being detected at environmentally low concentrations across mussel sampling locations, our data showed that these compounds are measurable, and are still bioaccumulating in the tissue of lower trophic-level organisms across the Great Lakes. Equally important, this study provides insight into the persistent nature of these legacy organic compounds detected in mussel tissue, and the level of contamination that is still occurring at inshore, nearshore and offshore Great Lakes mussel sampling locations. From a biomonitoring perspective, our mussel legacy contaminant data in conjunction with legacy organic contaminant tissue data from other Great Lakes biomonitoring programs [GLFMSP, Tree swallows (Tachycineta bicolor; Custer et al., 2021), and The Great Lakes Herring Gull Monitoring Program (GLHGMP); Hammond et al., 2024] can provide a better understanding of the spatiotemporal patterns and trophic transfer of these contaminants across the Great Lakes food-web (Edwards et al., 2024b; Kimbrough et al., 2014).

*Table 13. Summary of site-based trend analysis for legacy organic contaminants detected in dreissenid mussel tissue at long-term/historic MWP sites (*bold italicized) sampled in 2022; D = decreasing trend, I = increasing trend, and (--) = no trend. Individual sites are listed by their general location (associated river/lake region) sampled. Lakes are listed geographically from west to east across the Great Lakes Basin.*

Sampling Location	Site	Total Butyltins	Total Chlordanes	Total Chloro-benzenes	Chlor-pyrifos	Total DDTs	Total Dieldrins	Total Endosulfans	Total HCHs	Mirex	Total PAHs	Total PCBs
Lake Michigan	***GBBS**	--	--	--	--	D	--	--	--	--	D	D
	***LMCB**	--	--	--	--	D	--	--	--	--	D	I
	***LMHB**	--	D	--	--	D	--	--	--	--	--	D
	***LMHM**	D	--	--	--	D	--	--	--	--	I	D
	***LMNC**	--	D	--	--	I	--	--	--	--	D	I
Lake Huron	***LHBR**	--	--	--	--	--	--	--	--	--	D	D
	***LHTB**	--	I	--	--	D	--	--	--	--	I	I
Lake St. Clair	***LSAB**	--	--	--	--	D	--	--	--	--	D	D
Lake Erie	***LEAB**	--	--	--	--	D	--	--	--	--	D	D
	***LEDK**	--	--	--	--		--	--	--	--	D	D
	***LELR**		D	--		D	--	--	--	--	D	I
	***LERB**	--	--	--	--	--	--	--	--	--	D	I
	***LESP**	--	--	--	--	D	--	--	--	--	D	D
Lake Ontario	***LOOC**	--	--	--	--	D	--	--	--	--	D	D
	***LOOS**	--	--	--	--	D	--	--	--	--	D	D
	***LORC**	--	--	--	--	D	--	--	--	--	D	D

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APPENDICES

*Table A1. Percent dry (% Dry) values for mussel tissue samples at each site collected in the Great Lakes Basin in 2022. Individual sites are listed by their general location (associated lake/river) sampled. Long-term/historic MWP sites (*__bold italicized__) sampled in 2022 are highlighted below. Lakes are listed geographically from west to east across the Great Lakes Basin.*

Sampling Location	Site	% Dry of Tissue Samples*	% Lipid (Dry)
Lake Michigan	***GBBS**	7.94	9.40
	Indianajo_Waterway	10.6	9.76
	LMAG.0	14.8	18.7
	LMGF.03	10.9	11.1
	LMKW.0	13.6	17.1
	LMKZ	17.2	15.5
	LMMB.0	12.3	0.27
	LMMB.01	15.2	20.3
	LMMB.04	14.8	16.1
	LMML.01	10.3	11.0
	LMML.02	9.86	11.1
	LMML.03	12.8	11.9
	***LMMQ**	12.7	8.56
	LMMQ.03b	12.3	8.23
	LMMW.0	14.5	14.4
	LMMW.01	9.84	14.1
	LMPS.0	13.9	22.2
	LMSJ	13.5	12.9
	LMSR.01	16.5	21.9
	LMSR.02	17.4	23.0
	LMSR.03	17.2	23.6
	LMTR.0	14.5	18.2
	LMWH	12.0	8.15
	LMWH.01	10.7	10.7
	LMWH.03	11.5	12.5
	LMWL	13.6	10.4
	***LMCB**	11.9	12.1
	***LMHB**	14.6	11.5
	***LMHM**	8.90	9.83
	***LMNC**	10.9	12.5

** conc. (ng/g ww) = conc. (ng/g dw) x (% dry / 100)*

*Table A1 (cont). Percent dry (% Dry) values for mussel tissue samples at each site collected in the Great Lakes Basin in 2022. Individual sites are listed by their general location (associated lake/river) sampled. Long-term/historic MWP sites (**bold italicized**) sampled in 2022 are highlighted below. Lakes are listed geographically from west to east across the Great Lakes Basin.*

Sampling Location	Site	% Dry of Tissue Samples*	% Lipid (Dry)
Lake Huron	***LHBR**	11.8	11.8
	LHEB	9.56	9.60
	LHLB	11.7	11.9
	***LHTB**	10.1	13.7
	LHTB.01	9.56	10.9
	LHTB.03	**10.1**	**11.2**
	LHTB.05	**11.1**	**11.5**
Lake St. Clair	LCCR	9.31	9.53
	***LSAB**	11.8	8.43
Lake Erie	LEAB	10.7	11.5
	LEAR.01	9.08	9.80
	LEAR.02	9.76	8.67
	LEAR.03	9.13	10.0
	LEBR.01	8.96	10.6
	LEBR.02	9.21	10.8
	LEBR.03	9.92	10.3
	LECR.01	11.0	8.54
	LECR.02	10.9	11.2
	LECR.03	11.9	11.0
	***LEDK**	7.89	8.88
	***LELR**	8.57	11.2
	LEMR.03	10.2	7.16
	LEPB.06	7.80	9.82
	LEPB.07	8.40	10.6
	LEPB.08	8.50	10.2
	***LERB**	10.6	8.88
	***LESP**	15.1	10.1
Niagara River	NRNF.02	7.77	7.80
Lake Ontario	LOCV.0	7.55	8.16
	LOFC.0	9.04	7.70
	LOGR.0	10.9	11.0
	LOOR.1	13.9	8.45
	***LOOC**	12.3	9.07
	***LOOS**	**8.75**	**7.32**
	***LORC**	**8.34**	**9.67**
	***LOSL**	**11.74**	**7.98**

** conc. (ng/g ww) = conc. (ng/g dw) x (% dry / 100)*

Appendices

Table A2. US FDA Action and Tolerance levels and US EPA Screening Values (SVs) for chemical contaminants in fish and shellfish (ng/g wet weight) compared to the maximum detected concentration in the Great Lakes in 2022 dataset (FDA, 2011; EPA, 2000).

Contaminant Group	Compounds	Maximum Detected Value	US FDA Action Level	US FDA Tolerance Level	US EPA Recreational Fishers Values - Noncarcinogenic	US EPA Recreational Fishers Values - Carcinogenic	US EPA Subsistence Fishers Values - Noncarcinogenic	US EPA Subsistence Fishers Values - Carcinogenic
Total Butyltins	Tributyltin	1.32			1200		147	
Total Chlordanes	Alpha-Chlordane	2.28	300			114		14.0
	Gamma-Chlordane							
	Cis-Nonachlor							
	Trans-Nonachlor							
	Oxychlordane							
Total Chlordanes 2	Heptachlor	0.00	300			4.39		0.54
	Heptachlor Epoxide							
Total Chlorobenzenes	Hexachlorobenzene	0.295				250		3.07
Total DDTs	2,4'-DDT	7.49	5000			117		14.4
	4,4'-DDT							
	2,4'-DDE							
	4,4'-DDE							
	2,4'-DDD							
	4,4'-DDD							
Total Dieldrins	Aldrin	0.00	300			2.50		0.307
	Dieldrin	0.00	300			2.50		0.307
Total Dieldrins 2	Endrin	0.00			1200		147	
Total Endosulfans	Endosulfan I	0.00			24000		2949	
	Endosulfan II							
Total HCHs	Lindane / γ-HCH	0.00				30.7		3.78
Mirex	Mirex	1.14	100		800		98	
Total PAHs*	15 compounds**	130.9				5.47		0.673
Total PCBs	18 congeners***	50.6		2000		20		2.45

** The EPA recommends that tissue samples be analyzed for 15 PAH compounds and that a potency-weighted total concentration be calculated for each sample for comparison with the recommended SVs for benzo(a)pyrene.*

*** Acenaphthene, Acenaphthylene, Anthracene, Benz[a]anthracene, Benzo[a]pyrene, Benzo[b]fluoranthene, Benzo[g,h,i]perylene, Benzo[k]fluoranthene, Chrysene, Dibenzo[a,h] anthracene, Fluoranthene, Fluorene, Indeno[1,2,3-c,d]pyrene, Phenanthrene, Pyrene*

**** 8, 18, 28, 44, 52, 66, 77, 101, 105, 118, 126, 128, 138, 153, 169, 170, 180, 187*

Appendices

*Table A3. Total concentrations of contaminant groups with exceedances in the Great Lakes Basin in 2022 compared to the US EPA Screening Values (SVs) for chemical contaminants in fish and shellfish (ng/g wet weight) (US EPA, 2000). Individual sites are listed by their general location (associated lake/river) sampled. Long-term/historic MWP sites (***bold italicized**) sampled in 2022 are highlighted below. Lakes are listed geographically from west to east across the Great Lakes Basin.*

Sampling Location	Site	Total PAHs Concentration	PAHs Flag	Total PCBs Concentration	PCBs Flag
Lake Michigan	***GBBS**	2.80	*	0.97	--
	Indianajo_Waterway	8.12	**	0.64	--
	LMAG.0	--	--	1.62	--
	LMGF.03	--	**	1.65	--
	LMKW.0	12.5	**	1.08	--
	LMKZ	15.5	**	5.24	*
	LMMB.0	71.7	**	5.94	*
	LMMB.01	130.9	**	14.9	*
	LMMB.04	105.4	**	5.56	*
	LMML.01	4.11	*	0.75	--
	LMML.02	3.78	*	0.48	--
	LMML.03	5.88	*	0.68	--
	***LMMQ**	3.79	*	0.75	--
	LMMQ.03b	1.44	*	0.50	
	LMMW.0	28.6	**	9.33	*
	LMMW.01	2.09	*	0.66	--
	LMPS.0	7.17	**	2.11	--
	LMSJ	19.6	**	3.11	*
	LMSR.01	33.9	**	50.5	**
	LMSR.02	36.3	**	47.5	**
	LMSR.03	31.4	**	37.6	**
	LMTR.0	16.8	**	1.26	--
	LMWH	11.2	**	1.02	--
	LMWH.01	13.2	**	1.20	--
	LMWH.03	14.0	**	0.89	--
	LMWL	2.45	*	0.28	--
	***LMCB**	7.23	**	0.74	--
	***LMHB**	0.00	--	0.90	--
	***LMHM**	7.20	**	0.63	--
	***LMNC**	12.6	**	2.67	*

** exceeds the US EPA Subsistence Fishers Screening Value*

*** exceeds the US EPA Recreational Fishers Screening Value*

*Table A3 (cont). Total concentrations of contaminant groups with exceedances in the Great Lakes Basin in 2022 compared to the US EPA Screening Values (SVs) for chemical contaminants in fish and shellfish (ng/g wet weight) (US EPA, 2000). Individual sites are listed by their general location (associated lake/river) sampled. Long-term/historic MWP sites (**bold italicized**) sampled in 2022 are highlighted below. Lakes are listed geographically from west to east across the Great Lakes Basin.*

Sampling Location	Site	Total PAHs Concentration	PAHs Flag	Total PCBs Concentration	PCBs Flag
Lake Huron	***LHBR**	0.93	*	0.15	--
	LHEB	0.80	*	0.17	--
	LHLB	0.67	--	0.12	--
	***LHTB**	1.31	*	0.14	--
	LHTB.01	5.41	*	0.23	--
	LHTB.03	0.85	*	0.09	--
	LHTB.05	2.81	*	0.30	--
Lake St. Clair	LCCR	3.04	*	0.29	--
	***LSAB**	3.64	*	0.23	--
Lake Erie	***LEAB**	2.45	*	0.92	--
	LEAR.01	5.48	**	0.57	--
	LEAR.02	4.26	*	0.59	--
	LEAR.03	3.59	*	0.52	--
	LEBR.01	6.69	**	0.00	--
	LEBR.02	5.68	**	1.15	--
	LEBR.03	4.74	*	0.97	
	LECR.01	24.1	**	2.74	*
	LECR.02	30.7	**	3.98	*
	LECR.03	36.8	**	2.12	--
	***LEDK**	1.12	*	0.15	--
	***LELR**	3.29	*	0.92	--
	LEMR.03	1.67	*	1.52	--
	LEPB.06	29.8	**	1.82	--
	LEPB.07	46.0	**	1.51	--
	LEPB.08	33.4	**	1.88	--
	***LERB**	2.39	*	0.73	--
	***LESP**	14.3	**	6.13	*
Niagara River	NRNF.02	3.32	*	0.11	--
Lake Ontario	LOCV.0	0.63		0.07	--
	LOFC.0	2.09	*	0.19	--
	LOGR.0	27.9	**	0.79	--
	LOOR.1	7.35	**	0.74	--
	***LOOC**	2.29	*	0.30	--
	***LOOS**	1.78	*	0.15	--
	***LORC**	3.96	*	0.25	--
	***LOSL**	1.29	*	0.16	--

** exceeds the US EPA Subsistence Fishers Screening Value*

*** exceeds the US EPA Recreational Fishers Screening Value*

Appendices

*Table A4. Breakdown of cluster analysis for dreissenid mussel tissue in the Great Lakes Basin respective to one another in this 2022 study. The first section of the table is the cluster value assigned for each chemical class (0 = not detected, 1 = low, 2 = medium, 3 = high). The second section of the table is the calculation conducted to normalize the chemical class cluster sums by the number of chemical classes assessed at each site. The final column is the overall chemical contamination cluster rank assigned to each site (0 = not detected, 1 = low, 2 = medium, 3 = high, 4 = very high). Total ChlBenz = Total Chlorobenzenes. Total Endo. = Total Endosulfans. Long-term/historic MWP sites (***bold italicized**) sampled in 2022 are highlighted below. Lakes are listed geographically from west to east across the Great Lakes Basin.*

Sampling Location	Site	Total Butyltins	Total Chlordanes	Total ChlBenz	Chlor-pyrifos	Total DDTs	Total Dieldrins	Total Endo.	Total HCHs	Mirex	Total PAHs	Total PCBs	Cluster Sum	#Classes Analyzed	Normalized Cluster	Overall Cluster
Lake Michigan	***GBBS**	o	0	0	0	1	0	0	0	0	1	1	3	11	9.1	1
	Indianajo_Waterway	0	1	0	0	0	0	0	0	0	1	1	3	11	9.1	1
	LMAG.0	0	0	1	0	1	0	0	0	0	1	1	4	11	12.1	1
	LMGF.03	0	0	0	0	1	0	0	0	0	2	1	4	11	12.1	1
	LMKW.0	0	0	1	0	1	0	0	0	0	1	1	4	11	12.1	1
	LMKZ	0	0	0	0	2	0	0	0	0	1	2	5	11	15.2	2
	LMMB.0	0	2	3	0	2	0	0	0	0	3	2	12	11	36.4	3
	LMMB.01	0	3	0	0	3	0	0	0	0	3	2	11	11	33.3	3
	LMMB.04	0	2	1	0	2	0	0	0	0	3	2	10	11	30.3	3
	LMML.01	0	1	0	0	1	0	0	0	0	1	1	4	11	12.1	1
	LMML.02	0	1	0	0	1	0	0	0	0	1	1	4	11	12.1	1
	LMML.03	0	1	0	0	1	0	0	0	0	1	1	4	11	12.1	1
	***LMMQ**	0	0	0	0	0	0	0	0	0	1	1	2	11	6.1	1
	LMMQ.03b	0	0	0	0	0	0	0	0	0	1	1	2	11	6.1	1
	LMMW.0	0	1	0	0	1	0	0	0	0	2	2	6	11	18.2	2
	LMMW.01	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	1
	LMPS.0	0	2	0	0	2	0	0	0	0	1	1	6	11	18.2	2
	LMSJ	0	2	0	0	2	0	0	0	2	1	1	8	11	24.2	3
	LMSR.01	1	3	2	0	2	0	0	0	0	2	3	13	11	39.4	3
	LMSR.02	0	3	2	0	2	0	0	0	0	2	3	12	11	36.4	3
	LMSR.03	0	2	3	0	2	0	0	0	3	2	3	15	11	45.4	4
	LMTR.0	0	2	3	0	1	0	0	0	0	1	1	8	11	24.2	2
	LMWH	0	0	0	0	2	0	0	0	0	1	1	4	11	12.1	1
	LMWH.01	0	1	0	0	2	0	0	0	0	1	1	5	11	15.2	1
	LMWH.03	0	1	0	0	2	0	0	0	0	1	1	5	11	15.2	2
	LMWL	0	1	0	0	1	0	0	0	1	1	1	5	11	15.2	2
	***LMCB**	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	1
	***LMHB**	0	1	0	0	1	0	0	0	0	1	1	4	11	12.1	1
	***LMHM**	2	0	0	0	1	0	0	0	0	2	1	6	11	18.2	2
	***LMNC**	0	1	0	0	3	0	0	0	0	1	1	6	11	18.2	2

*Table A4 (cont). Breakdown of cluster analysis for dreissenid mussel tissue in the Great Lakes Basin respective to one another in this 2022 study. The first section of the table is the cluster value assigned for each chemical class (0 = not detected, 1 = low, 2 = medium, 3 = high). The second section of the table is the calculation conducted to normalize the chemical class cluster sums by the number of chemical classes assessed at each site. The final column is the overall chemical contamination cluster rank assigned to each site (0 = not detected, 1 = low, 2 = medium, 3 = high, 4 = very high). Total ChlBenz = Total Chlorobenzenes. Total Endo. = Total Endosulfans. Long-term/historic MWP sites (***bold italicized**) sampled in 2022 are highlighted below. Lakes are listed geographically from west to east across the Great Lakes Basin.*

Sampling Location	Site	Total Butyltins	Total Chlordanes	Total ChlBenz	Chlor-pyrifos	Total DDTs	Total Dieldrins	Total Endo.	Total HCHs	Mirex	Total PAHs	Total PCBs	Cluster Sum	#Classes Analyzed	Normalized Cluster	Overall Cluster
	***LHBR**	0	0	0	0	0	0	0	0	0	1	1	2	11	6.1	1
	LHEB	0	0	0	0	0	0	0	0	0	1	1	2	11	6.1	1
Lake	LHLB	0	0	0	0	0	0	0	0	0	1	1	2	11	6.1	1
Huron	***LHTB**	0	1	0	0	1	0	0	0	0	1	1	4	11	12.1	1
	LHTB.01	0	0	0	0	0	0	0	0	0	1	1	2	11	6.1	1
	LHTB.03	0	0	0	0	0	0	0	0	0	1	1	2	11	6.1	2
	LHTB.05	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	2
Lake	LCCR	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	1
St. Clair	***LSAB**	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	1
	***LEAB**	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	1
	LEAR.01	0	1	2	0	1	0	0	3	0	1	1	9	11	27.3	3
	LEAR.02	0	1	0	0	1	0	0	0	0	1	1	4	11	12.1	1
	LEAR.03	0	0	2	0	0	0	0	0	0	1	1	4	11	12.1	1
	LEBR.01	0	2	0	0	1	0	0	0	0	1	1	5	11	15.2	2
	LEBR.02	0	2	0	0	1	0	0	0	0	1	1	5	11	15.2	2
	LEBR.03	0	1	0	0	1	0	0	0	0	1	1	4	11	12.1	1
Lake	LECR.01	0	2	0	0	1	0	0	0	0	2	1	6	11	18.2	2
Erie	LECR.02	0	3	0	0	1	0	1	0	0	2	1	8	11	24.2	2
	LECR.03	0	1	0	0	1	0	0	0	0	2	1	5	11	15.2	2
	***LEDK**	0	0	0	0	0	0	0	0	0	1	1	2	11	6.1	1
	***LELR**	0	2	0	0	1	0	0	0	0	1	1	5	11	15.2	2
	LEMR.03	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	1
	LEPB.06	0	2	0	0	1	0	0	0	0	2	1	6	11	18.2	2
	LEPB.07	3	0	0	0	1	0	0	0	0	3	1	8	11	24.2	2
	LEPB.08	0	0	0	0	1	0	0	0	0	2	1	4	11	12.1	1
	***LERB**	0	0	0	0	0	0	0	0	0	1	1	2	11	6.1	1
	***LESP**	0	0	0	0	1	0	0	0	0	1	2	4	11	12.1	1

*Table A4 (cont). Breakdown of cluster analysis for dreissenid mussel tissue in the Great Lakes Basin respective to one another in this 2022 study. The first section of the table is the cluster value assigned for each chemical class (0 = not detected, 1 = low, 2 = medium, 3 = high). The second section of the table is the calculation conducted to normalize the chemical class cluster sums by the number of chemical classes assessed at each site. The final column is the overall chemical contamination cluster rank assigned to each site (0 = not detected, 1 = low, 2 = medium, 3 = high, 4 = very high). Total ChlBenz = Total Chlorobenzenes. Total Endo. = Total Endosulfans. Long-term/historic MWP sites (***bold italicized**) sampled in 2022 are highlighted below. Lakes are listed geographically from west to east across the Great Lakes Basin.*

Sampling Location	Site	Total Butyltins	Total Chlordanes	Total ChlBenz	Chlor-pyrifos	Total DDTs	Total Dieldrins	Total Endo.	Total HCHs	Mirex	Total PAHs	Total PCBs	Cluster Sum	#Classes Analyzed	Normalized Cluster	Overall Cluster
Niagara River	NRNF.02	0	1	0	0	0	0	0	0	0	1	1	3	11	9.1	1
Lake Ontario	LOCV.0	0	0	0	0	0	0	0	0	0	1	1	2	11	6.1	1
	LOFC.0	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	1
	LOGR.0	1	1	0	0	1	0	0	0	0	2	1	6	11	18.2	2
	LOOR.1	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	1
	***LOOC**	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	1
	***LOOS**	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	1
	***LORC**	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	1
	***LOSL**	0	0	0	0	0	0	0	0	0	1	1	2	11	6.1	1

*Table A5. Breakdown of cluster analysis for dreissenid mussel tissue in the Great Lakes Basin in 2022 respective to all historic MWP contamination analyzed in dreissenid mussels. The first section of the table is the cluster value assigned for each chemical class (0 = not detected, 1 = low, 2 = medium, 3 = high). The second section of the table is the calculation conducted to normalize the chemical class cluster sums by number of chemical classes assessed at each site. The final column is the overall chemical contamination cluster rank assigned to each site (0 = not detected, 1 = low, 2 = medium, 3 = high, 4 = very high). Total ChlBenz = Total Chlorobenzenes. Total Endo. = Total Endosulfans. Long-term/historic MWP sites (***bold italicized**) sampled in 2022 are highlighted below. Lakes are listed geographically from west to east across the Great Lakes Basin.*

Sampling Location	Site	Total Butyltins	Total Chlordanes	Total ChlBenz	Chlor-pyrifos	Total DDTs	Total Dieldrins	Total Endo.	Total HCHs	Mirex	Total PAHs	Total PCBs		Cluster Sum	#Classes Analyzed	Normalized Cluster		Overall Cluster
	***GBBS**	0	0	0	0	0	0	0	0	0	1	1		2	11	6.1		1
	Indianajo_Waterway	0	1	0	0	0	0	0	0	0	2	1		4	11	12.1		1
	LMAG.0	0	0	1	0	0	0	0	0	0	2	1		4	11	12.1		1
	LMGF.03	0	0	0	0	0	0	0	0	0	2	1		3	11	9.1		1
	LMKW.0	0	0	1	0	0	0	0	0	0	2	1		4	11	12.1		1
	LMKZ	0	0	0	0	0	0	0	0	0	2	1		3	11	6.1		1
	LMMB.0	0	1	1	0	0	0	0	0	0	3	2		7	11	21.2		2
	LMMB.01	0	1	0	0	0	0	0	0	0	3	3		7	11	21.2		2
	LMMB.04	0	1	1	0	0	0	0	0	0	3	1		6	11	18.2		2
	LMML.01	0	1	0	0	0	0	0	0	0	1	1		3	11	9.1		1
	LMML.02	0	1	0	0	0	0	0	0	0	1	1		3	11	9.1		1
	LMML.03	0	1	0	0	0	0	0	0	0	1	1		3	11	9.1		1
	***LMMQ**	0	0	0	0	0	0	0	0	0	1	1		2	11	6.1		1
Lake	LMMQ.03b	0	0	0	0	0	0	0	0	0	2	1		2	11	6.1		1
Michigan	LMMW.0	0	1	0	0	0	0	0	0	0	2	1		4	11	12.1		1
	LMMW.01	0	1	0	0	0	0	0	0	0	1	1		3	11	9.1		2
	LMPS.0	0	1	0	0	0	0	0	0	0	1	1		3	11	9.1		1
	LMSJ	0	1	0	0	0	0	0	0	1	2	1		5	11	15.2		2
	LMSR.01	1	1	1	0	0	0	0	0	0	2	3		8	11	24.2		2
	LMSR.02	0	1	1	0	0	0	0	0	0	3	3		8	11	24.2		2
	LMSR.03	0	1	1	0	0	0	0	0	2	2	2		8	11	24.2		2
	LMTR.0	0	1	1	0	0	0	0	0	0	2	1		6	11	18.2		2
	LMWH	0	0	0	0	0	0	0	0	0	2	1		3	11	9.1		1
	LMWH.01	0	1	0	0	0	0	0	0	0	2	1		4	11	12.1		1
	LMWH.03	0	1	0	0	0	0	0	0	0	2	1		4	11	12.1		1
	LMWL	0	1	0	0	0	0	0	0	1	1	1		4	11	12.1		1
	***LMCB**	0	0	0	0	0	0	0	0	0	1	1		2	11	6.1		1
	***LMHB**	0	1	0	0	0	0	0	0	0	1	1		3	11	9.1		1
	***LMHM**	1	0	0	0	0	0	0	0	0	2	1		4	11	12.1		1
	***LMNC**	0	1	0	0	0	0	0	0	0	2	1		4	11	12.1		1

*Table A5 (cont). Breakdown of cluster analysis for dreissenid mussel tissue in the Great Lakes Basin in 2022 respective to all historic MWP contamination analyzed in dreissenid mussels. The first section of the table is the cluster value assigned for each chemical class (0 = not detected, 1 = low, 2 = medium, 3 = high). The second section of the table is the calculation conducted to normalize the chemical class cluster sums by number of chemical classes assessed at each site. The final column is the overall chemical contamination cluster rank assigned to each site (0 = not detected, 1 = low, 2 = medium, 3 = high, 4 = very high). Total ChlBenz = Total Chlorobenzenes. Total Endo. = Total Endosulfans. Long-term/ historic MWP sites (*bold italicized) sampled in 2022 are highlighted below. Lakes are listed geographically from west to east across the Great Lakes Basin.*

Sampling Location	Site	Total Butyltins	Total Chlordanes	Total ChlBenz	Chlor-pyrifos	Total DDTs	Total Dieldrins	Total Endo.	Total HCHs	Mirex	Total PAHs	Total PCBs	Cluster Sum	#Classes Analyzed	Normalized Cluster	Overall Cluster
	***LHBR**	0	0	0	0	0	0	0	0	0	1	1	2	11	6.1	1
	LHEB	0	0	0	0	0	0	0	0	0	2	1	3	11	9.1	1
Lake	LHLB	0	0	0	0	0	0	0	0	0	1	1	2	11	6.1	1
Huron	***LHTB**	0	1	0	0	1	0	0	0	0	2	1	5	11	15.2	2
	LHTB.01	0	0	0	0	0	0	0	0	0	2	1	3	11	9.1	1
	LHTB.03	0	0	0	0	0	0	0	0	0	2	1	3	11	9.1	1
	LHTB.05	0	0	0	0	1	0	0	0	0	2	1	4	11	12.1	1
Lake	LCCR	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	1
St. Clair	***LSAB**	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	1
	***LEAB**	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	1
	LEAR.01	0	1	1	0	1	0	0	3	0	1	1	8	11	24.2	2
	LEAR.02	0	1	0	0	1	0	0	0	0	1	1	4	11	12.1	1
	LEAR.03	0	0	1	0	0	0	0	0	0	1	1	3	11	9.1	1
	LEBR.01	0	0	0	0	0	0	0	0	0	2	0	2	11	6.1	1
	LEBR.02	0	1	0	0	1	0	0	0	0	1	1	4	11	12.1	1
	LEBR.03	0	1	0	0	1	0	0	0	0	1	1	4	11	12.1	1
Lake	LECR.01	0	1	0	0	1	0	0	0	0	2	1	5	11	15.2	2
Erie	LECR.02	0	1	0	0	1	0	1	0	0	3	1	7	11	21.2	2
	LECR.03	0	1	0	0	1	0	0	0	0	3	1	6	11	18.1	2
	***LEDK**	0	0	0	0	0	0	0	0	0	1	1	2	11	6.1	1
	***LELR**	0	1	0	0	0	0	0	0	0	1	1	3	11	9.1	1
	LEMR.03	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	1
	LEPB.06	0	1	0	0	1	0	0	0	0	3	1	6	11	18.1	2
	LEPB.07	1	0	0	0	1	0	0	0	0	3	1	6	11	18.1	2
	LEPB.08	0	0	0	0	1	0	0	0	0	3	1	5	11	15.2	2
	***LERB**	0	0	0	0	0	0	0	0	0	1	1	2	11	6.1	1
	***LESP**	0	0	0	0	1	0	0	0	0	2	1	4	11	12.1	1

*Table A5 (cont). Breakdown of cluster analysis for dreissenid mussel tissue in the Great Lakes Basin in 2022 respective to all historic MWP contamination analyzed in dreissenid mussels. The first section of the table is the cluster value assigned for each chemical class (0 = not detected, 1 = low, 2 = medium, 3 = high). The second section of the table is the calculation conducted to normalize the chemical class cluster sums by number of chemical classes assessed at each site. The final column is the overall chemical contamination cluster rank assigned to each site (0 = not detected, 1 = low, 2 = medium, 3 = high, 4 = very high). Total ChlBenz = Total Chlorobenzenes. Total Endo. = Total Endosulfans. Long-term/ historic MWP sites (***bold italicized**) sampled in 2022 are highlighted below. Lakes are listed geographically from west to east across the Great Lakes Basin.*

Sampling Location	Site	Total Butyltins	Total Chlordanes	Total ChlBenz	Chlor-pyrifos	Total DDTs	Total Dieldrins	Total Endo.	Total HCHs	Mirex	Total PAHs	Total PCBs		Cluster Sum	#Classes Analyzed	Normalized Cluster		Overall Cluster
Niagara River	NRNF.02	0	1	0	0	0	0	0	0	0	1	1		3	11	9.1		1
Lake Ontario	LOCV.0	0	0	0	0	0	0	0	0	0	1	1		2	11	6.1		1
	LOFC.0	0	0	0	0	1	0	0	0	0	1	1		3	11	9.1		1
	LOGR.0	1	1	0	0	1	0	0	0	0	1	1		5	11	15.2		2
	LOOR.1	0	0	0	0	1	0	0	0	0	1	1		3	11	9.1		1
	***LOOC**	0	0	0	0	1	0	0	0	0	1	1		3	11	9.1		1
	***LOOS**	0	0	0	0	1	0	0	0	0	1	1		3	11	9.1		1
	***LORC**	0	0	0	0	1	0	0	0	0	1	1		3	11	9.1		1
	***LOSL**	0	0	0	0	0	0	0	0	0	1	1		2	11	6.1		1

Appendices

Table A6. Results of Spearman's Rank tests on site specific temporal trends in mussel tissue for sites sampled in the Great Lakes in 2022.

Chemical Group	Site	p value	rho value	Increasing / Decreasing
Total Butyltins	LMHM	*p* = 0.6228	rho = -0.2571	D
Total Chlordanes	LMHB	*p* = 0.0208	rho = -0.7857	D
Total Chlordanes	LMNC	*p* = 0.8665	rho = 0.0714	D
Total Chlordanes	LHTB	*p* = 0.1361	rho = -0.5749	I
Total Chlordanes	LELR	*p* = 0.6272	rho = -0.1758	D
Total DDTs	GBBS	*p* = 0.0003	rho = -0.9030	D
Total DDTs	LMCB	*p* = 0.0048	rho = -0.9429	D
Total DDTs	LMHB	*p* = 0.0053	rho = -0.8333	D
Total DDTs	LMHM	*p* = 0.6228	rho = -0.2571	D
Total DDTs	LMNC	*p* = 0.3655	rho = -0.3212	I
Total DDTs	LHTB	*p* = 0.2604	rho = -0.4524	D
Total DDTs	LSAB	*p* = 0.1605	rho = -0.5929	D
Total DDTs	LEAB	*p* = 0.0128	rho = -0.7182	D
Total DDTs	LELR	*p* = 0.0353	rho = -0.6364	D
Total DDTs	LESP	*p* = 0.0319	rho = -0.6455	D
Total DDTs	LOOC	*p* = 0.2006	rho = -0.4182	D
Total DDTs	LOOS	*p* = 0.0319	rho = -0.6455	D
Total DDTs	LORC	*p* = 0.1601	rho = -0.4545	D
Total PAHs	GBBS	*P* = 0.0133	rho = -0.7455	D
Total PAHs	LMCB	*p* = 0.0048	rho = -0.9429	D
Total PAHs	LMHM	*p* = 0.3287	rho = -0.4857	I
Total PAHs	LMNC	*p* = 0.3848	rho = -0.3091	D
Total PAHs	LHBR	*p* = 0.0579	rho = -0.6905	D
Total PAHs	LHTB	*p* = 0.6103	rho = -0.2143	I
Total PAHs	LSAB	*p* = 0.6228	rho = -0.2571	D
Total PAHs	LEAB	*p* = 0.0084	rho = -0.7455	D
Total PAHs	LEDK	*p* = 0.1797	rho = -0.4364	D
Total PAHs	LELR	*p* = 0.0956	rho = -0.5273	D
Total PAHs	LERB	*p* = 0.2861	rho = -0.4	D
Total PAHs	LESP	*p* = 0.0111	rho = -0.7576	D
Total PAHs	LOOC	*p* = 0.7293	rho = -0.1182	D
Total PAHs	LOOS	*p* = 0.2589	rho = -0.3727	D
Total PAHs	LORC	*p* = 0.9365	rho = -0.0273	D

Table A6 (cont). Results of Spearman's Rank tests on site specific temporal trends in mussel tissue for sites sampled in the Great Lakes in 2022.

Chemical Group	Site	p value	rho value	Increasing / Decreasing
Total PCBs	GBBS	*p* = 0.0092	rho = -0.7697	D
Total PCBs	LMCB	*p* = 0.0724	rho = -0.7714	I
Total PCBs	LMHB	*p* = 0.0029	rho = -0.8303	D
Total PCBs	LMHM	*p* = 0.2657	rho = -0.5429	D
Total PCBs	LMNC	*p* = 0.2931	rho = -0.3697	I
Total PCBs	LHBR	*p* = 0.2242	rho = -0.45	D
Total PCBs	LHTB	*p* = 0.7358	rho = -0.1429	I
Total PCBs	LSAB	*p* = -0.1429	rho = -0.6786	D
Total PCBs	LEAB	*p* = 0.1025	rho = -0.5182	D
Total PCBs	LEDK	*p* = 0.1602	rho = -0.4545	D
Total PCBs	LELR	*p* = 0.4011	rho = -0.2818	I
Total PCBs	LERB	*p* = 0.0075	rho = -0.7818	I
Total PCBs	LESP	*p* = 0.0426	rho = -0.6182	D
Total PCBs	LOOC	*p* = 0.0165	rho = -0.7	D
Total PCBs	LOOS	*p* = 0.0353	rho = -0.6364	D
Total PCBs	LORC	*p* = 0.0389	rho = -0.6273	D



U.S. Department of Commerce

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National Ocean Service

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