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NATIONAL STATUS AND TRENDS, MUSSEL WATCH PROGRAM
A 2022 Assessment of Trace Metals in Dreissenid Mussels in the Great Lakes Basin







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United States Department of Commerce

National Oceanic and Atmospheric Administration

National Ocean Service

EXECUTIVE SUMMARY

In 2022, the National Oceanic and Atmospheric Administration (NOAA) Mussel Watch Program (MWP) conducted an assessment of the presence, distribution, and concentrations of trace metals in mussels (*Dreissena polymorpha* and *Dreissena rostriformis bugensis*, collectively referred to as dreissenid mussels) from the Great Lakes Basin. The national MWP utilizes a sentinel-based monitoring approach by collecting and analyzing bivalves as surrogates for coastal water pollution. 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. Dreissenid mussel tissue samples from 66 monitoring sites across the Great Lakes Basin from Wisconsin to New York were analyzed in this study. Samples were analyzed for 14 trace metals including aluminum (Al), arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), lead (Pb), manganese (Mn), mercury (Hg), nickel (Ni), selenium (Se), silver (Ag), tin (Sn), and zinc (Zn).

The results indicated that trace metal concentrations varied in bivalve tissue collected across the Great Lakes Basin in 2022. The accumulation of metals in organisms is often analyte and location dependent and can be influenced by the natural presence of many metals in the environment. In this study, the results indicated that the presence and concentration of a specific metal are heavily influenced by its chemistry, sources, fate, and transport. The information presented here is useful to regional managers because of the long-term nature and geographic spread of the dataset, which could compliment data from other regional monitoring programs.

The MWP provides unique data that is essential for assessing the health of the Nation's coastal waters through temporal and spatial evaluation of chemical contamination. Studies such as this not only provide the needed data and information for the MWP but also address contamination data gaps that are relevant for coastal management. Therefore, coastal and watershed managers within the Great Lakes Basin states would benefit from this study as they develop long-term policies to protect their managed ecosystems.

KEY FINDINGS

- All 14 metals (aluminum, arsenic, cadmium, chromium, copper, iron, lead, manganese, mercury, nickel, selenium, silver, tin, and zinc) were detected at 100% of sites.
- Regionally, aluminum, arsenic, cadmium, chromium, manganese, mercury, nickel, selenium, and zinc showed significantly decreasing temporal trends.
- Regionally, copper and tin showed significantly increasing temporal trends.
- Increasing site-specific temporal trends were seen in copper, iron, lead, silver, and tin.
- Sites GBBS (copper, silver, tin) and LERB (copper, silver, tin) each had 3 instances of increasing site-specific temporal trends, indicating that these two sites may be exposed to additional inputs of select trace metals.
- Lake St. Clair appeared to have elevated levels of copper, lead, tin, and zinc compared to other lakes in 2022.
- Lake Ontario appeared to have elevated levels of silver compared to other lakes in 2022.
- Lake Michigan appeared to have elevated levels of iron compared to other lakes in 2022.

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COMMONLY USED ACRONYMS

AOC	Areas of Concern
dw	dry weight
GLERL	Great Lakes Environmental Research Laboratory
GLRI	Great Lakes Restoration Initiative
g	gram
LAMPS	Lakewide Action and Management Plans for the Great Lakes
MDL	method detection limit
MWP	Mussel Watch Program
NCCOS	National Centers for Coastal Ocean Science
NOAA	National Oceanic and Atmospheric Administration
NS&T	National Status and Trends
SD	standard deviation
µg	microgram
U.S.	United States
U.S. EPA	United States Environmental Protection Agency
U.S. FDA	United States Food and Drug Administration

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. 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.

The MWP monitoring sites are located along all U.S. coastlines, including Alaska, the Great Lakes, Hawaii, and in territories such as Puerto Rico. As a result of this extensive 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 (*Dreissena polymorpha*) and quagga mussels (*Dreissena rostriformis bugensis*) 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) redesigned 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 strengthen 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).

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 enables 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

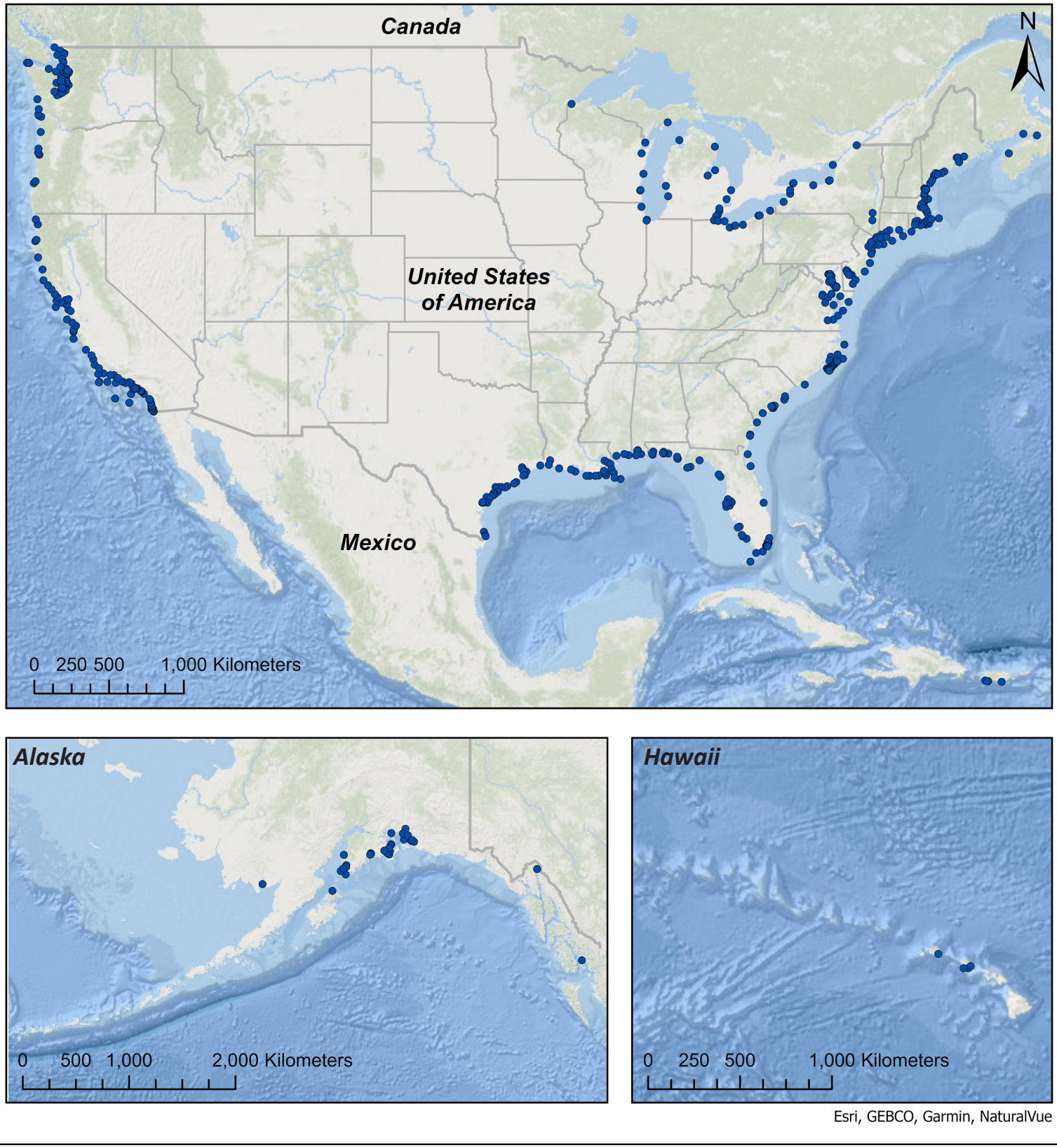


Figure 1. National Mussel Watch long-term monitoring sites.

2.0 INTRODUCTION

The Mussel Watch Program (MWP) maintains a network of long-term monitoring sites within the Laurentian Great Lakes, which were analyzed in this study. Monitoring sites (from west to east) included the freshwater coastline of Wisconsin, Illinois, Indiana, Michigan, Ohio, Pennsylvania, and New York. The Laurentian Great Lakes constitute one of the largest freshwater systems in the world, with an estimated 21% of Earth's surface freshwater (~ 90% of the United States' freshwater; Fields, 2005; Sponberg, 2009). The Great Lakes cover a drainage basin of over 244,000 km^2 (94,000 mi^2), which is home to over 35 million people (Breffle et al., 2013; Danz et al., 2007). The region's watershed, which drains 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 Lakes' 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 driver behind coastal water quality and ecosystem impairment within the Great Lakes Basin (Hull et al., 2015; Kiesling et al., 2019).

Since 1992, NOAA has monitored bivalves and sediment in the Great Lakes Basin as part of its National Status and Trends (NS&T) Program, with the MWP serving as a principal means of monitoring and assessment, establishing one of the most spatially robust biomonitoring data sets in the region (Edwards et al., 2024a, 2024b; Kimbrough et al., 2008, 2018). Additionally, supplemental research and monitoring efforts were conducted within the umbrella of the Great Lakes Restoration Initiative (GLRI) from 2010 to 2018. Regional monitoring and research programs, specifically the Lakewide Action and Management Plans for the Great Lakes (LAMPS) and the NOAA Great Lakes Environmental Research Laboratory (GLERL), provide additional data in the Great Lakes Basin that can be used to supplement data collected through the MWP and independently support local management efforts in addition to providing direction on defining Areas of Concern (AOC) in the region (Goodspeed, et al., 2014). Programs such as these, which value the collection and assessment of long-term water quality monitoring, have provided relevant data and information to coastal managers and the scientific community with a primary focus on legacy organic contaminants. These legacy pollutants are routinely monitored and regulated and include trace elements (i.e., trace metals) and persistent organic pollutants such as butyltins, chlordanes, chlorobenzenes, dichlorodiphenyltrichloroethane (DDTs), dieldrins, endosulfans, hexachlorocyclohexane (HCHs), mirex, polycyclic aromatic hydrocarbons (PAHs), and polychlorinated biphenyls (PCBs). Trace metals occur naturally in Earth's crust, so their sources of contamination are not always anthropogenic or of concern, and high concentrations can often be linked to natural enrichment or biogenic sources.

In 2022, the MWP leveraged partnerships with federal, state, academic, and nonprofit collaborators to conduct a comprehensive assessment of contaminants within the Great Lakes Basin (Figure 2). The study was designed within the MWP regional monitoring approach framework, which balances the importance of a large geographic site network with the cost of broad contaminant surveys. The objectives of this study were to 1) assess the magnitude and distribution of trace metals historically assessed by the MWP; 2) compare contamination in the Great Lakes Basin in 2022 to previous studies in the same region and nationally; and 3) make data electronically available to coastal resource managers in the Great Lakes region. 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 trace metals detected in dreissenid mussels within the Great Lakes Basin. This information is important for developing integrated approaches for contaminant prioritization, mitigation strategies, ecological risk assessments, and suitable mechanisms for identifying sources of detected trace metals that are likely "hotspots" within the Great Lakes Basin.

3.0 METHODS

3.1 Study Area and Sampling Design

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 trace metals in 2022. Collectively, 30 sites were sampled in Lake Michigan, 7 sites were sampled in Lake Huron, 2 sites were sampled in Lake

Methods

St. Clair, 18 sites were sampled in Lake Erie, 1 site was sampled in the Niagara River, and 8 sites were sampled in Lake Ontario and the St. Lawrence River.

Long-term MWP Great Lakes monitoring sites were selected in locations with abundant bivalve populations to enable repetitive sampling and to provide information about the degree of chemical contamination in the Great Lakes Basin area over time. These sites were not randomly selected nor designed to target specific pollution sources. Additional sites in the Great Lakes Basin were purposefully selected in the U.S. Environmental Protection Agency's (U.S. EPA) designated Areas of Concern (AOC: areas designated for restoration due in part to historical environmental contamination; Hartig et al., 2020; e.g., Niagara River AOC, Milwaukee Estuary AOC), 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) 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.

Sample collection was successful at 66 out of 72 attempted sites and was conducted with the support of local collaborators including the NOAA Great Lakes Environmental Research Laboratory (GLERL), members of the NOAA Corps to serve as scuba divers, and personnel working to support the Lakewide Action and Management Plans for the Great Lakes (LAMPS) through the U.S. EPA, following standard protocols utilized by the MWP (Apeti et al., 2012) in August – September 2022 (Figure 2). For bivalve collection, divers harvested *in situ* dreissenid mussels from established populations. Once collected, 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). In this study, dreissenid mussels from 66 sites were collected and analyzed for 14 trace metals.

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 the analytical methods for trace metals analyzed in this study by TDI Brooks International can be found in Kimbrough et al. (2007). Laboratory results were subjected to regular NS&T performance-based quality assessment and quality control procedures to ensure data accuracy and precision. Trace metals data are presented in micrograms per gram of dry tissue (µg/g dw). Chemical contaminant analyses have been conducted by multiple laboratories over time; however, inter-laboratory calibrations have been conducted to ensure data quality and consistency, and no significant concerns regarding data comparability have been identified (Christopher et al., 2012). In the MWP, trace metal contaminants have been analyzed by TDI-Brooks International since 2000, before being taken over by the NOAA Ecotoxicology Branch at the Hollings Marine Laboratory in 2014. 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://arcg.is/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 4.5.0 (R Core Team, 2025), Microsoft Excel (Microsoft Corporation, 2024), ArcGIS (ESRI, 2011), and JMP12 Software (JMP, 2022). Concentrations of all compounds were blank corrected and any values below the Method Detection Limit (MDL) were categorized as undetected and were assigned a value of 0. The MDL is defined as the lowest concentration able to be detected by the analytical instrument or method with 99% confidence by the analytical instrument or method (as defined by the EPA in CFR Title 40 Appendix B to Part 136). Temporal trends for each metal from sites in 2022 were statistically analyzed at site and regional scales using a Spearman's Rank test. Spearman's Rank correlation was used to evaluate whether concentrations co-varied predictably as a function of time (Zar, 1999), thereby indicating a significant increasing or decreasing temporal trend. The 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 concentration. Regional temporal trends were also depicted using a three-point moving average of the yearly mean for all sites combined to smooth out short-term fluctuations and better identify trends. Additionally, all data from sites assessed in the Great Lakes from 1992 - 2022 were compared using a Kruskal-Wallis test followed by a Dunn's post hoc test to determine contamination differences of trace metals between lakes.

Methods

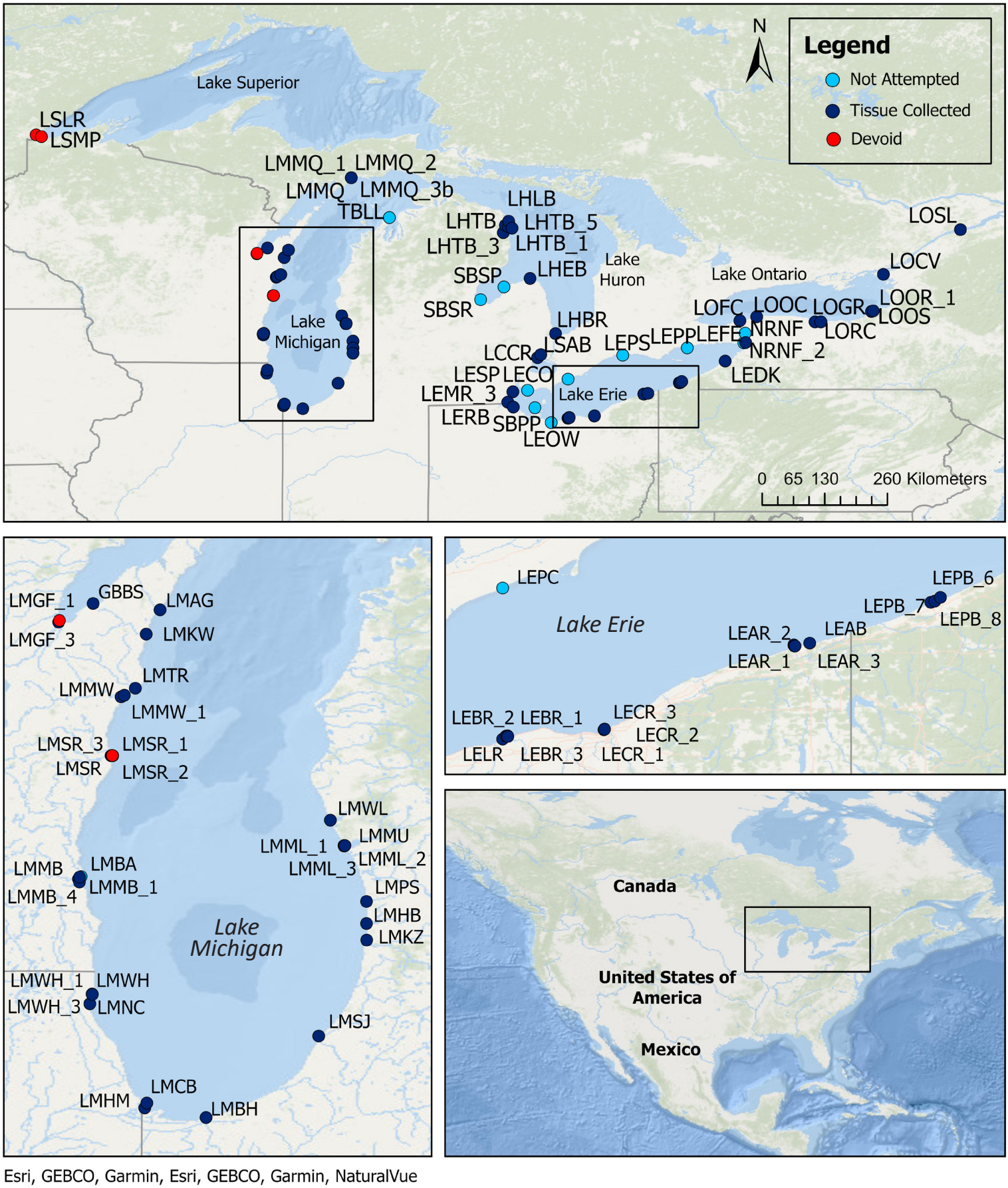


Figure 2. Map of Mussel Watch sites in the Great Lakes Basin sampled in 2022 and their respective 2022 collection status. "Devoid" indicates that the site was attempted for sampling but no mussels were present.

Methods

Additionally, contamination levels detected at sites in 2022 were compared to regional long-term NOAA NS&T monitoring data. The NS&T data used for comparison comprises dreissenid mussel tissue sample concentrations collected by the national MWP since the program's establishment in the Great Lakes Basin in 1992. This comparison was performed using a multivariate cluster analysis (the Ward method) on the concentrations detected for each compound. This analysis clusters contaminant concentrations into significantly different groups, such that values within a group are more like each other than any other value of a different group. The categories derived from the clusters were not representative measurements that have exceeded any regulatory thresholds; rather, they denoted concentrations that were significantly higher than the preceding category. For each contaminant class, sites were clustered into three groups to represent high, medium, and low contamination relative to all data collected within the MWP. If the resulting high cluster had fewer than 10 data points, those points were temporarily removed as "outliers" (due to the large sample size of this dataset), the data were re-clustered, and the outliers were added back in to the high cluster before the resulting boxplots were created. Clustering results were reported in a map on the Summary page of each compound. Any instances of "not detected" contaminants were differentiated from low contamination on the map.

To provide further context, cluster analysis results for all metals at each site assessed in this study were summarized in two heatmaps showing (1) cluster results of the 2022 study sites respective to concentrations found within the sites assessed in this study, and (2) cluster results of the 2022 study sites respective to regional long-term NOAA NS&T monitoring data. The clustering process for comparing contaminant data between sites only analyzed within the 2022 Great Lakes Basin study was similar to the process described for the regionwide, historic clustering, with two exceptions to account for the smaller sample size. The concentrations were similarly clustered into groups of different contamination levels, but "outliers" were not removed (as described previously) and sites were not forced into three clusters (high, medium, and low) if they statistically defaulted to fewer than three clusters, in which case they were labeled as low and high groups.

After this cluster analysis, cluster values of each compound for each site for both datasets were summed and normalized by the maximum value possible at that site, taking the number of compounds assessed at each site and the maximum cluster value possible based on the cluster analysis results into account (i.e., (sum cluster values)/(# chemical classes analyzed * maximum cluster value possible) * 100) (Table A2; Table A3). These normalized values were then clustered using the Ward method to generate groups of sites with statistically different degrees of overall contamination and were presented in a map. The 2022 sites only dataset was, again, treated slightly differently than the regionwide, historic dataset due to the difference in sample size. The regionwide, historic dataset was clustered into five groups of sites with statistically different degrees of overall contamination (not detected, low, medium, high, and very high). In contrast, the 2022 sites only dataset was not forced into five clusters if it statistically defaulted to fewer than five clusters, in which case they were labeled just as not detected, low, medium, and high groups.

*Table 1. Mussel Watch sites selected for the 2022 Great Lakes survey. ● = site was analyzed for metals in 2022 in dreissenid mussel tissue, **X** = sampling was attempted but collection was unsuccessful (i.e., "Devoid"), ●* sampling was successful but not enough tissue for metals analysis, no icon = sampling was not attempted in 2022. Int = International site collected by collaborator. An attempt was made to sample 72 / 87 sites in the Great Lakes Basin region in 2022, but only 66 of those 72 sites yielded bivalves to be analyzed for metals.*

Site	State	Lake	Specific Location	Latitude	Longitude	Study Type	Tissue Sampled?
LSLR	MN	Lake Superior	St Louis River	46.74652	-92.12393		**X**
LSMP	MN	Lake Superior	Minnesota Point	46.71094	-92.02261	Long-term	**X**
GBBS	WI	Lake Michigan	Green Bay Bayshore Park	44.6373	-87.80862	Long-term	●
LMAG	WI	Lake Michigan	Algoma	44.60111	-87.41889		●
LMBA	WI	Lake Michigan	Milwaukee Alternate	43.0462	-87.8793	Long-term	
LMBH	IN	Lake Michigan	Burns Harbor	41.64187	-87.1508		●
LMCB	IN	Lake Michigan	Calumet Breakwater	41.72725	-87.49527	Long-term	●
LMGF_1	WI	Lake Michigan	Green Bay Fox River	44.53901	-88.00517		**X**
LMGF_3	WI	Lake Michigan	Green Bay Fox River	44.52942	-88.00944		●
LMHB	MI	Lake Michigan	Holland Breakwater	42.77325	-86.21493	Long-term	●
LMHM	IN	Lake Michigan	Hammond Marina	41.69863	-87.50772	Long-term	●

Methods

*Table 1 cont. Mussel Watch sites selected for the 2022 Great Lakes survey. ● = site was analyzed for metals in 2022 in dreissenid mussel tissue, **X** = sampling was attempted but collection was unsuccessful (i.e., "Devoid"), ●* sampling was successful but not enough tissue for metals analysis, no icon = sampling was not attempted in 2022. Int = International site collected by collaborator. An attempt was made to sample 72 / 87 sites in the Great Lakes Basin region in 2022, but only 66 of those 72 sites yielded bivalves to be analyzed for metals.*

Site	State	Lake	Specific Location	Latitude	Longitude	Study Type	Tissue Sampled?
LMKW	WI	Lake Michigan	Kewaunee River	44.4591	-87.49912		●
LMKZ	MI	Lake Michigan	Kalamazoo River	42.6766	-86.2141		●
LMMB	WI	Lake Michigan	Milwaukee Bay	43.0321	-87.89504	Long-term	●
LMMB_1	WI	Lake Michigan	Milwaukee Bay	43.01425	-87.88818		●
LMMB_4	WI	Lake Michigan	Milwaukee Bay	43.0432	-87.88784		●
LMML_1	MI	Lake Michigan	Muskegon Lake	43.22719	-86.34676		●
LMML_2	MI	Lake Michigan	Muskegon Lake	43.22396	-86.34513		●
LMML_3	MI	Lake Michigan	Muskegon Lake	43.22793	-86.34193		●
LMMQ	MI	Lake Michigan	Manistique River	45.94578	-86.2497		●
LMMQ_1	MI	Lake Michigan	Manistique River	45.94543	-86.24798		●*
LMMQ_2	MI	Lake Michigan	Manistique River	45.94655	-86.24705		●*
LMMQ_3b	MI	Lake Michigan	Manistique River	45.94543	-86.24913		●
LMMU	MI	Lake Michigan	Muskegon	43.22776	-86.34655	Long-term	
LMMW	WI	Lake Michigan	Manitowoc	44.09338	-87.64553		●
LMMW_1	WI	Lake Michigan	Manitowoc	44.10377	-87.62707		●
LMNC	IL	Lake Michigan	North Chicago	42.30469	-87.82731	Long-term	●
LMPS	MI	Lake Michigan	Port Sheldon	42.90167	-86.21542		●
LMSJ	MI	Lake Michigan	Saint Joseph River	42.11651	-86.49393		●
LMSR	WI	Lake Michigan	Sheboygan River	43.7516	-87.69733		**X**
LMSR_1	WI	Lake Michigan	Sheboygan River	43.75085	-87.69489		●
LMSR_2	WI	Lake Michigan	Sheboygan River	43.75121	-87.70107		●
LMSR_3	WI	Lake Michigan	Sheboygan River	43.75066	-87.70377		●
LMTR	WI	Lake Michigan	Two Rivers	44.14326	-87.56232		●
LMWH	IL	Lake Michigan	Waukegan Harbor	42.36269	-87.81498		●
LMWH_1	IL	Lake Michigan	Waukegan Harbor	42.36146	-87.81448		●
LMWH_3	IL	Lake Michigan	Waukegan Harbor	42.36038	-87.81357		●
LMWL	MI	Lake Michigan	White Lake	43.37531	-86.4264		●
TBLL	MI	Lake Michigan	Traverse Bay Leelanau State Park	45.2065	-85.53763	Long-term	
LHBR	MI	Lake Huron	Black River Canal	43.04306	-82.43775	Long-term	●
LHEB	MI	Lake Huron	Eagle Bay (Port Austin)	44.07058	-82.91579		●
LHLB	MI	Lake Huron	Lincoln Bay (Rockport)	45.1354	-83.31392		●
LHTB	MI	Lake Huron	Thunder Bay	44.92219	-83.4135	Long-term	●
LHTB_1	MI	Lake Huron	Thunder Bay	45.06152	-83.37555		●
LHTB_3	MI	Lake Huron	Thunder Bay	45.00603	-83.24983		●
LHTB_5	MI	Lake Huron	Thunder Bay	45.03468	-83.31802		●
SBSP	MI	Lake Huron	Saginaw Bay Sandpoint	43.9098	-83.4002	Long-term	
SBSR	MI	Lake Huron	Saginaw Bay Saginaw River	43.67301	-83.83523	Long-term	

Methods

*Table 1 cont. Mussel Watch sites selected for the 2022 Great Lakes survey. ● = site was analyzed for metals in 2022 in dreissenid mussel tissue, **X** = sampling was attempted but collection was unsuccessful (i.e., "Devoid"), ●* sampling was successful but not enough tissue for metals analysis, no icon = sampling was not attempted in 2022. Int = International site collected by collaborator. An attempt was made to sample 72 / 87 sites in the Great Lakes Basin region in 2022, but only 66 of those 72 sites yielded bivalves to be analyzed for metals.*

Site	State	Lake	Specific Location	Latitude	Longitude	Study Type	Tissue Sampled?
LCCR	MI	Lake St Clair	Clinton River	42.59448	-82.77219		●
LSAB	MI	Lake St. Clair	Anchor Bay	42.64916	-82.71128	Long-term	●
LEAB	OH	Lake Erie	Ashtabula	41.92554	-80.71922	Long-term	●
LEAR_1	OH	Lake Erie	Ashtabula River	41.9118	-80.79386		●
LEAR_2	OH	Lake Erie	Ashtabula River	41.91751	-80.7961		●
LEAR_3	OH	Lake Erie	Ashtabula River	41.91193	-80.79009		●
LEBR_1	OH	Lake Erie	Black River	41.47349	-82.18278		●
LEBR_2	OH	Lake Erie	Black River	41.47731	-82.18349		●
LEBR_3	OH	Lake Erie	Black River	41.47581	-82.19013		●
LECO	Int	Lake Erie	Colchester	41.983	-82.961	Long-term	
LECR_1	OH	Lake Erie	Cuyahoga River	41.5082	-81.71321		●
LECR_2	OH	Lake Erie	Cuyahoga River	41.50961	-81.71366		●
LECR_3	OH	Lake Erie	Cuyahoga River	41.50642	-81.71595		●
LEDK	NY	Lake Erie	Dunkirk	42.52973	-79.27715	Long-term	●
LEFE	Int	Lake Erie	Fort Erie	42.872	-78.93	Long-term	
LELR	OH	Lake Erie	Lorain	41.46119	-82.207	Long-term	●
LEMR_3	OH	Lake Erie	Maumee River	41.76183	-83.32868		●
LEOW	OH	Lake Erie	Old Woman Creek	41.38447	-82.51771	Long-term	
LEPB_6	PA	Lake Erie	Presque Isle Bay	42.12978	-80.11325		●
LEPB_7	PA	Lake Erie	Presque Isle Bay	42.12347	-80.13227		●
LEPB_8	PA	Lake Erie	Presque Isle Bay	42.14758	-80.08572		●
LEPC	Int	Lake Erie	Port Crewe	42.193	-82.207	Long-term	
LEPP	Int	Lake Erie	Peacock Point	42.775	-79.982	Long-term	
LEPS	Int	Lake Erie	Port Stanley	42.638	-81.186	Long-term	
LERB	OH	Lake Erie	Reno Beach	41.67351	-83.22718	Long-term	●
LESP	MI	Lake Erie	Stony Point	41.95752	-83.23349	Long-term	●
NRNF	NY	Lake Erie	Niagara River Niagara Falls	43.05161	-78.90188	Long-term	
NRNF_2	NY	Lake Erie	Niagara River Niagara Falls	42.87422	-78.89532		●
SBPP	OH	Lake Erie	Peach Orchard Pt.	41.65987	-82.82505	Long-term	
LOCV	NY	Lake Ontario	Cape Vincent	44.14457	-76.32458	Long-term	●
LOFC	NY	Lake Ontario	Fourmile Creek	43.28401	-79.00198		●
LOGR	NY	Lake Ontario	Genesee River	43.25997	-77.60244		●
LOOC	NY	Lake Ontario	Olcott	43.35521	-78.68813	Long-term	●
LOOR_1	NY	Lake Ontario	Oswego River	43.46526	-76.51595		●
LOOS	NY	Lake Ontario	Oswego	43.45281	-76.55081	Long-term	●
LORC	NY	Lake Ontario	Rochester	43.25983	-77.49559	Long-term	●
LOSL	NY	Lake Ontario	St Lawrence River	44.98034	-74.89086		●

4.0 RESULTS - ALUMINUM (Al)

4.1 Aluminum Chemical Description

Aluminum is widely distributed in the environment and is the most abundant metal in the Earth's crust (ATSDR, 2008). As a common crustal element not bioaccumulated to a significant degree in shellfish, aluminum was measured in this study, in part, to help assess whether elevated concentrations of other metals were associated with tissue uptake or with ingestion of contaminated particles (LeBlanc et al., 2011). Aluminum is highly reactive and is never found as a free metal; instead, it is combined with other elements, mostly oxygen, silicon, and fluorine (LeBlanc et al., 2011). These compounds are typically found in soil, minerals, rocks, and clays (LeBlanc et al., 2011). Aluminum occurs naturally in soil, water, and air. High levels in the environment can be caused by the mining and processing of aluminum ores or the production of aluminum metals, alloys, and compounds. Small amounts of aluminum are released into the environment from coal-fired power plants and incinerators. Products that contain aluminum include beverage cans, pots and pans, airplanes, siding and roofing, foil, explosives, fireworks, antacids, aspirin, food additives, antiperspirants, and cosmetics (ATSDR, 2008).

Human exposure to aluminum is widespread and typically occurs by the consumption of processed foods including flour, baking powder, coloring agents, or anticaking agents (ATSDR, 2008). An average U.S. adult consumes approximately 7-9 mg of aluminum per day (ATSDR, 2008). Occupational exposure is the primary cause of exposure to high levels of aluminum in humans. High levels of exposure can result in lung problems (largely addressed by wearing masks) and decreased nervous system function. Accretion of aluminum happens in humans with kidney disease, as it is not removed through the urinary tract like normal, and can sometimes result in the development of bone or brain diseases (ATSDR, 2008).

Aluminum cannot be degraded in the environment; it can only change its form, attach to or separate from other particles, or undergo atmospheric deposition (ATSDR, 2008). However, very small aluminum particles can stay in the air for many days. Most aluminum-containing compounds do not dissolve in water unless the water is acidic or very alkaline (ATSDR, 2008).

4.2 Magnitude and Distribution of Aluminum in Dreissenid Mussel Tissue in 2022

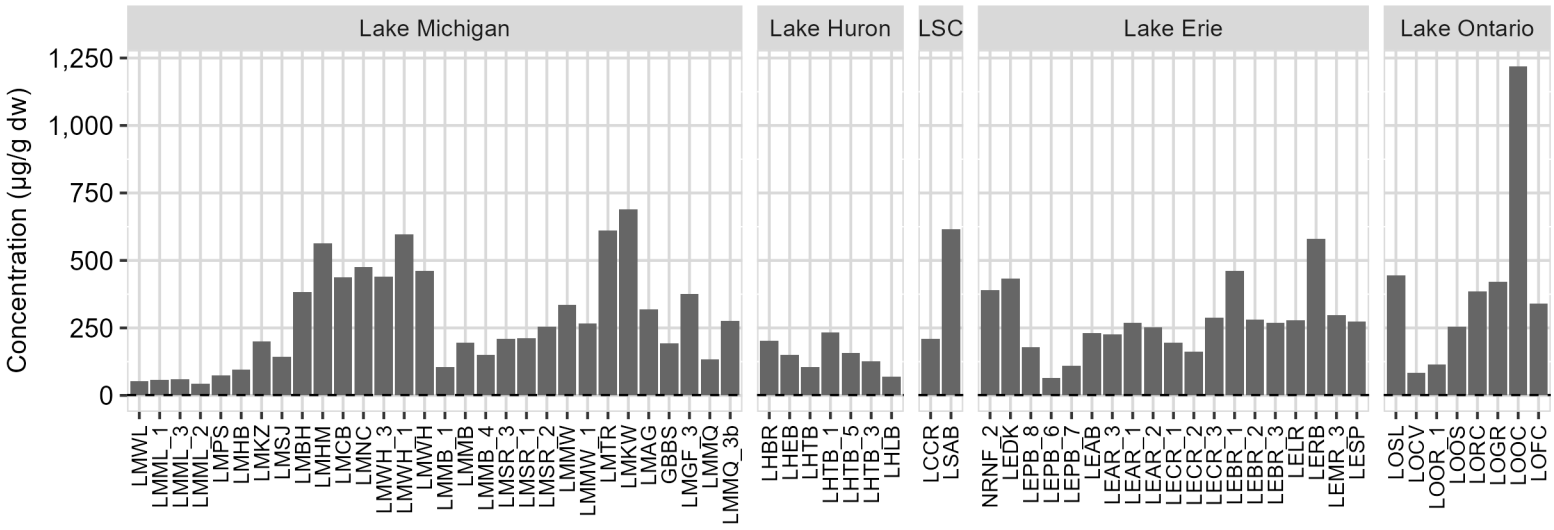


Figure 3. Bar graph showing magnitude of aluminum concentrations (µg/g dw) detected in dreissenid mussel tissue in the Great Lakes Basin in 2022. Dotted line represents the minimum weight corrected MDL. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair.

Results - Aluminum (Al)

4.3 Historical Context of Aluminum Magnitude and Distribution in Dreissenid Mussel Tissue

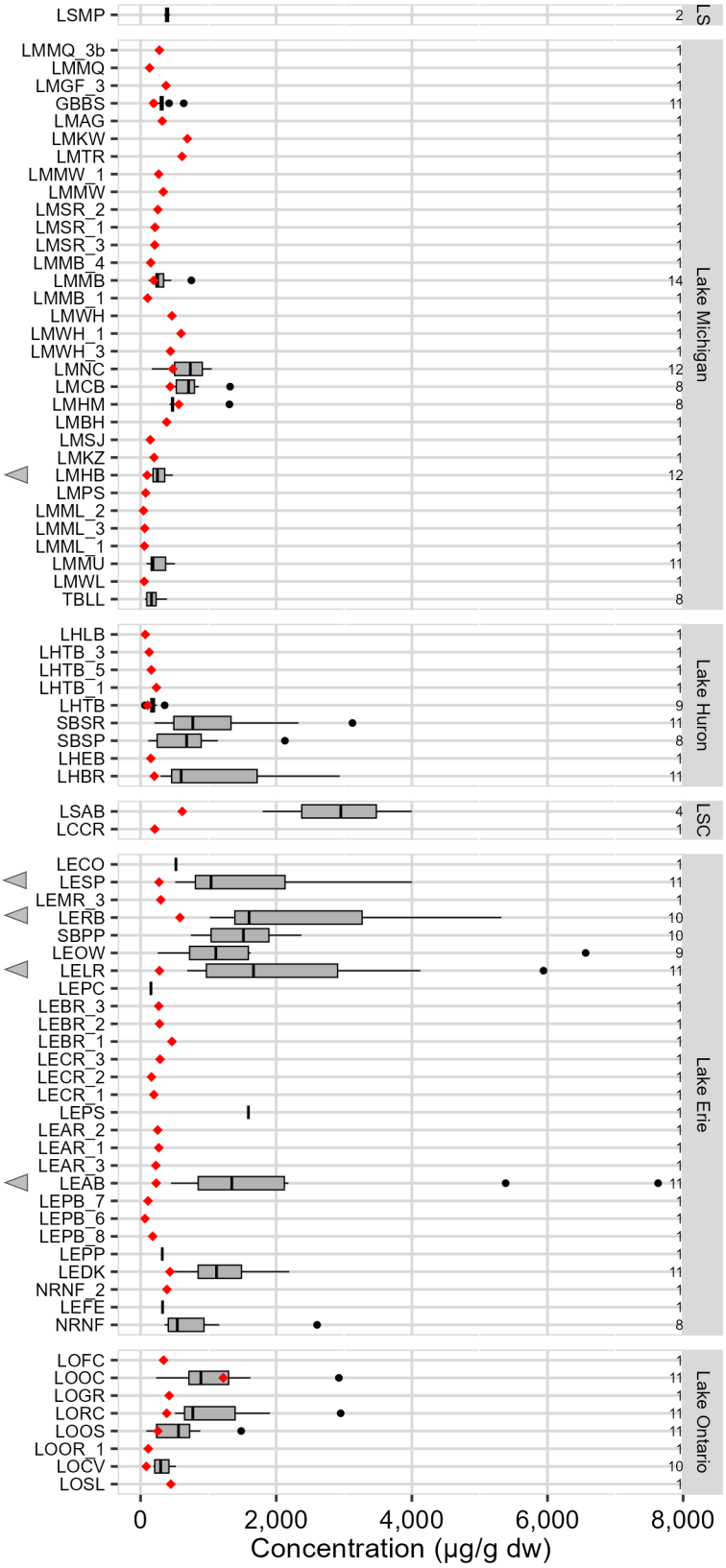


Figure 4. Aluminum concentrations (µg/g dw) in dreissenid mussel tissue in 2022 (red diamonds) compared to boxplots of the historic aluminum concentrations for each site (1992- 2008) and results of the trend analysis showing significant increasing (▻) or decreasing (◅) site-specific contamination trends over time. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair; LS = Lake Superior. The number of years each site has been sampled since 1992 is noted at the right of the plot for each site.

Results - Aluminum (Al)

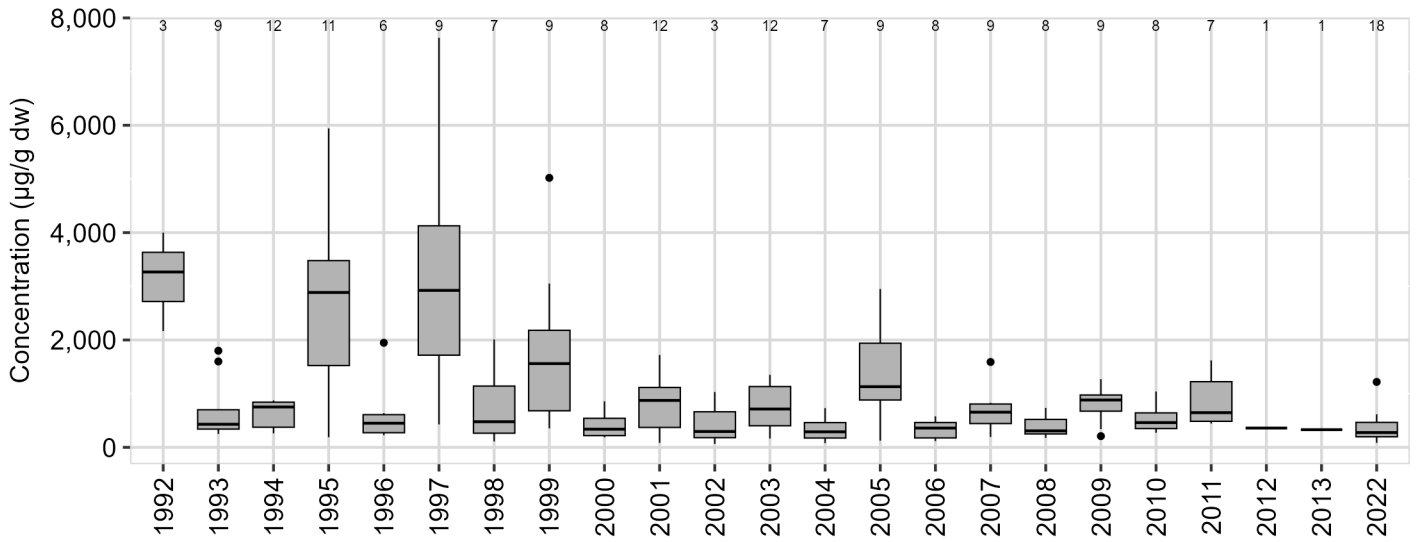


Figure 5. Boxplots representing the historic aluminum concentrations (µg/g dw) in dreissenid mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot.

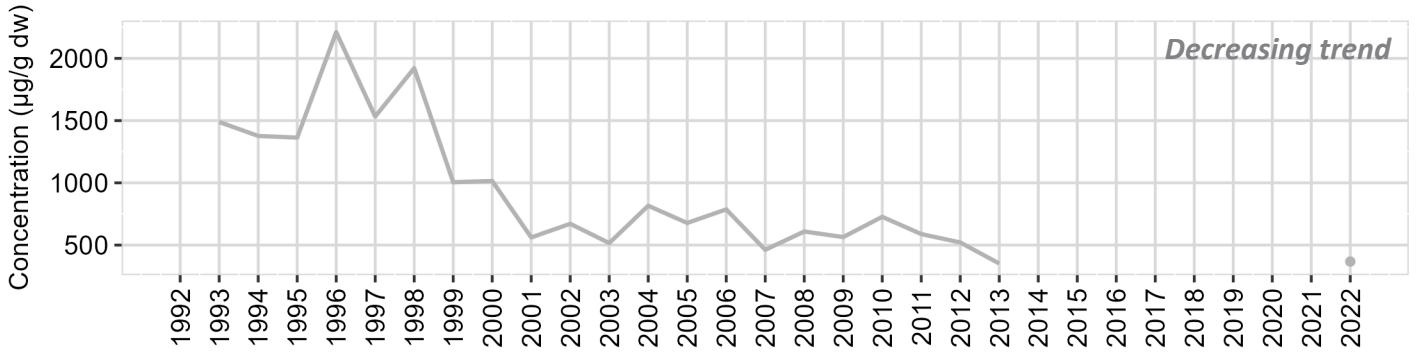


Figure 6. Three-point moving average of the yearly mean aluminum concentrations (µg/g dw) in dreissenid mussel tissue in the sites analyzed in this study. Average concentration for 2022 is included as a standalone point for reference.

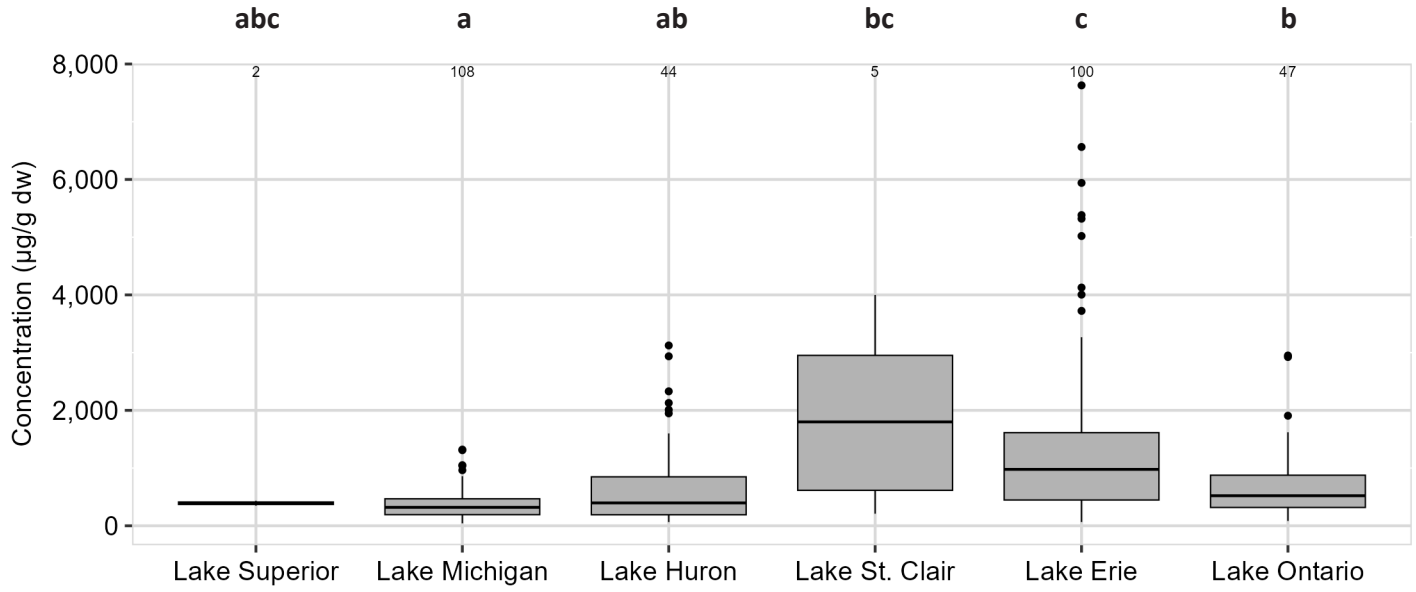


Figure 7. Aluminum concentrations (µg/g dw) measured in dreissenid mussel tissue in the Great Lakes from 1992 - 2022. Lakes and major rivers including connecting channels are listed geographically from west to east across the Great Lakes Basin. The number of samples collected in each lake is noted at the top of the plot. Letters above each lake indicate significant differences (at α = 0.05).

Results - Aluminum (Al)

4.4 Aluminum Summary

2022 Dreissenid Mussel Tissue Results:

- Al was detected at 100% of 66 sites surveyed (Figure 3).
- Al concentration descriptive statistics (Figure 3):
  - Range: 42.80 – 1,218.55 µg/g dw
  - Minimum Al concentration was detected at LMML_2
  - Maximum Al concentration was detected at LOOC
  - Median: 254.00 µg/g dw
  - Mean ± SD: 284.52 ± 197.78 µg/g dw
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster respective to 2022 data only:
  - 67% of sites in low cluster (42.80 - 298.00 µg/g dw)
  - 32% of sites in medium cluster (319.21 - 690.20 µg/g dw)
  - 1% of sites in high cluster (1,218.55 - 1,218.55 µg/g dw)

Historic Context for Dreissenid Mussel Tissue (18 out of 66 sites had historic data to analyze for trend analyses):

- 11% of sites surveyed (2/18) were above their historic median concentrations in 2022 (Figure 4).
- Regionally, there was a significant decreasing temporal trend of Al concentrations in dreissenid mussels in the Great Lakes Basin ($p = 0.01$, $rho = -0.53$) (Figure 5; Figure 6).
- 5 sites (LMHB, LESP, LERB, LELR, LEAB) in the Great Lakes Basin showed significant decreasing temporal trends of Al concentrations at $\alpha = 0.05$ (Figure 4; Table A4).
- 0 sites in the Great Lakes Basin showed significant increasing temporal trends of Al concentrations at $\alpha = 0.05$ (Figure 4; Table A4).
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster compared to historic data (Figure 8):
  - 100% of sites in low cluster (42.80 - 1270.00 µg/g dw)
  - 0% of sites in medium cluster (1310.00 - 2602.64 µg/g dw)
  - 0% of sites in high cluster (2885.06 - 7630.00 µg/g dw)

General Observations:

- Al concentrations were ubiquitous but at comparatively low concentrations in the Great Lakes Basin in dreissenid mussel tissue in 2022, likely reflecting natural ambient concentrations (Figure 8).
- Regionally, Al concentrations have significantly decreased over time in dreissenid mussel tissue, indicating decreasing inputs of Al into the environment (Figure 6).
- The post-hoc analysis evaluating differences between lakes indicates that Lake Superior is a statistical bridge, showing no significant difference to any other lake. In contrast, the other lakes demonstrate a gradient of relationships, being significantly similar to some and significantly different from others. Generally, the highest concentrations of Al appear to be in lakes St. Clair and Erie, possibly due to their proximity to Detroit, MI (Figure 7).

Results - Aluminum (Al)

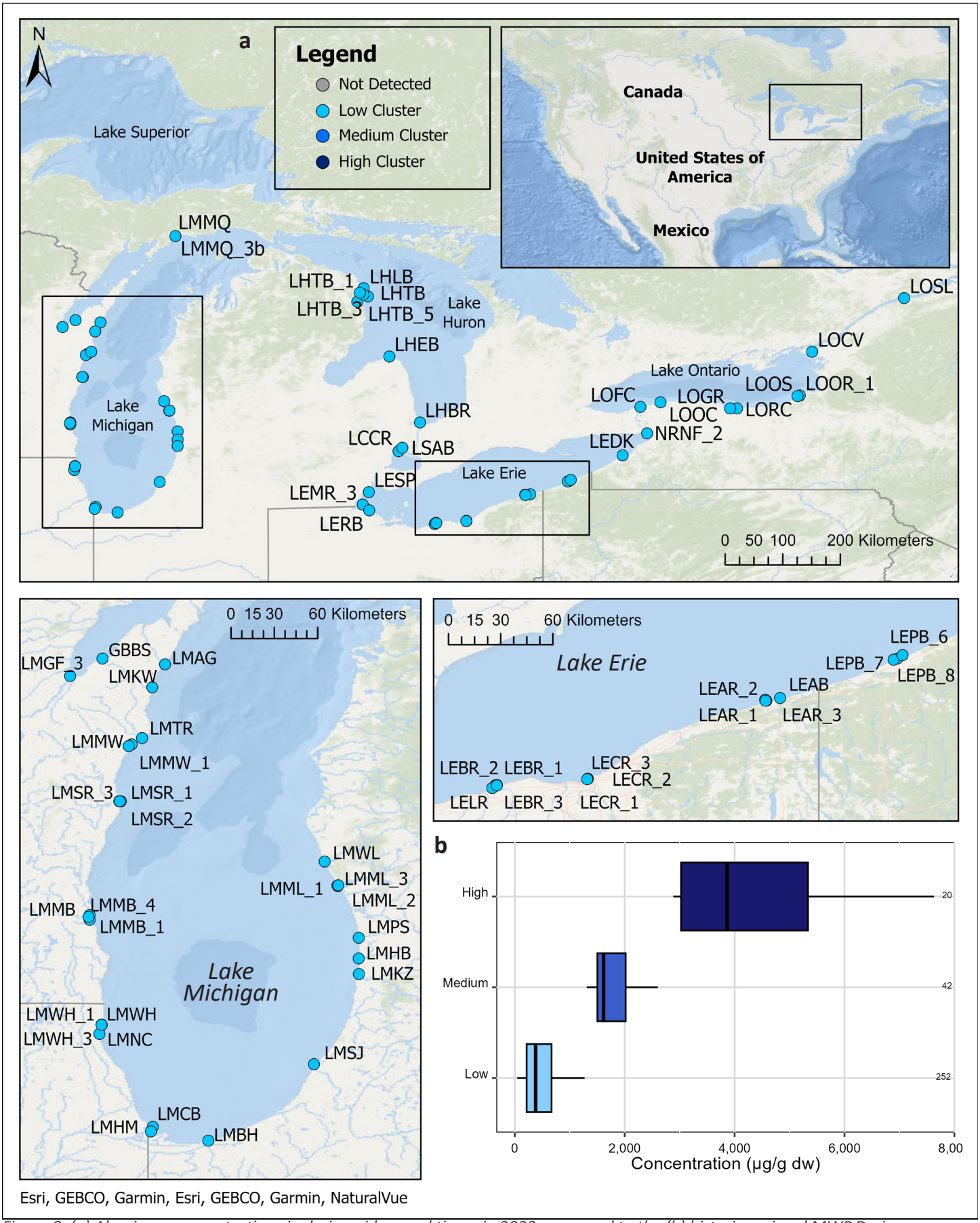


Figure 8. (a) Aluminum concentrations in dreissenid mussel tissue in 2022 compared to the (b) historic regional MWP Dreissena *species aluminum concentrations (µg/g dw). Boxplot colors correspond to cluster groups delineated in the map legend. 81 regional sites sampled between 1992 - 2022 for a total of 314 dreissenid mussel samples.*

5.0 RESULTS - ARSENIC (As)

5.1 Arsenic Chemical Description

Arsenic is found at high levels in the environment as a result of both natural sources and industrial production. It is naturally present in low concentrations in the Earth's crust and in rock, soil, water, and air, and is highly prevalent in the bedrock along the Mid-Atlantic Coast. Additionally, 30% of atmospheric arsenic comes from volcanoes and other natural sources, and a significant source responsible for apparent elevated levels of arsenic in the Nation is natural crustal rock, which varies by region (Welch et al., 1988). Generally, in trend analysis, continuous natural sources are associated with neither decreasing nor increasing trends (Kimbrough et al., 2008). Industrial products that contain arsenic include preserved wood, semiconductors, pesticides, defoliants, pigments, antifouling paints, and veterinary medicines. In recent years, as much as 90% of arsenic used in industrial production was for wood preservation (ATSDR, 2007a). Atmospheric sources of arsenic, including smelting, fossil fuel combustion, power generation, and pesticide application, contribute to high environmental levels of arsenic (ATSDR, 2007a).

Arsenic is an essential trace nutrient for many animals; however, dietary requirements for humans are unknown (Uthus, 1992). Arsenic is toxic at high concentrations to fish, birds and plants. In animals and humans, prolonged chronic exposure is linked to cancer (Goyer, 1986). Inorganic arsenic, the most toxic form, represents approximately 10% of total arsenic in bivalves. Less harmful organic forms, such as arsenobetaine, predominate in seafood (Edmonds and Francesconi, 1977, 1988, 1993; Phillips, 1990; U.S. FDA, 1993). The MWP measures total arsenic, including both the inorganic and the organic forms. Safety guidance levels for arsenic in fish and shellfish are no longer listed by the U.S. FDA (U.S. FDA, 2011).

Centuries of human activities have changed the natural biogeochemical cycle of arsenic, resulting in contamination of land, water, and air. Movement of arsenic into coastal and estuarine water occurs primarily from river runoff and atmospheric deposition (Welch et al., 1988).

5.2 Magnitude and Distribution of Arsenic in Dreissenid Mussel Tissue in 2022

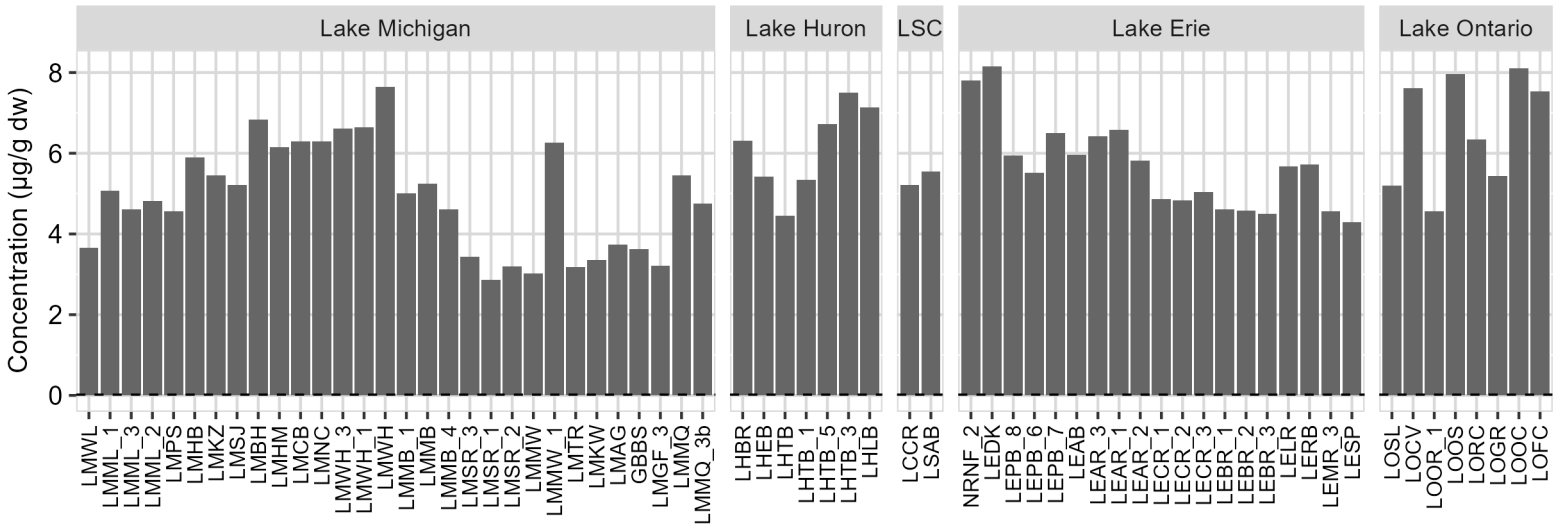


Figure 9. Bar graph showing magnitude of arsenic concentrations (µg/g dw) detected in dreissenid mussel tissue in the Great Lakes Basin in 2022. Dotted line represents the minimum weight corrected MDL. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair.

Results - Arsenic (As)

5.3 Historical Context of Arsenic Magnitude and Distribution in Dreissenid Mussel Tissue

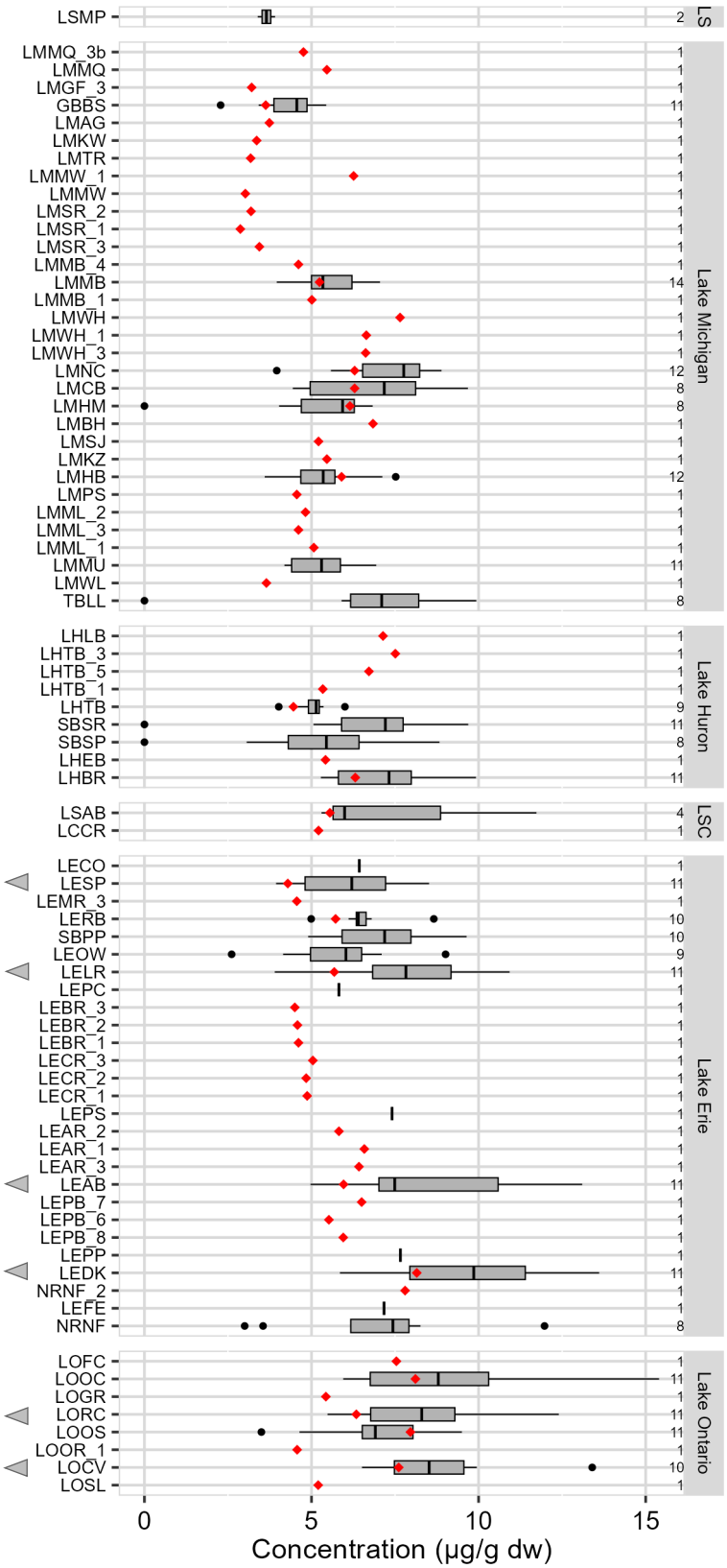


Figure 10. Arsenic concentrations (µg/g dw) in dreissenid mussel tissue in 2022 (red diamonds) compared to boxplots of the historic arsenic concentrations for each site (1992 - 2008) and results of the trend analysis showing significant increasing (▻) or decreasing (◅) site-specific contamination trends over time. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair; LS = Lake Superior. The number of years each site has been sampled since 1992 is noted at the right of the plot for each site.

Results - Arsenic (As)

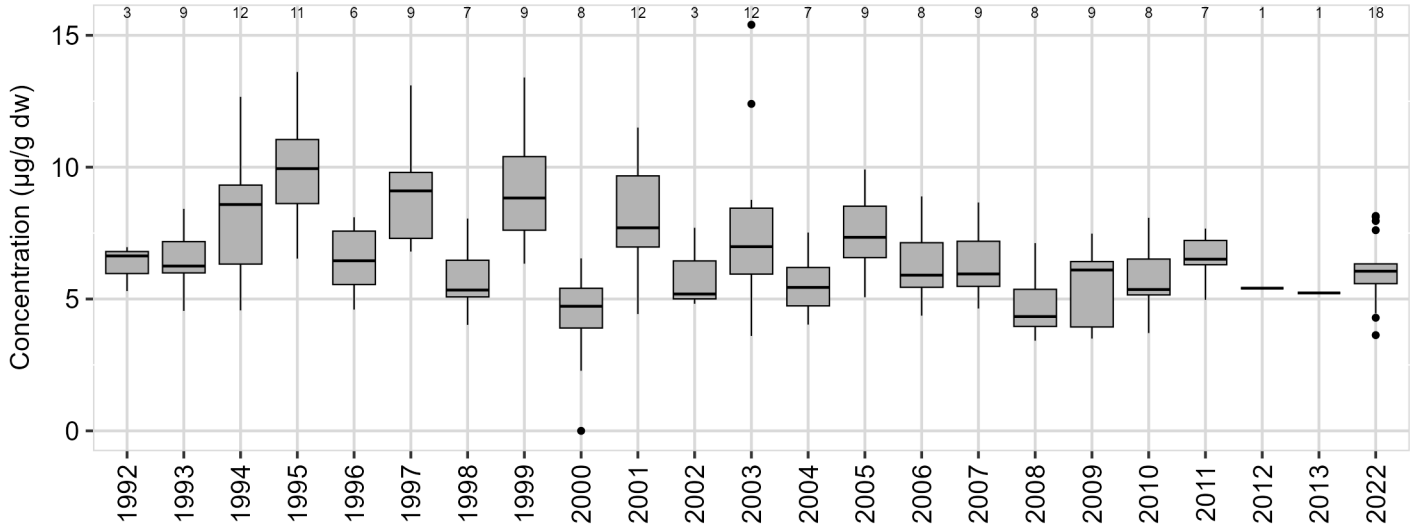


Figure 11. Boxplots representing the historic arsenic concentrations (µg/g dw) in dreissenid mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot.

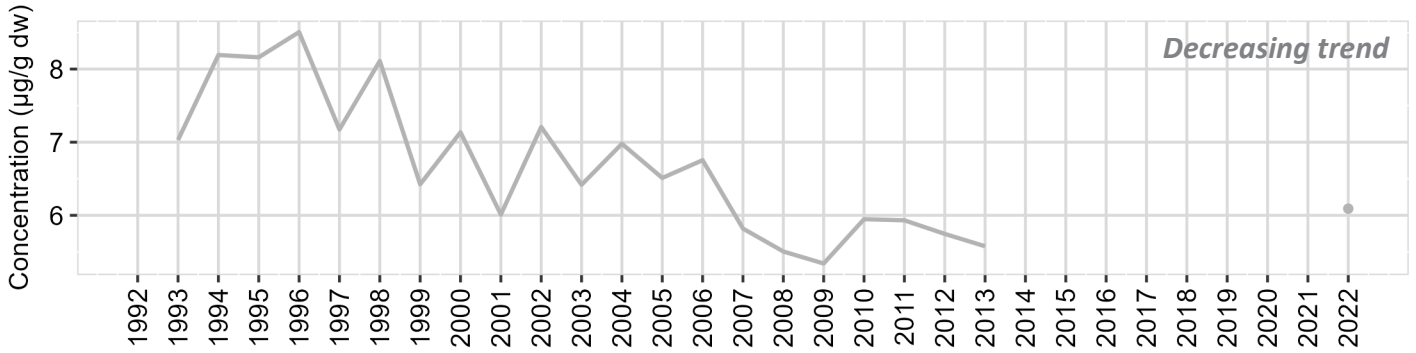


Figure 12. Three-point moving average of the yearly mean arsenic concentrations (µg/g dw) in dreissenid mussel tissue in the sites analyzed in this study. Average concentration for 2022 is included as a standalone point for reference.

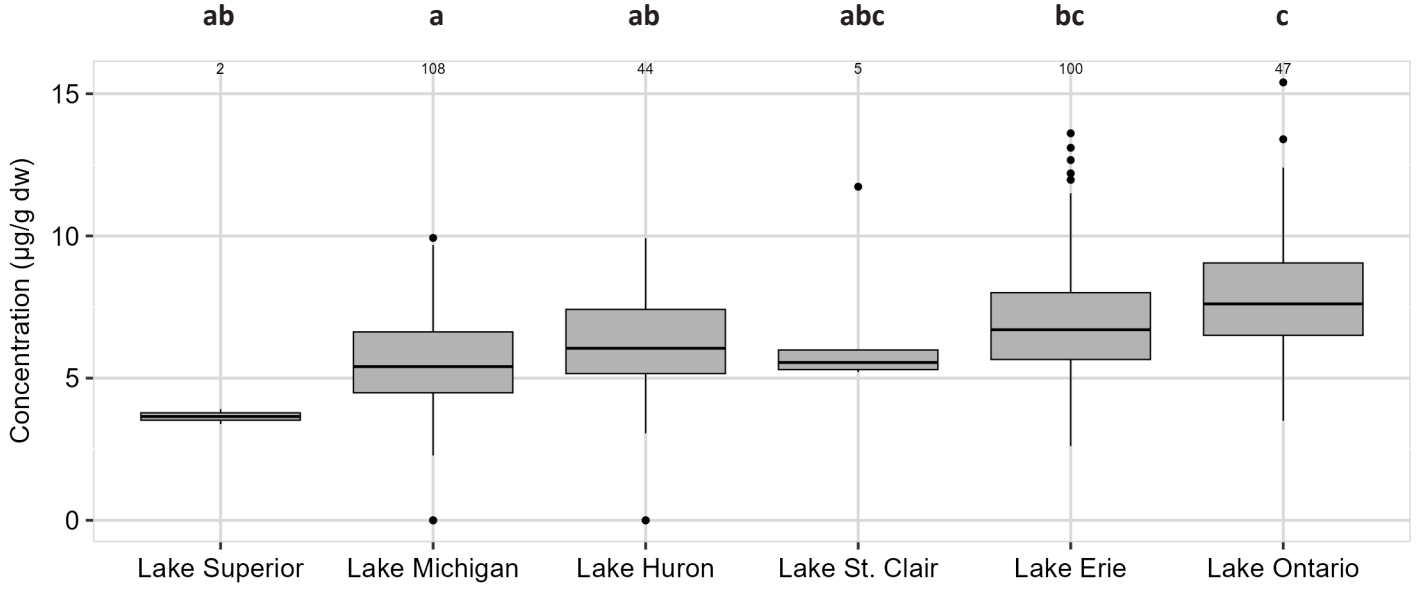


Figure 13. Arsenic concentrations (µg/g dw) measured in dreissenid mussel tissue in the Great Lakes from 1992 - 2022. Lakes and major rivers including connecting channels are listed geographically from west to east across the Great Lakes Basin. The number of samples collected in each lake is noted at the top of the plot. Letters above each lake indicate significant differences (at α = 0.05).

Results - Arsenic (As)

5.4 Arsenic Summary

2022 Dreissenid Mussel Tissue Results:

- As was detected at 100% of 66 sites surveyed (Figure 9).
- As concentration descriptive statistics (Figure 9):
  - Range: 2.87 – 8.15 µg/g dw
  - Minimum As concentration was detected at LMSR_1
  - Maximum As concentration was detected at LEDK
  - Median: 5.42 µg/g dw
  - Mean ± SD: 5.46 ± 1.35 µg/g dw
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster respective to 2022 data only:
  - 15% of sites in low cluster (2.87 - 3.74 µg/g dw)
  - 52% of sites in medium cluster (4.29 - 5.96 µg/g dw)
  - 33% of sites in high cluster (6.15 - 8.15 µg/g dw)

Historic Context for Dreissenid Mussel Tissue (18 out of 66 sites had historic data to analyze for trend analyses):

- 17% of sites surveyed (3/18) were above their historic median concentrations in 2022 (Figure 10).
- Regionally, there was a significant decreasing temporal trend of As concentrations in dreissenid mussels in the Great Lakes Basin ($p = 0.01$, $rho = -0.53$) (Figure 11; Figure 12).
- 6 sites (LESP, LELR, LEAB, LEDK, LORC, LOCV) in the Great Lakes Basin showed significant decreasing temporal trends of As concentrations at $\alpha = 0.05$ (Figure 10; Table A4).
- 0 sites in the Great Lakes Basin showed significant increasing temporal trends of As concentrations at $\alpha = 0.05$ (Figure 10; Table A4).
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster compared to historic data (Figure 14):
  - 47% of sites in low cluster (0.00 - 5.24 µg/g dw)
  - 47% of sites in medium cluster (5.28 - 7.70 µg/g dw)
  - 6% of sites in high cluster (7.76 - 15.40 µg/g dw)

General Observations:

- As concentrations were ubiquitous and at comparatively low to medium concentrations in the Great Lakes Basin in dreissenid mussel tissue in 2022, likely reflecting naturally higher ambient concentrations (Figure 14).
  - A few instances of comparatively high concentrations were found in Lake Erie (LEDK, NRNF_2) and Lake Ontario (LOOC, LOOS), possibly indicating local inputs of As in that area (Figure 14).
- Regionally, As concentrations have significantly decreased over time in dreissenid mussel tissue, indicating decreasing inputs of As into the environment (Figure 12).
- The post-hoc analysis evaluating differences between lakes indicates that Lake St. Clair is a statistical bridge, showing no significant difference to any other lake. In contrast, the other lakes demonstrate a gradient of relationships, being significantly similar to some and significantly different from others. Generally, there appears to be a trend of As increasing in concentration going from west to east across the Great Lakes Basin, potentially indicating accumulation or differences in regional geology (Figure 13).

Results - Arsenic (As)

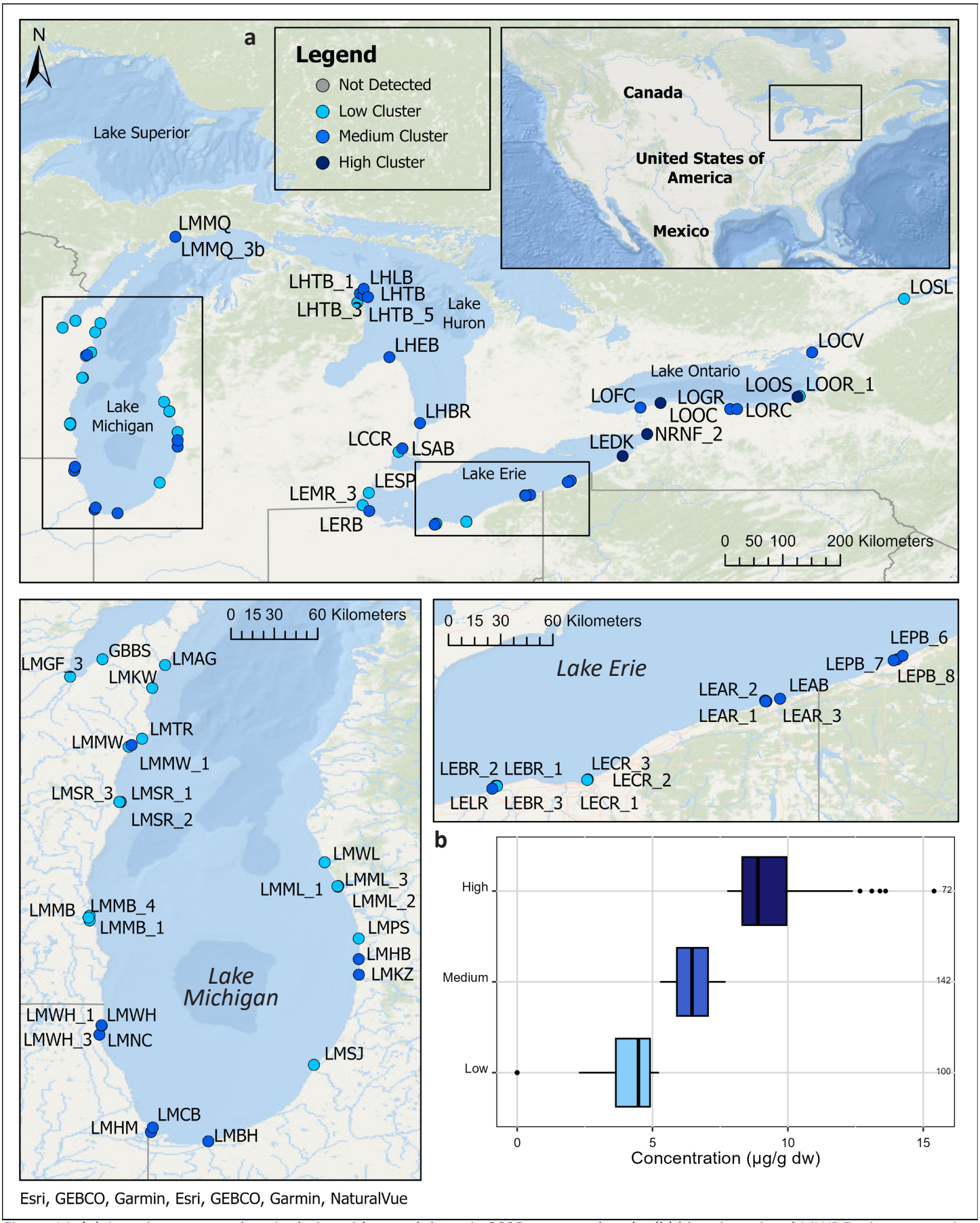


Figure 14. (a) Arsenic concentrations in dreissenid mussel tissue in 2022 compared to the (b) historic regional MWP Dreissena *species arsenic concentrations (µg/g dw). Boxplot colors correspond to cluster groups delineated in the map legend. 81 regional sites sampled between 1992 - 2022 for a total of 314 dreissenid mussel samples.*

6.0 RESULTS - CADMIUM (Cd)

6.1 Cadmium Chemical Description

Cadmium found in coastal and estuarine environments can be linked to both natural and anthropogenic sources (Roesijadi, 1984). Cadmium occurs naturally in the Earth's crust as complex oxides and sulfides in ores (Plachy, 2003), and natural sources can be linked to river runoff from cadmium rich soils, leaching from bedrock, and upwelling from marine sediment deposits (Sokolova et al., 2005). Industrial sources and uses include zinc, lead, and copper production, electroplating and galvanizing, smelting, mining, fossil fuel burning, waste slag, and sewage sludge (ATSDR, 2012a; U.S. FDA, 1993). In addition to abundant industrial applications, other products that contain cadmium include batteries, color pigments, plastics, and phosphate fertilizers. Generally, anthropogenic emissions exceed natural emissions, and elevated cadmium levels are primarily found in freshwater-dominated estuaries (e.g., the Mississippi Delta, the Great Lakes, and the Chesapeake Bay; Sokolova et al., 2005).

Cadmium is a non-essential, toxic element to humans and various biota. Cadmium is toxic to fish, especially salmonid species and juveniles, and chronic exposure can reduce growth. Cadmium has been shown to impair development and reproduction in several invertebrate species and osmoregulation in herring larvae (ATSDR, 2012a; Eisler, 1985). Respiration and consumption are the two major exposure pathways for humans to cadmium, with exposure to high levels resulting primarily from occupational exposure. Safety guidance levels for cadmium in fish and shellfish are no longer listed by the U.S. FDA (U.S. FDA, 2011).

As a result of fossil fuel burning, erosion, and biological activities, cadmium becomes airborne and is transported by atmospheric processes (ATSDR, 2012a). Land-based runoff, river transport, and ocean upwelling are the primary conveyors of cadmium into coastal environments.

6.2 Magnitude and Distribution of Cadmium in Dreissenid Mussel Tissue in 2022

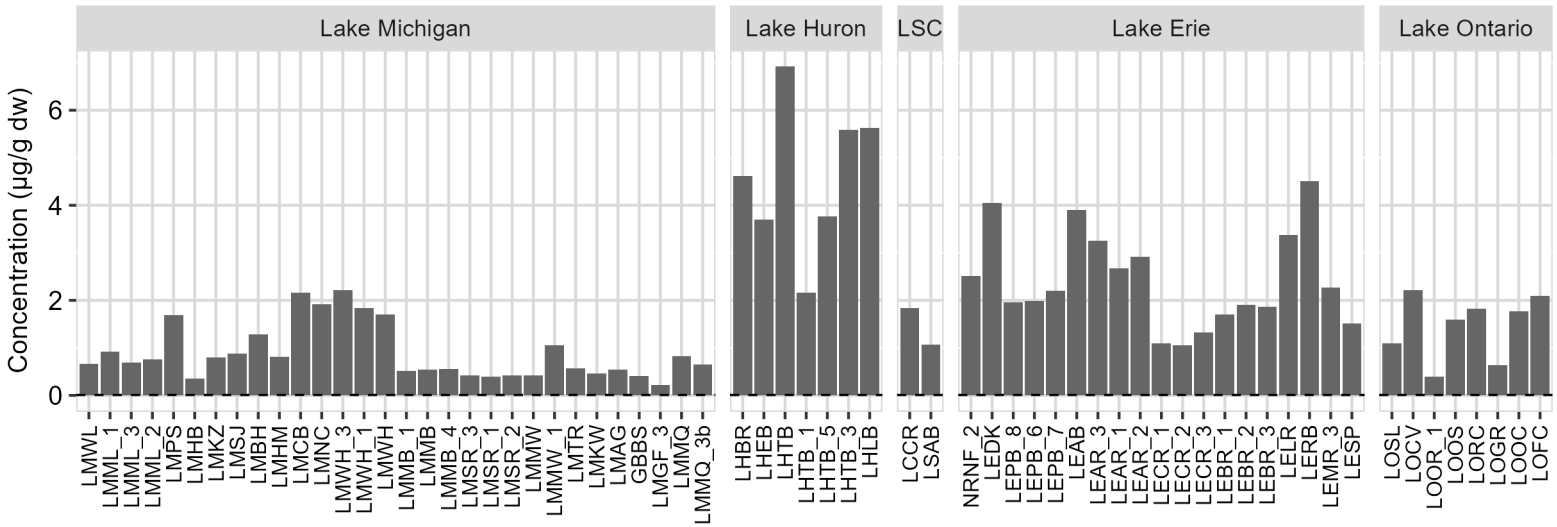


Figure 15. Bar graph showing magnitude of cadmium concentrations (µg/g dw) detected in dreissenid mussel tissue in the Great Lakes Basin in 2022. Dotted line represents the minimum weight corrected MDL. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair.

6.3 Historical Context of Cadmium Magnitude and Distribution in Dreissenid Mussel Tissue

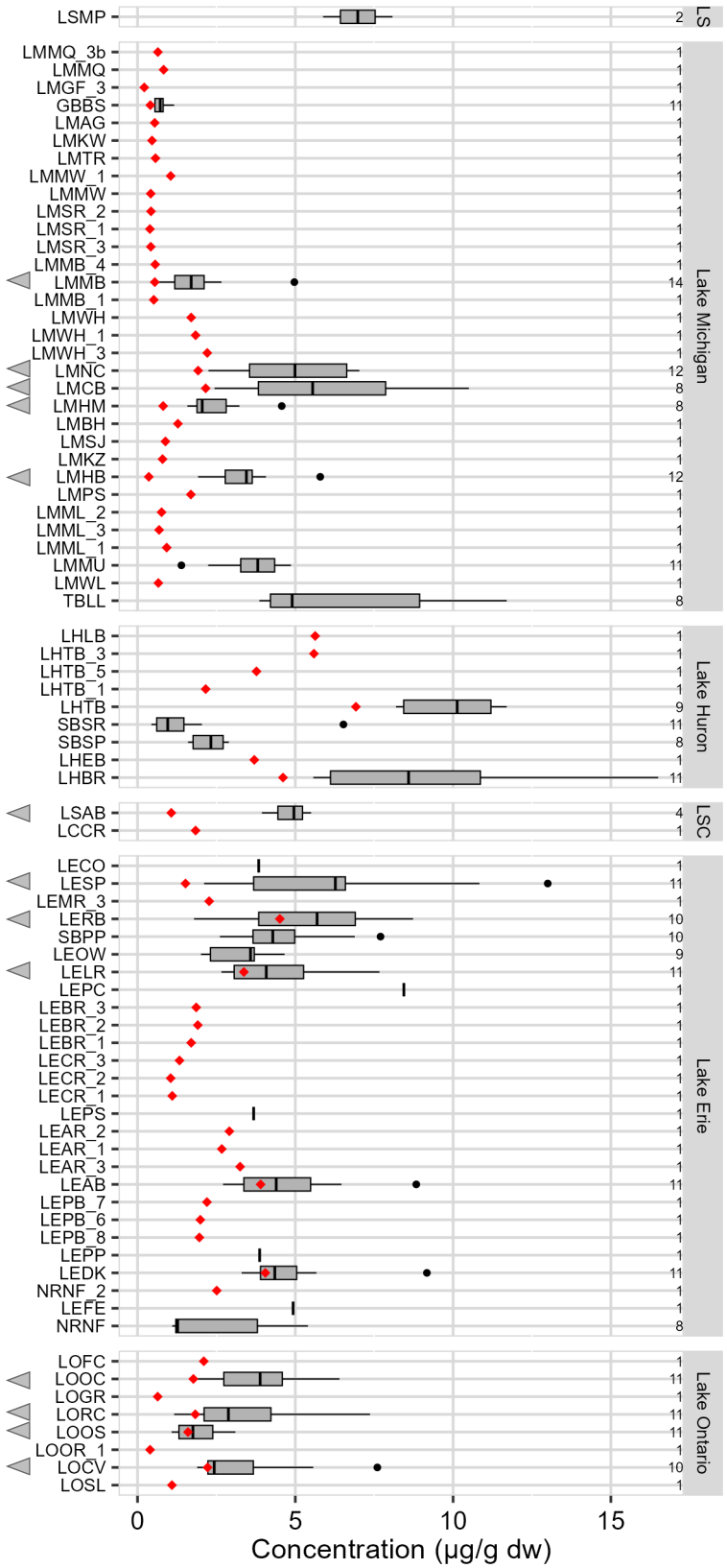


Figure 16. Cadmium concentrations (µg/g dw) in dreissenid mussel tissue in 2022 (red diamonds) compared to boxplots of the historic cadmium concentrations for each site (1992 - 2008) and results of the trend analysis showing significant increasing (▶) or decreasing (◀) site-specific contamination trends over time. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair; LS = Lake Superior. The number of years each site has been sampled since 1992 is noted at the right of the plot for each site.

Results - Cadmium (Cd)

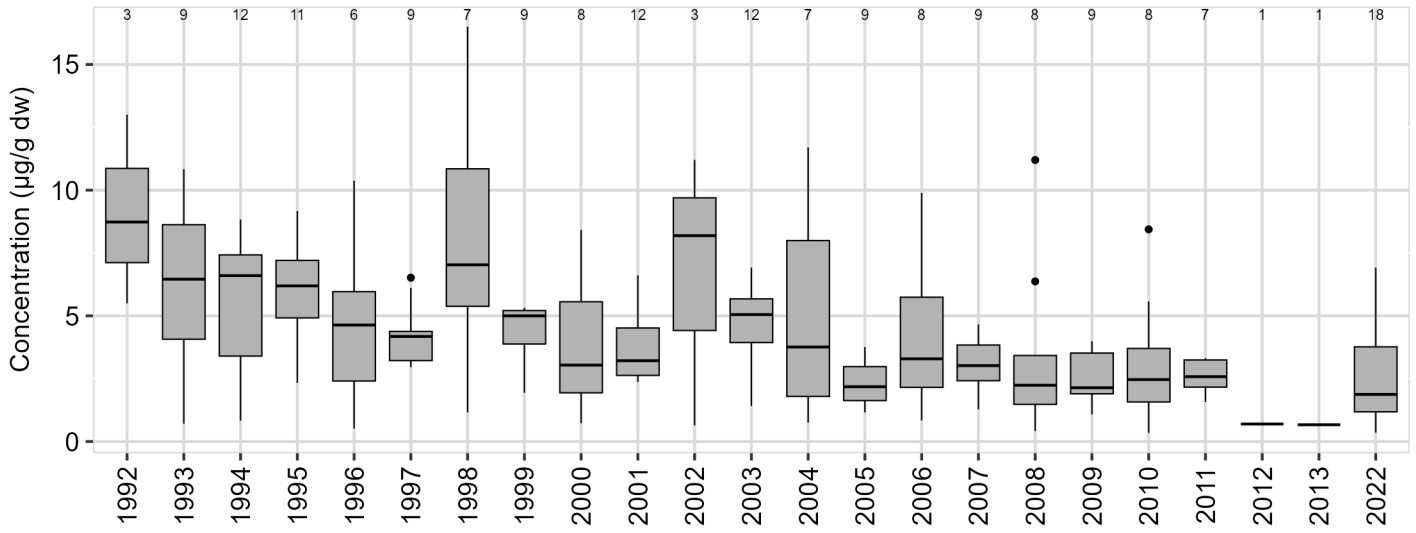


Figure 17. Boxplots representing the historic cadmium concentrations (µg/g dw) in dreissenid mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot.

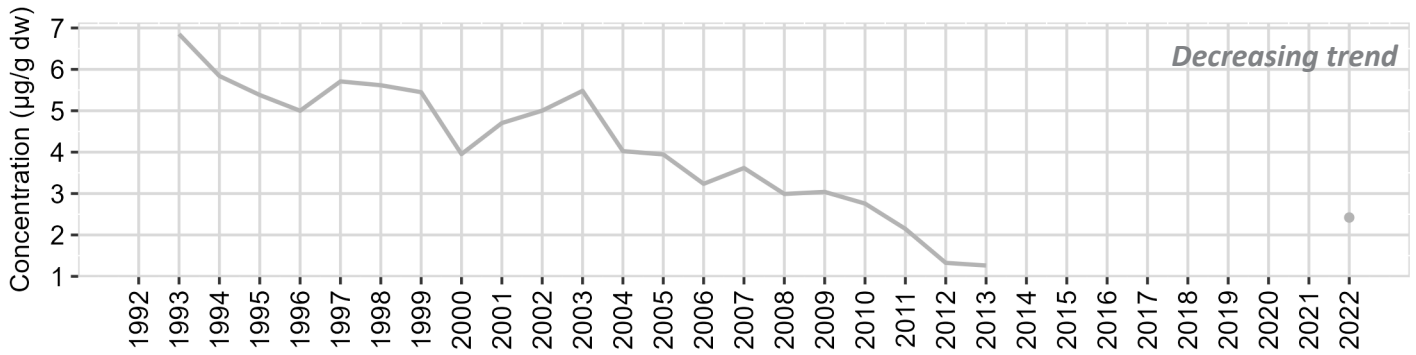


Figure 18. Three-point moving average of the yearly mean cadmium concentrations (µg/g dw) in dreissenid mussel tissue in the sites analyzed in this study. Average concentration for 2022 is included as a standalone point for reference.

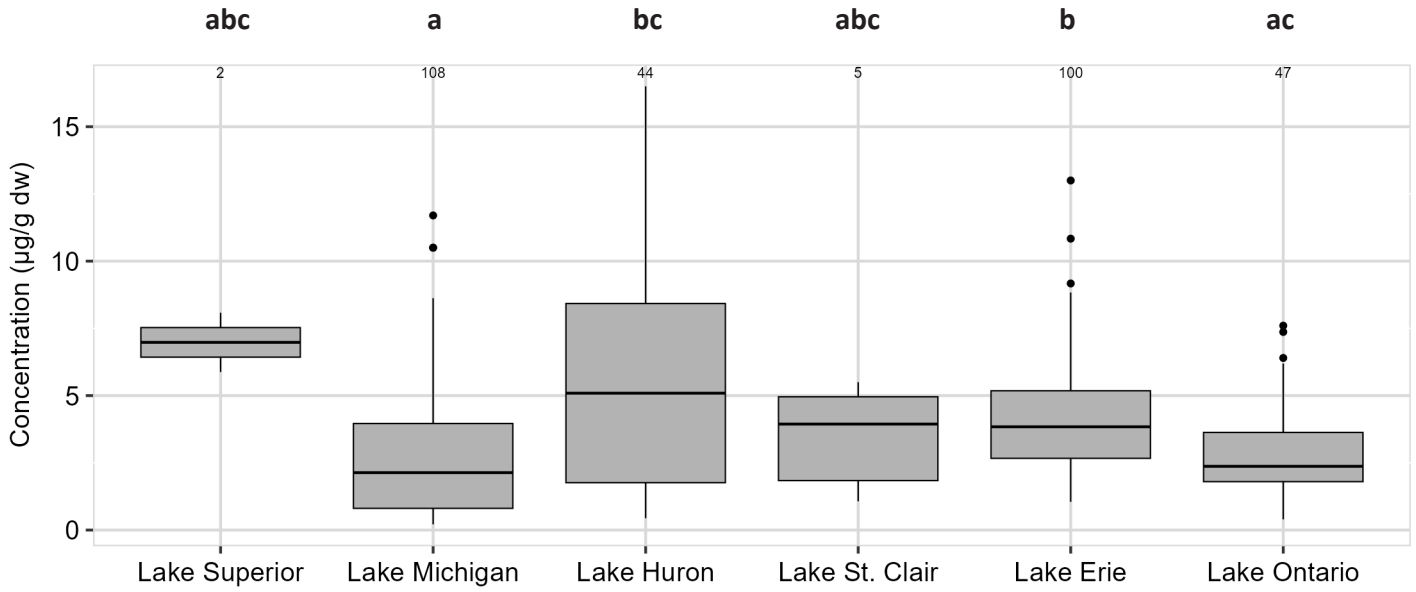


Figure 19. Cadmium concentrations (µg/g dw) measured in dreissenid mussel tissue in the Great Lakes from 1992 - 2022. Lakes and major rivers including connecting channels are listed geographically from west to east across the Great Lakes Basin. The number of samples collected in each lake is noted at the top of the plot. Letters above each lake indicate significant differences (at α = 0.05).

6.4 Cadmium Summary

2022 Dreissenid Mussel Tissue Results:

- Cd was detected at 100% of 66 sites surveyed (Figure 15).
- Cd concentration descriptive statistics (Figure 15):
  - Range: 0.21 – 6.92 µg/g dw
  - Minimum Cd concentration was detected at LMGF_3
  - Maximum Cd concentration was detected at LHTB
  - Median: 1.65 µg/g dw
  - Mean ± SD: 1.81 ± 1.44 µg/g dw
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster respective to 2022 data only:
  - 47% of sites in low cluster (0.21 - 1.33 µg/g dw)
  - 36% of sites in medium cluster (1.52 - 2.91 µg/g dw)
  - 17% of sites in high cluster (3.25 - 6.92 µg/g dw)

Historic Context for Dreissenid Mussel Tissue (18 out of 66 sites had historic data to analyze for trend analyses):

- 0% of sites surveyed (0/18) were above their historic median concentrations in 2022 (Figure 16).
- Regionally, there was a significant decreasing temporal trend of Cd concentrations in dreissenid mussels in the Great Lakes Basin ($p < 0.001$, rho = -0.84) (Figure 17; Figure 18).
- 13 sites (LMMB, LMNC, LMCB, LMHM, LMHB, LSAB, LESP, LERB, LELR, LOOC, LORC, LOOS, LOCV) in the Great Lakes Basin showed significant decreasing temporal trends of Cd concentrations at $\alpha = 0.05$ (Figure 16; Table A4).
- 0 sites in the Great Lakes Basin showed significant increasing temporal trends of Cd concentrations at $\alpha = 0.05$ (Figure 16; Table A4).
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster compared to historic data (Figure 20):
  - 86% of sites in low cluster (0.21 - 3.37 µg/g dw)
  - 14% of sites in medium cluster (3.45 - 9.23 µg/g dw)
  - 0% of sites in high cluster (9.89 - 16.50 µg/g dw)

General Observations:

- Cd concentrations were ubiquitous but at comparatively low to medium concentrations in the Great Lakes Basin in dreissenid mussel tissue in 2022, likely reflecting natural ambient concentrations (Figure 20).
- Regionally, Cd concentrations have significantly decreased over time in dreissenid mussel tissue, indicating decreasing inputs of Cd into the environment (Figure 18).
- The post-hoc analysis evaluating differences between lakes indicates that lakes Superior and St. Clair are statistical bridges, showing no significant difference to any other lake. In contrast, the other lakes demonstrate a gradient of relationships, being significantly similar to some and significantly different from others. Generally, there appears to be a trend of decreasing Cd concentration going from west to east across the Great Lakes Basin, potentially indicating accumulation or differences in regional geology (Figure 19).

Results - Cadmium (Cd)

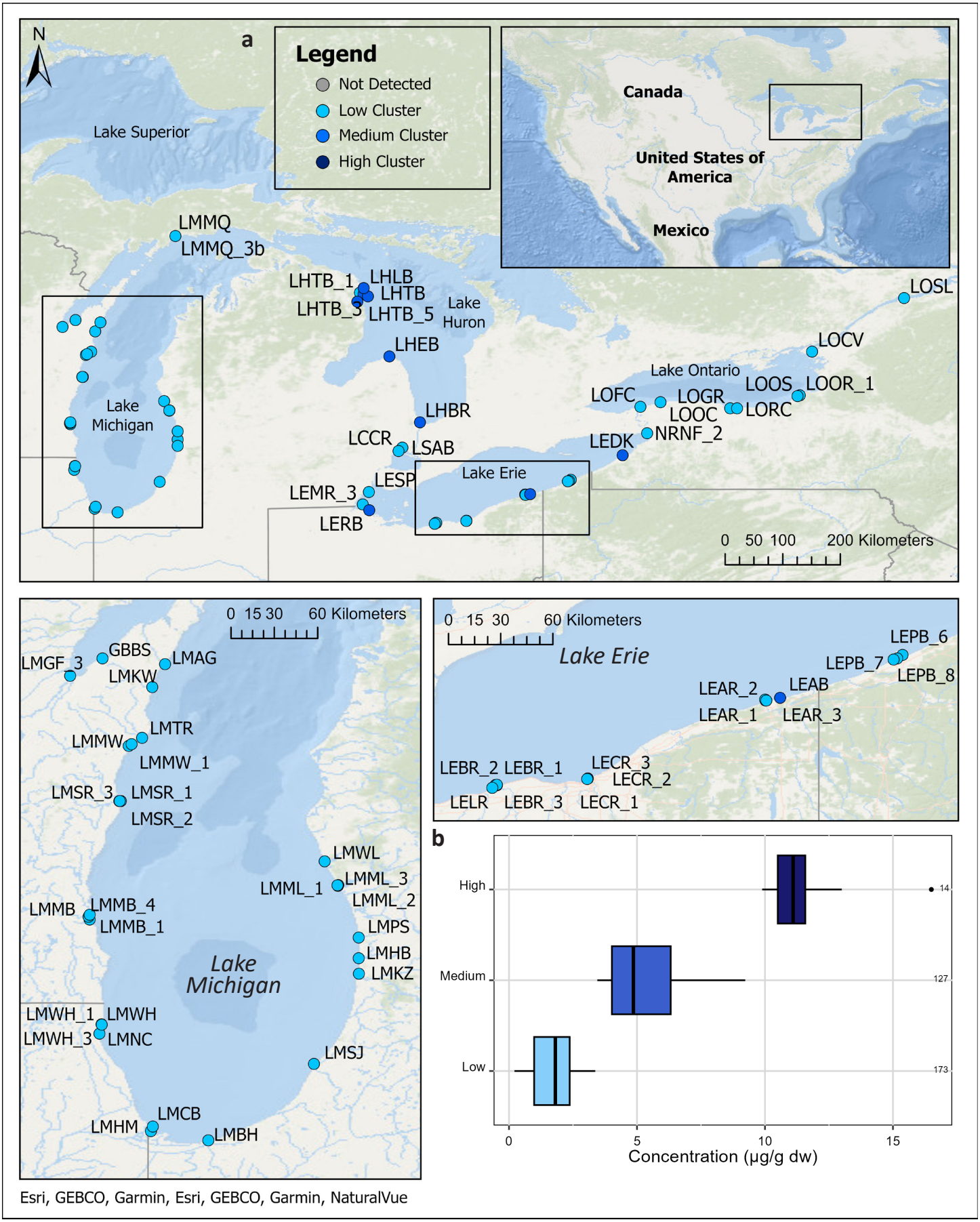


Figure 20. (a) Cadmium concentrations in dreissenid mussel tissue in 2022 compared to the (b) historic regional MWP Dreissena *species cadmium concentrations (µg/g dw). Boxplot colors correspond to cluster groups delineated in the map legend. 81 regional sites sampled between 1992 - 2022 for a total of 314 dreissenid mussel samples.*

7.0 RESULTS - CHROMIUM (Cr)

7.1 Chromium Chemical Description

Chromium is a naturally occurring metal found in rocks, animals, plants, and soils in three main forms: chromium(0), chromium(III), and chromium(VI). Chromium is widely used in industries involved in electroplating, leather tanning, textile production, and the manufacture of chromium-based products, including treated wood, tanned leather, stainless steel cookware, and metal-on-metal hip replacements (ATSDR, 2012b). Chromium can also be released into the environment from the burning of natural gas, oil, or coal (ATSDR, 2012b). The U.S. EPA has established a maximum contaminant level of 0.1 mg/L for total chromium in drinking water and sought voluntary monitoring of chromium(VI) in drinking water by municipalities. The Occupational Safety and Health Administration (OSHA) has set a legal limit for atmospheric exposure over an 8-hour workday of 1.0 mg/m³ for chromium, 0.005 mg/m³ for chromium(VI), and 0.5 mg/m³ for chromium(III).

Chromium is an essential trace nutrient, but can be toxic at high levels. Exposure to chromium can result from inhalation, ingestion, and physical contact. Inhalation primarily occurs near industries that use or manufacture chromium, or through cigarette smoke. Ingestion typically occurs through drinking or bathing water. The general population is most likely to be exposed to trace levels of chromium in foods, including fruits, vegetables, nuts, beverages, and meats, but exposure at high levels occurs primarily resulting from occupational exposure (ATSDR, 2012b). In humans, exposure to chromium primarily impacts the respiratory tract, specifically causing irritation to nasal lining, nasal congestion, and breathing problems. In animals, ingestion of chromium(VI) has been shown to cause adverse effects on the stomach, small intestine, blood, and male reproductive system. The International Agency for Research on Cancer (IARC) has determined that chromium(VI) compounds are carcinogenic to humans (ATSDR, 2012b). In animals, some studies show that exposure to high doses during pregnancy may cause miscarriage, low birth weight, and some changes in the development of the skeleton and reproductive system (ATSDR, 2012b).

Chromium does not typically remain in the atmosphere, but is deposited into the soil and water. In soil and water, chromium can change forms depending on external conditions (ATSDR, 2012b).

7.2 Magnitude and Distribution of Chromium in Dreissenid Mussel Tissue in 2022

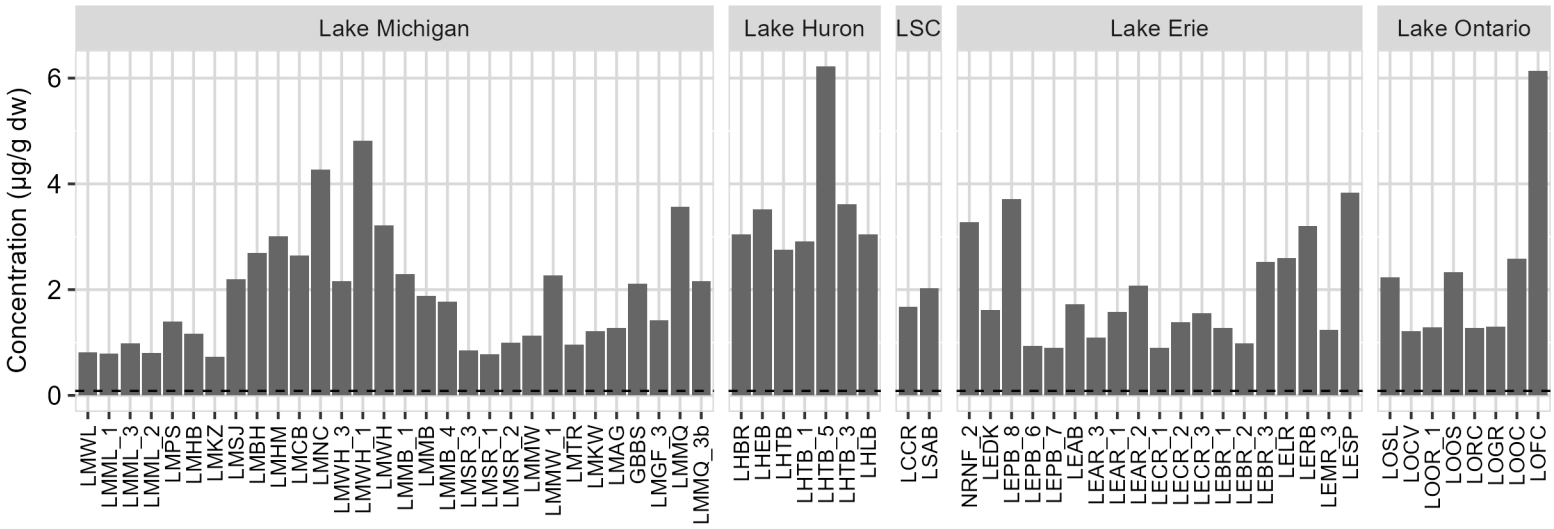


Figure 21. Bar graph showing magnitude of chromium concentrations (µg/g dw) detected in dreissenid mussel tissue in the Great Lakes Basin in 2022. Dotted line represents the minimum weight corrected MDL. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair.

7.3 Historical Context of Chromium Magnitude and Distribution in Dreissenid Mussel Tissue

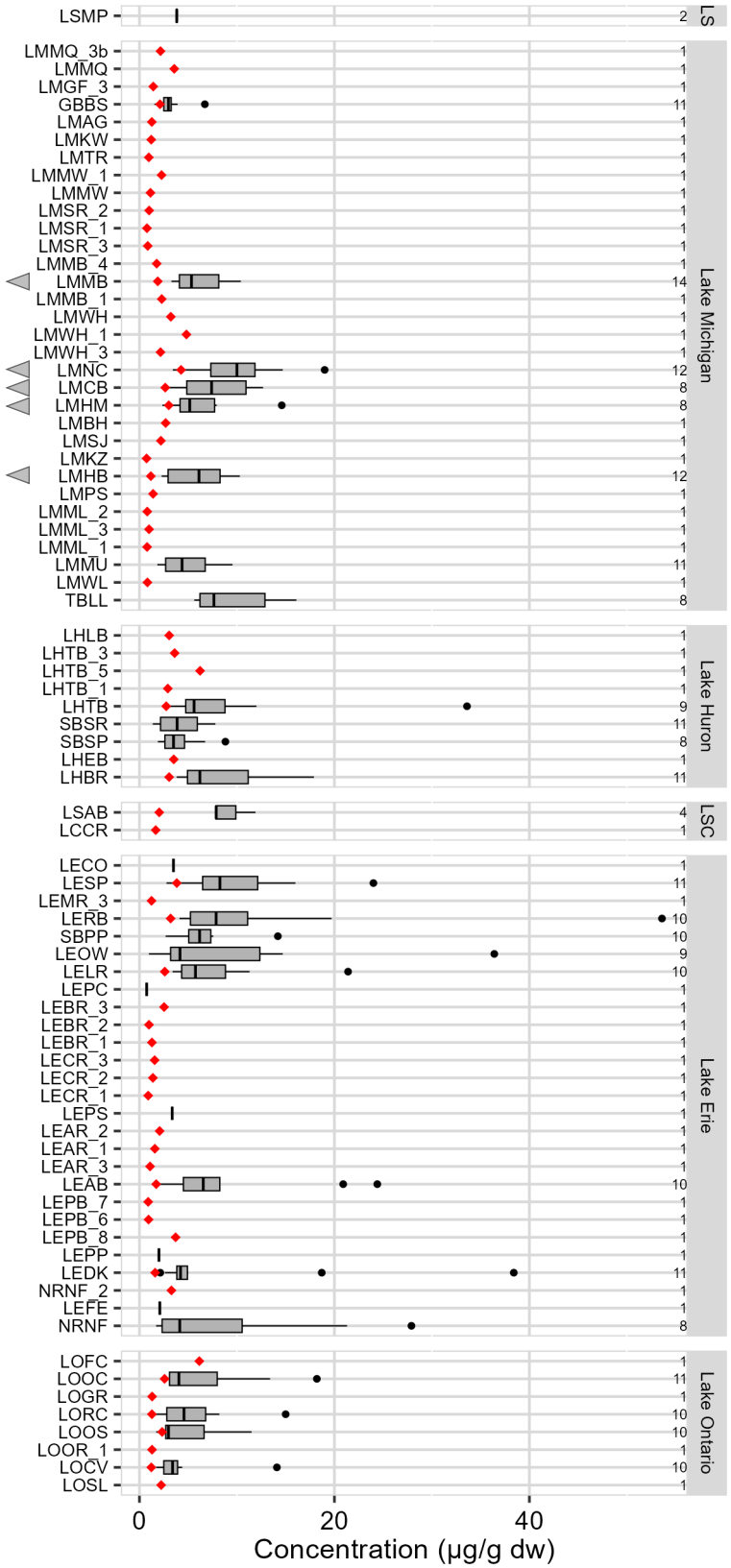


Figure 22. Chromium concentrations (µg/g dw) in dreissenid mussel tissue in 2022 (red diamonds) compared to boxplots of the historic chromium concentrations for each site (1992 - 2008) and results of the trend analysis showing significant increasing (▷) or decreasing (◁) site-specific contamination trends over time. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair; LS = Lake Superior. The number of years each site has been sampled since 1992 is noted at the right of the plot for each site. Sites LORC, LELR, and LEAB in 2005 (177.00 µg/g dw, 126.00 µg/g dw, 106.00 µg/g dw, respectively) and site LOOS in 2003 (75.9 µg/g dw) were removed to aid in visualization.

Results - Chromium (Cr)

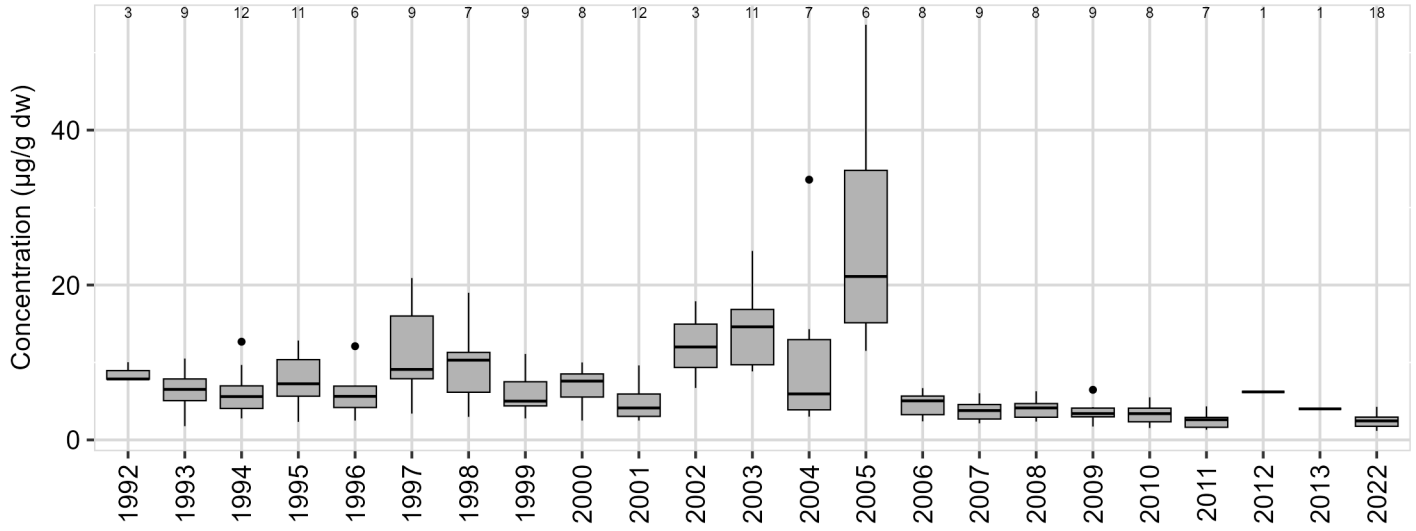


Figure 23. Boxplots representing the historic chromium concentrations (µg/g dw) in dreissenid mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot. Sites LORC, LELR, and LEAB in 2005 (177.00 µg/g dw, 126.00 µg/g dw, 106.00 µg/g dw, respectively) and site LOOS in 2003 (75.9 µg/g dw) were removed to aid in visualization.

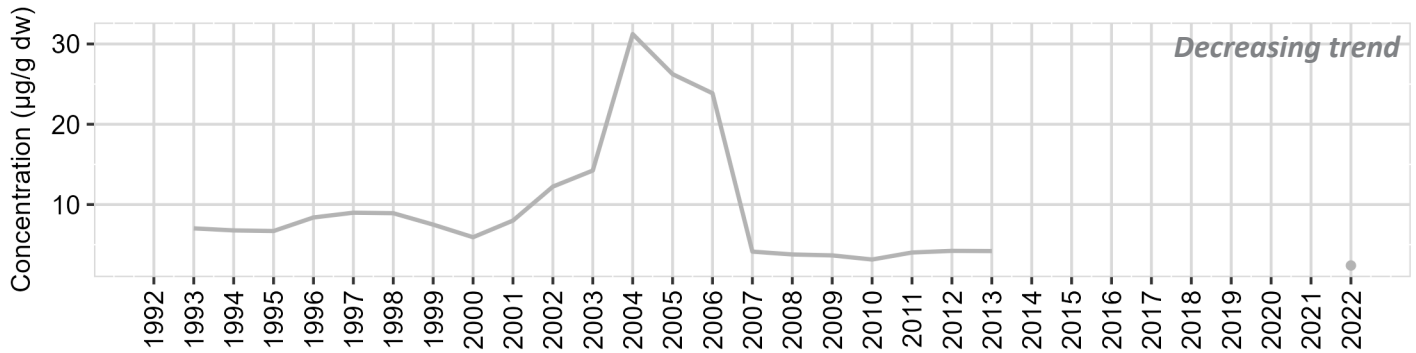


Figure 24. Three-point moving average of the yearly mean chromium concentrations (µg/g dw) in dreissenid mussel tissue in the sites analyzed in this study. Average concentration for 2022 is included as a standalone point for reference.

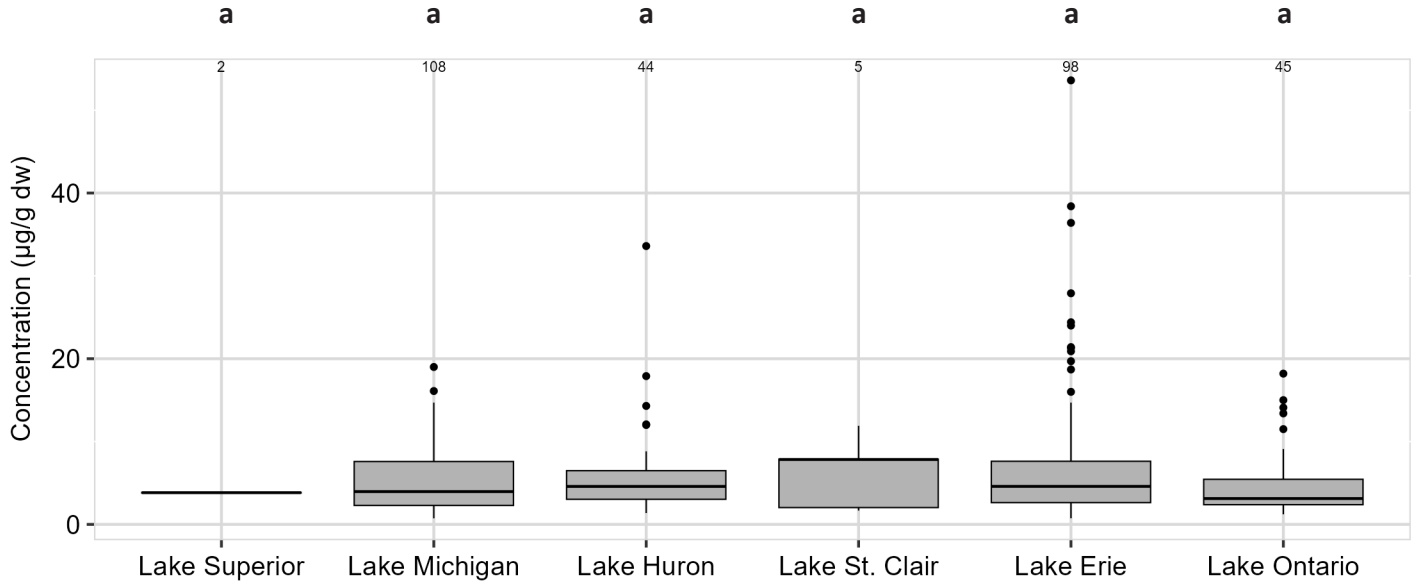


Figure 25. Chromium concentrations (µg/g dw) measured in dreissenid mussel tissue in the Great Lakes from 1992 - 2022. Lakes and major rivers including connecting channels are listed geographically from west to east across the Great Lakes Basin. The number of samples collected in each lake is noted at the top of the plot. Letters above each lake indicate significant differences (at α = 0.05). Sites LORC, LELR, and LEAB in 2005 (177.00 µg/g dw, 126.00 µg/g dw, 106.00 µg/g dw, respectively) and site LOOS in 2003 (75.9 µg/g dw) were removed to aid in visualization.

Results - Chromium (Cr)

7.4 Chromium Summary

2022 Dreissenid Mussel Tissue Results:

- Cr was detected at 100% of 66 sites surveyed (Figure 21).
- Cr concentration descriptive statistics (Figure 21):
  - Range: 0.73 – 6.22 µg/g dw
  - Minimum Cr concentration was detected at LMKZ
  - Maximum Cr concentration was detected at LHTB_5
  - Median: 1.83 µg/g dw
  - Mean ± SD: 2.12 ± 1.22 µg/g dw
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster respective to 2022 data only:
  - 67% of sites in low cluster (0.73 - 2.33 µg/g dw)
  - 30% of sites in medium cluster (2.53 - 4.82 µg/g dw)
  - 3% of sites in high cluster (6.14 - 6.22 µg/g dw)

Historic Context for Dreissenid Mussel Tissue (18 out of 66 sites had historic data to analyze for trend analyses):

- 0% of sites surveyed (0/18) were above their historic median concentrations in 2022 (Figure 22).
- Regionally, there was a significant decreasing temporal trend of Cr concentrations in dreissenid mussels in the Great Lakes Basin ($p = 0.004$, $rho = -0.57$) (Figure 23; Figure 24).
- 5 sites (LMMB, LMNC, LMCB, LMHM, LMHB) in the Great Lakes Basin showed significant decreasing temporal trends of Cr concentrations at $\alpha = 0.05$ (Figure 22; Table A4).
- 0 sites in the Great Lakes Basin showed significant increasing temporal trends of Cr concentrations at $\alpha = 0.05$ (Figure 22; Table A4).
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster compared to historic data (Figure 26):
  - 97% of sites in low cluster (0.73 - 5.16 µg/g dw)
  - 3% of sites in medium cluster (5.23 - 16.10 µg/g dw)
  - 0% of sites in high cluster (17.90 - 177.00 µg/g dw)

General Observations:

- Cr concentrations were ubiquitous but at comparatively low concentrations in the Great Lakes Basin in dreissenid mussel tissue in 2022, likely reflecting natural ambient concentrations (Figure 26).
  - Two instances of comparatively medium concentrations were found in Lake Huron (LHTB_5) and in Lake Ontario (LOFC), possibly indicating local inputs of Cr in those areas (Figure 26).
- Regionally, Cr concentrations have significantly decreased over time in dreissenid mussel tissue, indicating decreasing inputs of Cr into the environment (Figure 24).
- The post-hoc analysis evaluating differences between lakes indicates that all lakes are statistically similar, showing no significant differences between lakes (Figure 25).

Results - Chromium (Cr)

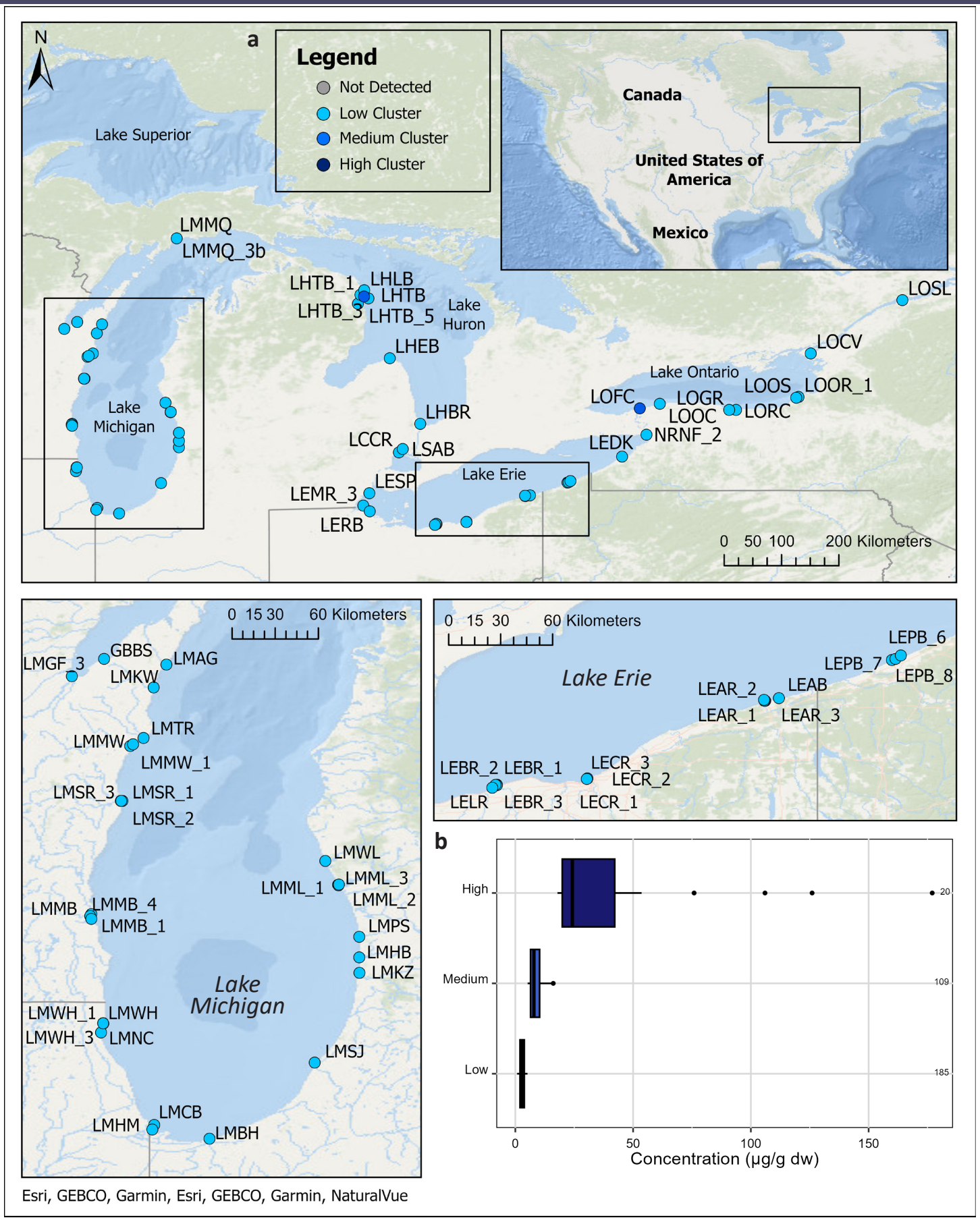


Figure 26. (a) Chromium concentrations in dreissenid mussel tissue in 2022 compared to the (b) historic regional MWP Dreissena *species chromium concentrations (µg/g dw). Boxplot colors correspond to cluster groups delineated in the map legend. 81 regional sites sampled between 1992 - 2022 for a total of 314 dreissenid mussel samples.*

8.0 RESULTS - COPPER (Cu)

8.1 Copper Chemical Description

Copper is a naturally occurring element that is ubiquitous in the environment. Anthropogenic sources include: mining, manufacturing, agriculture, sewage sludge, antifouling paint, fungicides, wood preservatives, and vehicle brake pads (ATSDR, 2004; Denier van der Gon et al., 2007). The U.S. ranks third in the world for utilization and second in production. The U.S. EPA phase-out of chromated copper arsenate (CCA) wood preservatives and the 1980s restrictions on tributyltin marine antifouling paint has stimulated a transition to copper-based wood preservatives and marine antifouling paints.

Trace amounts of copper are an essential nutrient for plants and animals (Eisler, 1998). High concentrations of copper can be toxic to aquatic organisms, with juvenile fishes and invertebrates much more sensitive than adults (Eisler, 1998). Elevated levels of copper can impact aquatic organisms including the functioning of gills, reproduction, and development (Eisler, 1998). Although copper is not highly toxic to humans, chronic effects can occur from prolonged exposure to high doses, and include eye irritation and damage to the digestive tract (ATSDR, 2004). There is no recommended U.S. FDA safety level for copper in fish and fish products (U.S. FDA, 2020).

The most common form of copper in water is Cu (II), which is mostly found bound to organic matter. Transport of copper to coastal and estuarine water occurs as a result of runoff and river transport. Atmospheric transport (Denier van der Gon et al., 2007) and deposition of particulate copper into surface waters may also be significant sources of copper to coastal waters.

8.2 Magnitude and Distribution of Copper in Dreissenid Mussel Tissue in 2022

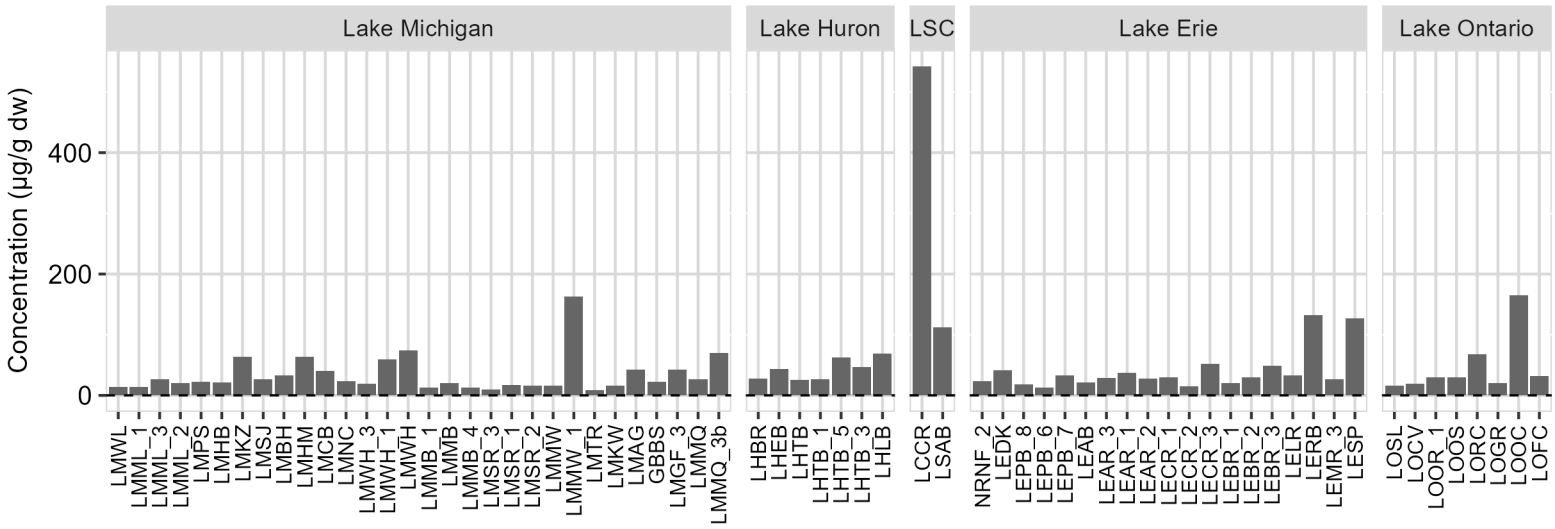


Figure 27. Bar graph showing magnitude of copper concentrations (µg/g dw) detected in dreissenid mussel tissue in the Great Lakes Basin in 2022. Dotted line represents the minimum weight corrected MDL. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair.

8.3 Historical Context of Copper Magnitude and Distribution in Dreissenid Mussel Tissue

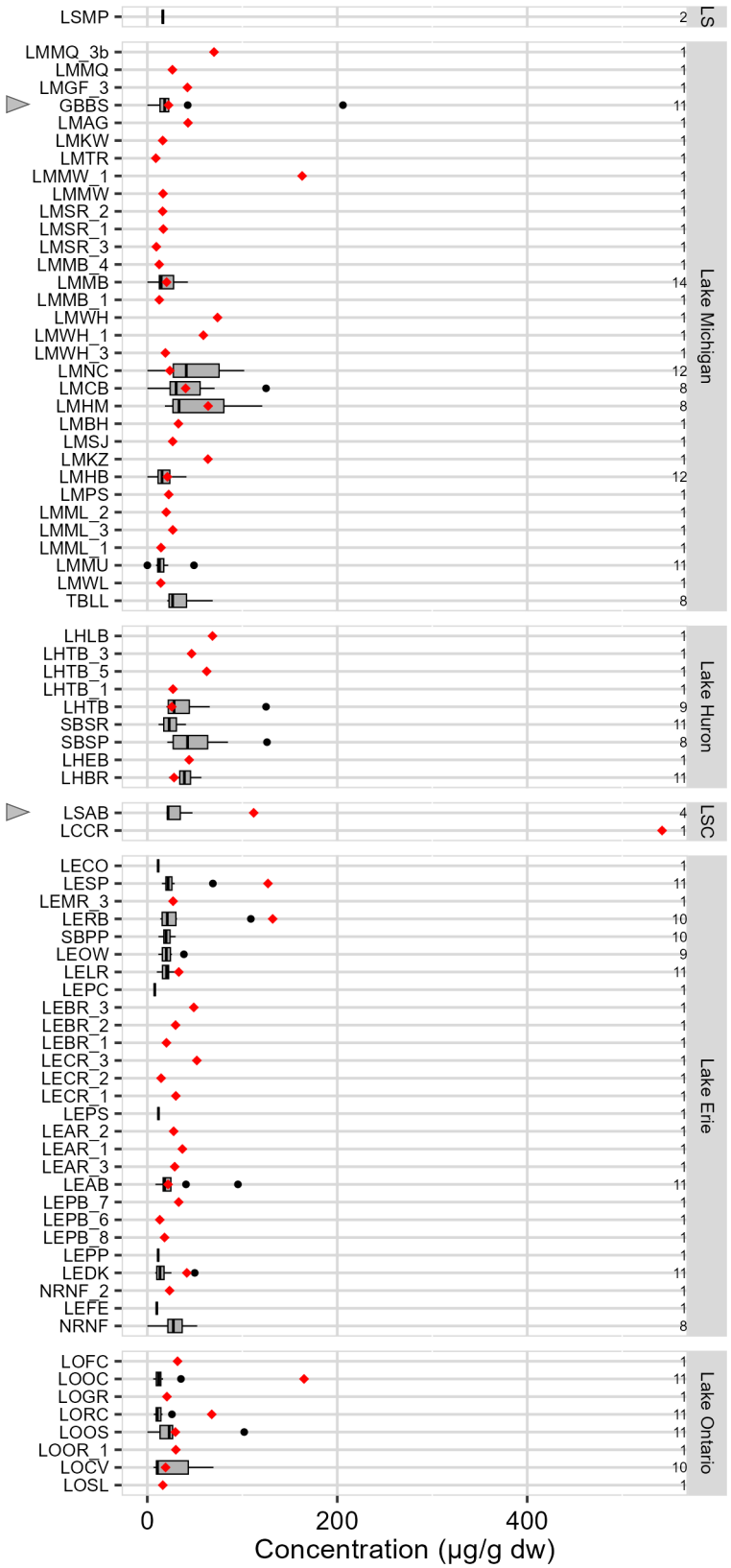


Figure 28. Copper concentrations (µg/g dw) in dreissenid mussel tissue in 2022 (red diamonds) compared to boxplots of the historic copper concentrations for each site (1992 - 2008) and results of the trend analysis showing significant increasing (▶) or decreasing (◀) site-specific contamination trends over time. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair; LS = Lake Superior. The number of years each site has been sampled since 1992 is noted at the right of the plot for each site.

Results - Copper (Cu)

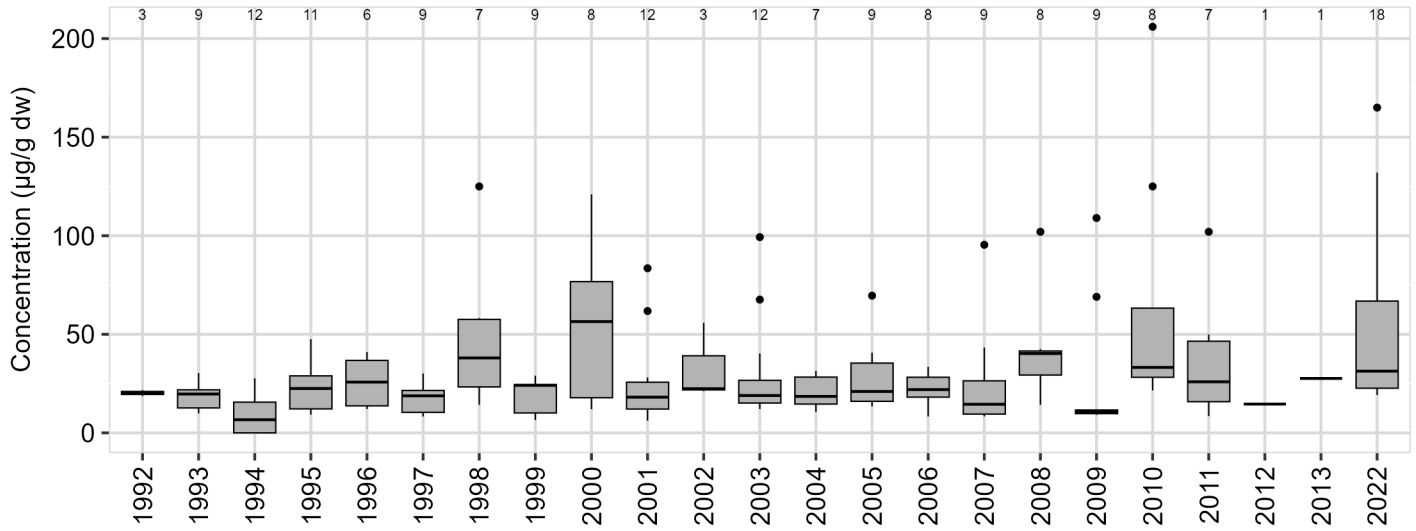


Figure 29. Boxplots representing the historic copper concentrations (µg/g dw) in dreissenid mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot.



Figure 30. Three-point moving average of the yearly mean copper concentrations (µg/g dw) in dreissenid mussel tissue in the sites analyzed in this study. Average concentration for 2022 is included as a standalone point for reference.

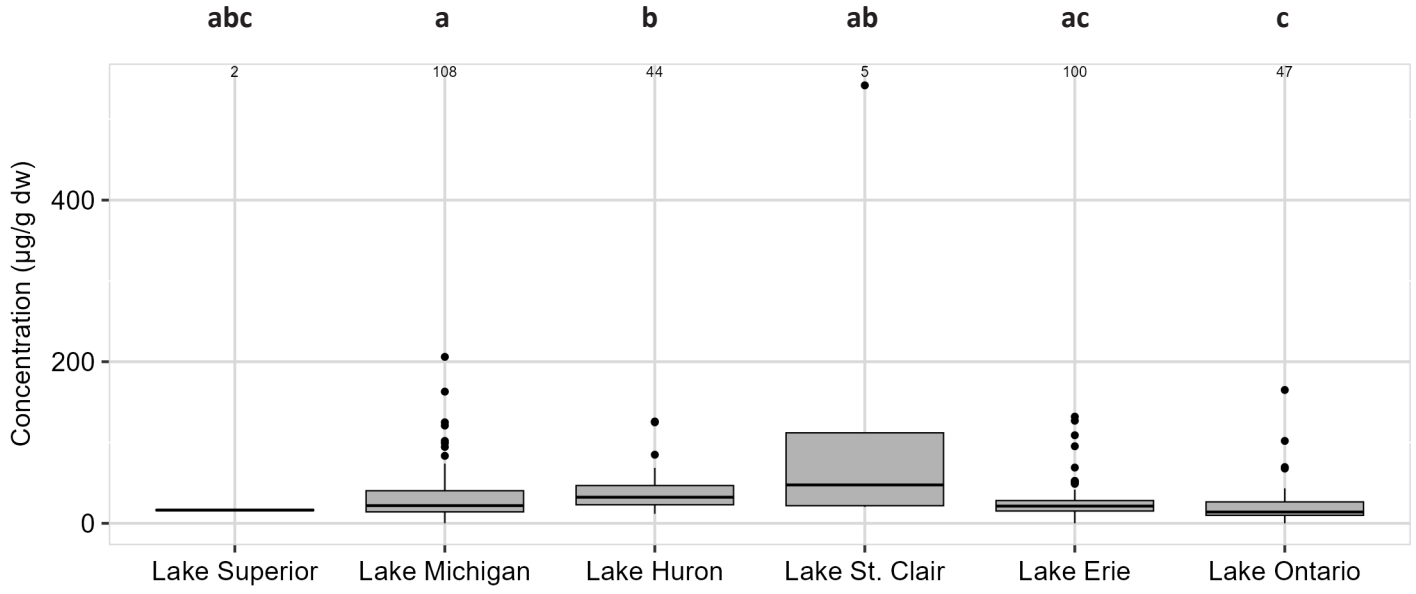


Figure 31. Copper concentrations (µg/g dw) measured in dreissenid mussel tissue in the Great Lakes from 1992 - 2022. Lakes and major rivers including connecting channels are listed geographically from west to east across the Great Lakes Basin. The number of samples collected in each lake is noted at the top of the plot. Letters above each lake indicate significant differences (at α = 0.05).

Results - Copper (Cu)

8.4 Copper Summary

2022 Dreissenid Mussel Tissue Results:

- Cu was detected at 100% of 66 sites surveyed (Figure 27).
- Cu concentration descriptive statistics (Figure 27):
  - Range: 8.94 – 542.00 µg/g dw
  - Minimum Cu concentration was detected at LMTR
  - Maximum Cu concentration was detected at LCCR
  - Median: 27.90 µg/g dw
  - Mean ± SD: 47.18 ± 70.42 µg/g dw
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster respective to 2022 data only:
  - 91% of sites in low cluster (8.94 - 73.90 µg/g dw)
  - 8% of sites in medium cluster (112.00 - 165.00 µg/g dw)
  - 1% of sites in high cluster (542.00 - 542.00 µg/g dw)

Historic Context for Dreissenid Mussel Tissue (18 out of 66 sites had historic data to analyze for trend analyses):

- 83% of sites surveyed (15/18) were above their historic median concentrations in 2022 (Figure 28).
- Regionally, there was a significant increasing regional temporal trend of Cu concentrations in dreissenid mussels in the Great Lakes Basin ($p = 0.03$, $rho = 0.46$) (Figure 29; Figure 30).
- 0 sites in the Great Lakes Basin showed significant decreasing temporal trends of Cu concentrations at $\alpha = 0.05$ (Figure 28; Table A4).
- 2 sites (GBBS, LSAB) in the Great Lakes Basin showed significant increasing temporal trends of Cu concentrations at $\alpha = 0.05$ (Figure 28; Table A4).
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster compared to historic data (Figure 32):
  - 36% of sites in low cluster (0.00 - 23.00 µg/g dw)
  - 55% of sites in medium cluster (23.40 - 84.90 µg/g dw)
  - 9% of sites in high cluster (94.50 - 542.00 µg/g dw)

General Observations:

- Cu concentrations were ubiquitous and were detected across a large range of concentrations in the Great Lakes Basin in dreissenid mussel tissue in 2022, likely reflecting naturally higher ambient concentrations with additional localized inputs (Figure 32).
  - A few instances of comparatively high concentrations were found in Lake Michigan (LMMW_1), Lake St. Clair (LCCR, LSAB), Lake Erie (LERB, LESP), and Lake Ontario (LOOC), possibly indicating local inputs of Cu in those areas (Figure 32).
- Regionally, Cu concentrations have significantly increased over time in dreissenid mussel tissue, indicating increasing inputs of Cu into the environment (Figure 30).
- Cu was notably high at site LCCR in 2022, with its concentration being an outlier even when compared to all historic region-wide data (Figure 28).
- The post-hoc analysis evaluating differences between lakes indicates that Lake Superior is a statistical bridge, showing no significant difference to any other lake. In contrast, the other lakes demonstrate a gradient of relationships, being significantly similar to some and significantly different from others. Generally, concentrations of Cu are relatively similar across lakes with a few high outliers that differentiate the groups from one another (Figure 31).

Results - Copper (Cu)

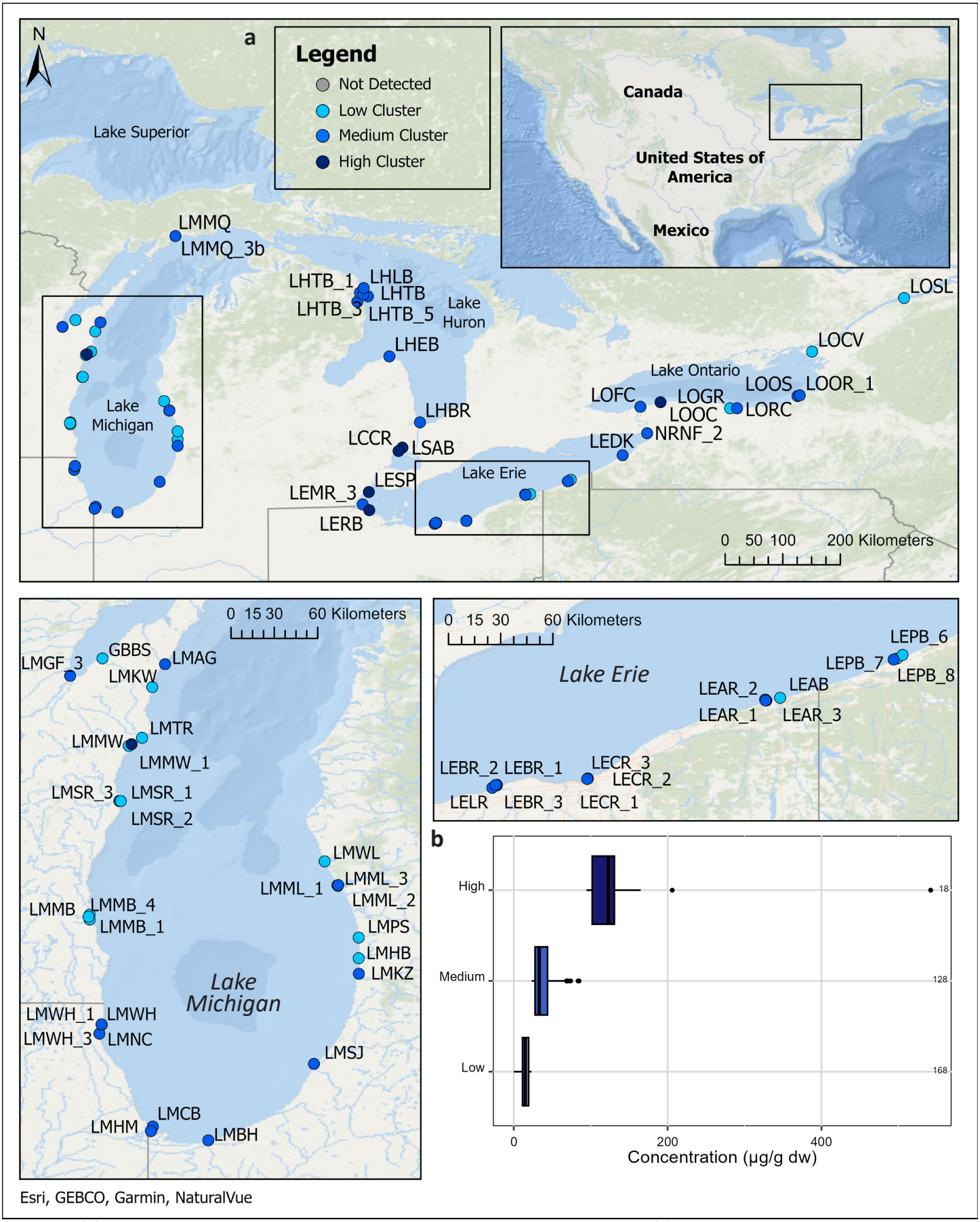


Figure 32. (a) Copper concentrations in dreissenid mussel tissue in 2022 compared to the (b) historic regional MWP Dreissena *species copper concentrations (µg/g dw). Boxplot colors correspond to cluster groups delineated in the map legend. 81 regional sites sampled between 1992 - 2022 for a total of 314 dreissenid mussel samples.*

9.0 RESULTS - IRON (Fe)

9.1 Iron Chemical Description

Iron is the second most abundant metal in the Earth's crust, but tends to have low concentrations in water due to low solubility (Xing and Liu, 2011). Iron concentrations are generally low in natural freshwater bodies, but contamination from anthropogenic sources can occur and result in eutrophication and algal blooms (Xing and Liu, 2011). Natural sources of iron in the environment primarily come from weathered rocks and soils. In contrast, anthropogenic sources of iron input primarily result from fossil fuel combustion and the incorporation of iron into foods and nutritional supplements (Liu et al., 2022). More recently, increasing usage of iron-based nanoparticles for in situ soil and groundwater remediation, biomedical applications, and water treatment, has raised concerns about their environmental behavior and ecological effects (Lei et al., 2018).

Iron is an essential nutrient for humans, with adverse effects caused both by deficiency and exposure to high concentrations (U.S. EPA, 2006). Iron deficiency, known as anemia, can cause fatigue, weakness, and cold extremities in humans. Conversely, exposure to high levels of iron in animals and humans typically causes injury to the gastrointestinal mucosa, leading to nausea, vomiting, abdominal pain, and diarrhea (Yuen and Becker, 2022). Exposure at these levels is typically caused by accidental ingestion (Lei et al., 2018). At the cellular level, exposure to high levels of iron can impair cellular metabolism in the heart, liver, and central nervous system (Yuen and Becker, 2022). In the environment, the toxicity of iron-based nanoparticles is increasingly observed (Lei et al., 2018).

Iron-based nanoparticles undergo physical and chemical transformations influenced by environmental factors, including pH, ions, dissolved oxygen, natural organic matter, and biota (Lei et al., 2018). Iron in the environment is often taken up by both terrestrial and aquatic plants, to the extent that excess iron is often stored as iron plaque to use in times of iron deficiency (Lei et al., 2018). Potentially harmful environmental effects of iron are a direct result of pollution caused by iron ore mining (Lei et al., 2018).

9.2 Magnitude and Distribution of Iron in Dreissenid Mussel Tissue in 2022

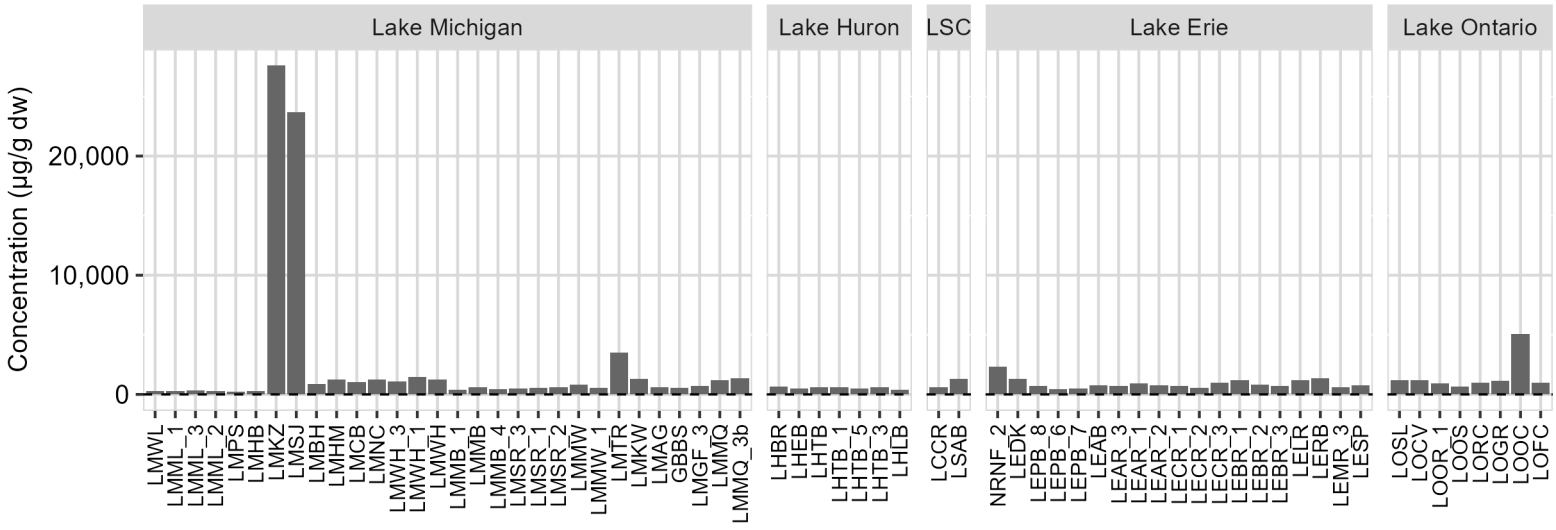


Figure 33. Bar graph showing magnitude of iron concentrations (µg/g dw) detected in dreissenid mussel tissue in the Great Lakes Basin in 2022. Dotted line represents the minimum weight corrected MDL. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair.

9.3 Historical Context of Iron Magnitude and Distribution in Dreissenid Mussel Tissue

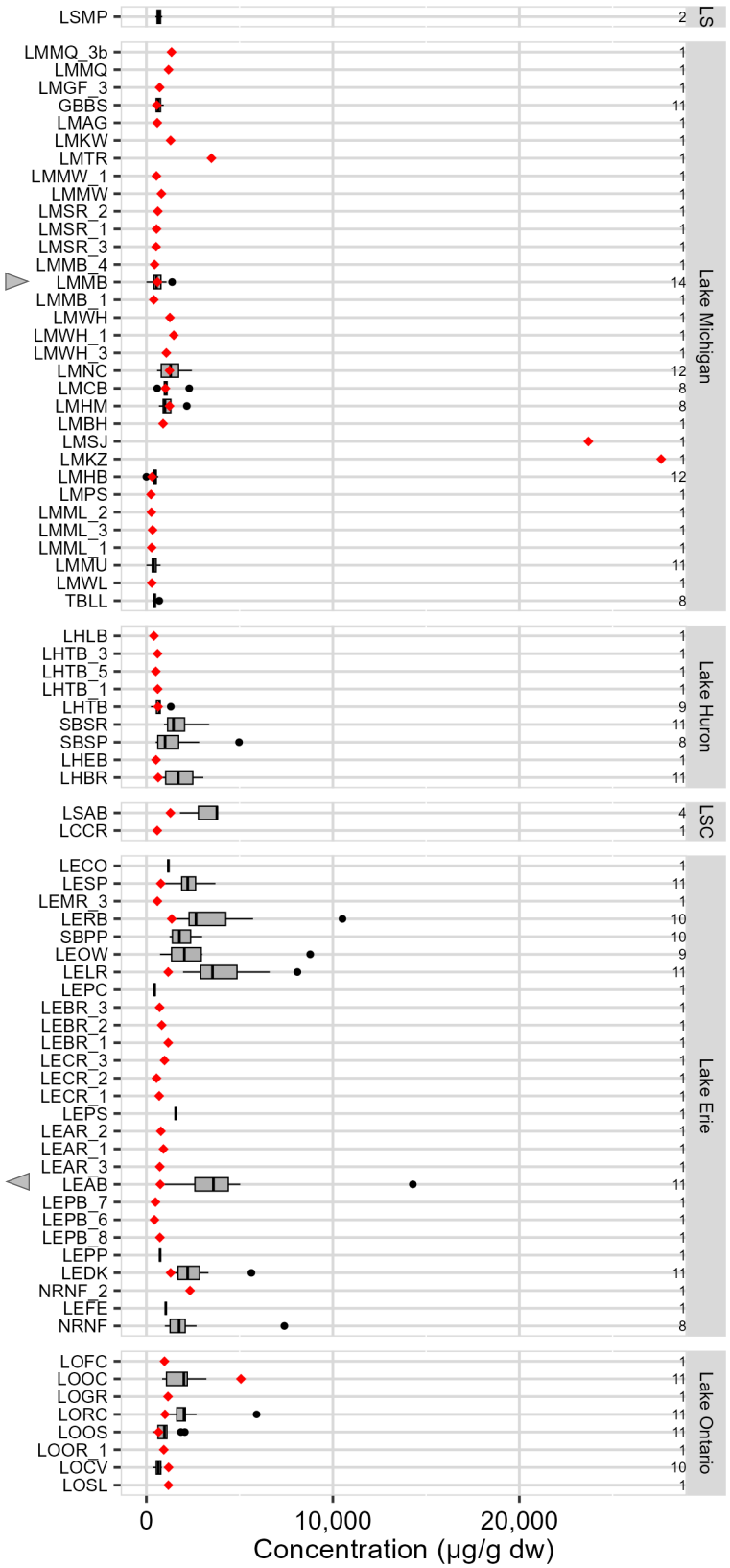


Figure 34. Iron concentrations (µg/g dw) in dreissenid mussel tissue in 2022 (red diamonds) compared to boxplots of the historic iron concentrations for each site (1992 - 2008) and results of the trend analysis showing significant increasing (►) or decreasing (◄) site-specific contamination trends over time. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair; LS = Lake Superior. The number of years each site has been sampled since 1992 is noted at the right of the plot for each site.

Results - Iron (Fe)

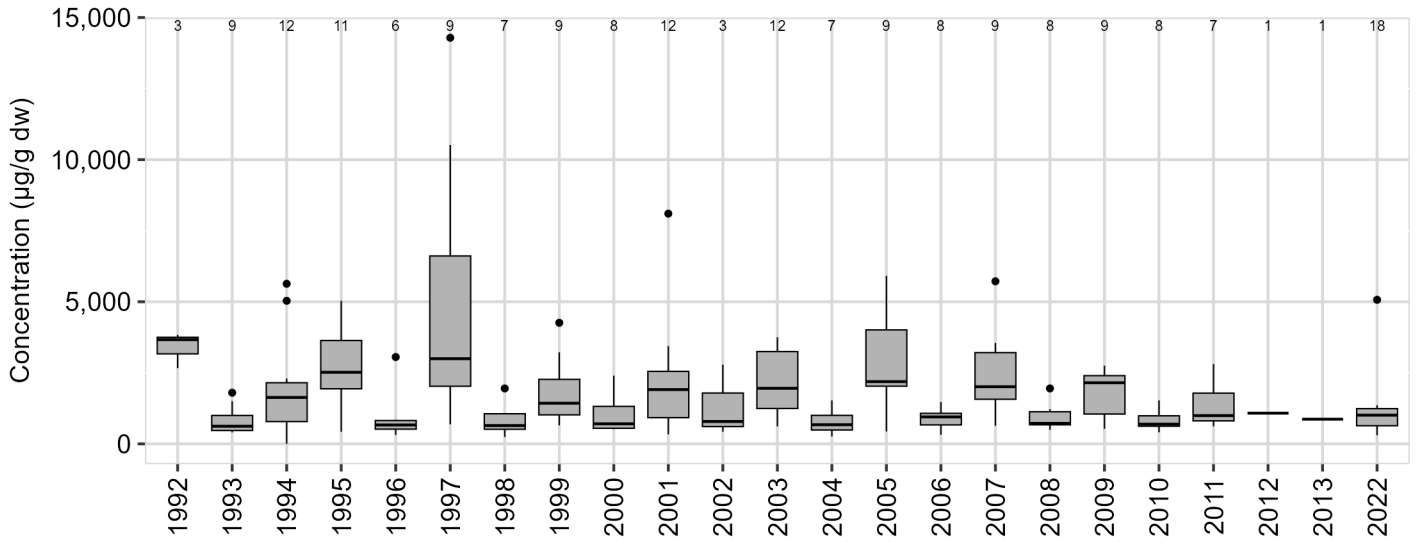


Figure 35. Boxplots representing the historic iron concentrations (µg/g dw) in dreissenid mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot.

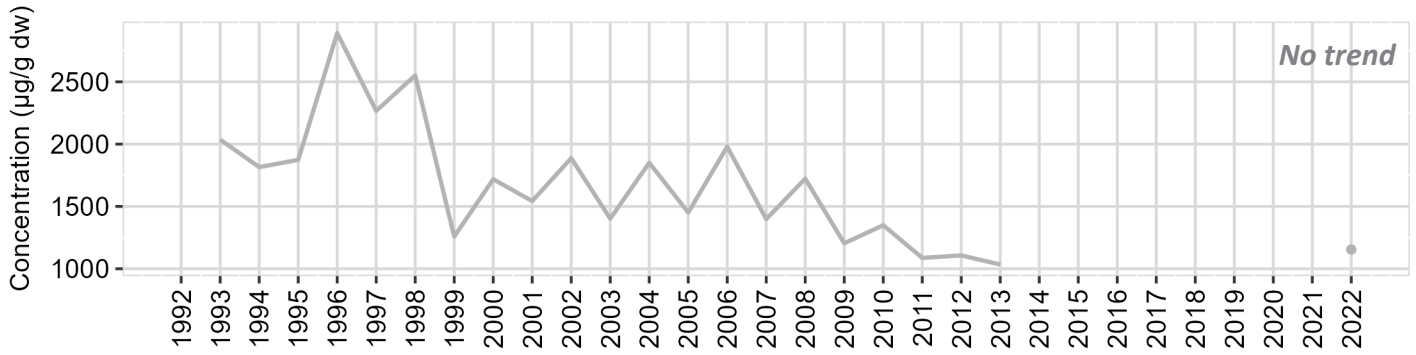


Figure 36. Three-point moving average of the yearly mean iron concentrations (µg/g dw) in dreissenid mussel tissue in the sites analyzed in this study. Average concentration for 2022 is included as a standalone point for reference.

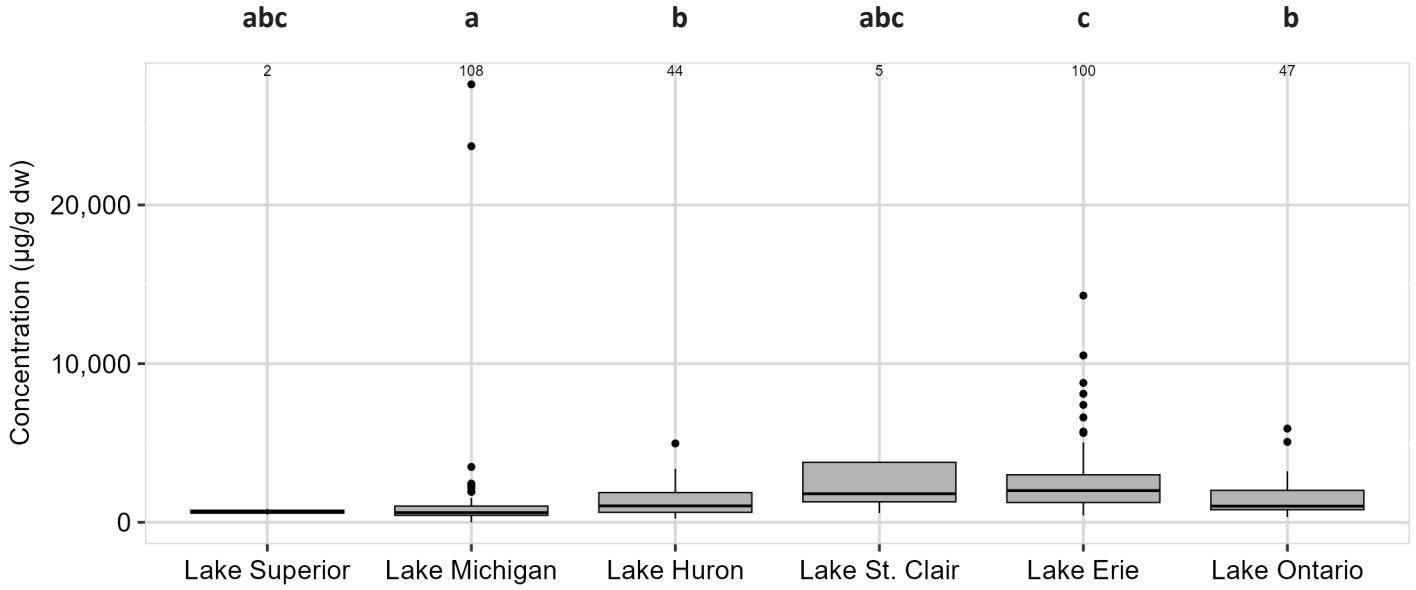


Figure 37. Iron concentrations (µg/g dw) measured in dreissenid mussel tissue in the Great Lakes from 1992 - 2022. Lakes and major rivers including connecting channels are listed geographically from west to east across the Great Lakes Basin. The number of samples collected in each lake is noted at the top of the plot. Letters above each lake indicate significant differences (at α = 0.05).

Results - Iron (Fe)

9.4 Iron Summary

2022 Dreissenid Mussel Tissue Results:

- Fe was detected at 100% of 66 sites surveyed (Figure 33).
- Fe concentration descriptive statistics (Figure 33):
  - Range: 245.00 – 27,600.00 µg/g dw
  - Minimum Fe concentration was detected at LMPS
  - Maximum Fe concentration was detected at LMKZ
  - Median: 734.82 µg/g dw
  - Mean ± SD: 1,667.15 ± 4,345.74 µg/g dw
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster respective to 2022 data only:
  - 92% of sites in low cluster (245.00 - 1,470.00 µg/g dw)
  - 5% of sites in medium cluster (2,339.82 - 5,070.00 µg/g dw)
  - 3% of sites in high cluster (23,700.00 - 27,600.00 µg/g dw)

Historic Context for Dreissenid Mussel Tissue (18 out of 66 sites had historic data to analyze for trend analyses):

- 22% of sites surveyed (4/18) were above their historic median concentrations in 2022 (Figure 34).
- Regionally, there was no significant temporal trend of Fe concentrations in dreissenid mussels in the Great Lakes Basin ($p = 0.18$, $rho = -0.29$) (Figure 35; Figure 36).
- 1 site (LEAB) in the Great Lakes Basin showed a significant decreasing temporal trend of Fe concentrations at $\alpha = 0.05$ (Figure 34; Table A4).
- 1 site (LMMB) in the Great Lakes Basin showed a significant increasing temporal trend of Fe concentrations at $\alpha = 0.05$ (Figure 34; Table A4).
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster compared to historic data (Figure 38):
  - 92% of sites in low cluster (0.00 - 1720.00 µg/g dw)
  - 2% of sites in medium cluster (1800.00 - 2552.00 µg/g dw)
  - 6% of sites in high cluster (2659.55 - 27600.00 µg/g dw)

General Observations:

- Fe concentrations were ubiquitous but at comparatively low concentrations in the Great Lakes Basin in dreissenid mussel tissue in 2022, likely reflecting natural ambient concentrations (Figure 38).
  - A few instances of comparatively high concentrations were found in Lake Michigan (LMKZ, LMSJ, LMTR), Lake Erie (NRNF_2), and Lake Ontario (LOOC), possibly indicating local inputs of Fe in those areas (Figure 38).
- Regionally, Fe concentrations have not changed significantly over time in dreissenid mussel tissue, indicating no obvious anthropogenic inputs of Fe into the environment (Figure 36).
- Fe was notably high at sites LMKZ and KMSJ in 2022, with their concentrations being outliers even when compared to all historic region-wide data (Figure 34).
- The post-hoc analysis evaluating differences between lakes indicates that lakes Superior and St. Clair are statistical bridges, showing no significant difference to any other lake. In contrast, the other lakes demonstrate a gradient of relationships, being significantly similar to some and significantly different from others. Generally, the highest concentrations of Fe appear to be located in lakes St. Clair and Erie, possibly due to their proximity to Detroit, MI (Figure 37).

Results - Iron (Fe)

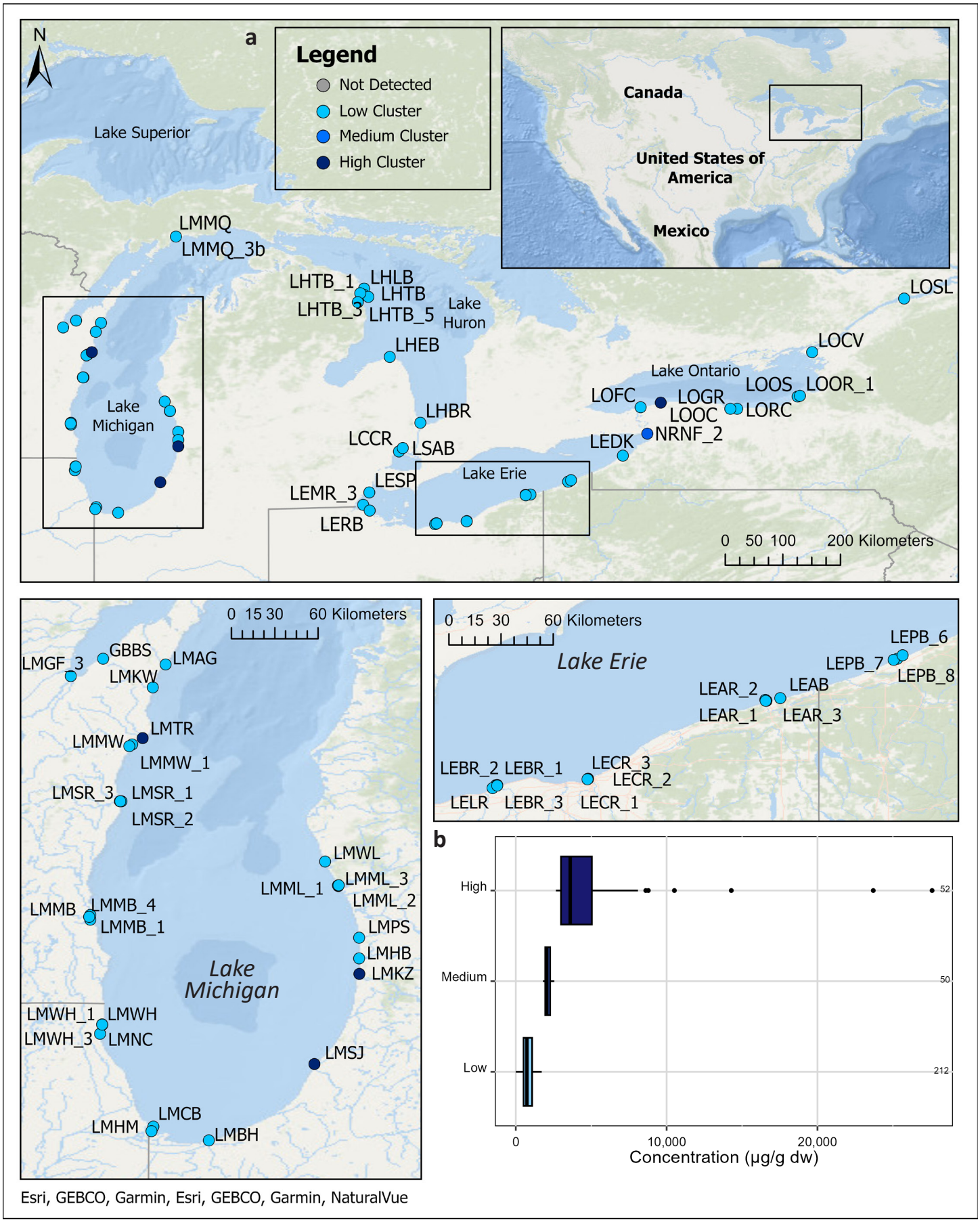


Figure 38. (a) Iron concentrations in dreissenid mussel tissue in 2022 compared to the (b) historic regional MWP Dreissena *species iron concentrations (µg/g dw). Boxplot colors correspond to cluster groups delineated in the map legend. 81 regional sites sampled between 1992 - 2022 for a total of 314 dreissenid mussel samples.*

10.0 RESULTS - LEAD (Pb)

10.1 Lead Chemical Description

Lead is a ubiquitous metal that occurs naturally in the Earth's crust. Release of lead into coastal waters is primarily linked to wastewater discharge, river runoff, atmospheric deposition and natural weathering of rock. Lead can be found in air, soil, and surface water (ATSDR, 2007b). Environmental levels of lead increased worldwide over the past century because of leaded gasoline use (ATSDR, 2007b). Significant reductions in source and load resulted from the regulation of lead in gasoline and lead-based paints. High levels found in the environment are usually linked to anthropogenic activities such as manufacturing processes, paint and pigment, solder, ammunition, plumbing, incineration, and fossil fuel burning. In the communications industry, lead is still used extensively as protective sheathing for underground and underwater cables, including transoceanic cable systems (USGS, 2008).

Lead has no biological use and is toxic to many organisms, including humans. Exposure of fish to elevated concentrations of lead results in neurological deformities and black fins in fish (Mance, 1987). Lead primarily affects the nervous system, resulting in decreased mental performance and inhibited mental development in humans. Exposure to lead may also cause brain and kidney damage and cancer (IARC, 2006). Safety guidance levels for lead in fish and shellfish are no longer listed by the U.S. FDA (U.S. FDA, 2011).

Loadings of lead into coastal waters are primarily linked to wastewater discharge, river runoff, atmospheric deposition and natural weathering of rock (ATSDR, 2007b). Lead can be found in air, soil and surface water (ATSDR, 2007b).

10.2 Magnitude and Distribution of Lead in Dreissenid Mussel Tissue in 2022

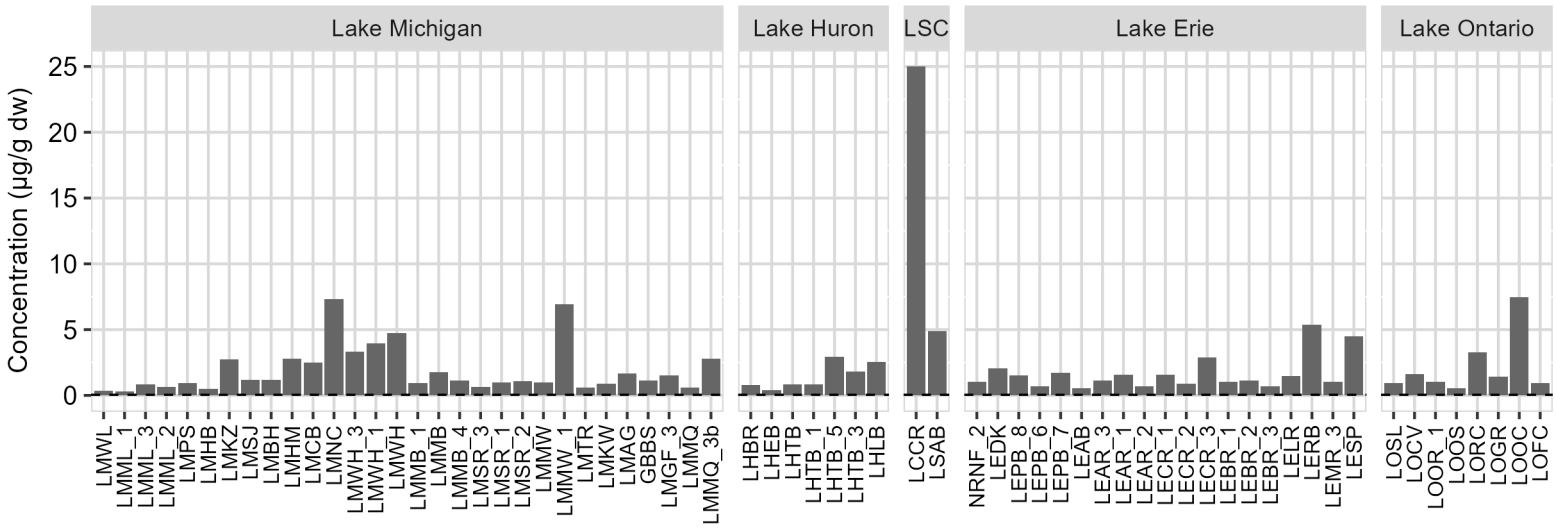


Figure 39. Bar graph showing magnitude of lead concentrations (µg/g dw) detected in dreissenid mussel tissue in the Great Lakes Basin in 2022. Dotted line represents the minimum weight corrected MDL. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair.

10.3 Historical Context of Lead Magnitude and Distribution in Dreissenid Mussel Tissue

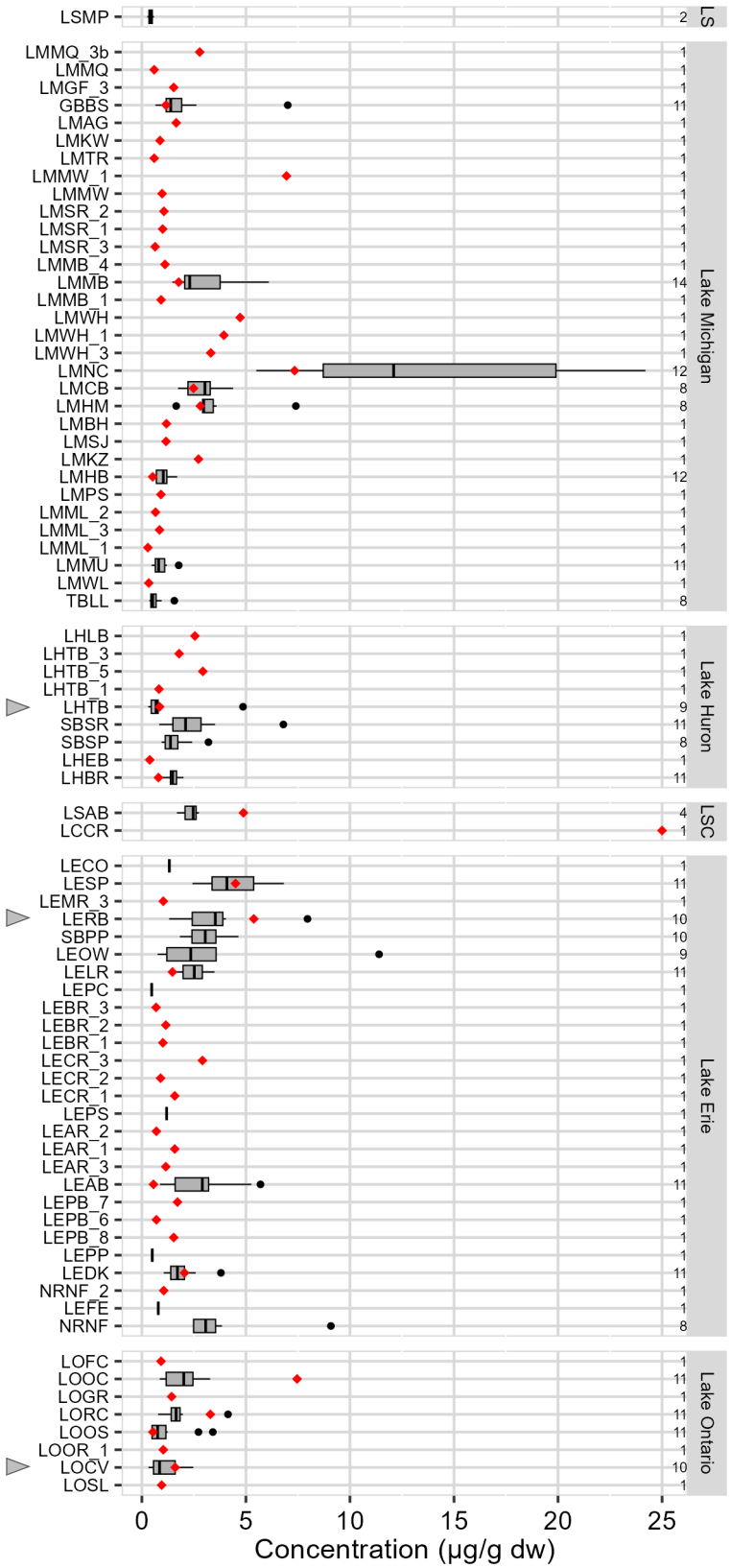


Figure 40. Lead concentrations (µg/g dw) in dreissenid mussel tissue in 2022 (red diamonds) compared to boxplots of the historic lead concentrations for each site (1992 - 2008) and results of the trend analysis showing significant increasing (►) or decreasing (◄) site-specific contamination trends over time. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair; LS = Lake Superior. The number of years each site has been sampled since 1992 is noted at the right of the plot for each site.

Results - Lead (Pb)

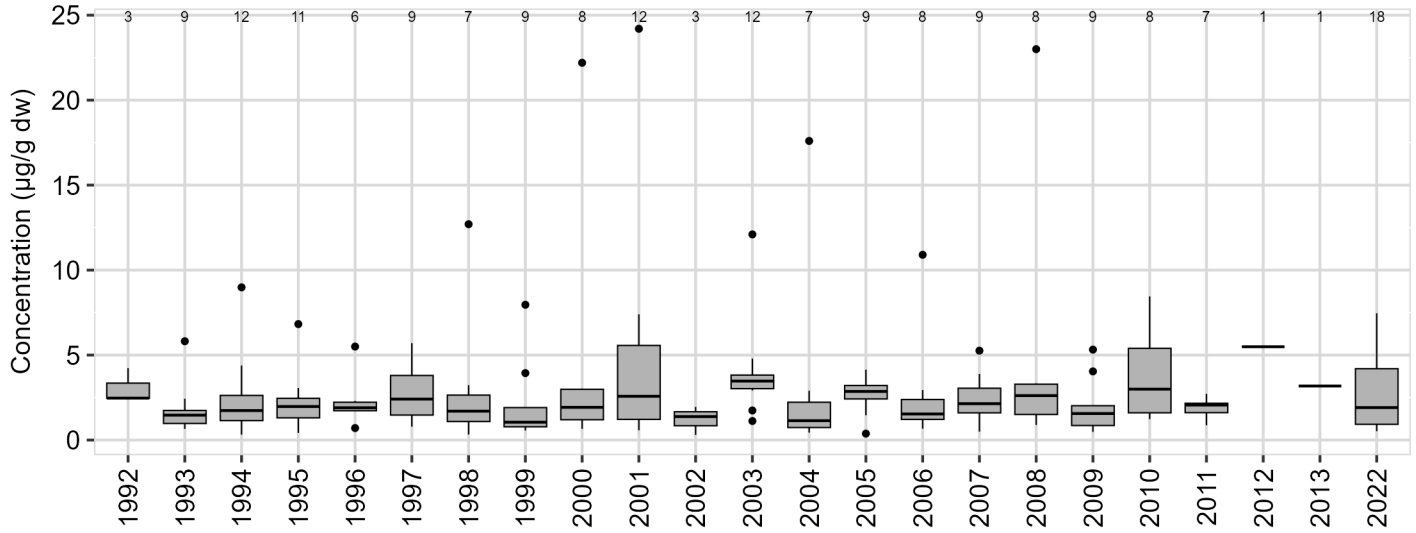


Figure 41. Boxplots representing the historic lead concentrations (µg/g dw) in dreissenid mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot.



Figure 42. Three-point moving average of the yearly mean lead concentrations (µg/g dw) in dreissenid mussel tissue in the sites analyzed in this study. Average concentration for 2022 is included as a standalone point for reference.

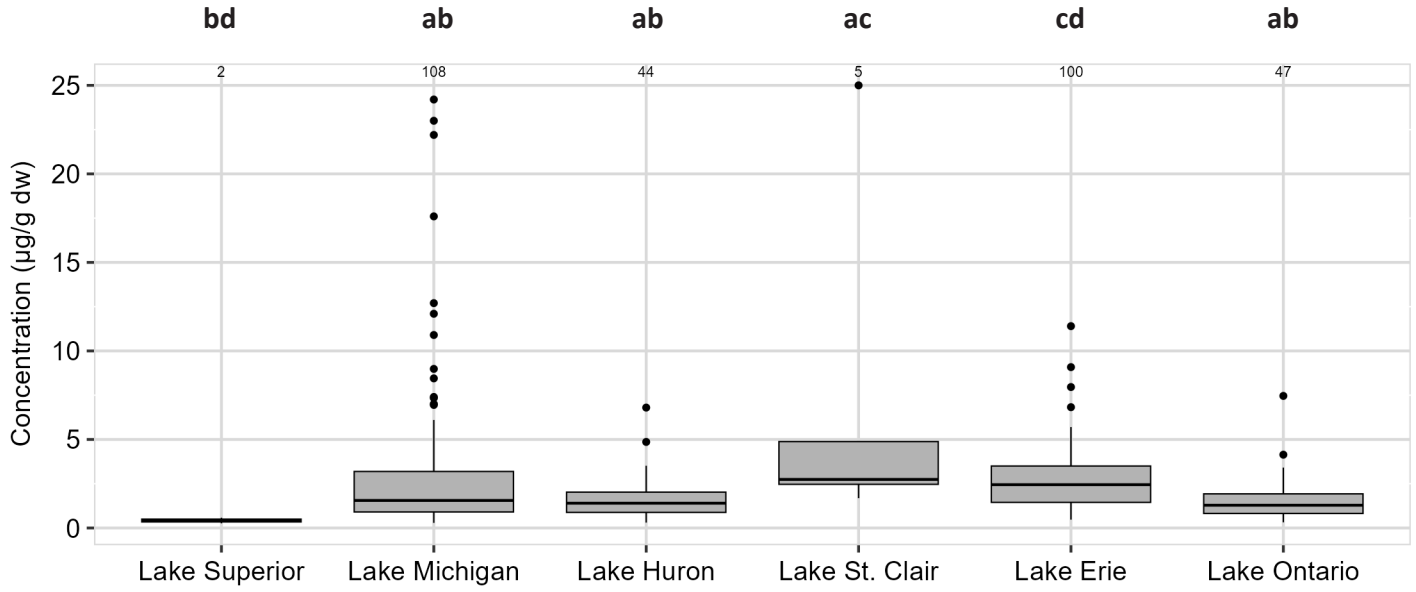


Figure 43. Lead concentrations (µg/g dw) measured in dreissenid mussel tissue in the Great Lakes from 1992 - 2022. Lakes and major rivers including connecting channels are listed geographically from west to east across the Great Lakes Basin. The number of samples collected in each lake is noted at the top of the plot. Letters above each lake indicate significant differences (at α = 0.05).

Results - Lead (Pb)

10.4 Lead Summary

2022 Dreissenid Mussel Tissue Results:

- Pb was detected at 100% of 66 sites surveyed (Figure 39).
- Pb concentration descriptive statistics (Figure 39):
  - Range: 0.29 – 25.00 µg/g dw
  - Minimum Pb concentration was detected at LMML_1
  - Maximum Pb concentration was detected at LCCR
  - Median: 1.15 µg/g dw
  - Mean ± SD: 2.21 ± 3.30 µg/g dw
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster respective to 2022 data only:
  - 88% of sites in low cluster (0.29 - 3.94 µg/g dw)
  - 11% of sites in medium cluster (4.50 - 7.46 µg/g dw)
  - 1% of sites in high cluster (25.00 - 25.00 µg/g dw)

Historic Context for Dreissenid Mussel Tissue (18 out of 66 sites had historic data to analyze for trend analyses):

- 44% of sites surveyed (8/18) were above their historic median concentrations in 2022 (Figure 40).
- Regionally, there was no significant temporal trend of Pb concentrations in dreissenid mussels in the Great Lakes Basin ($p = 0.19$, $rho = 0.28$) (Figure 41; Figure 42).
- 0 sites in the Great Lakes Basin showed significant decreasing temporal trends of Pb concentrations at $\alpha = 0.05$ (Figure 40; Table A4).
- 3 sites (LHTB, LERB, LOCV) in the Great Lakes Basin showed significant increasing temporal trends of Pb concentrations at $\alpha = 0.05$ (Figure 40; Table A4).
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster compared to historic data (Figure 44):
  - 76% of sites in low cluster (0.26 - 2.63 µg/g dw)
  - 14% of sites in medium cluster (2.68 - 4.50 µg/g dw)
  - 10% of sites in high cluster (4.64 - 25.00 µg/g dw)

General Observations:

- Pb concentrations were ubiquitous and at comparatively low to medium concentrations in the Great Lakes Basin in dreissenid mussel tissue in 2022, likely reflecting naturally higher ambient concentrations (Figure 44).
  - A few instances of comparatively high concentrations were found in Lake Michigan (LMMW_1, LMNC, LMWH), Lake St. Clair (LCCR, LSAB), Lake Erie (LERB), and Lake Ontario (LOOC), possibly indicating local inputs of Pb in those areas (Figure 44).
- Regionally, Pb concentrations have not changed significantly over time in dreissenid mussel tissue, indicating no obvious anthropogenic inputs of Pb into the environment (Figure 42).
- Pb was notably high at site LCCR in 2022, with its concentration being an outlier even when compared to all historic region-wide data (Figure 40).
- The post-hoc analysis evaluating differences between lakes indicates that there are no statistical bridges, with all lakes demonstrating a gradient of relationships, being significantly similar to some and significantly different from others. Generally, there appears to be a lot of overlap in Pb concentrations between lakes across the Great Lakes Basin (Figure 43).

Results - Lead (Pb)

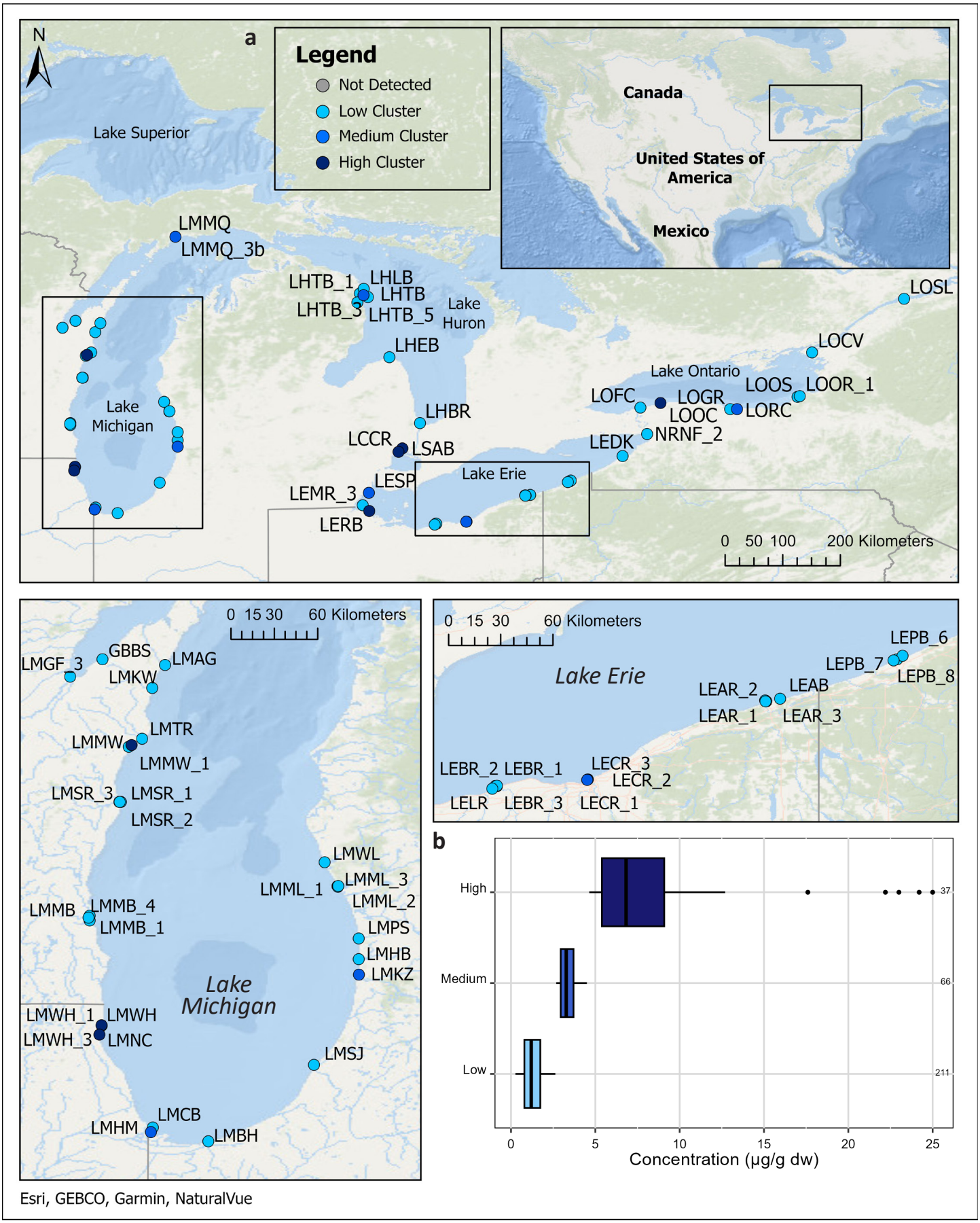


Figure 44. (a) Lead concentrations in dreissenid mussel tissue in 2022 compared to the (b) historic regional MWP Dreissena *species lead concentrations (µg/g dw). Boxplot colors correspond to cluster groups delineated in the map legend. 81 regional sites sampled between 1992 - 2022 for a total of 314 dreissenid mussel samples.*

11.0 RESULTS - MANGANESE (Mn)

11.1 Manganese Chemical Description

Manganese is found in the environment both naturally and from anthropogenic sources including industrial production, pharmaceuticals, and gasoline (ATSDR, 2012c). Manganese occurs naturally in Earth's crust combined with other substances such as oxygen, sulfur, and chlorine, and is also a normal component of air, soil, water, and food. Humans are routinely exposed to low levels of manganese through drinking water, ground water, and soil. Anthropogenic sources of manganese include use in steel production to improve the hardness, stiffness, and strength of carbon steel, stainless steel, high-temperature steel, tool steel, cast iron, and superalloys (ATSDR, 2012c). Manganese can also be found in products such as fireworks, dry-cell batteries, fertilizer, paints, medical imaging agents, cosmetics, and gasoline. Atmospheric manganese can result from industrial activities, mining, automobile exhaust, and cigarette smoke (ATSDR, 2012c).

While manganese is an essential nutrient, it can have adverse effects at high doses (ATSDR, 2012c). Typically, manganese enters the body through inhalation, ingestion, and dermal contact. Primarily, humans are exposed to manganese through manganese-containing nutritional supplements or food, especially grains, beans, nuts, and tea. Additionally, occupations such as welding or steel-making may increase the chances of high manganese exposure. High exposure to manganese often affects the nervous system, leading to behavioral changes and slow, clumsy movements – referred to as “manganism” (ATSDR, 2012c). Additional observed effects include lung irritation resulting in pneumonia and adverse effects on the male reproductive system (ATSDR, 2012c). Typically, the concentrations required to produce these negative effects are twenty thousand times higher than concentrations normally found in the environment. The EPA has established that exposure to manganese in drinking water at concentrations of 1 mg/L for 1 - 10 days is not expected to cause any adverse effects, and OSHA set a legal limit of 5 mg/m³ manganese in air averaged over an 8-hour workday (ATSDR, 2012c).

Manganese does not break down in the environment and can only change form or become attached or separated from particles (ATSDR, 2012c). The chemical state of manganese and the type of soil determine how fast it moves through the soil and how much is retained in the soil. Most manganese in water tends to attach to particles or settle into the sediment and may be significantly bioconcentrated at lower trophic levels and in fish. Manganese found in gasoline additives may degrade in the environment quickly when exposed to sunlight, releasing manganese (ATSDR, 2012c).

11.2 Magnitude and Distribution of Manganese in Dreissenid Mussel Tissue in 2022

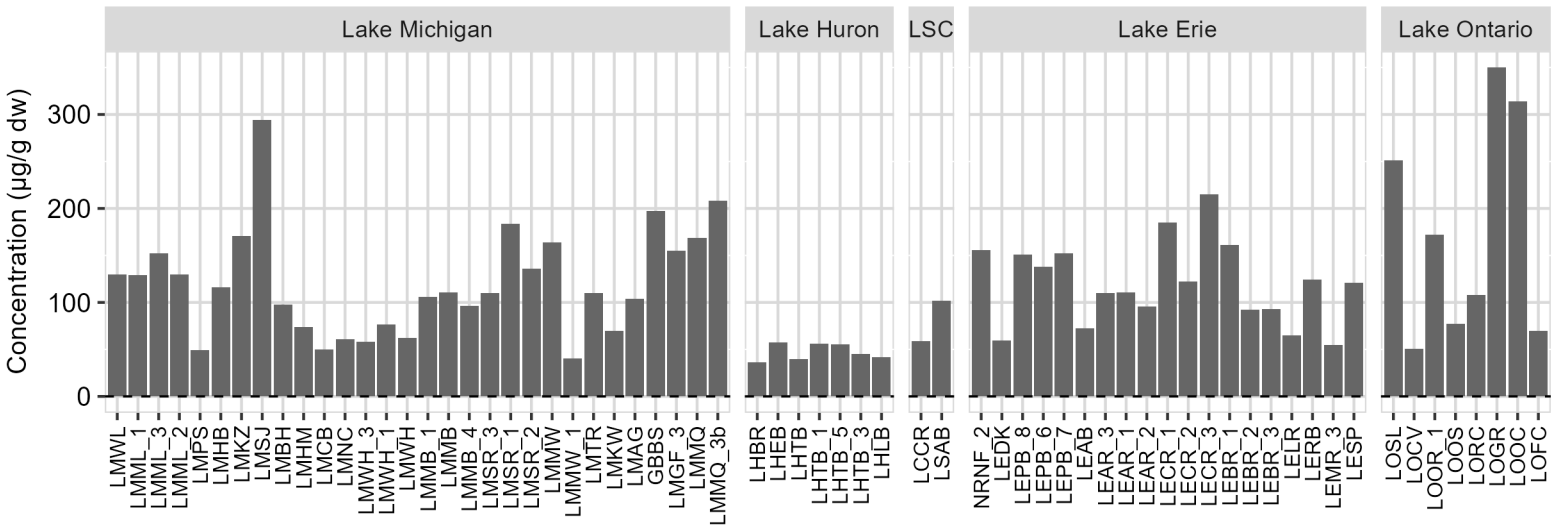


Figure 45. Bar graph showing magnitude of manganese concentrations (µg/g dw) detected in dreissenid mussel tissue in the Great Lakes Basin in 2022. Dotted line represents the minimum weight corrected MDL. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair.

Results - Manganese (Mn)

11.3 Historical Context of Manganese Magnitude and Distribution in Dreissenid Mussel Tissue

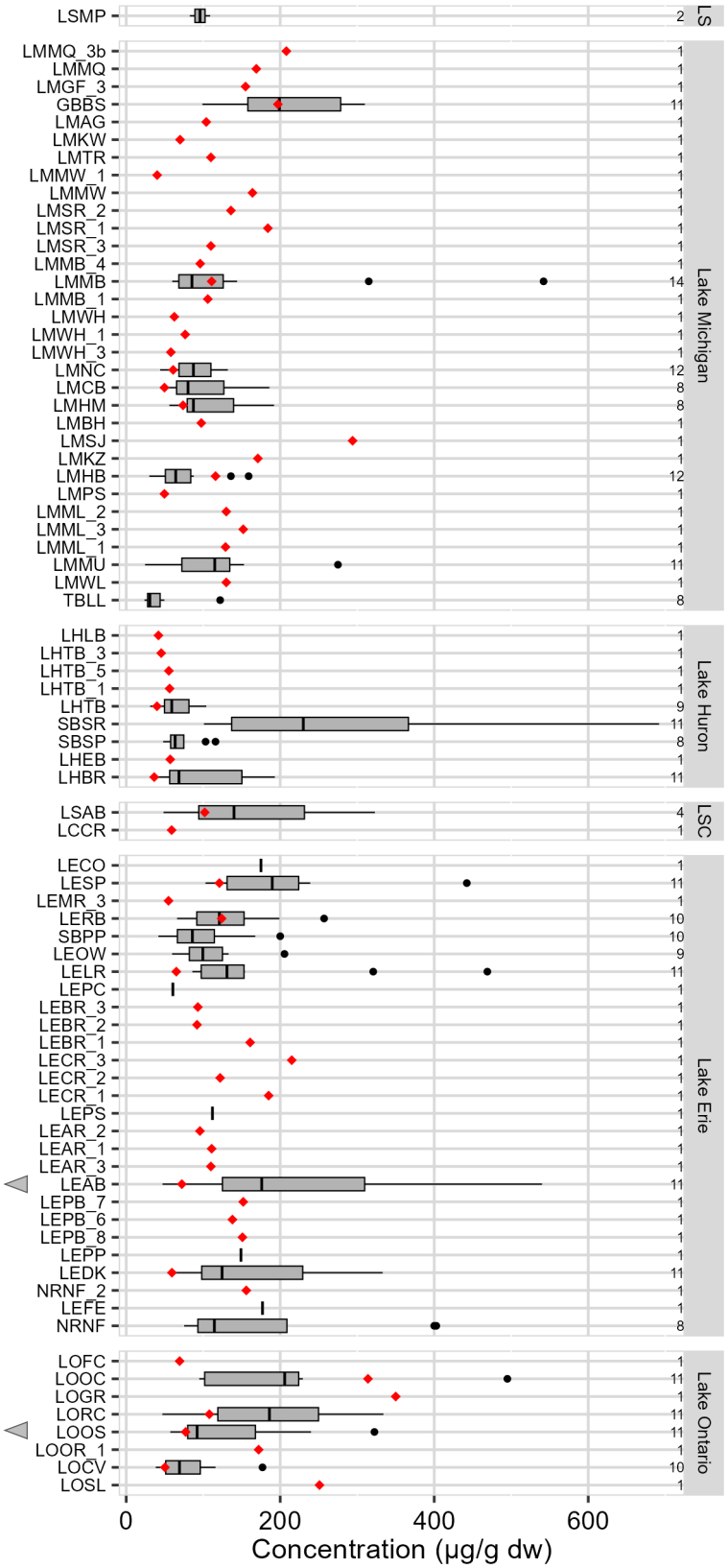


Figure 46. Manganese concentrations (µg/g dw) in dreissenid mussel tissue in 2022 (red diamonds) compared to boxplots of the historic manganese concentrations for each site (1992 - 2008) and results of the trend analysis showing significant increasing (▶) or decreasing (◀) site-specific contamination trends over time. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair; LS = Lake Superior. The number of years each site has been sampled since 1992 is noted at the right of the plot for each site.

Results - Manganese (Mn)

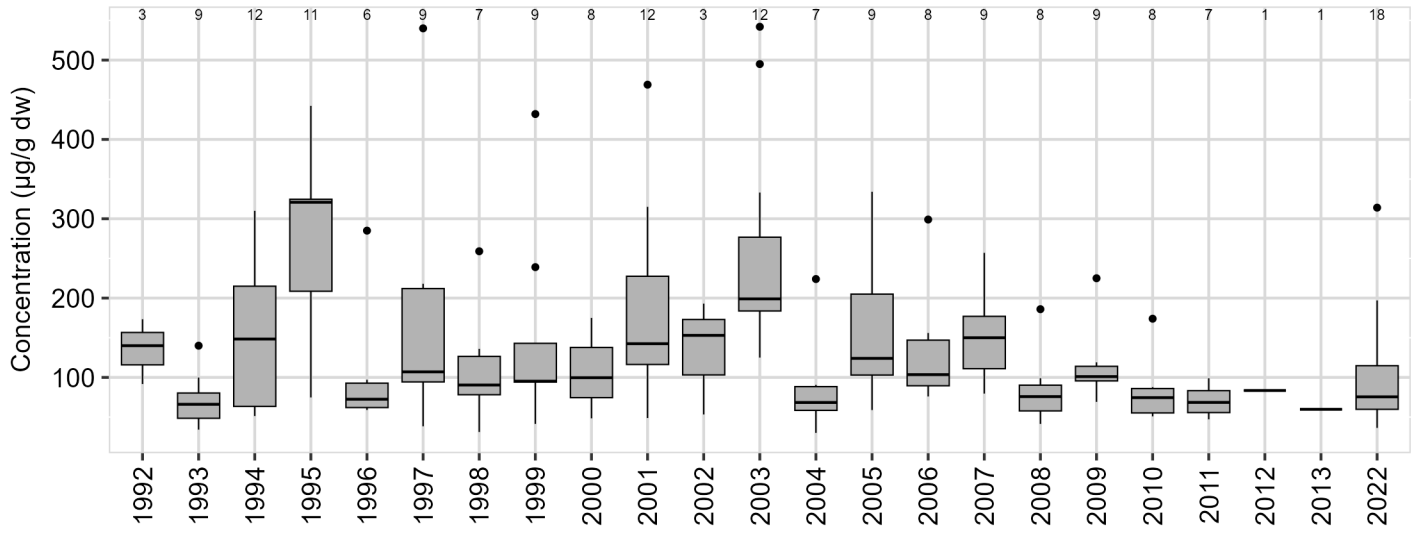


Figure 47. Boxplots representing the historic manganese concentrations (µg/g dw) in dreissenid mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot.

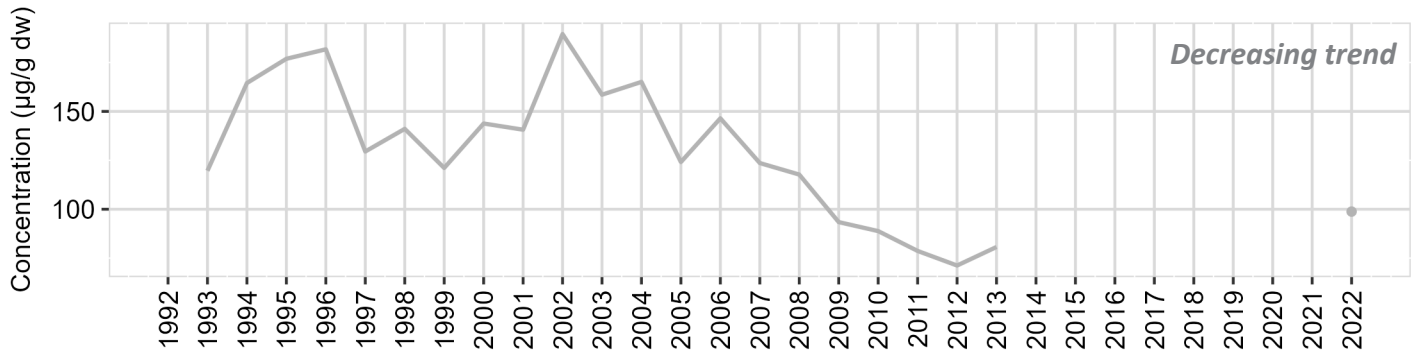


Figure 48. Three-point moving average of the yearly mean manganese concentrations (µg/g dw) in dreissenid mussel tissue in the sites analyzed in this study. Average concentration for 2022 is included as a standalone point for reference.

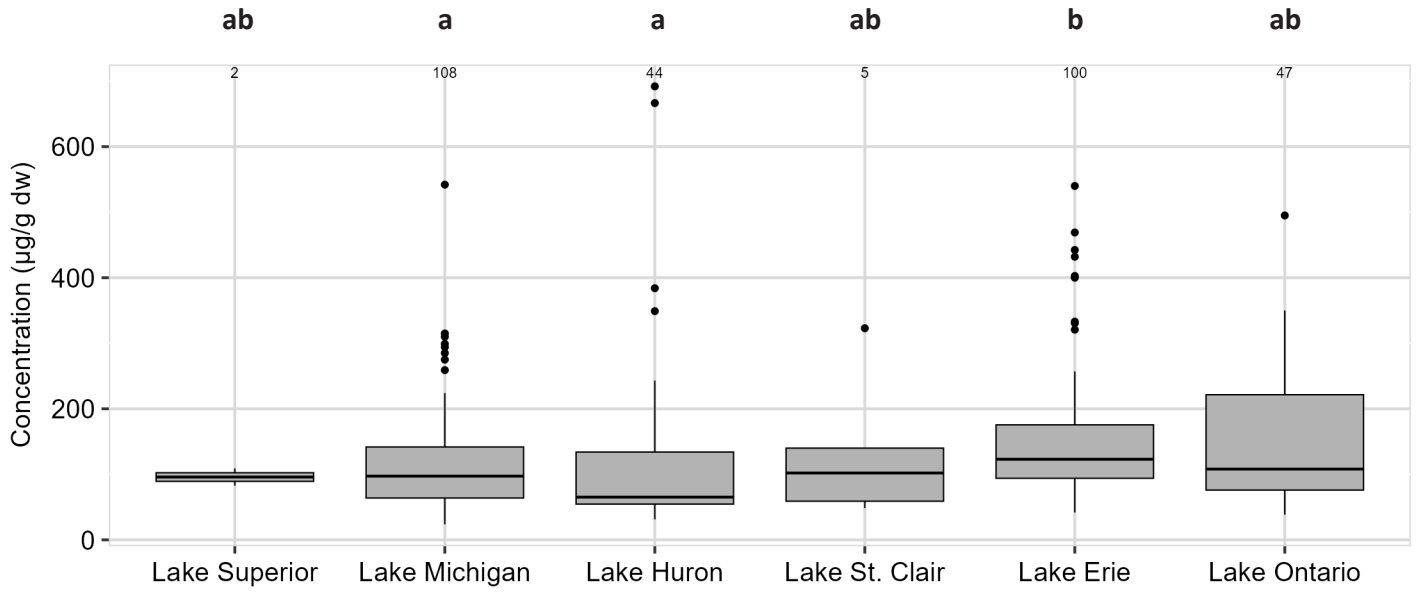


Figure 49. Manganese concentrations (µg/g dw) measured in dreissenid mussel tissue in the Great Lakes from 1992 - 2022. Lakes and major rivers including connecting channels are listed geographically from west to east across the Great Lakes Basin. The number of samples collected in each lake is noted at the top of the plot. Letters above each lake indicate significant differences (at α = 0.05).

Results - Manganese (Mn)

11.4 Manganese Summary

2022 Dreissenid Mussel Tissue Results:

- Mn was detected at 100% of 66 sites surveyed (Figure 45).
- Mn concentration descriptive statistics (Figure 45):
  - Range: 36.37 – 350.00 µg/g dw
  - Minimum Mn concentration was detected at LHBR
  - Maximum Mn concentration was detected at LOGR
  - Median: 108.99 µg/g dw
  - Mean ± SD: 117.80 ± 66.54 µg/g dw
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster respective to 2022 data only:
  - 71% of sites in low cluster (36.37 - 137.97 µg/g dw)
  - 24% of sites in medium cluster (150.97 - 251.00 µg/g dw)
  - 5% of sites in high cluster (294.00 - 350.00 µg/g dw)

Historic Context for Dreissenid Mussel Tissue (18 out of 66 sites had historic data to analyze for trend analyses):

- 22% of sites surveyed (4/18) were above their historic median concentrations in 2022 (Figure 46).
- Regionally, there was a significant decreasing temporal trend of Mn concentrations in dreissenid mussels in the Great Lakes Basin ($p = 0.02$, $rho = -0.48$) (Figure 47; Figure 48).
- 2 sites (LEAB, LOOS) in the Great Lakes Basin showed significant decreasing temporal trends of Mn concentrations at $\alpha = 0.05$ (Figure 46; Table A4).
- 0 sites in the Great Lakes Basin showed significant increasing temporal trends of Mn concentrations at $\alpha = 0.05$ (Figure 46; Table A4).
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster compared to historic data (Figure 50):
  - 68% of sites in low cluster (23.70 - 133.00 µg/g dw)
  - 27% of sites in medium cluster (136.00 - 259.00 µg/g dw)
  - 5% of sites in high cluster (275.00 - 716.87 µg/g dw)

General Observations:

- Mn concentrations were ubiquitous and were detected across a large range of concentrations in the Great Lakes Basin in dreissenid mussel tissue in 2022, likely reflecting naturally higher ambient concentrations with additional localized inputs (Figure 50).
  - A few instances of comparatively high concentrations were found in Lake Michigan (LMSJ) and Lake Ontario (LOOC, LOGR), possibly indicating local inputs of Mn in those areas (Figure 50).
- Regionally, Mn concentrations have significantly decreased over time in dreissenid mussel tissue, indicating decreasing inputs of Mn into the environment (Figure 48).
- The post-hoc analysis evaluating differences between lakes indicates that there are no statistical bridges, with all lakes demonstrating a gradient of relationships, being significantly similar to some and significantly different from others. Generally, higher concentrations of Mn were found in Lake Erie, while lower concentrations of Mn were found in lakes Michigan and Huron (Figure 49).

Results - Manganese (Mn)

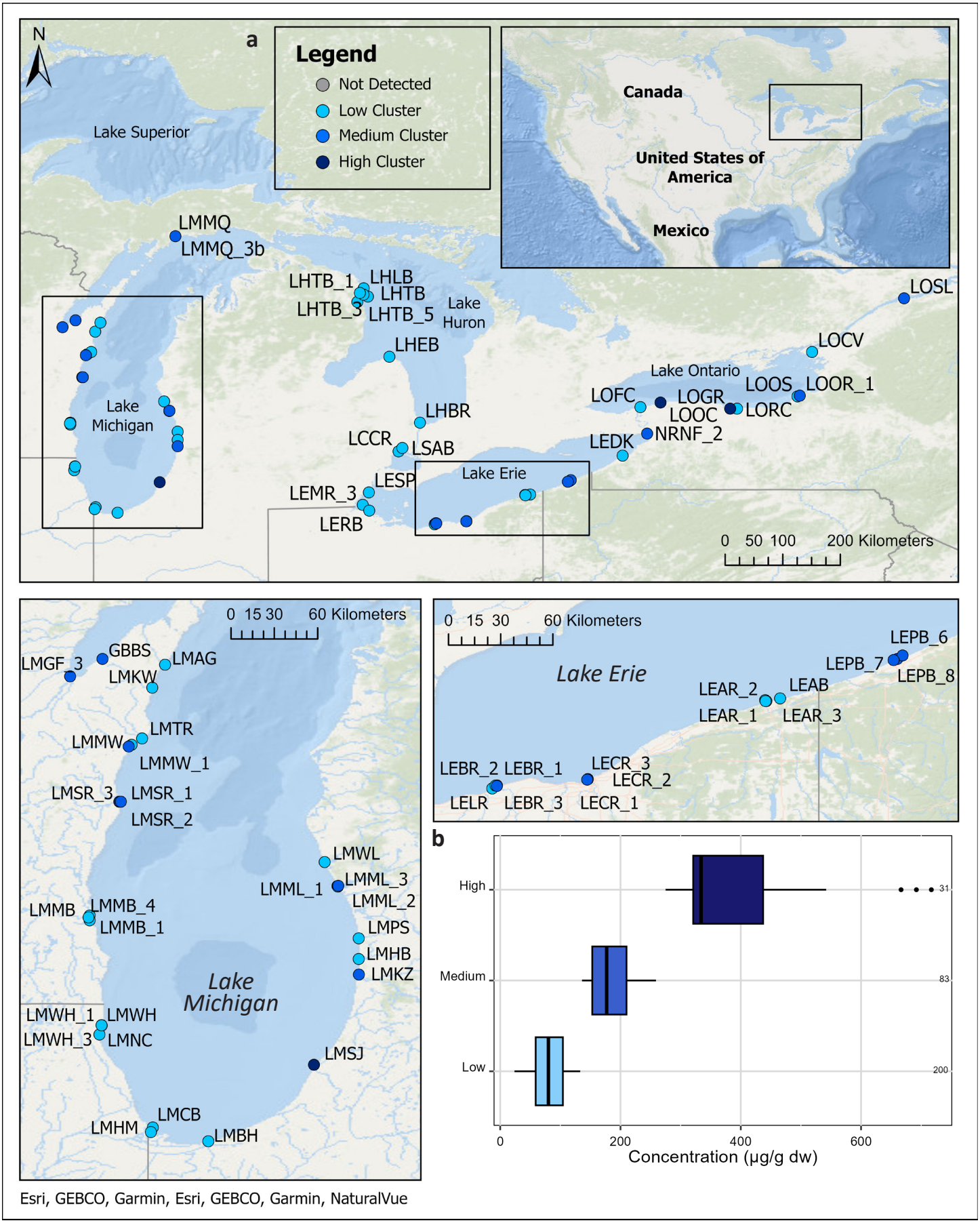


Figure 50. (a) Manganese concentrations in dreissenid mussel tissue in 2022 compared to the (b) historic regional MWP Dreissena *species manganese concentrations (µg/g dw). Boxplot colors correspond to cluster groups delineated in the map legend. 81 regional sites sampled between 1992 - 2022 for a total of 314 dreissenid mussel samples.*

12.0 RESULTS - MERCURY (Hg)

12.1 Mercury Chemical Description

Mercury is naturally found as mercuric sulfide from forest fires, crustal ores, fossil fuels, and volcanoes. Anthropogenic sources of mercury include mining, gold refining, coal fired power plants, and the wood pulp industry. Elevated levels occur as a result of human activity (ATSDR, 2012d). In the US, coal-fired electric turbines, municipal and medical waste incinerators, mining, landfills, and sewage sludge are the primary sources of mercury emissions into the air.

Mercury is a highly toxic, non-essential trace metal that occurs naturally. Mercury is a human neurotoxin that also affects the kidneys and developing fetuses. The most common human exposure route for mercury is the consumption of contaminated food. The U.S. FDA has not established a safety level for mercury but has set a safety level of 1.0 ppm wet weight for methyl mercury, the form most likely to impact animals and humans (U.S. FDA, 2011). The MWP measures total mercury, of which methyl mercury is only one component. Children, pregnant women, or women likely to become pregnant are advised to avoid consumption of swordfish, shark, king mackerel, and tilefish, and should limit consumption to fish and shellfish recommended by U.S. FDA and U.S. EPA (U.S. EPA, 1979; U.S. FDA, 2011). Mercury exposure can also reduce growth and reproduction rates in other phyla such as copepods (Eisler, 1987).

In the environment, mercury may change forms between elemental, inorganic and organic (ATSDR, 2012d). Natural sinks, such as sediment and soil, represent the largest environmental source of mercury (ATSDR, 2012d). Estimates suggest that wet and dry deposition accounts for 50-90% of the mercury load to many estuaries, making atmospheric transport a significant source of mercury worldwide (ATSDR, 2012d). Long range atmospheric transport is responsible for the presence of mercury at or above background levels in surface waters in remote areas (ATSDR, 2012d).

12.2 Magnitude and Distribution of Mercury in Dreissenid Mussel Tissue in 2022

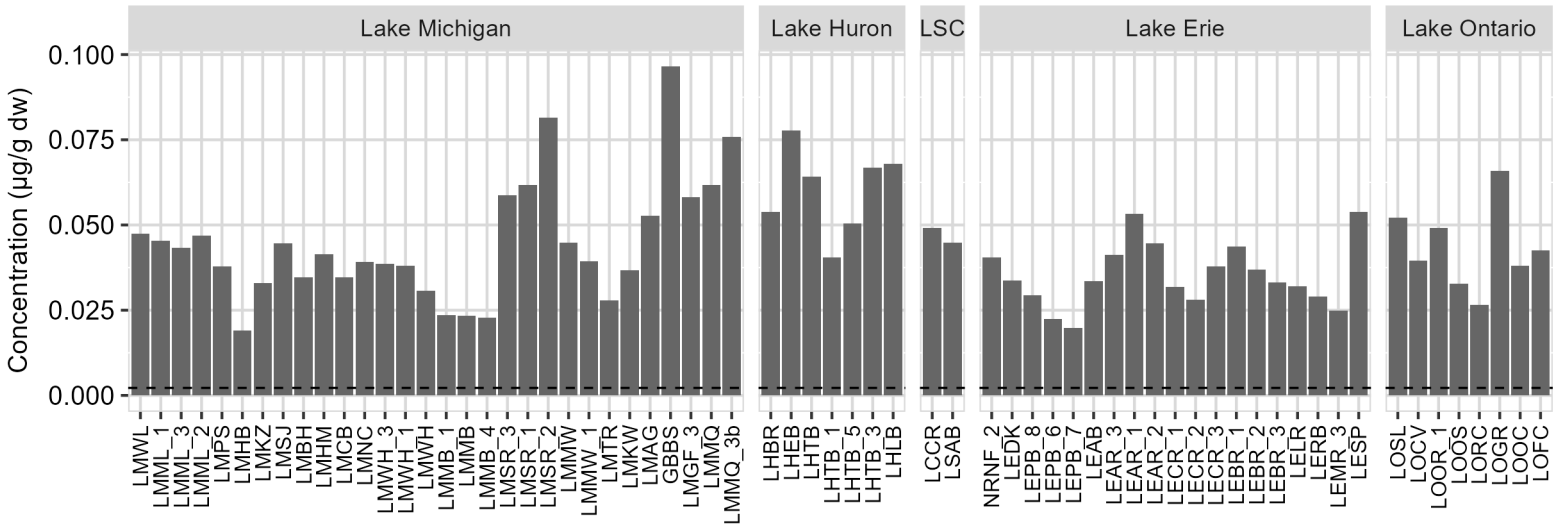


Figure 51. Bar graph showing magnitude of mercury concentrations (µg/g dw) detected in dreissenid mussel tissue in the Great Lakes Basin in 2022. Dotted line represents the minimum weight corrected MDL. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair.

12.3 Historical Context of Mercury Magnitude and Distribution in Dreissenid Mussel Tissue

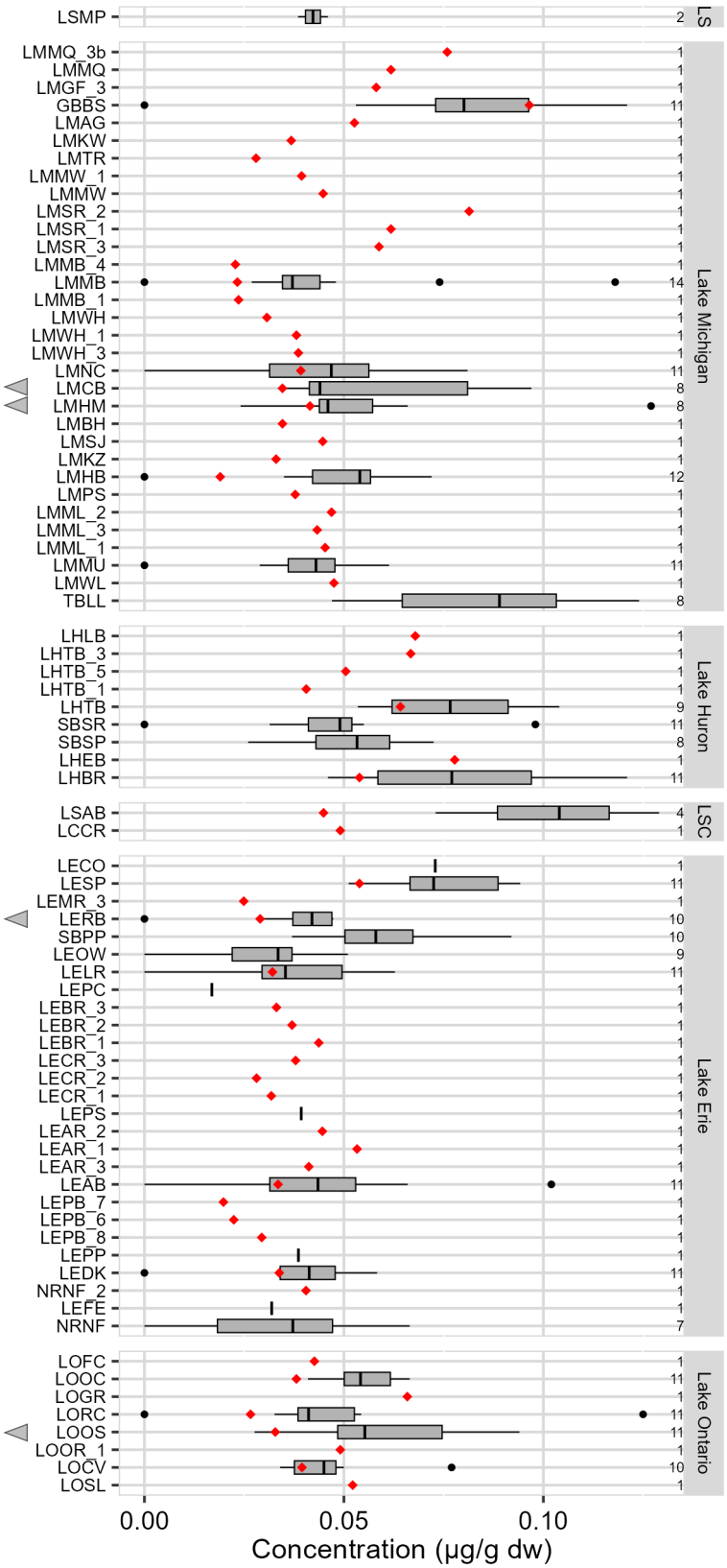


Figure 52. Mercury concentrations (µg/g dw) in dreissenid mussel tissue in 2022 (red diamonds) compared to boxplots of the historic mercury concentrations for each site (1992 - 2008) and results of the trend analysis showing significant increasing (▶) or decreasing (◀) site-specific contamination trends over time. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair; LS = Lake Superior.The number of years each site has been sampled since 1992 is noted at the right of the plot for each site. Site LMNC in 1998 (0.27 µg/g dw) was removed to aid in visualization.

Results - Mercury (Hg)

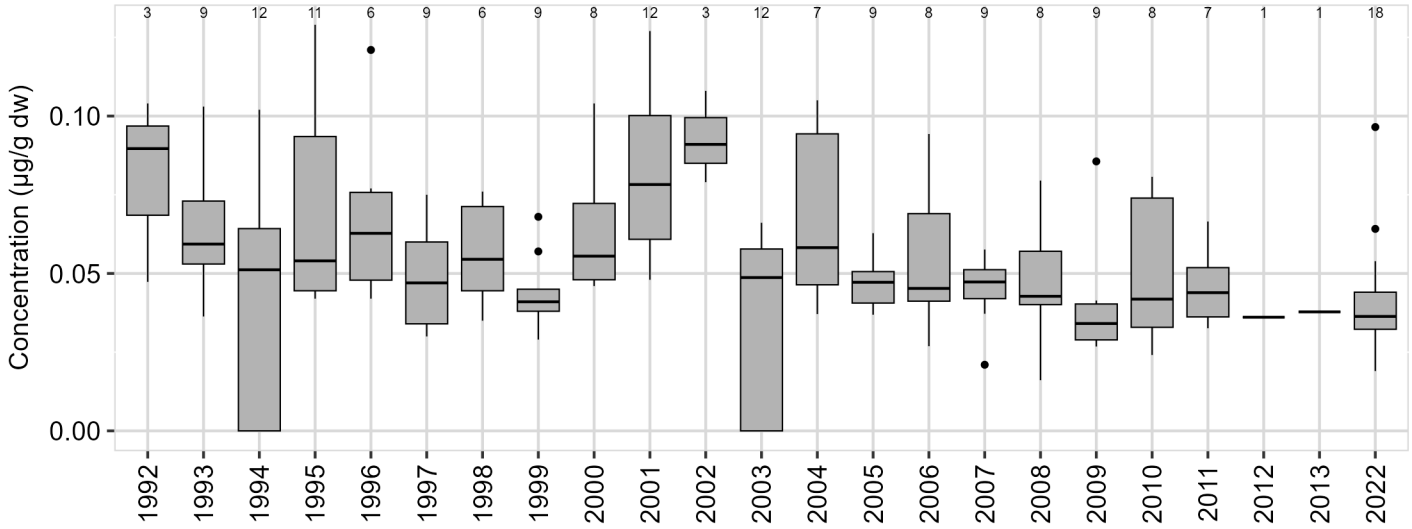


Figure 53. Boxplots representing the historic mercury concentrations (µg/g dw) in dreissenid mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot. Site LMNC in 1998 (0.27 µg/g dw) was removed to aid in visualization.

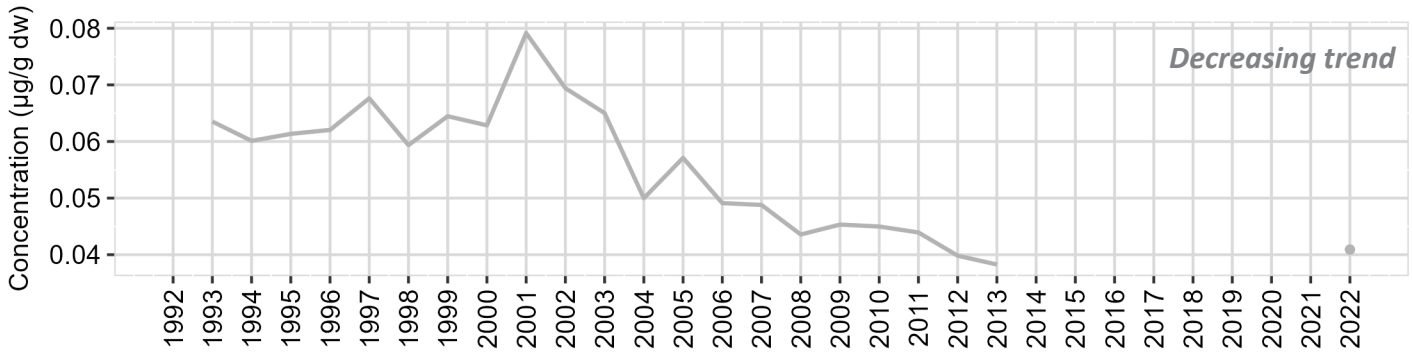


Figure 54. Three-point moving average of the yearly mean mercury concentrations (µg/g dw) in dreissenid mussel tissue in the sites analyzed in this study. Average concentration for 2022 is included as a standalone point for reference.

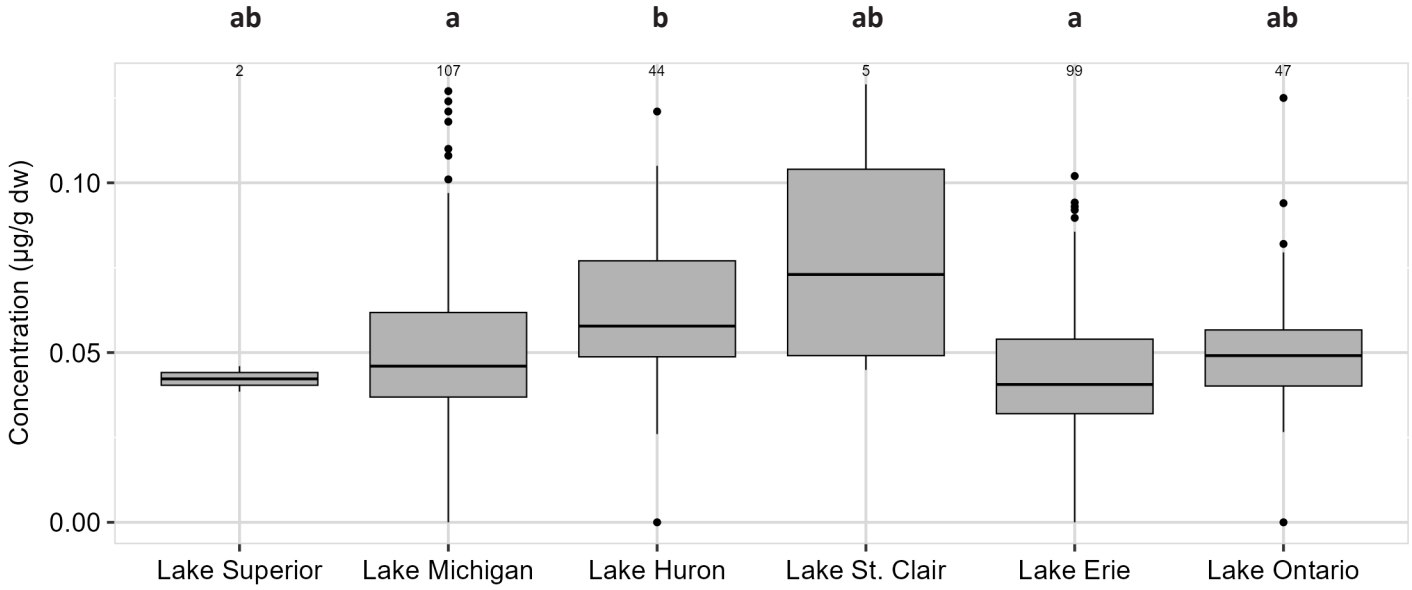


Figure 55. Mercury concentrations (µg/g dw) measured in dreissenid mussel tissue in the Great Lakes from 1992 - 2022. Lakes and major rivers including connecting channels are listed geographically from west to east across the Great Lakes Basin. The number of samples collected in each lake is noted at the top of the plot. Letters above each lake indicate significant differences (at α = 0.05). Site LMNC in 1998 (0.27 µg/g dw) was removed to aid in visualization.

12.4 Mercury Summary

2022 Dreissenid Mussel Tissue Results:

- Hg was detected at 100% of 66 sites surveyed (Figure 51).
- Hg concentration descriptive statistics (Figure 51):
  - Range: 0.02 – 0.10 µg/g dw
  - Minimum Hg concentration was detected at LMHB
  - Maximum Hg concentration was detected at GBBS
  - Median: 0.04 µg/g dw
  - Mean ± SD: 0.04 ± 0.02 µg/g dw
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster respective to 2022 data only:
  - 67% of sites in low cluster (0.02 - 0.05 µg/g dw)
  - 27% of sites in medium cluster (0.05 - 0.07 µg/g dw)
  - 6% of sites in high cluster (0.08 - 0.10 µg/g dw)

Historic Context for Dreissenid Mussel Tissue (18 out of 66 sites had historic data to analyze for trend analyses):

- 6% of sites surveyed (1/18) were above their historic median concentrations in 2022 (Figure 52).
- Regionally, there was a significant decreasing temporal trend of Hg concentrations in dreissenid mussels in the Great Lakes Basin ($p = 0.004$, $rho = -0.57$) (Figure 53; Figure 54).
- 4 sites (LMCB, LMHM, LERB, LOOS) in the Great Lakes Basin showed significant decreasing temporal trends of Hg concentrations at $\alpha = 0.05$ (Figure 52; Table A4).
- 0 sites in the Great Lakes Basin showed significant increasing temporal trends of Hg concentrations at $\alpha = 0.05$ (Figure 52; Table A4).
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster compared to historic data (Figure 56):
  - 74% of sites in low cluster (0.00 - 0.05 µg/g dw)
  - 24% of sites in medium cluster (0.05 - 0.09 µg/g dw)
  - 2% of sites in high cluster (0.09 - 0.27 µg/g dw)

General Observations:

- Hg concentrations were ubiquitous and at comparatively low to medium concentrations in the Great Lakes Basin in dreissenid mussel tissue in 2022, likely reflecting naturally higher ambient concentrations (Figure 56).
  - One comparatively high concentration was found in Lake Michigan (GBBS), possibly indicating local inputs of Hg in that area (Figure 56).
- Regionally, Hg concentrations have significantly decreased over time in dreissenid mussel tissue, indicating decreasing inputs of Hg into the environment (Figure 54).
- The post-hoc analysis evaluating differences between lakes indicates that there are no statistical bridges, with all lakes demonstrating a gradient of relationships, being significantly similar to some and significantly different from others. Generally, higher concentrations of Hg were found in Lake Huron, while lower concentrations of Hg were found in lakes Michigan and Erie (Figure 55).

Results - Mercury (Hg)

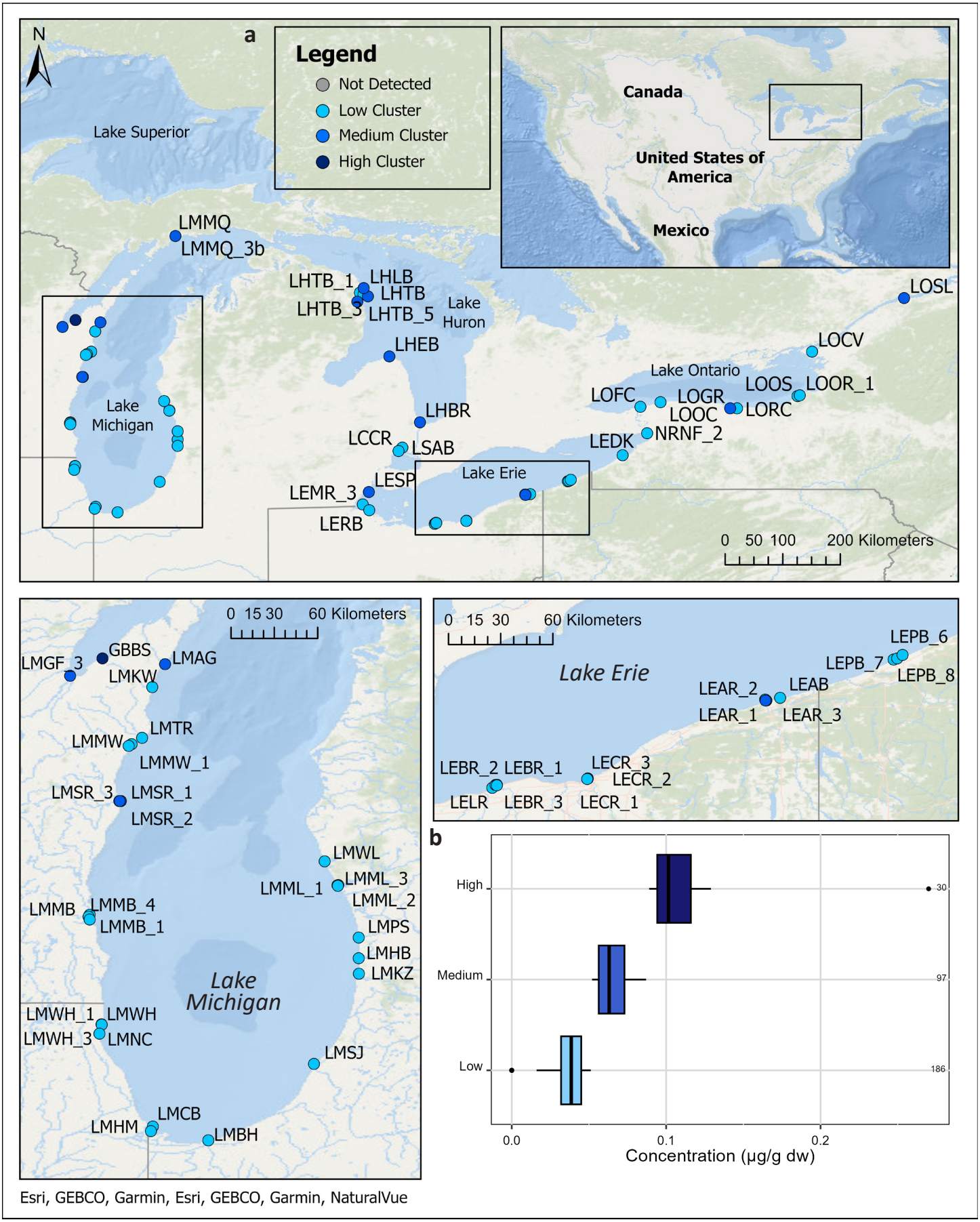


Figure 56. (a) Mercury concentrations in dreissenid mussel tissue in 2022 compared to the (b) historic regional MWP Dreissena *species mercury concentrations (µg/g dw). Boxplot colors correspond to cluster groups delineated in the map legend. 81 regional sites sampled between 1992 - 2022 for a total of 314 dreissenid mussel samples.*

13.0 RESULTS - NICKEL (Ni)

13.1 Nickel Chemical Description

Nickel is naturally occurring and widely distributed in the environment. It exists in alloy form in combination with other metals and as a soluble element (ATSDR, 2005a). Naturally, nickel is derived from weathering rocks and soil and is transported to streams and rivers by runoff. River and stream inputs of nickel are the largest sources for oceans and coastal waters. Anthropogenic sources of nickel include stainless steel, nickel-cadmium batteries, pigments, computers, wire, coinage, and electroplating (ATSDR, 2005a). Atmospheric sources are usually not significant, except in the Great Lakes, where atmospheric input of nickel accounts for 60-80% of the total anthropogenic input to Lake Superior and 20-70% of total inputs to Lakes Erie and Ontario (Nriagu et al., 1995). In Lakes Erie and Ontario, most of the nickel is derived from municipal and industrial wastewaters and thus is likely to be complexed with organic matter (Sweet et al., 1998).

Nickel is a biologically essential trace element for animals and plants and is widely distributed in the environment at very low concentrations (ATSDR, 2005a). Exposure to large doses of nickel can cause serious health effects, such as bronchitis, and long-term exposure can result in cancer. Food is the major source of human exposure to nickel (ATSDR, 2005a). Safety guidance levels for nickel in fish and shellfish are no longer listed by the U.S. FDA (U.S. FDA, 2011).

Nickel derived from weathering rocks and soil is transported to streams and rivers by runoff. It accumulates in sediment and becomes inert when it is incorporated into minerals. There is no evidence that nickel biomagnifies in the food chain (McGeer et al., 2003; Suedel et al., 1994). Complexation reduces the chemical reactivity and bioavailability of Ni and may account for the high concentrations and long residence times in the water column (Nriagu et al., 1995).

13.2 Magnitude and Distribution of Nickel in Dreissenid Mussel Tissue in 2022

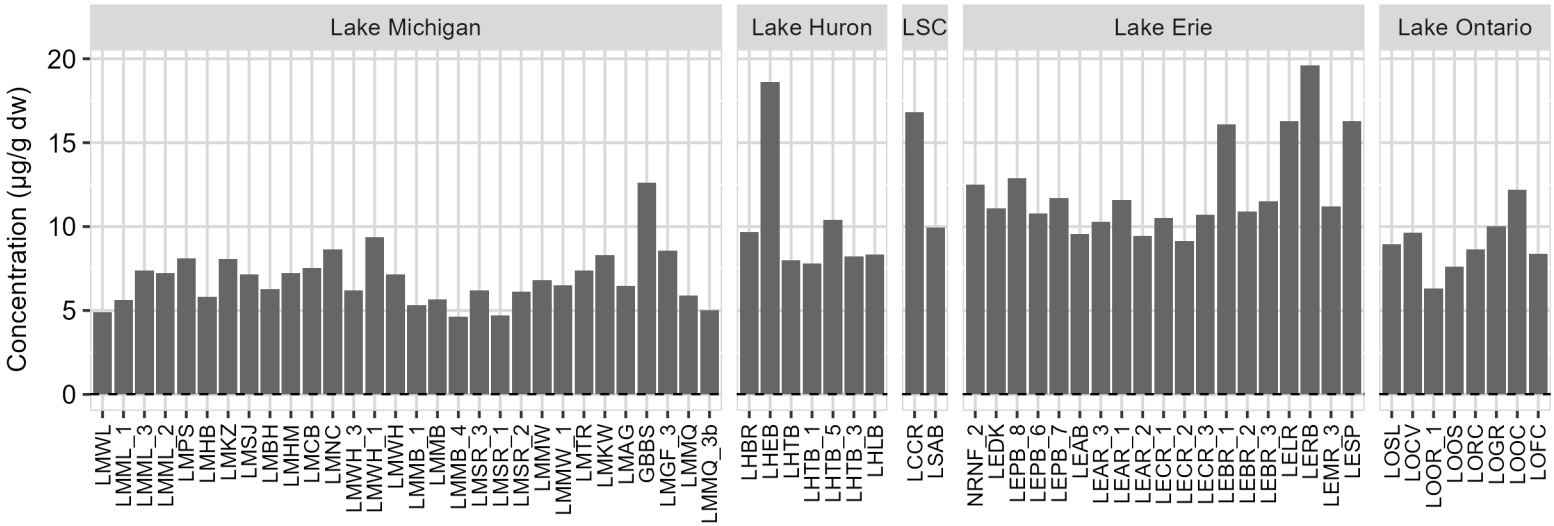


Figure 57. Bar graph showing magnitude of nickel concentrations (µg/g dw) detected in dreissenid mussel tissue in the Great Lakes Basin in 2022. Dotted line represents the minimum weight corrected MDL. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair.

13.3 Historical Context of Nickel Magnitude and Distribution in Dreissenid Mussel Tissue

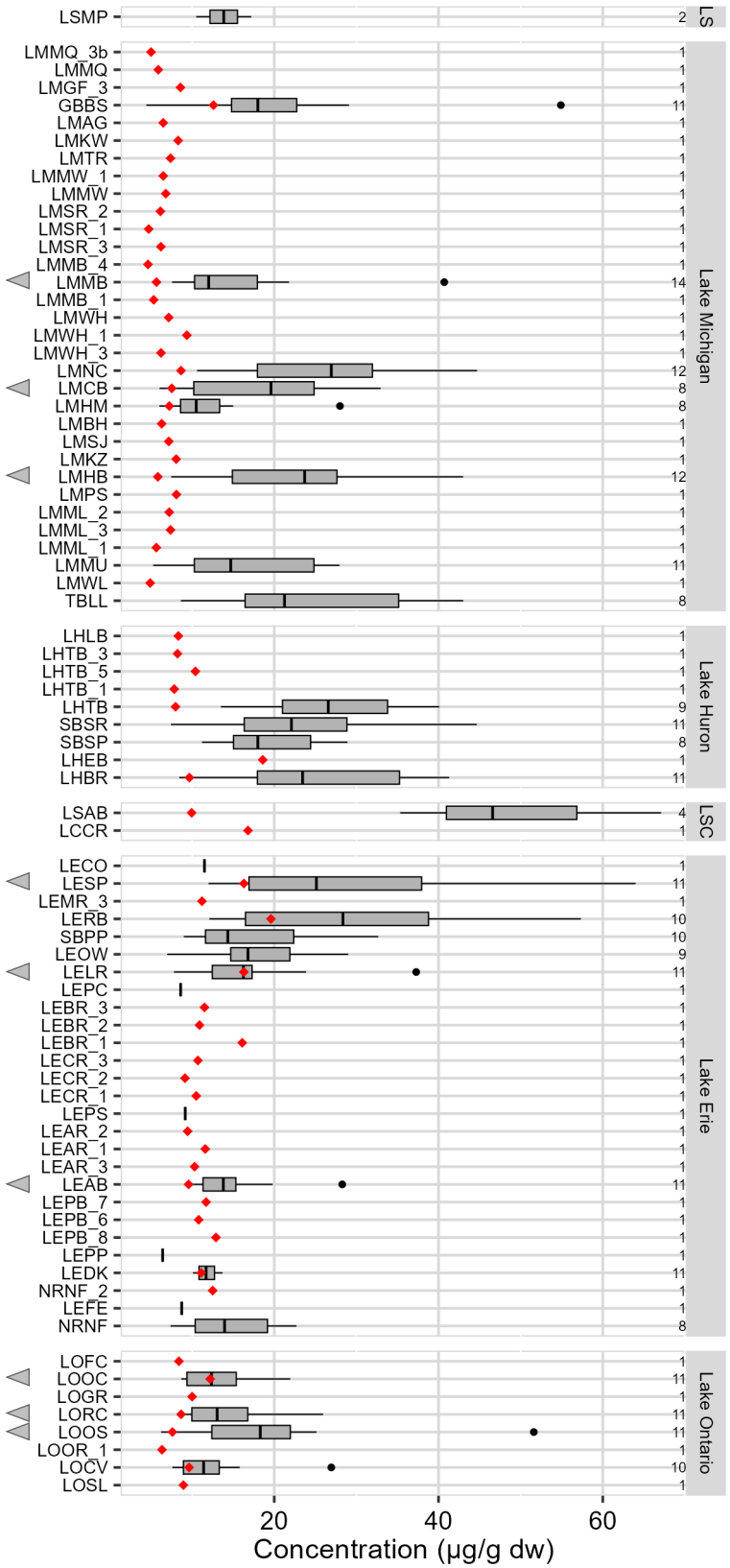


Figure 58. Nickel concentrations (µg/g dw) in dreissenid mussel tissue in 2022 (red diamonds) compared to boxplots of the historic nickel concentrations for each site (1992 - 2008) and results of the trend analysis showing significant increasing (►) or decreasing (◄) site-specific contamination trends over time. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair; LS = Lake Superior. The number of years each site has been sampled since 1992 is noted at the right of the plot for each site.

Results - Nickel (Ni)

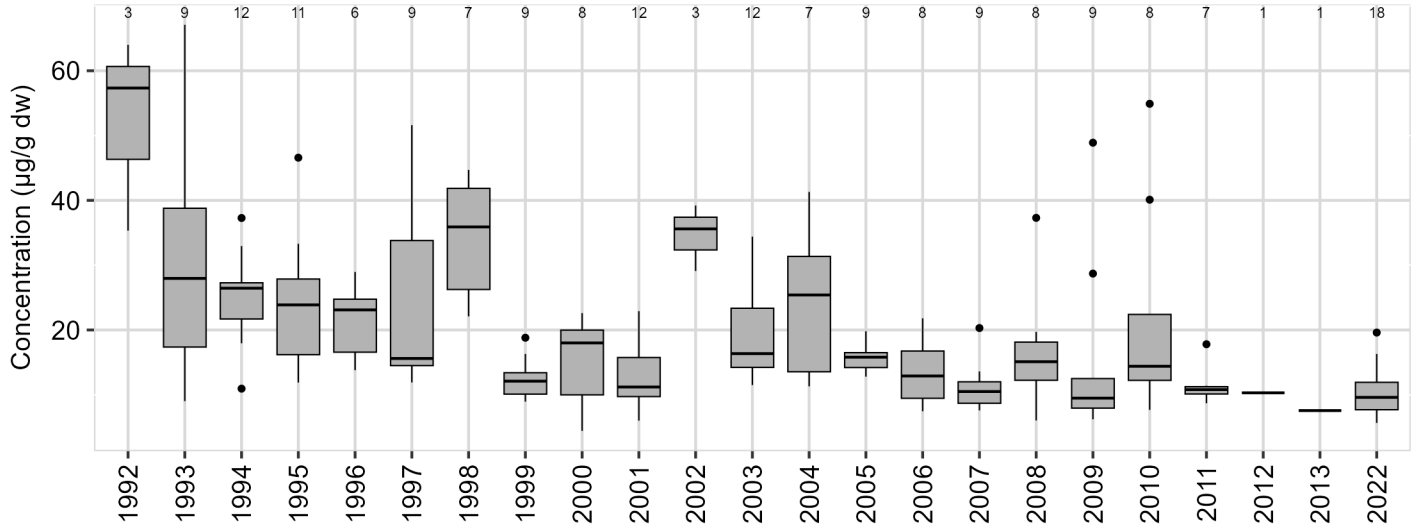


Figure 59. Boxplots representing the historic nickel concentrations (µg/g dw) in dreissenid mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot.

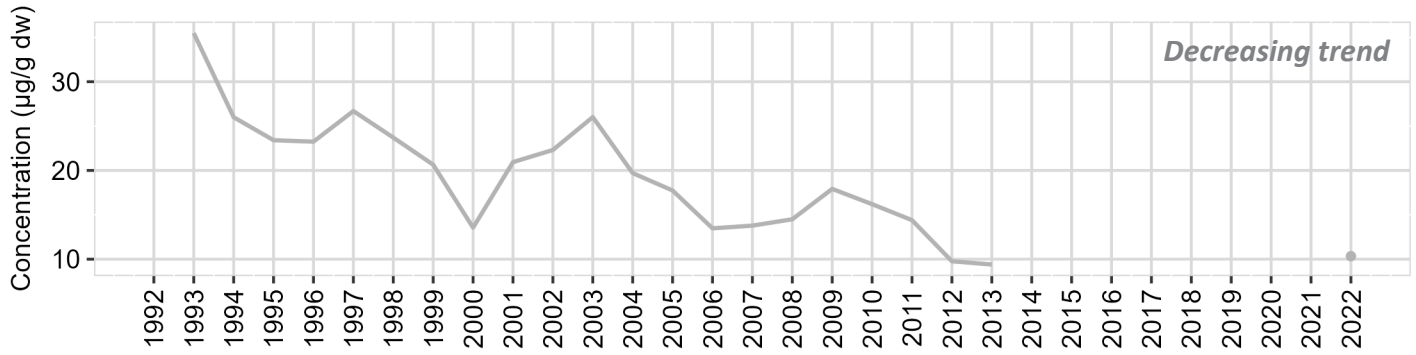


Figure 60. Three-point moving average of the yearly mean nickel concentrations (µg/g dw) in dreissenid mussel tissue in the sites analyzed in this study. Average concentration for 2022 is included as a standalone point for reference.

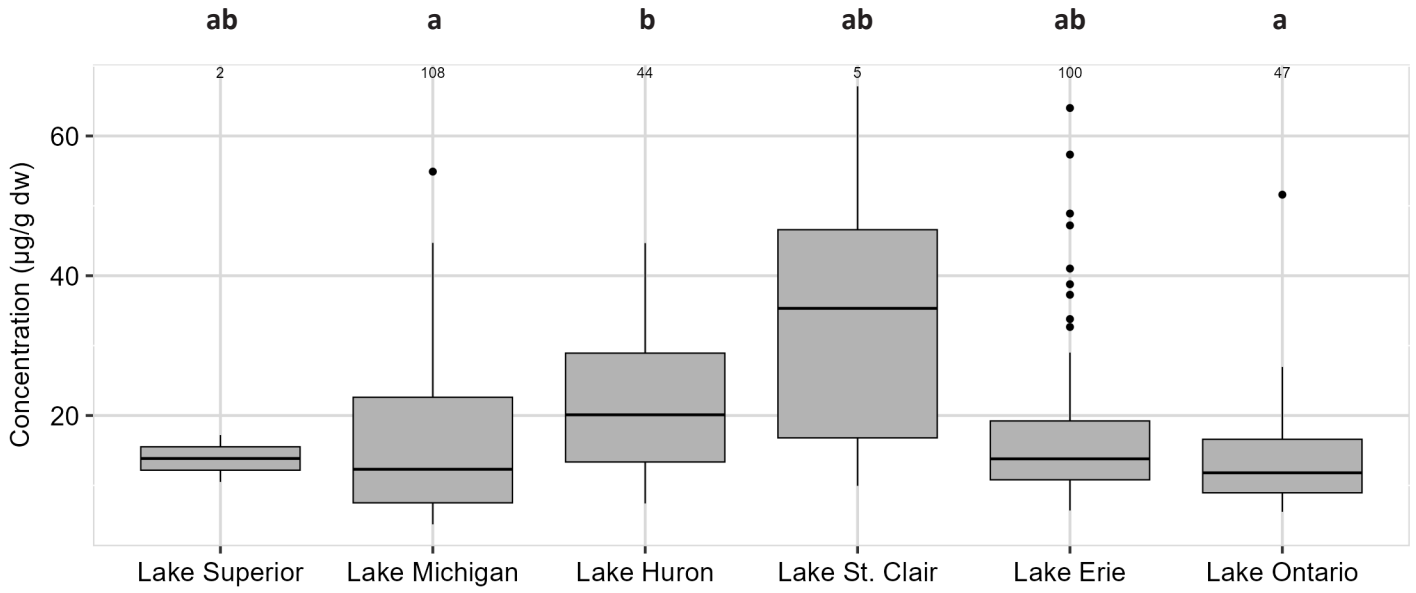


Figure 61. Nickel concentrations (µg/g dw) measured in dreissenid mussel tissue in the Great Lakes from 1992 - 2022. Lakes and major rivers including connecting channels are listed geographically from west to east across the Great Lakes Basin. The number of samples collected in each lake is noted at the top of the plot. Letters above each lake indicate significant differences (at α = 0.05).

Results - Nickel (Ni)

13.4 Nickel Summary

2022 Dreissenid Mussel Tissue Results:

- Ni was detected at 100% of 66 sites surveyed (Figure 57).
- Ni concentration descriptive statistics (Figure 57):
  - Range: 4.63 – 19.60 µg/g dw
  - Minimum Ni concentration was detected at LMMB_4
  - Maximum Ni concentration was detected at LERB
  - Median: 8.49 µg/g dw
  - Mean ± SD: 9.22 ± 3.35 µg/g dw
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster respective to 2022 data only:
  - 55% of sites in low cluster (4.63 - 8.66 µg/g dw)
  - 36% of sites in medium cluster (8.94 - 12.90 µg/g dw)
  - 9% of sites in high cluster (16.10 - 19.60 µg/g dw)

Historic Context for Dreissenid Mussel Tissue (18 out of 66 sites had historic data to analyze for trend analyses):

- 6% of sites surveyed (1/18) were above their historic median concentrations in 2022 (Figure 58).
- Regionally, there was a significant decreasing temporal trend of Ni concentrations in dreissenid mussels in the Great Lakes Basin ($p < 0.001$, rho = -0.73) (Figure 59; Figure 60).
- 9 sites (LMMB, LMCB, LMHB, LESP, LELR, LEAB, LOOC, LORC, LOOS) in the Great Lakes Basin showed significant decreasing temporal trends of Ni concentrations at $\alpha = 0.05$ (Figure 58; Table A4).
- 0 sites in the Great Lakes Basin showed significant increasing temporal trends of Ni concentrations at $\alpha = 0.05$ (Figure 58; Table A4).
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster compared to historic data (Figure 62):
  - 100% of sites in low cluster (4.43 - 20.30 µg/g dw)
  - 0% of sites in medium cluster (20.95 - 30.90 µg/g dw)
  - 0% of sites in high cluster (32.67 - 67.10 µg/g dw)

General Observations:

- Ni concentrations were ubiquitous but at comparatively low concentrations in the Great Lakes Basin in dreissenid mussel tissue in 2022, likely reflecting natural ambient concentrations (Figure 62).
- Regionally, Ni concentrations have significantly decreased over time in dreissenid mussel tissue, indicating decreasing inputs of Ni into the environment (Figure 60).
- The post-hoc analysis evaluating differences between lakes indicates that there are no statistical bridges, with all lakes demonstrating a gradient of relationships, being significantly similar to some and significantly different from others. Generally, higher concentrations of Ni were found in Lake Huron, while lower concentrations of Ni were found in lakes Michigan and Ontario (Figure 61).

Results - Nickel (Ni)

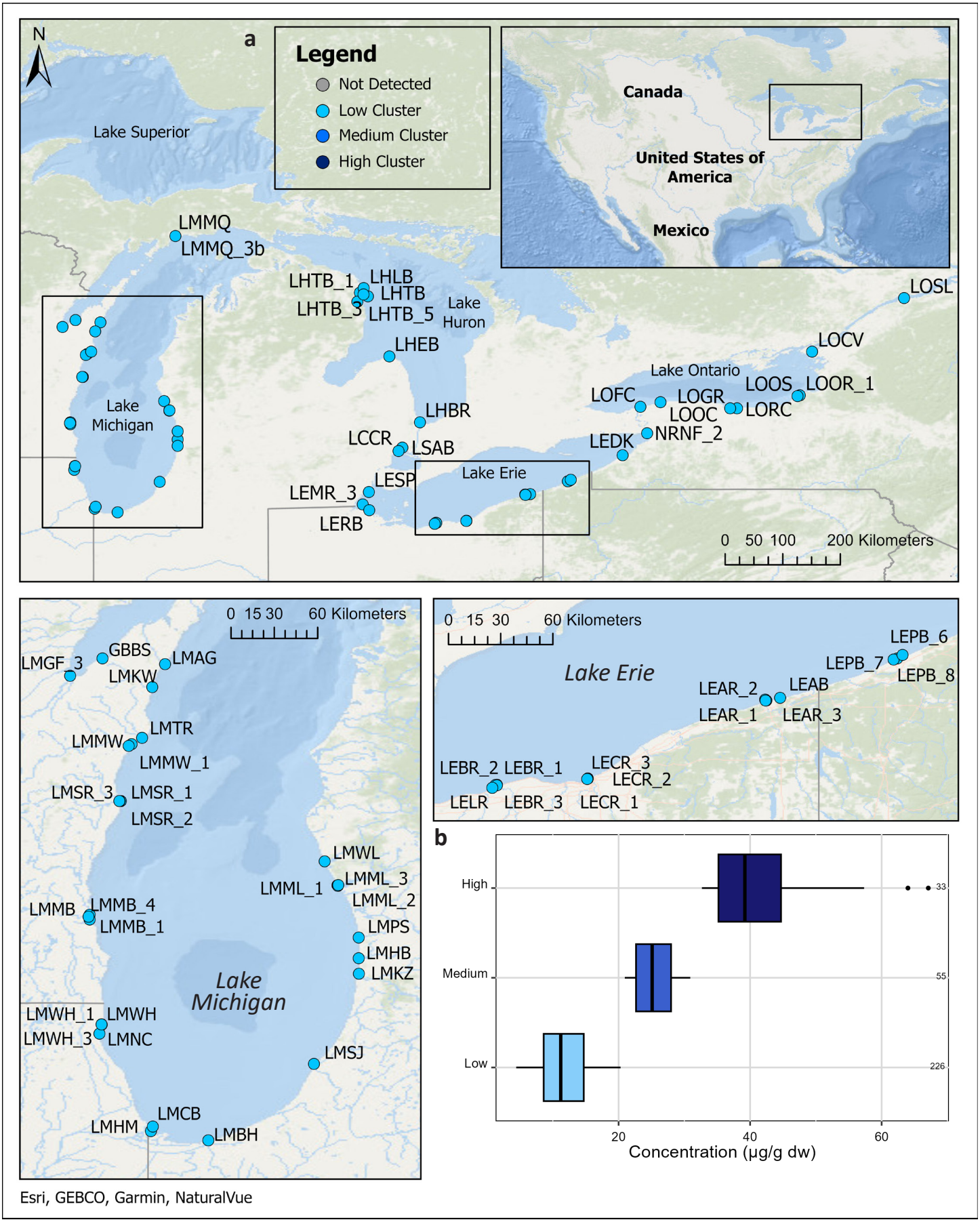


Figure 62. (a) Nickel concentrations in dreissenid mussel tissue in 2022 compared to the (b) historic regional MWP Dreissena *species nickel concentrations (µg/g dw). Boxplot colors correspond to cluster groups delineated in the map legend. 81 regional sites sampled between 1992 - 2022 for a total of 314 dreissenid mussel samples.*

14.0 RESULTS - SELENIUM (Se)

14.1 Selenium Chemical Description

Selenium naturally occurs in the Earth's crust and is commonly found in rocks and soil (ATSDR, 2003). Weathering of rocks and soil can result in low levels of selenium in water and in the air as dust-like particles. Additionally, volcanic eruptions may release atmospheric selenium. Selenium is also commercially produced as a byproduct of copper refining or from burning coal or oil. Other products that contain selenium compounds include photographic devices, gun bluing, plastics, paints, anti-dandruff shampoos, vitamin and mineral supplements, fungicides, glass, drug preparation, and livestock feed supplements (ATSDR, 2003). Regardless of source, selenium is not often found in its elemental form in the environment, but rather combined with sulfide, silver, copper, lead, or nickel minerals (ATSDR, 2003).

Selenium is an essential nutrient in low concentrations and is usually ingested by humans through drinking water, food, and air (ATSDR, 2003). Selenium is often contained in soils and varies by region, so depending on where food is grown, humans may be exposed to differing levels of selenium. Humans are not typically exposed to high selenium levels but this can vary with occupation, specifically for individuals employed in the metal industries, selenium-recovery processes, paint manufacturing, and special trades. When exposed at high levels, dizziness, fatigue, and irritation of mucous membranes can occur, and even mild exposure over long time periods can result in brittle hair, deformed nails, and even loss of feeling and control in extremities (called selenosis). Conditions this extreme usually occur in areas with locally high levels of selenium, where the population consumes locally produced food (ATSDR, 2003). For most people in the U.S. who eat food from many different areas, overexposure to selenium is unlikely to occur. There is no support that selenium causes cancer, except for one specific form called selenium sulfide, which has been shown to cause cancer in animals. FDA regulations allow a level of 50 ppb of selenium in bottled water and OSHA set the exposure limit for selenium compounds in the air for an 8-hour period at 0.2 mg selenium/m³ (ATSDR, 2003).

The forms and fate of selenium in soil depend primarily on soil pH and its interaction with oxygen (ATSDR, 2003). In the absence of oxygen when the soil is acidic, the amount of selenium that can enter plants and organisms should be low. Elemental selenium, which cannot dissolve in water, and other insoluble forms of selenium are less mobile and will usually remain in the soil, posing a smaller risk of exposure. Selenium compounds that can dissolve in water are sometimes highly mobile; thus, there is an increased risk of exposure. Some evidence indicates that selenium can be taken up by and bioaccumulate in tissues of aquatic organisms. Selenium concentrations in aquatic organisms have been a concern due to irrigation runoff in some areas of the U.S. (ATSDR, 2003).

14.2 Magnitude and Distribution of Selenium in Dreissenid Mussel Tissue in 2022

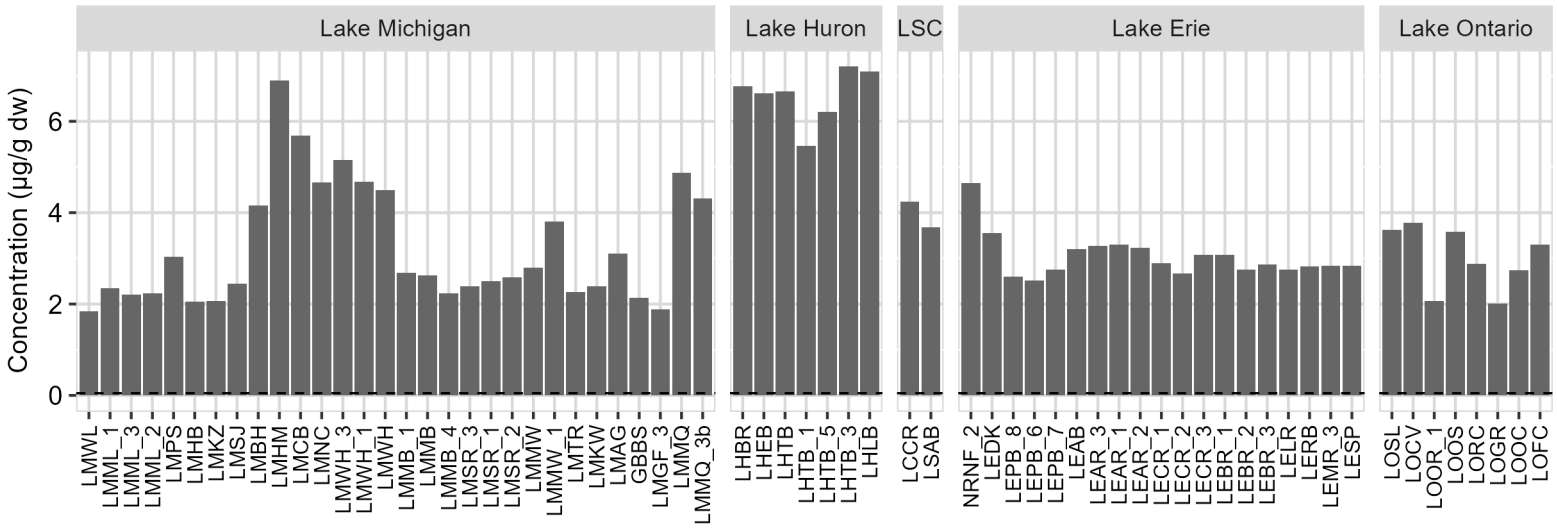


Figure 63. Bar graph showing magnitude of selenium concentrations (µg/g dw) detected in dreissenid mussel tissue in the Great Lakes Basin in 2022. Dotted line represents the minimum weight corrected MDL. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair.

14.3 Historical Context of Selenium Magnitude and Distribution in Dreissenid Mussel Tissue

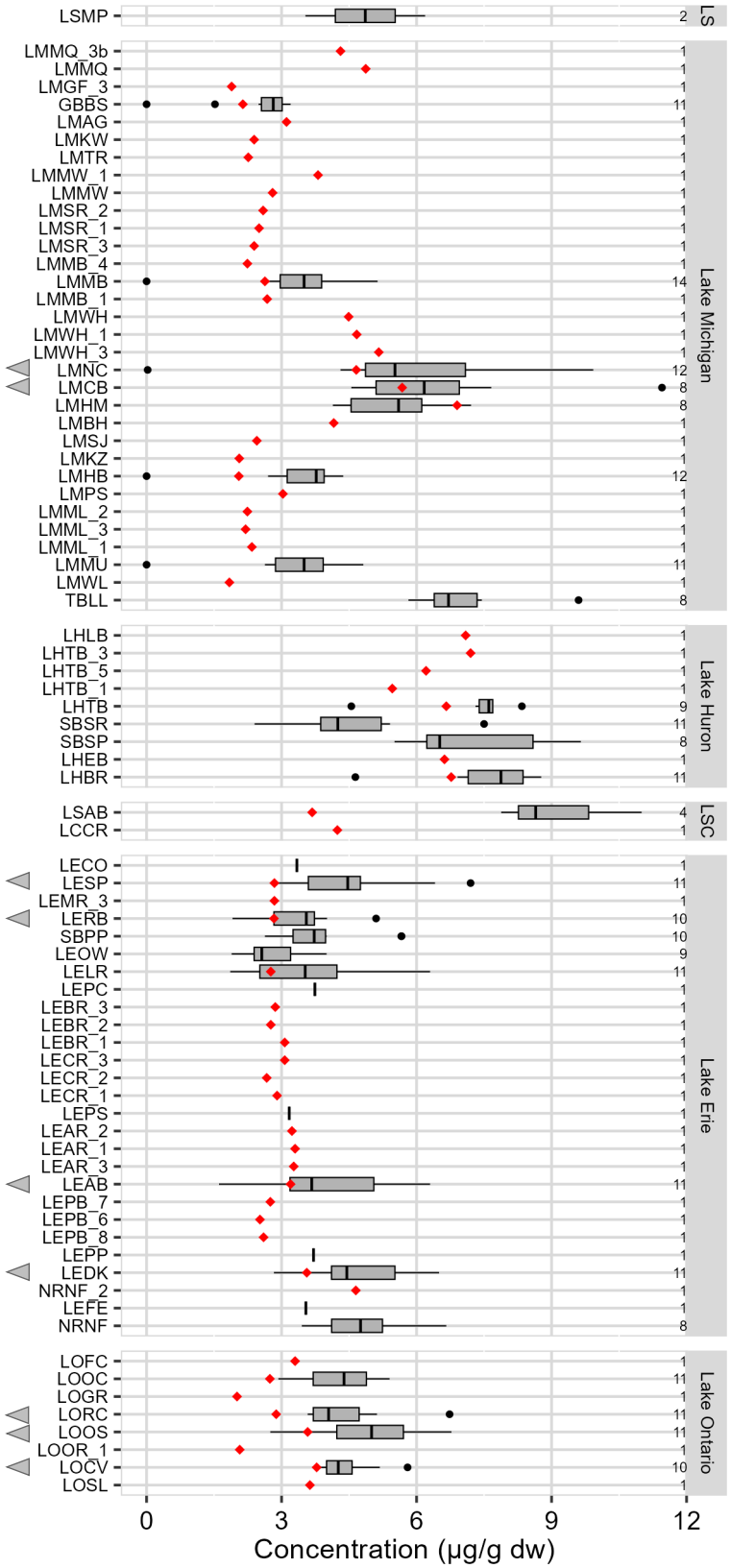


Figure 64. Selenium concentrations (µg/g dw) in dreissenid mussel tissue in 2022 (red diamonds) compared to boxplots of the historic selenium concentrations for each site (1992 - 2008) and results of the trend analysis showing significant increasing (▷) or decreasing (◁) site-specific contamination trends over time. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair; LS = Lake Superior. The number of years each site has been sampled since 1992 is noted at the right of the plot for each site.

Results - Selenium (Se)

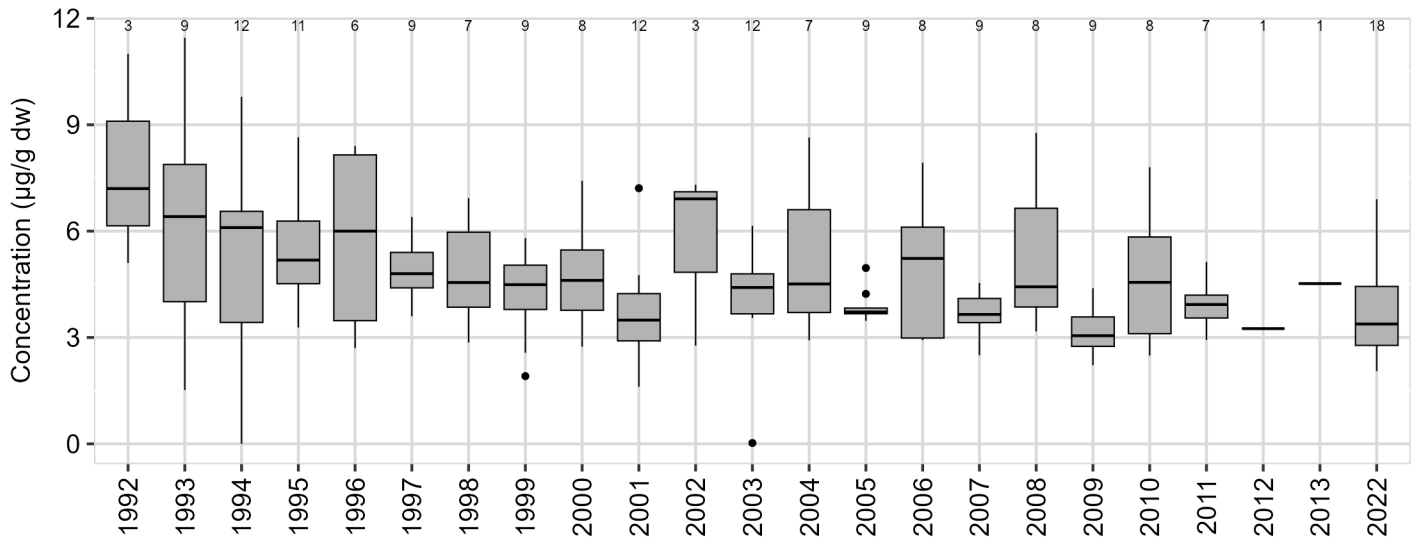


Figure 65. Boxplots representing the historic selenium concentrations (µg/g dw) in dreissenid mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot.

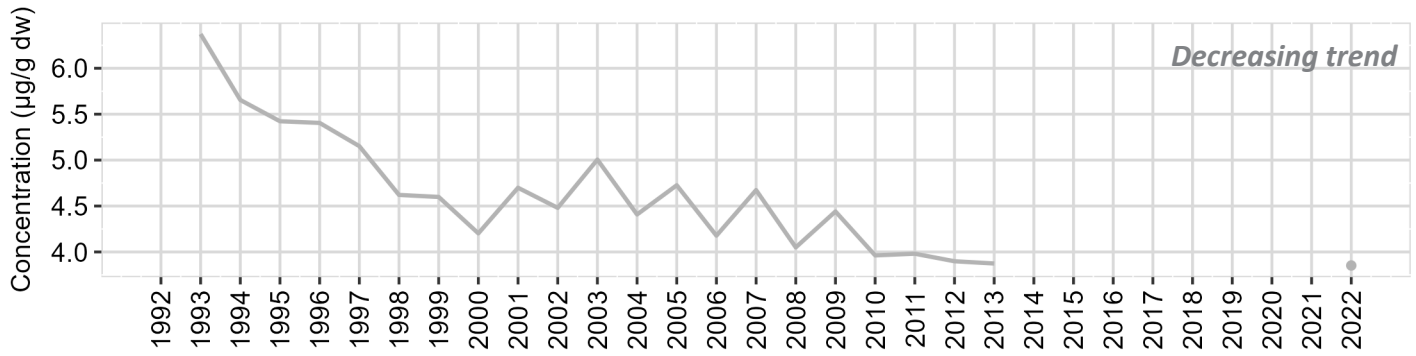


Figure 66. Three-point moving average of the yearly mean selenium concentrations (µg/g dw) in dreissenid mussel tissue in the sites analyzed in this study. Average concentration for 2022 is included as a standalone point for reference.

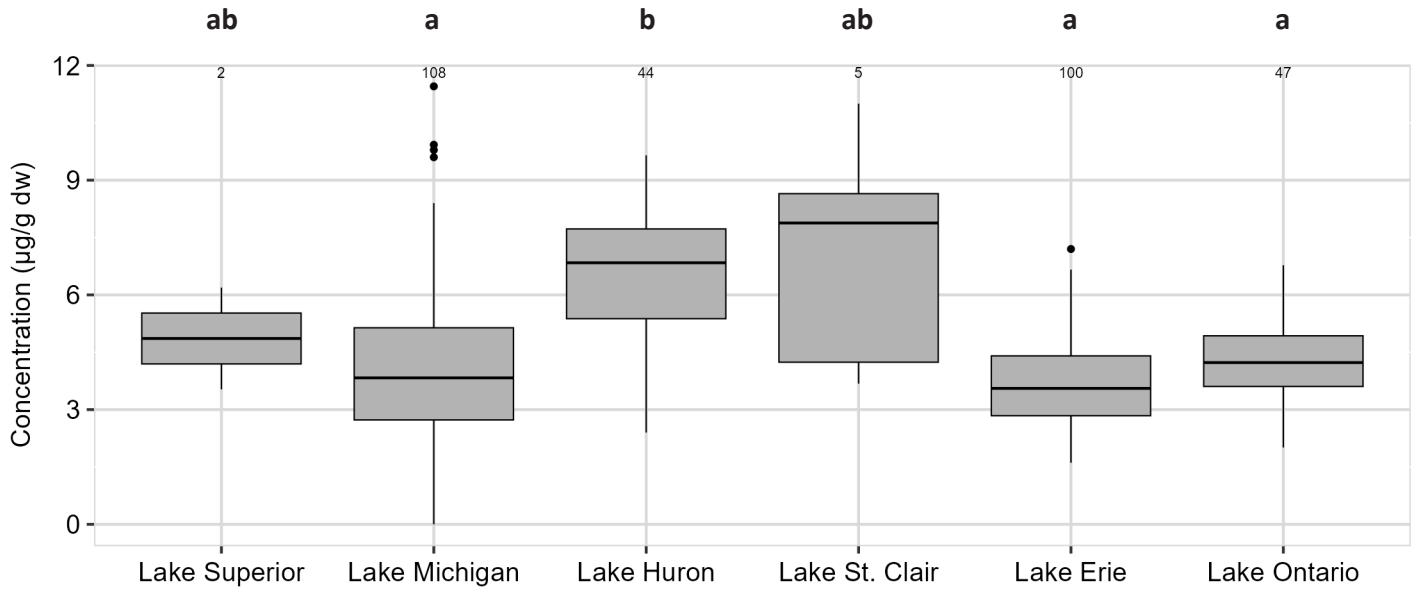


Figure 67. Selenium concentrations (µg/g dw) measured in dreissenid mussel tissue in the Great Lakes from 1992 - 2022. Lakes and major rivers including connecting channels are listed geographically from west to east across the Great Lakes Basin. The number of samples collected in each lake is noted at the top of the plot. Letters above each lake indicate significant differences (at α = 0.05).

Results - Selenium (Se)

14.4 Selenium Summary

2022 Dreissenid Mussel Tissue Results:

- Se was detected at 100% of 66 sites surveyed (Figure 63).
- Se concentration descriptive statistics (Figure 63):
  - Range: 1.84 – 7.20 µg/g dw
  - Minimum Se concentration was detected at LMWL
  - Maximum Se concentration was detected at LHTB_3
  - Median: 2.97 µg/g dw
  - Mean ± SD: 3.52 ± 1.44 µg/g dw
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster respective to 2022 data only:
  - 50% of sites in low cluster (1.84 - 2.90 µg/g dw)
  - 36% of sites in medium cluster (3.03 - 5.16 µg/g dw)
  - 14% of sites in high cluster (5.46 - 7.20 µg/g dw)

Historic Context for Dreissenid Mussel Tissue (18 out of 66 sites had historic data to analyze for trend analyses):

- 6% of sites surveyed (1/18) were above their historic median concentrations in 2022 (Figure 64).
- Regionally, there was a significant decreasing temporal trend of Se concentrations in dreissenid mussels in the Great Lakes Basin ($p < 0.001$, rho = -0.66) (Figure 65; Figure 66).
- 9 sites (LMNC, LMCB, LESP, LERB, LEAB, LEDK, LORC, LOOS, LOCV) in the Great Lakes Basin showed significant decreasing temporal trends of Se concentrations at $\alpha = 0.05$ (Figure 64; Table A4).
- 0 sites in the Great Lakes Basin showed significant increasing temporal trends of Se concentrations at $\alpha = 0.05$ (Figure 64; Table A4).
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster compared to historic data (Figure 68):
  - 56% of sites in low cluster (0.00 - 3.11 µg/g dw)
  - 33% of sites in medium cluster (3.17 - 5.95 µg/g dw)
  - 11% of sites in high cluster (6.08 - 11.45 µg/g dw)

General Observations:

- Se concentrations were ubiquitous and were detected across a large range of concentrations in the Great Lakes Basin in dreissenid mussel tissue in 2022, likely reflecting naturally higher ambient concentrations with additional localized inputs (Figure 68).
  - Some instances of comparatively high concentrations were found in Lake Michigan (LMHM) and Lake Huron (LHBR, LHEB, LHLB, LHTB, LHTB_3, LHTB_5), possibly indicating local inputs of Se in those areas (Figure 68).
- Regionally, Se concentrations have significantly decreased over time in dreissenid mussel tissue, indicating decreasing inputs of Se into the environment (Figure 66).
- The post-hoc analysis evaluating differences between lakes indicates that there are no statistical bridges, with all lakes demonstrating a gradient of relationships, being significantly similar to some and significantly different from others. Generally, higher concentrations of Se were found in Lake Huron, while lower concentrations of Se were found in lakes Michigan, Erie, and Ontario (Figure 67).

Results - Selenium (Se)

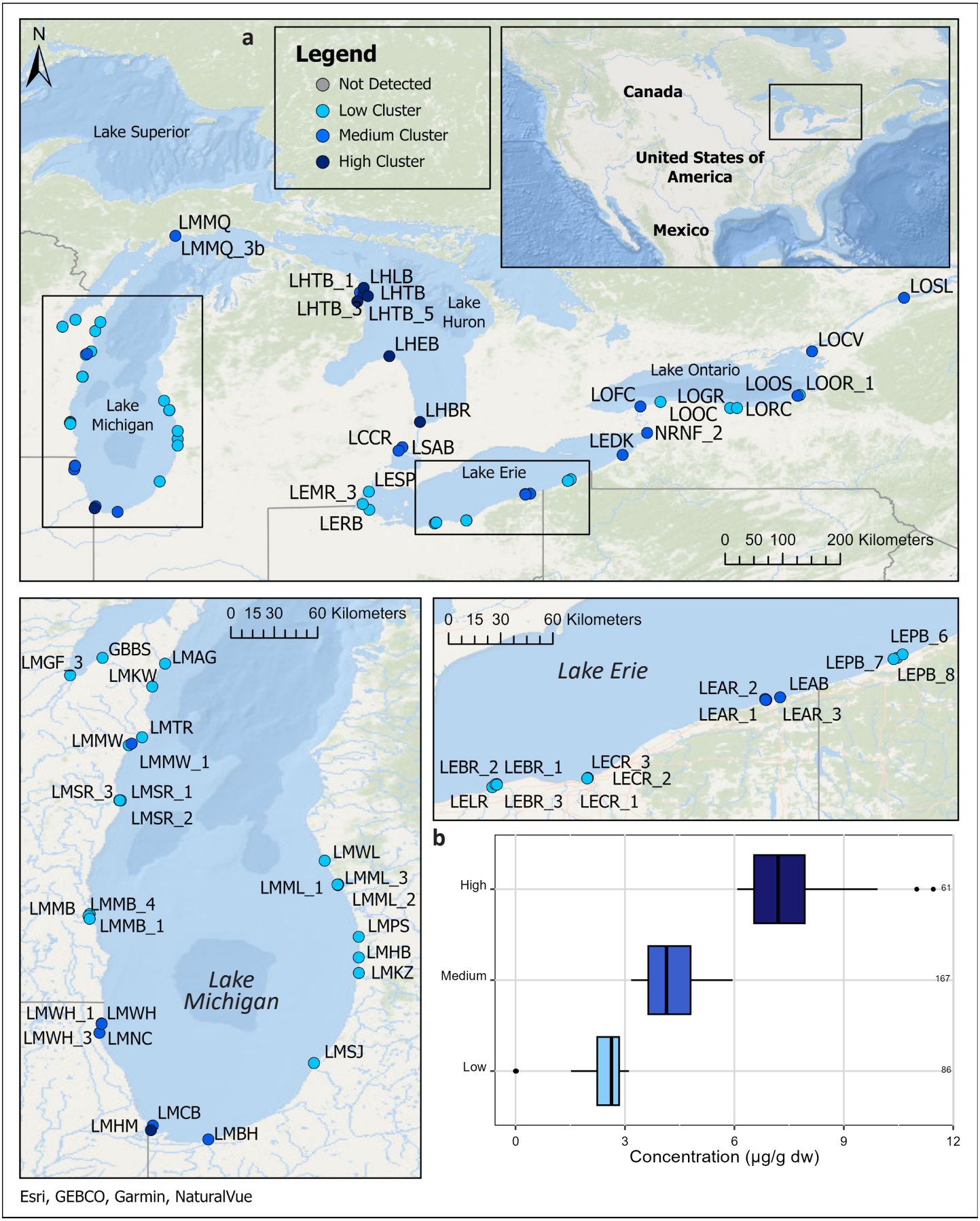


Figure 68. (a) Selenium concentrations in dreissenid mussel tissue in 2022 compared to the (b) historic regional MWP Dreissena *species selenium concentrations (µg/g dw). Boxplot colors correspond to cluster groups delineated in the map legend. 81 regional sites sampled between 1992 - 2022 for a total of 314 dreissenid mussel samples.*

15.0 RESULTS - SILVER (Ag)

15.1 Silver Chemical Description

Silver is a naturally occurring element but elevated levels of contamination are typically from anthropogenic sources (ATSDR, 1990). The natural weathering of silver-bearing rocks and soil by wind and rain releases large amounts of silver into the environment. Anthropogenically, products that contain silver are photographic technology, dental applications, electronics, silverware, jewelry, and solders. Photographic materials are the major anthropogenic source of silver that is released into the environment (ATSDR, 1990). Silver has been regulated in the U.S. since the 1960s, but the initial criteria were conservative because the realized risk of silver was not known at the time. The U.S. EPA downgraded silver from a primary to a secondary maximum contaminant level in the 1990s due to its low realized risk from exposure (Purcell, 2009).

Silver is not an essential nutrient to humans and is mostly harmless at low levels and infrequent exposure (ATSDR, 1990). Humans are generally exposed to silver through food and drinking water, and to a lesser extent, through the air. Silver can cause some areas of the skin and other body tissues to turn gray or gray-blue, a condition known as “argyria”. This condition can occur in humans who ingest or inhale silver compounds over a long period. Long or high exposure to silver compounds such as silver nitrate or silver oxide may cause breathing problems, lung and throat irritation, and stomach pain. These conditions typically manifest with occupational exposure (ATSDR, 1990). Skin contact with silver compounds can cause mild allergic reactions, such as rash, swelling, and inflammation in some people. Animal tests have shown that exposure to silver is unlikely to be life threatening unless very high amounts are swallowed, which is unlikely. Conversely, silver does have beneficial medicinal uses including as eye drops to prevent blindness, in salves for burn victims, and to kill bacteria in water (ATSDR, 1990).

Silver released into the environment may be carried long distances through air and water (ATSDR, 1990). Rain washes silver compounds out of many soils, eventually moving them into groundwater. Silver is stable and remains in the environment in one form or another until it is taken out again by people. Silver does not break down, but it can change its form by combining with other substances, and the form it is found in depends on environmental conditions (ATSDR, 1990).

15.2 Magnitude and Distribution of Silver in Dreissenid Mussel Tissue in 2022

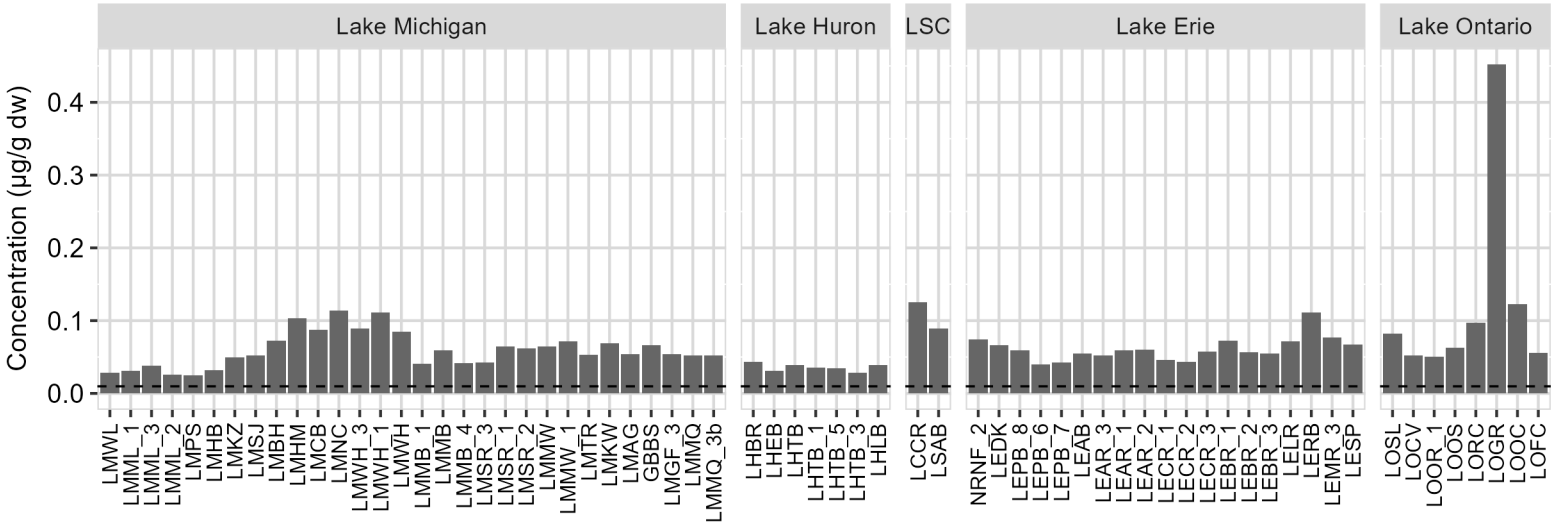


Figure 69. Bar graph showing magnitude of silver concentrations (µg/g dw) detected in dreissenid mussel tissue in the Great Lakes Basin in 2022. Dotted line represents the minimum weight corrected MDL. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair.

15.3 Historical Context of Silver Magnitude and Distribution in Dreissenid Mussel Tissue

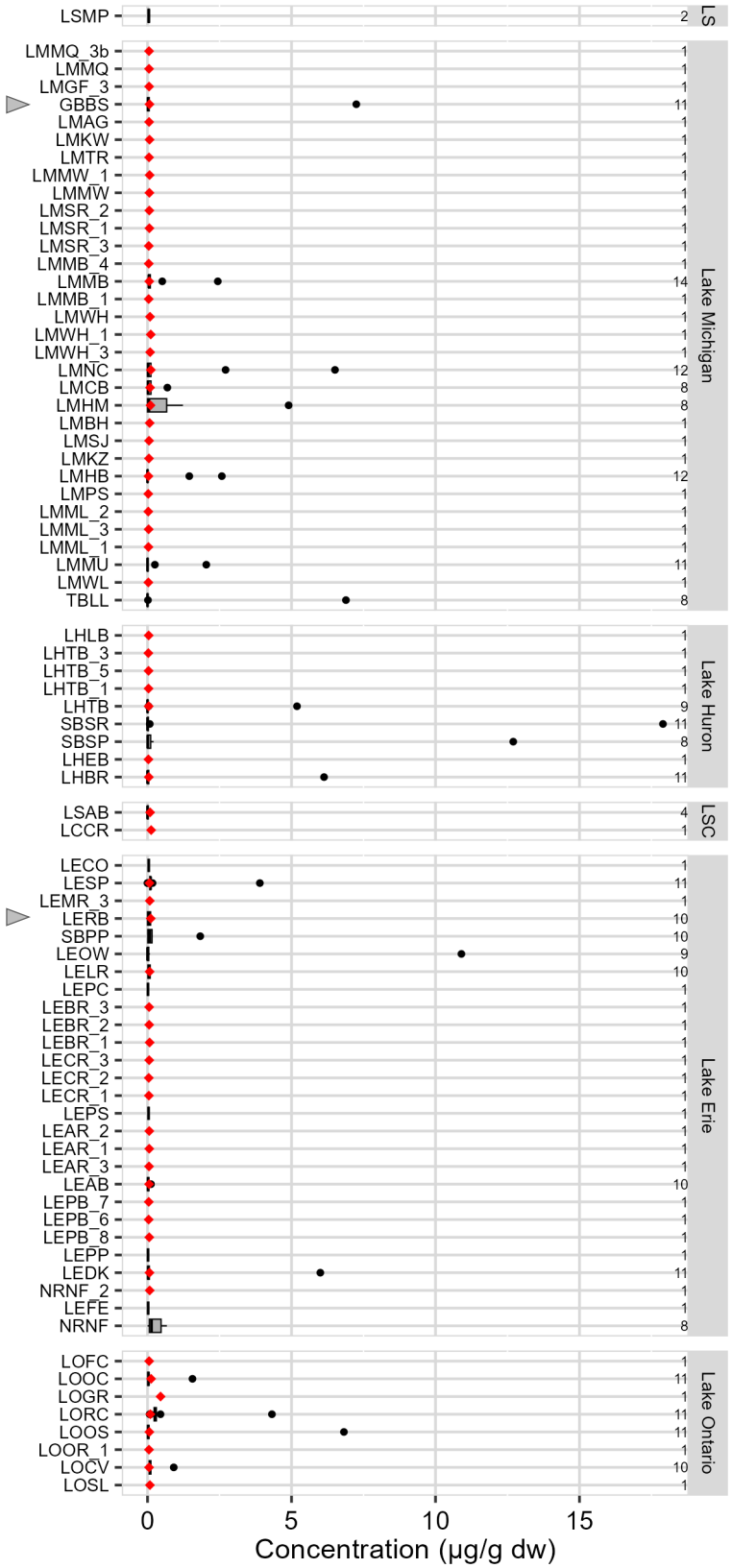


Figure 70. Silver concentrations (µg/g dw) in dreissenid mussel tissue in 2022 (red diamonds) compared to boxplots of the historic silver concentrations for each site (1992 - 2008) and results of the trend analysis showing significant increasing (▻) or decreasing (◅) site-specific contamination trends over time. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair; LS = Lake Superior. The number of years each site has been sampled since 1992 is noted at the right of the plot for each site. Sites LEAB and LELR in 2001 (81.8 µg/g dw, 69.3 µg/g dw, respectively) were removed to aid in visualization.

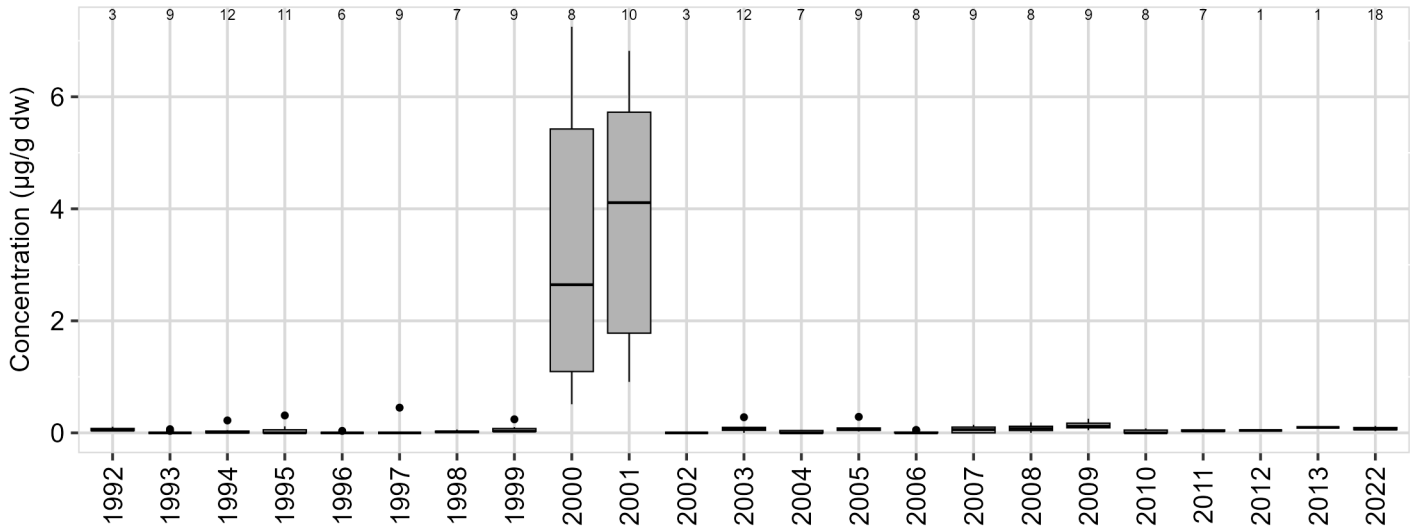


Figure 71. Boxplots representing the historic silver concentrations (µg/g dw) in dreissenid mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot. Sites LEAB and LELR in 2001 (81.8 µg/g dw, 69.3 µg/g dw, respectively) were removed to aid in visualization.

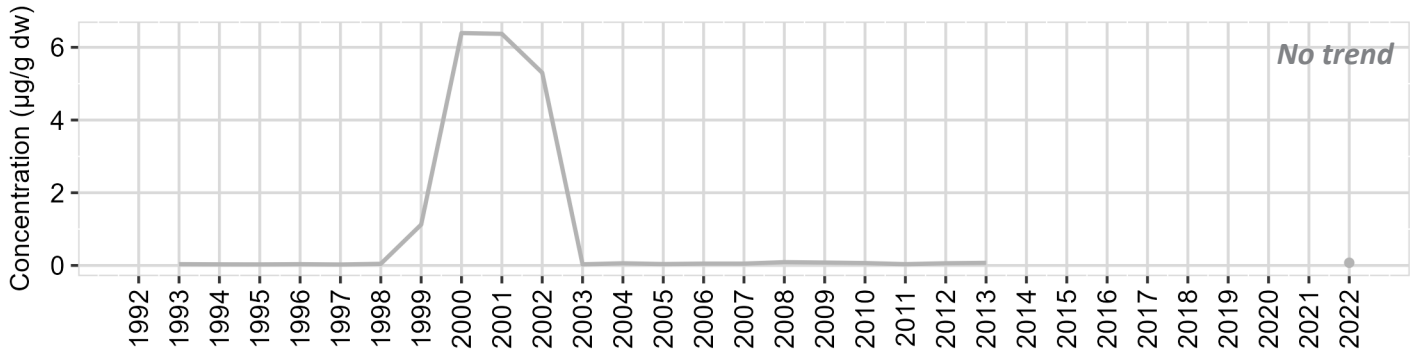


Figure 72. Three-point moving average of the yearly mean silver concentrations (µg/g dw) in dreissenid mussel tissue in the sites analyzed in this study. Average concentration for 2022 is included as a standalone point for reference.

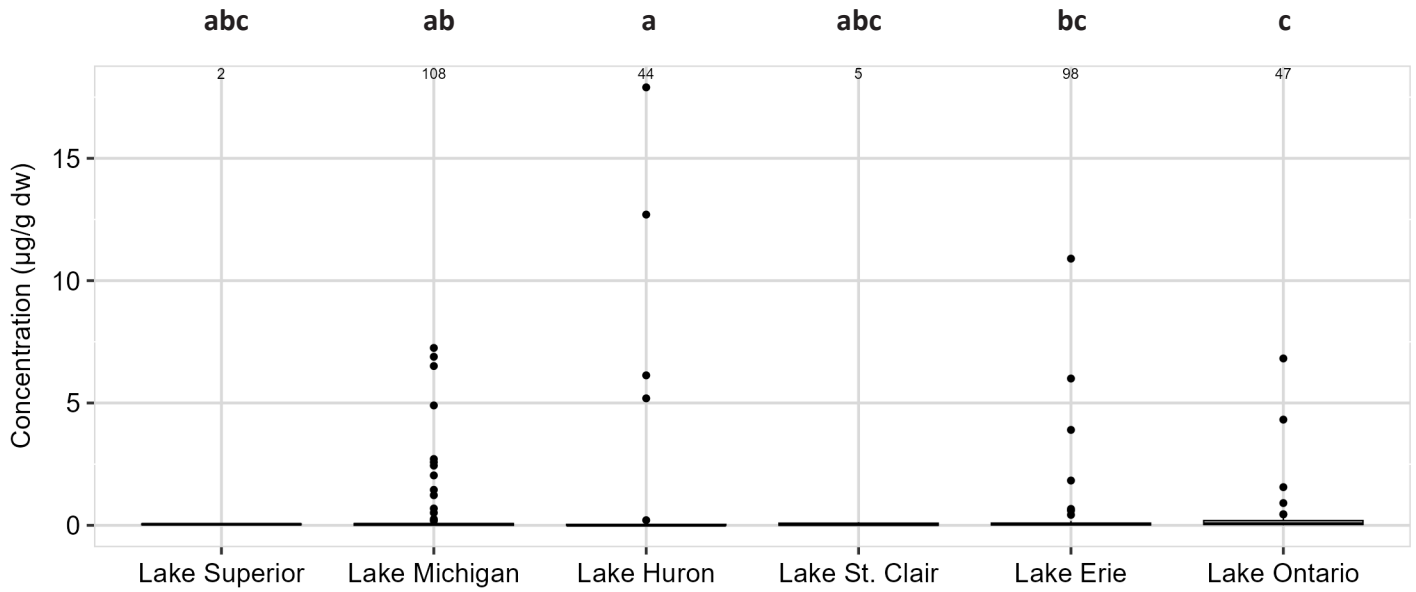


Figure 73. Silver concentrations (µg/g dw) measured in dreissenid mussel tissue in the Great Lakes from 1992 - 2022. Lakes and major rivers including connecting channels are listed geographically from west to east across the Great Lakes Basin. The number of samples collected in each lake is noted at the top of the plot. Letters above each lake indicate significant differences (at α = 0.05). Sites LEAB and LELR in 2001 (81.8 µg/g dw, 69.3 µg/g dw, respectively) were removed to aid in visualization.

Results - Silver (Ag)

15.4 Silver Summary

2022 Dreissenid Mussel Tissue Results:

- Ag was detected at 100% of 66 sites surveyed (Figure 69).
- Ag concentration descriptive statistics (Figure 69):
  - Range: 0.03 – 0.45 µg/g dw
  - Minimum Ag concentration was detected at LMPS
  - Maximum Ag concentration was detected at LOGR
  - Median: 0.06 µg/g dw
  - Mean ± SD: 0.07 ± 0.05 µg/g dw
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster respective to 2022 data only:
  - 80% of sites in low cluster (0.03 - 0.08 µg/g dw)
  - 18% of sites in medium cluster (0.08 - 0.12 µg/g dw)
  - 2% of sites in high cluster (0.45 - 0.45 µg/g dw)

Historic Context for Dreissenid Mussel Tissue (18 out of 66 sites had historic data to analyze for trend analyses):

- 83% of sites surveyed (15/18) were above their historic median concentrations in 2022 (Figure 70).
- Regionally, there was no significant temporal trend of Ag concentrations in dreissenid mussels in the Great Lakes Basin ($p = 0.21$, $rho = 0.27$) (Figure 71; Figure 72).
- 0 sites in the Great Lakes Basin showed significant decreasing temporal trends of Ag concentrations at $\alpha = 0.05$ (Figure 70; Table A4).
- 2 sites (GBBS, LERB) in the Great Lakes Basin showed significant increasing temporal trends of Ag concentrations at $\alpha = 0.05$ (Figure 70; Table A4).
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster compared to historic data (Figure 74):
  - 100% of sites in low cluster (0.00 - 2.71 µg/g dw)
  - 0% of sites in medium cluster (3.90 - 7.25 µg/g dw)
  - 0% of sites in high cluster (10.50 - 81.80 µg/g dw)

General Observations:

- Ag concentrations were ubiquitous but at comparatively low concentrations in the Great Lakes Basin in dreissenid mussel tissue in 2022, likely reflecting natural ambient concentrations (Figure 74).
- Regionally, Ag concentrations have not changed significantly over time in dreissenid mussel tissue, indicating no obvious anthropogenic inputs of Ag into the environment (Figure 72).
- The post-hoc analysis evaluating differences between lakes indicates that lakes Superior and St. Clair are statistical bridges, showing no significant difference to any other lake. In contrast, the other lakes demonstrate a gradient of relationships, being significantly similar to some and significantly different from others. Generally, the concentrations of Ag are relatively low with some high outliers found in all lakes, except Lake St. Clair (Figure 73).

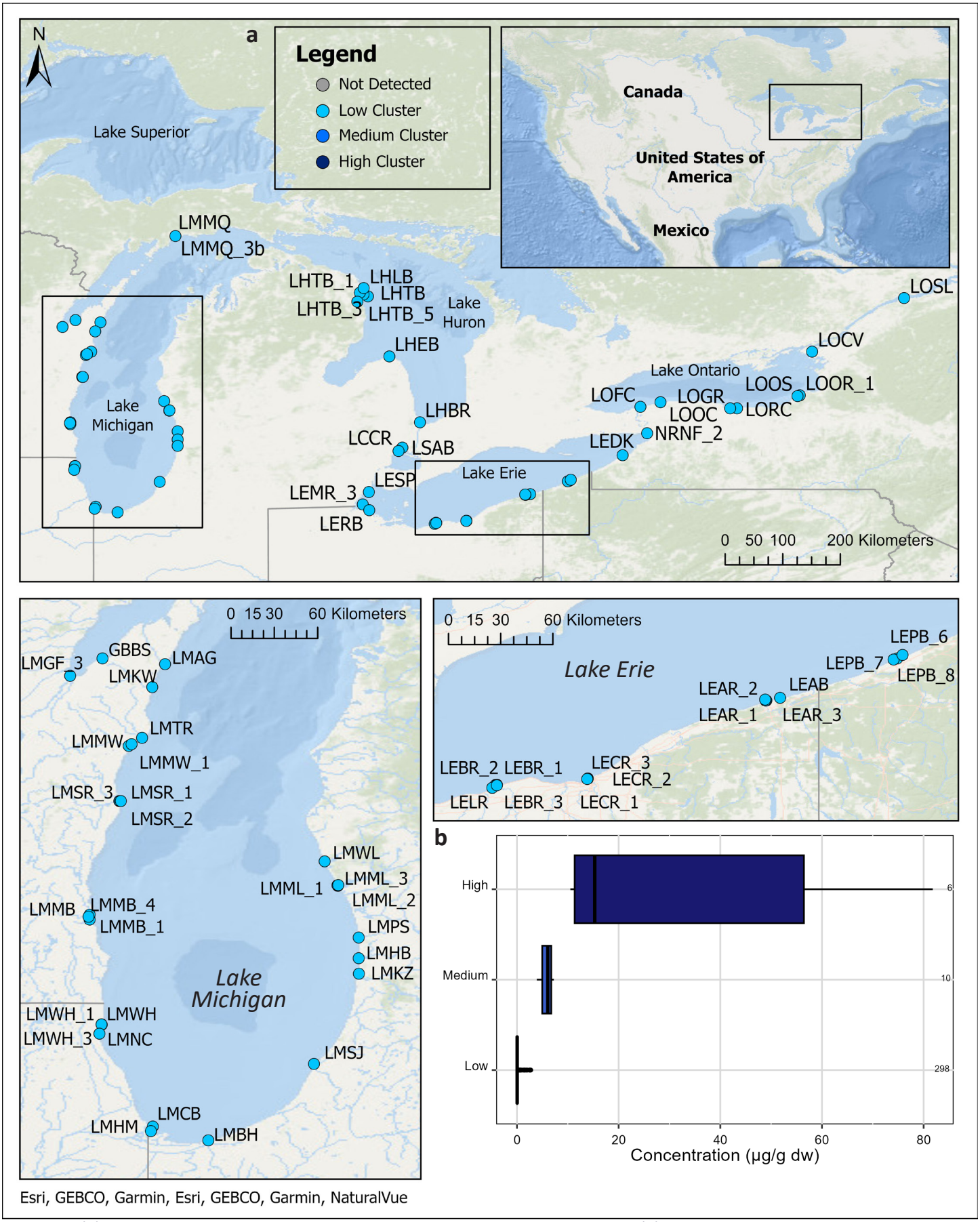


Figure 74. (a) Silver concentrations in dreissenid mussel tissue in 2022 compared to the (b) historic regional MWP Dreissena *species silver concentrations (µg/g dw). Boxplot colors correspond to cluster groups delineated in the map legend.81 regional sites sampled between 1992 - 2022 for a total of 314 dreissenid mussel samples.*

16.0 RESULTS - TIN (Sn)

16.1 Tin Chemical Description

Tin may be released to the environment from both natural and anthropogenic sources. Tin occurs in water in trace amounts naturally and can combine with other chemicals to form various inorganic and organic compounds. Sources of tin in coastal water and soil include manufacturing and processing facilities and mining. Tin has not been mined in the U.S. since 1993 (USGS, 2008); however, Canadian tin mining occurs in the Great Lakes region. Tin metal is mainly used as liner in cans for food and aerosols and is also used in paint, plastic, pesticide brass, bronze, and some soldering products (ATSDR, 2005b). Concentrations in unpolluted waters and the atmosphere are often near analytical detection limits.

Humans are exposed to elevated levels of tin by eating from tin-lined cans and by consuming contaminated seafood (ATSDR, 2005b). Exposure to elevated levels of tin compounds by humans leads to liver damage, kidney damage, and cancer. There is no U.S. FDA recommended guideline for tin in seafood.

Tin enters coastal waters bound to particulates and from riverine sources derived from soil and sediment erosion. Bioconcentration factors for inorganic tin were reported to be 1,900 and 3,000 for marine algae and fish (Seidel et al., 1980; Thompson et al., 1972). Inorganic tin can be transformed into organometallic forms by microbial methylation and is correlated with increasing organic content in sediment (Hadjispyrou et al., 1998). Tin is considered relatively immobile in the environment and is rarely detected in the atmosphere. It is mainly found in the atmosphere near industrial sources as particulates from combustion of fossil fuels and solid waste (Gerritse et al., 1982; WHO, 1980).

16.2 Magnitude and Distribution of Tin in Dreissenid Mussel Tissue in 2022

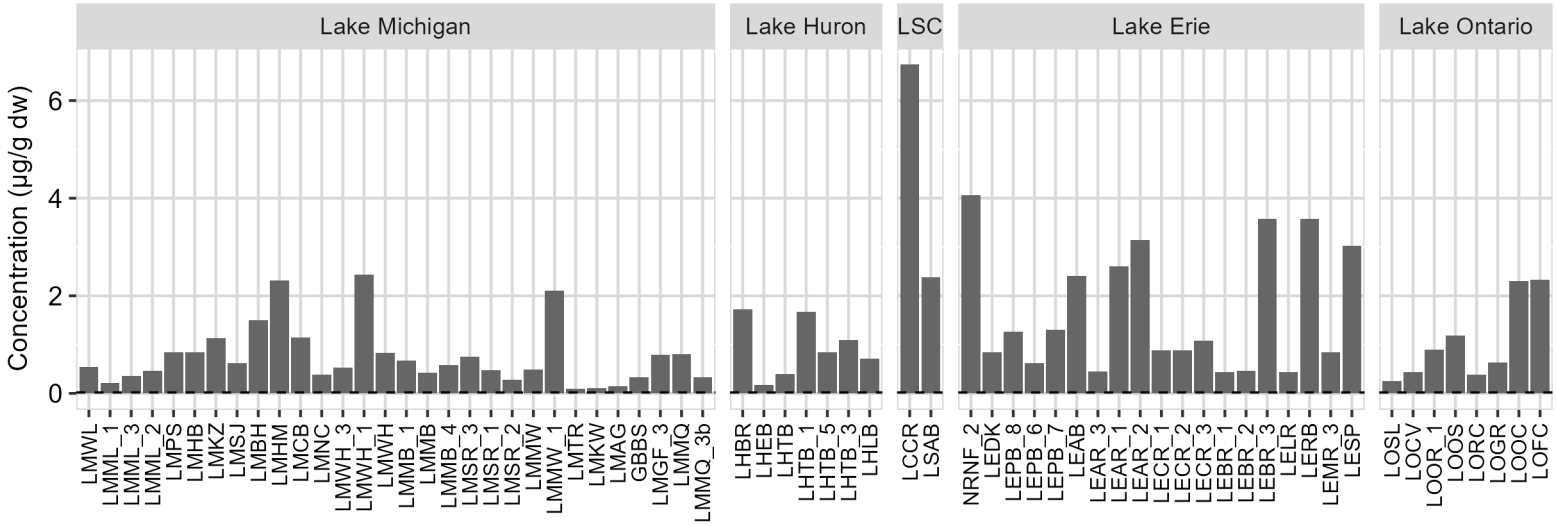


Figure 75. Bar graph showing magnitude of tin concentrations (µg/g dw) detected in dreissenid mussel tissue in the Great Lakes Basin in 2022. Dotted line represents the minimum weight corrected MDL. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair.

16.3 Historical Context of Tin Magnitude and Distribution in Dreissenid Mussel Tissue

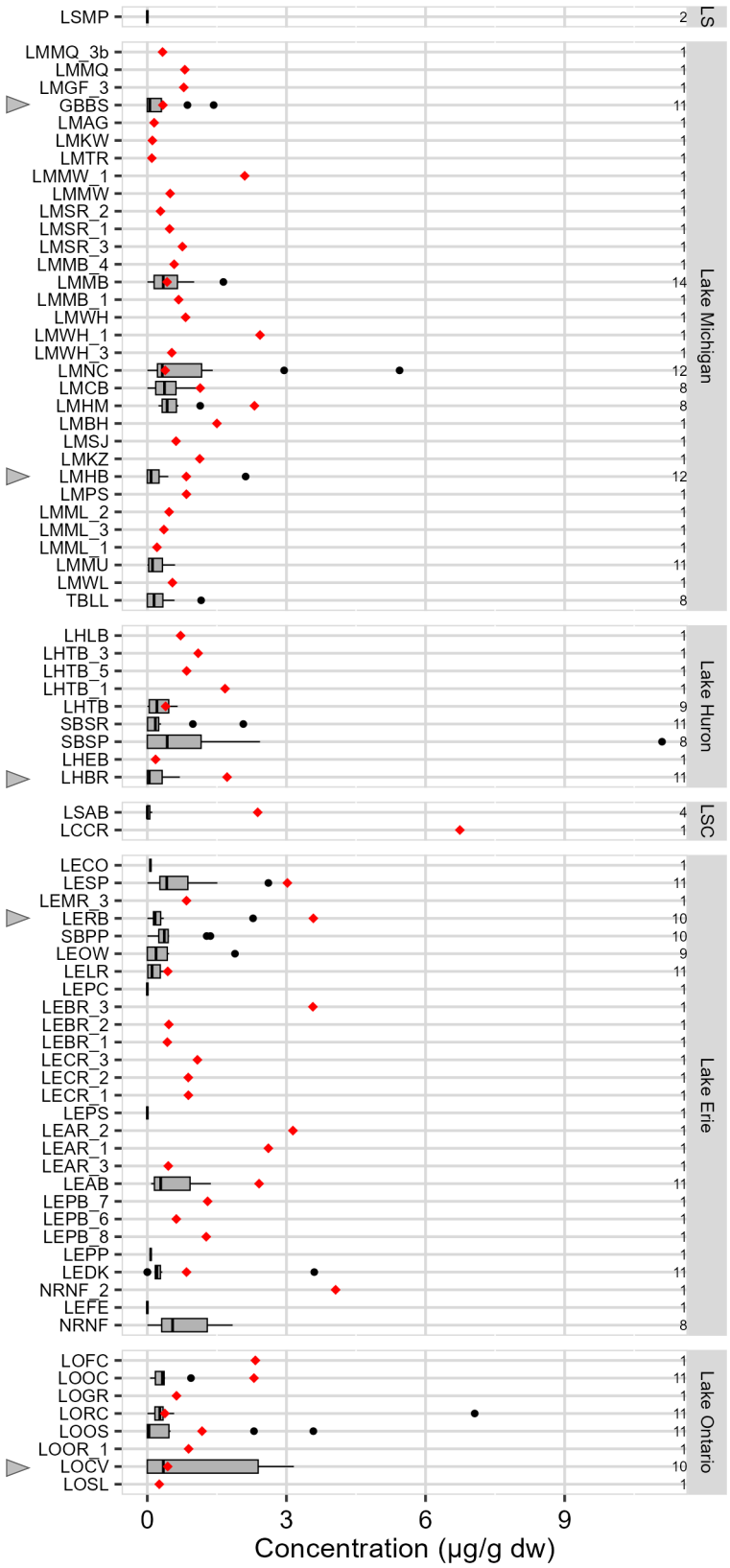


Figure 76. Tin concentrations (µg/g dw) in dreissenid mussel tissue in 2022 (red diamonds) compared to boxplots of the historic tin concentrations for each site (1992 - 2008) and results of the trend analysis showing significant increasing (▻) or decreasing (◅) site-specific contamination trends over time. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair; LS = Lake Superior. The number of years each site has been sampled since 1992 is noted at the right of the plot for each site.

Results - Tin (Sn)

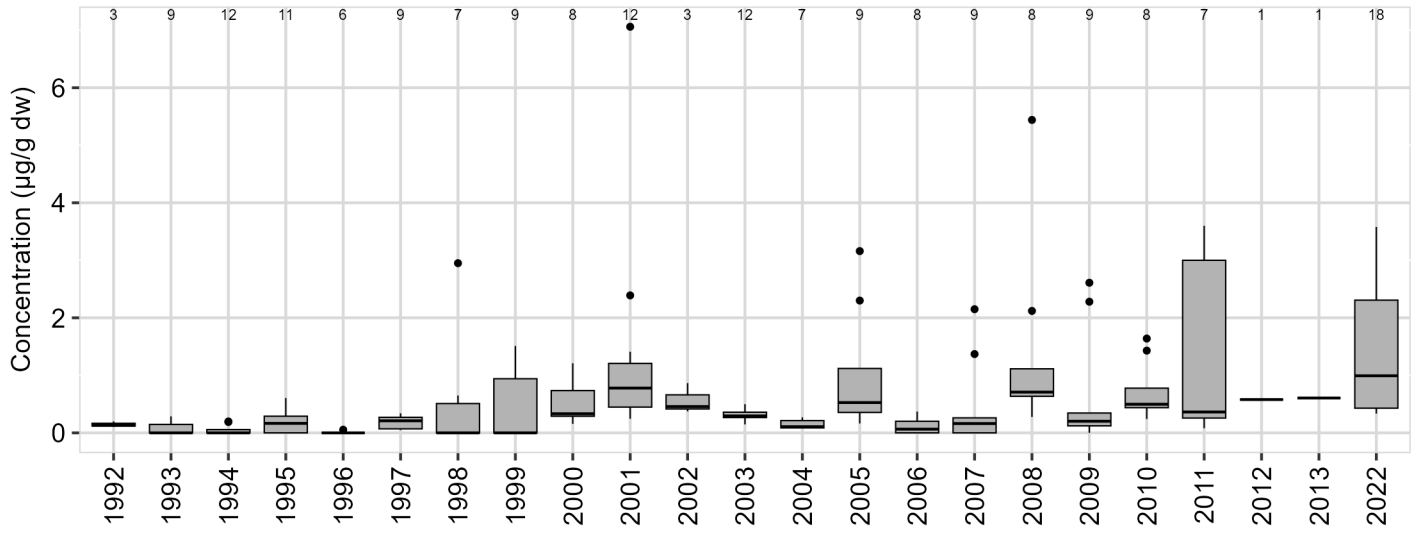


Figure 77. Boxplots representing the historic tin concentrations (µg/g dw) in dreissenid mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot.

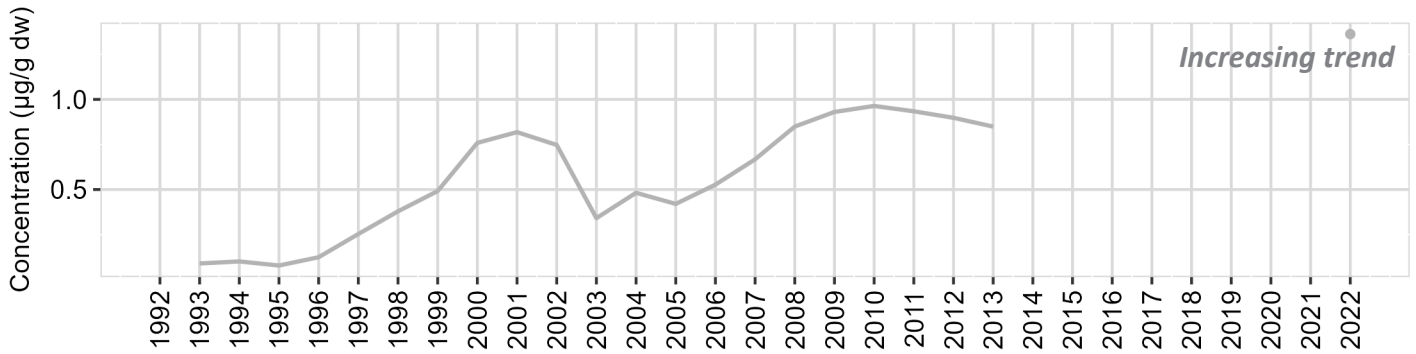


Figure 78. Three-point moving average of the yearly mean tin concentrations (µg/g dw) in dreissenid mussel tissue in the sites analyzed in this study. Average concentration for 2022 is included as a standalone point for reference.

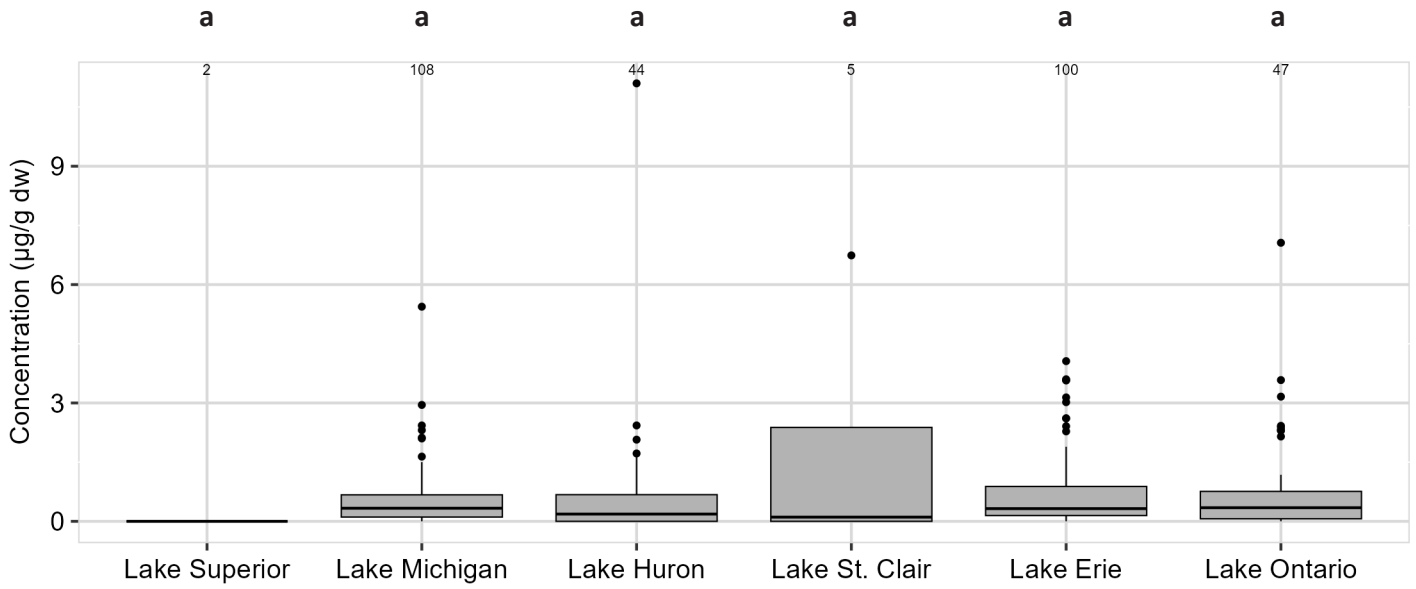


Figure 79. Tin concentrations (µg/g dw) measured in dreissenid mussel tissue in the Great Lakes from 1992 - 2022. Lakes and major rivers including connecting channels are listed geographically from west to east across the Great Lakes Basin. The number of samples collected in each lake is noted at the top of the plot. Letters above each lake indicate significant differences (at α = 0.05).

Results - Tin (Sn)

16.4 Tin Summary

2022 Dreissenid Mussel Tissue Results:

- Sn was detected at 100% of 66 sites surveyed (Figure 75).
- Sn concentration descriptive statistics (Figure 75):
  - Range: 0.10 – 6.74 µg/g dw
  - Minimum Sn concentration was detected at LMTR
  - Maximum Sn concentration was detected at LCCR
  - Median: 0.82 µg/g dw
  - Mean ± SD: 1.19 ± 1.17 µg/g dw
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster respective to 2022 data only:
  - 79% of sites in low cluster (0.10 - 1.72 µg/g dw)
  - 20% of sites in medium cluster (2.10 - 4.06 µg/g dw)
  - 1% of sites in high cluster (6.74 - 6.74 µg/g dw)

Historic Context for Dreissenid Mussel Tissue (18 out of 66 sites had historic data to analyze for trend analyses):

- 100% of sites surveyed (18/18) were above their historic median concentrations in 2022 (Figure 76).
- Regionally, there was a significant increasing temporal trend of Sn concentrations in dreissenid mussels in the Great Lakes Basin ($p < 0.001$, rho = 0.71) (Figure 77; Figure 78).
- 0 sites in the Great Lakes Basin showed significant decreasing temporal trends of Sn concentrations at $\alpha = 0.05$ (Figure 76; Table A4).
- 5 sites (GBBS, LMHB, LHBR, LERB, LOCV) in the Great Lakes Basin showed significant increasing temporal trends of Sn concentrations at $\alpha = 0.05$ (Figure 76; Table A4).
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster compared to historic data (Figure 80):
  - 33% of sites in low cluster (0.00 - 0.50 µg/g dw)
  - 46% of sites in medium cluster (0.52 - 1.89 µg/g dw)
  - 21% of sites in high cluster (2.07 - 11.10 µg/g dw)

General Observations:

- Sn concentrations were ubiquitous and were detected across a large range of concentrations in the Great Lakes Basin in dreissenid mussel tissue in 2022, likely reflecting naturally higher ambient concentrations with additional localized inputs (Figure 80).
  - Some instances of comparatively high concentrations were found in Lake Michigan (LMHM, LMMW_1, LMWH_1), Lake St. Clair (LCCR, LSAB), Lake Erie (LEAB, LEAR_1, LEAR_2, LEBR_3, LERB, LESP, NRNF_2), and Lake Ontario (LOFC, LOOC), possibly indicating local inputs of Sn in those areas (Figure 80).
- Regionally, Sn concentrations have significantly increased over time in dreissenid mussel tissue, indicating increasing inputs of Sn into the environment (Figure 78).
  - It is important to note; however, that increasing Sn concentrations are not a primary concern in the Great Lakes Basin, compared to more toxic metals.
- Sn was notably high at site LCCR in 2022, with its concentration being an outlier even when compared to all historic region-wide data (Figure 76).
- The post-hoc analysis evaluating differences between lakes indicates that all lakes are statistically similar, showing no significant differences between lakes (Figure 79).

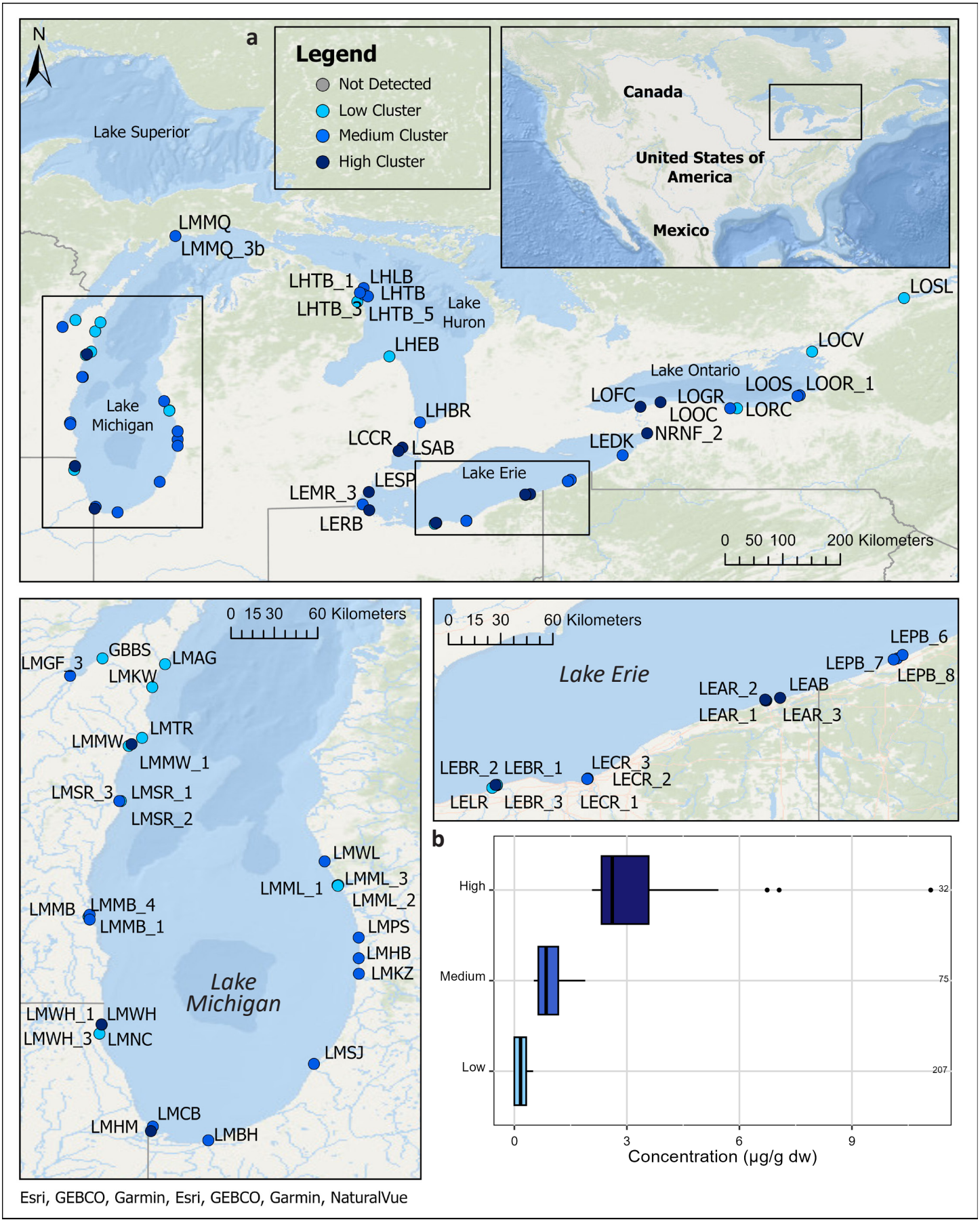


Figure 80. (a) Tin concentrations in dreissenid mussel tissue in 2022 compared to the (b) historic regional MWP Dreissena *species tin concentrations (µg/g dw). Boxplot colors correspond to cluster groups delineated in the map legend. 81 regional sites sampled between 1992 - 2022 for a total of 314 dreissenid mussel samples.*

17.0 RESULTS - ZINC (Zn)

17.1 Zinc Chemical Description

Zinc occurs naturally in the Earth's crust and can be released into the environment through natural rock weathering, but anthropogenic sources far exceed its natural ones. Major industrial sources include electroplating, smelting, and drainage from mining operations (Mirenda, 1986). The greatest use of zinc is as an anti-corrosive coating for iron and steel products (sheet and strip steel, tube and pipe, wire, and wire rope). Canada is one of the largest producers and exporters of zinc (Mirenda, 1986). The U.S. is the largest customer for Canadian refined zinc, and the automobile industry is the largest user of galvanized steel (Mirenda, 1986).

Zinc is an essential nutrient, and particles can be found in air, water, soil, and most foods. Human exposure to high doses of zinc may cause anemia or damage to the pancreas and kidneys (ATSDR, 2005c). However, zinc does not bioaccumulate in humans; therefore, toxic effects are uncommon and associated with excessively high doses. Fish exposed to low zinc concentrations can sequester it in some cases (McGeer et al., 2003). There is no FDA recommended safety level for zinc in fish and fish products.

Dissolved zinc occurs as the free hydrated ion and as dissolved complexes. Changes in water conditions (pH, redox potential, chemical speciation) can result in dissolution from or sorption to particles (U.S. EPA, 1979). In the air, zinc is primarily found in the oxidized form bound to particles. Zinc precipitates as zinc sulfide in anaerobic or reducing environments (such as wetlands) and thus is less mobile, but typically remains as a free ion at lower pHs (U.S. EPA, 1979). Zinc is found in all environmental media (air, water, soil, and biota) as a result of natural and anthropogenic activities.

17.2 Magnitude and Distribution of Zinc in Dreissenid Mussel Tissue in 2022

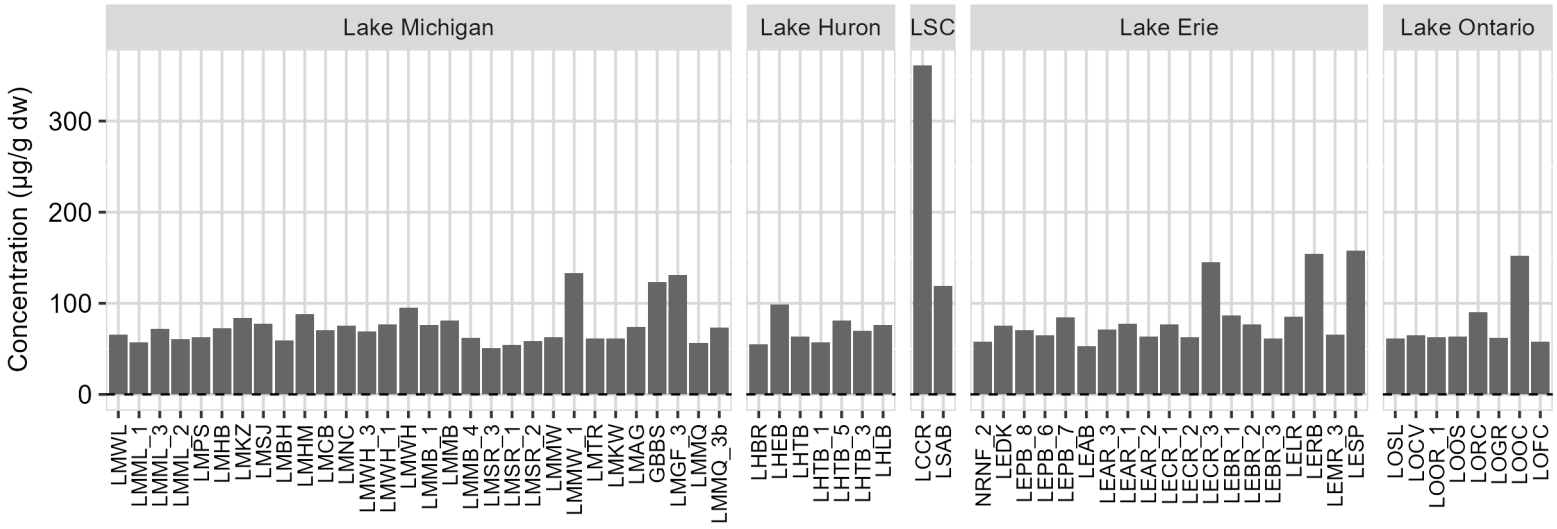


Figure 81. Bar graph showing magnitude of zinc concentrations (µg/g dw) detected in dreissenid mussel tissue in the Great Lakes Basin in 2022. Dotted line represents the minimum weight corrected MDL. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair.

17.3 Historical Context of Zinc Magnitude and Distribution in Dreissenid Mussel Tissue

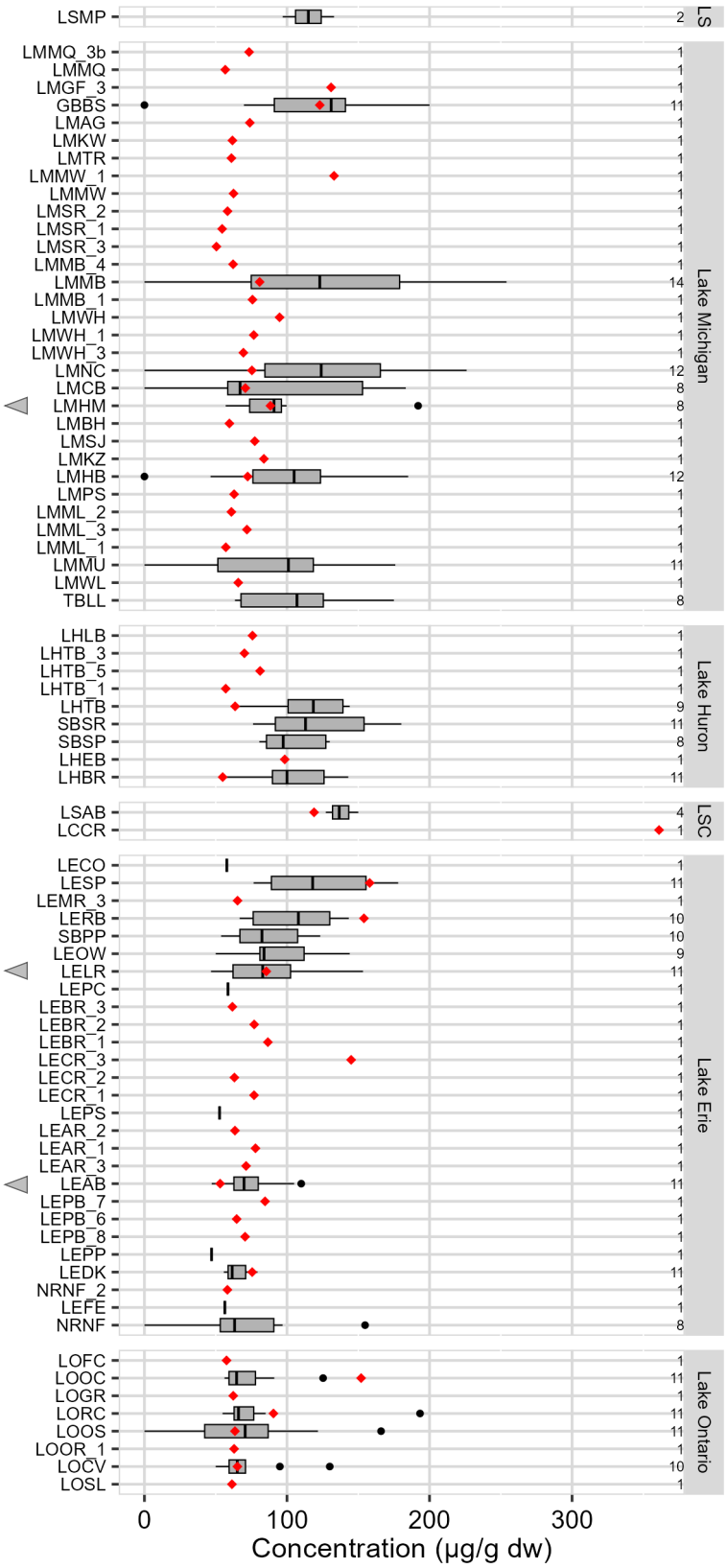


Figure 82. Zinc concentrations (µg/g dw) in dreissenid mussel tissue in 2022 (red diamonds) compared to boxplots of the historic zinc concentrations for each site (1992 - 2008) and results of the trend analysis showing significant increasing (▻) or decreasing (◅) site-specific contamination trends over time. Sites are listed geographically from west to east following the coastline. LSC = Lake St. Clair; LS = Lake Superior. The number of years each site has been sampled since 1992 is noted at the right of the plot for each site.

Results - Zinc (Zn)

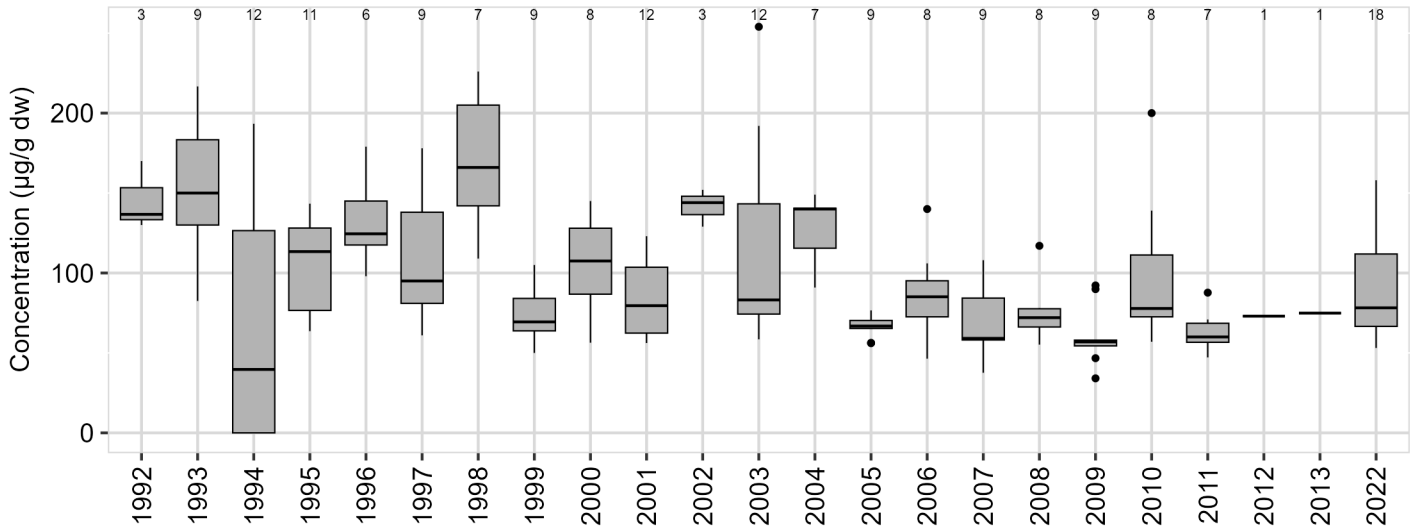


Figure 83. Boxplots representing the historic zinc concentrations (µg/g dw) in dreissenid mussel tissue of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot.

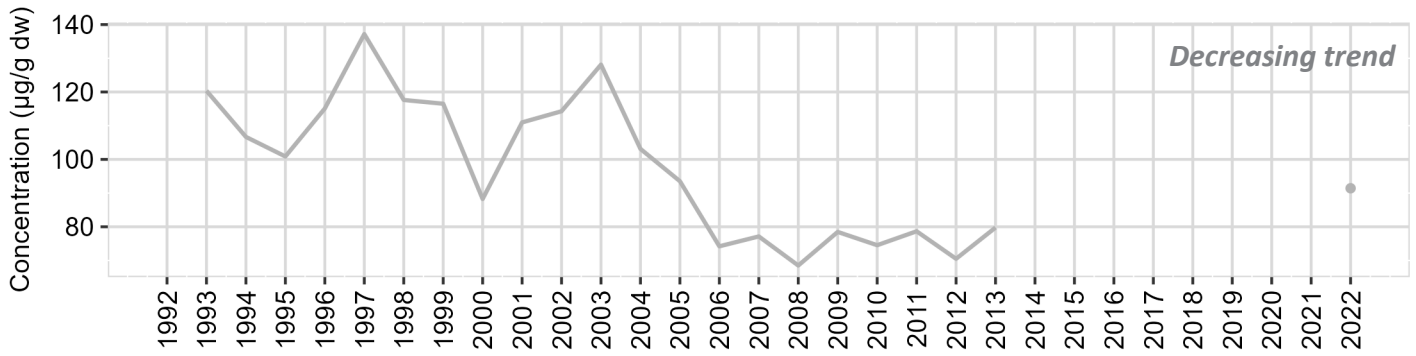


Figure 84. Three-point moving average of the yearly mean zinc concentrations (µg/g dw) in dreissenid mussel tissue in the sites analyzed in this study. Average concentration for 2022 is included as a standalone point for reference.

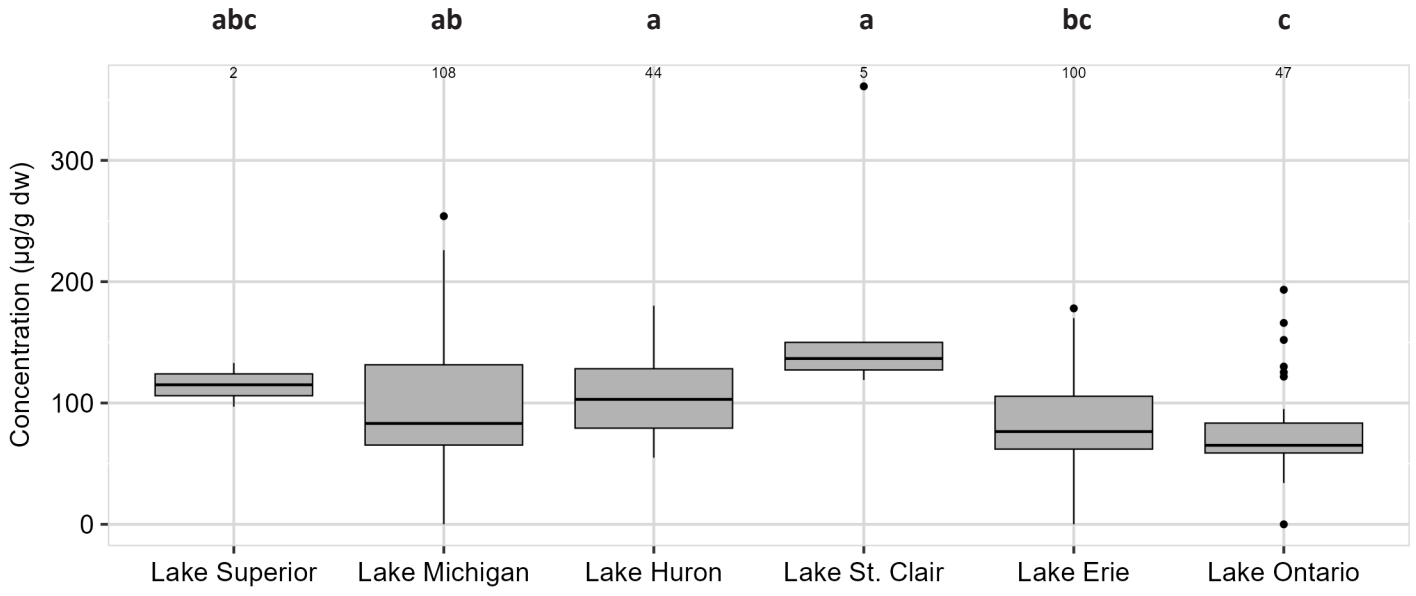


Figure 85. Zinc concentrations (µg/g dw) measured in dreissenid mussel tissue in the Great Lakes from 1992 - 2022. Lakes and major rivers including connecting channels are listed geographically from west to east across the Great Lakes Basin. The number of samples collected in each lake is noted at the top of the plot. Letters above each lake indicate significant differences (at α = 0.05).

17.4 Zinc Summary

2022 Dreissenid Mussel Tissue Results:

- Zn was detected at 100% of 66 sites surveyed (Figure 81).
- Zn concentration descriptive statistics (Figure 81):
  - Range: 50.50 – 361.00 µg/g dw
  - Minimum Zn concentration was detected at LMSR_3
  - Maximum Zn concentration was detected at LCCR
  - Median: 71.05 µg/g dw
  - Mean ± SD: 82.34 ± 43.24 µg/g dw
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster respective to 2022 data only:
  - 86% of sites in low cluster (50.50 - 98.42 µg/g dw)
  - 12% of sites in medium cluster (119.00 - 158.00 µg/g dw)
  - 2% of sites in high cluster (361.00 - 361.00 µg/g dw)

Historic Context for Dreissenid Mussel Tissue (18 out of 66 sites had historic data to analyze for trend analyses):

- 44% of sites surveyed (8/18) were above their historic median concentrations in 2022 (Figure 82).
- Regionally, there was a significant decreasing temporal trend of Zn concentrations in dreissenid mussels in the Great Lakes Basin ($p = 0.01$, $rho = -0.54$) (Figure 83; Figure 84).
- 3 sites (LMHM, LELR, LEAB) in the Great Lakes Basin showed significant decreasing temporal trends of Zn concentrations at $\alpha = 0.05$ (Figure 82; Table A4).
- 0 sites in the Great Lakes Basin showed significant increasing temporal trends of Zn concentrations at $\alpha = 0.05$ (Figure 82; Table A4).
- The percentage of sites in the Great Lakes Basin in 2022 grouped into each cluster compared to historic data (Figure 86):
  - 86% of sites in low cluster (0.00 - 102.00 µg/g dw)
  - 12% of sites in medium cluster (104.00 - 170.00 µg/g dw)
  - 2% of sites in high cluster (175.00 - 361.00 µg/g dw)

General Observations:

- Zn concentrations were ubiquitous and at comparatively low to medium concentrations in the Great Lakes Basin in dreissenid mussel tissue in 2022, likely reflecting naturally higher ambient concentrations (Figure 86).
  - One instance of a comparatively high concentration was found in Lake St. Clair (LCCR), possibly indicating local inputs of Zn in that area (Figure 86).
- Regionally, Zn concentrations have significantly decreased over time in dreissenid mussel tissue, indicating decreasing inputs of Zn into the environment (Figure 84).
- Zn was notably high at site LCCR in 2022, with its concentration being an outlier even when compared to all historic region-wide data (Figure 82).
- The post-hoc analysis evaluating differences between lakes indicates that Lake Superior is a statistical bridge, showing no significant difference to any other lake. In contrast, the other lakes demonstrate a gradient of relationships, being significantly similar to some and significantly different from others. Generally, it appears that Zn concentrations are higher in lakes Superior, Michigan, Huron, and St. Clair, while lower concentrations of Zn were found in lakes Erie and Ontario (Figure 85).

Results - Zinc (Zn)

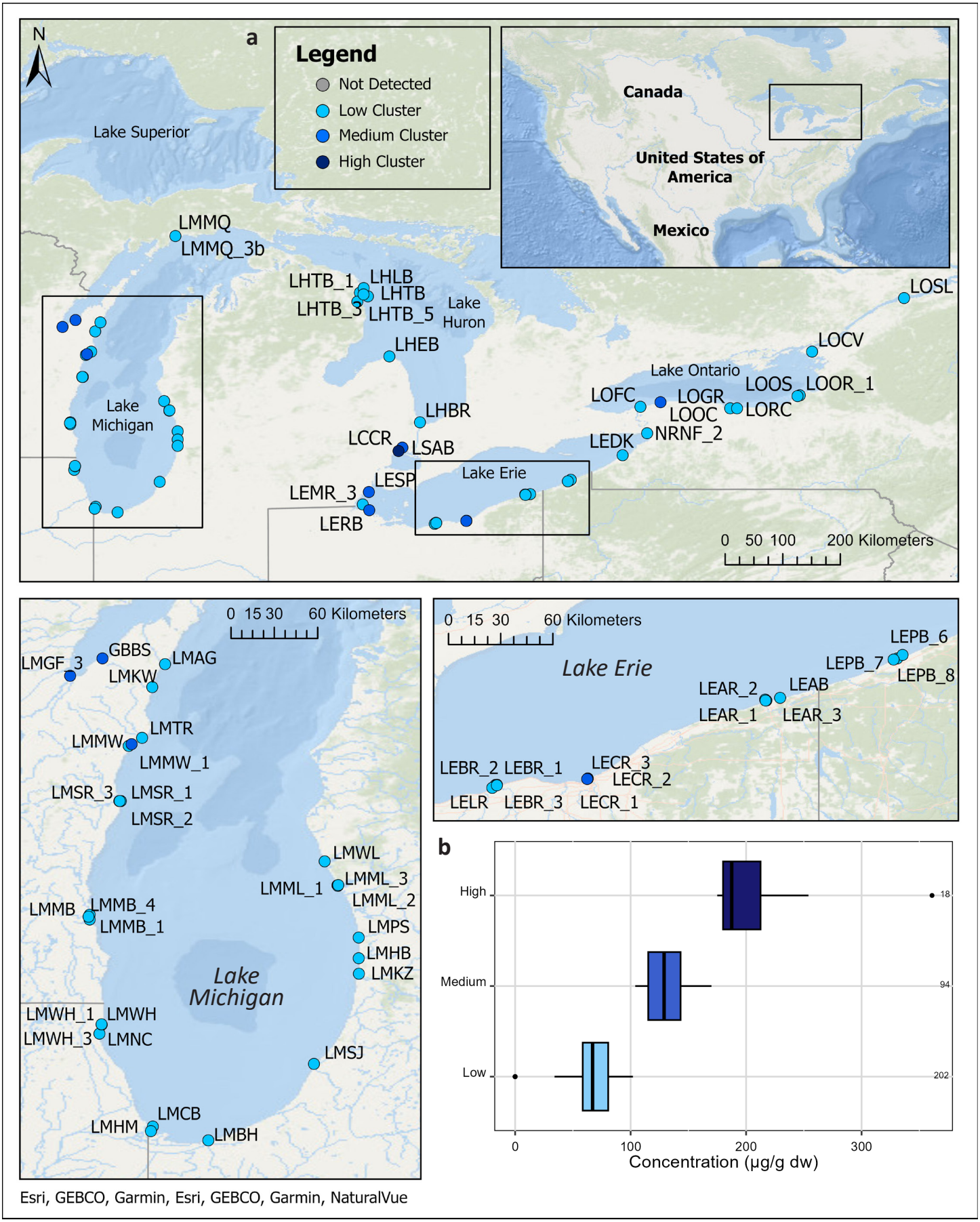


Figure 86. (a) Zinc concentrations in dreissenid mussel tissue in 2022 compared to the (b) historic regional MWP Dreissena *species zinc concentrations (µg/g dw). Boxplot colors correspond to cluster groups delineated in the map legend. 81 regional sites sampled between 1992 - 2022 for a total of 314 dreissenid mussel samples.*

18.0 SUMMARY

Mussels and oysters are good indicators of water quality; hence, they have been used worldwide as sentinel species for chemical pollution in aquatic systems. In this study, mussel tissue (*Dreissena polymorpha* and *Dreissena rostriformis bugensis*) samples were assessed for a suite of trace metals. The samples were collected at historic MWP monitoring sites and select additional sites in conjunction with the United States Geological Survey (USGS) within the Great Lakes Basin. Sample collection was conducted with the support of a network of local collaborators following standard protocols (Apeti et al., 2012). Trace metals were analyzed in dreissenid mussel tissue at 66 sites, 18 of which were historic MWP sites. Separate result summaries for each trace metal can be found in the summary subsection of each trace metal section within this document. This summary integrates all contamination results to assess overall contamination of sites in the Great Lakes Basin, both relative to the other sites analyzed within this study and to the larger historical MWP dataset. For both analyses, a multivariate cluster analysis (the Ward Method) was first conducted for each metal, and the results were presented in heatmaps, grouping sites into categories of not detected, low, medium, and high contamination (Figure 87; Figure 89). Then, a second multivariate cluster analysis (the Ward Method) was conducted using the total cluster value for each site for all metals to assign sites to overall categories with statistically different degrees of overall contamination (not detected, low, medium, high, and very high contamination; Figure 88; Figure 90). This two-pronged analysis was conducted (1) as respective of sites within this study (Figure 87; Figure 88) and (2) as respective to all historic MWP contamination analyzed in dreissenid mussels (Figure 89; Figure 90).

The first observation of note is that all trace metals are highly ubiquitous, with the results indicating that all metals analyzed in this study were detected at every tissue sampling location across the Great Lakes Basin in 2022 (Table 2; Table 3). Trace metals are naturally occurring crustal elements, so detecting them at a high proportion of sites is expected.

Table 2. Frequency of detection (% of total) of trace metals in dreissenid mussel tissue at sites in the Great Lakes Basin in 2022.

Contaminant Group	Sites with Detects	Sites Analyzed	Detection Frequency
Aluminum (Al)	66	66	100%
Arsenic (As)	66	66	100%
Cadmium (Cd)	66	66	100%
Chromium (Cr)	66	66	100%
Copper (Cu)	66	66	100%
Iron (Fe)	66	66	100%
Lead (Pb)	66	66	100%
Manganese (Mn)	66	66	100%
Mercury (Hg)	66	66	100%
Nickel (Ni)	66	66	100%
Selenium (Se)	66	66	100%
Silver (Ag)	66	66	100%
Tin (Sn)	66	66	100%
Zinc (Zn)	66	66	100%

For analyses conducted in dreissenid mussel tissue respective only to sites assessed in this 2022 study, 23% of sites were clustered into the low contamination category, 35% were clustered into the medium contamination category, 39% were clustered into the high contamination category, and 3% were clustered into the very high contamination category (Figure 88).

While it is useful to know which sites in the Great Lakes Basin are most contaminated relative to other sites in the region in the same year, it is equally important to put regional contamination into perspective with other available historic data. When compared to all trace metal data collected by the MWP for dreissenid mussels since 1992, 45% of sites were clustered into the low contamination category, 47% were clustered into the medium contamination category, 6% were clustered into the high contamination category, and 2% were clustered into the very high contamination category (Figure 90).

Summary

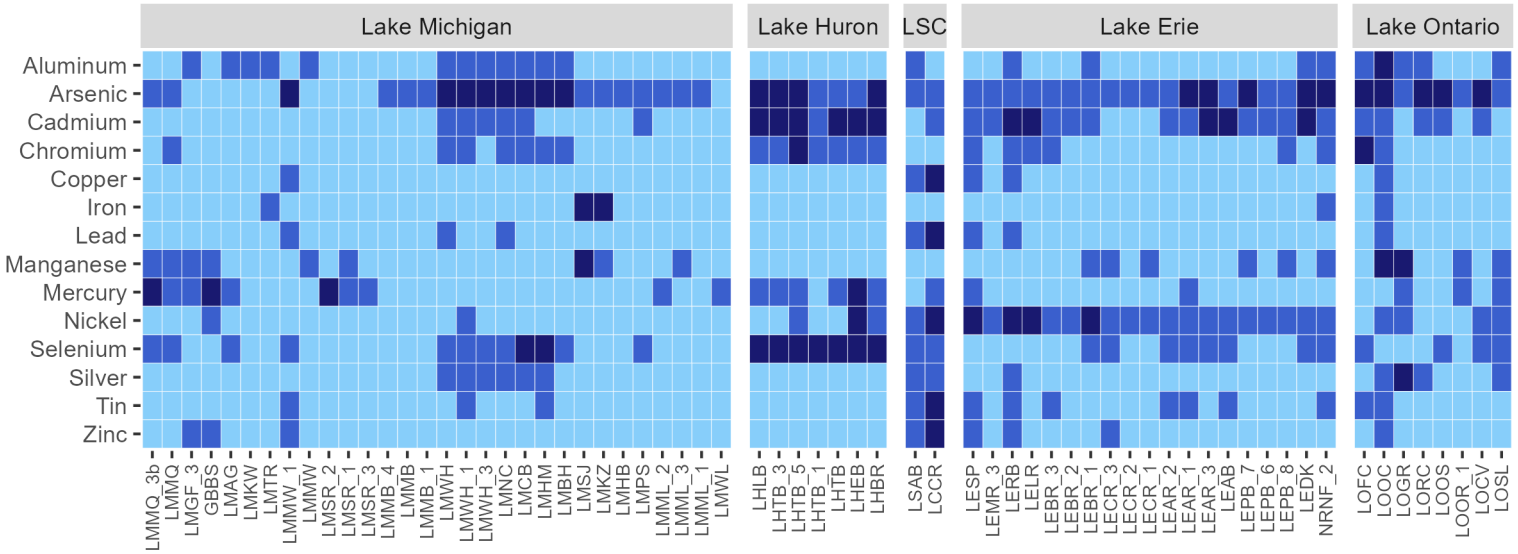


Figure 87. Cluster analysis heat map showing the distribution and magnitude of 2022 concentrations detected in the Great Lakes Basin in dreissenid mussel tissue, respective to one another in this study. Colors represent contamination magnitude as follows: low ( ), medium ( ), high ( ), and not detected ( ).

Regionally, most trace metals have shown decreasing temporal trends of metal concentrations, with a few exceptions. Copper and tin have shown regionally increasing temporal trends, and iron, lead, and silver have shown no temporal trend in the region (Table 4). Decreasing trends of cadmium, nickel, and selenium were found at more than 50% of historic MWP sites in the Great Lakes Basin, suggesting relatively widespread significant decreases in the concentrations of these metals throughout the region over time (Table 4). The analysis of trends seen in trace metals demonstrates the importance of long-standing monitoring programs and provides a point of reference for current sampling. These trends allow us to observe the effects of regulations on the concentrations of these contaminants in the environment, to put into perspective any recently detected concentrations, and to gain an understanding of how different proximate hydrological, abiotic, and anthropogenic characteristics may influence different metal concentrations. Further analysis and sampling may help determine the current point sources of trace metals in the region.

The different classifications of site contamination with respect to 2022 only, versus respective to nationwide NS&T data, support the need for consistent monitoring programs that can contextualize any given result. A successful monitoring program must both achieve an understanding of background contamination levels and capture the variability and range of possible contamination. Existing contaminant data have provided insight into the general background contamination including range, trends, and variability. Further monitoring should aim to continue analyzing the temporal trends of these contaminants on a regional scale (e.g., through periodic sampling and biennial sampling at select sites), but focus on areas of interest that could provide further insight into range, variation, and potential localized point sources of contamination. By understanding both temporal and spatial variations, a monitoring program can serve its purpose of assessing potential contaminant exposure. This study provides the necessary data and information on trace metals for the MWP and provides contamination data required by coastal resource managers as they develop long-term policies to protect the environmental services in this region.

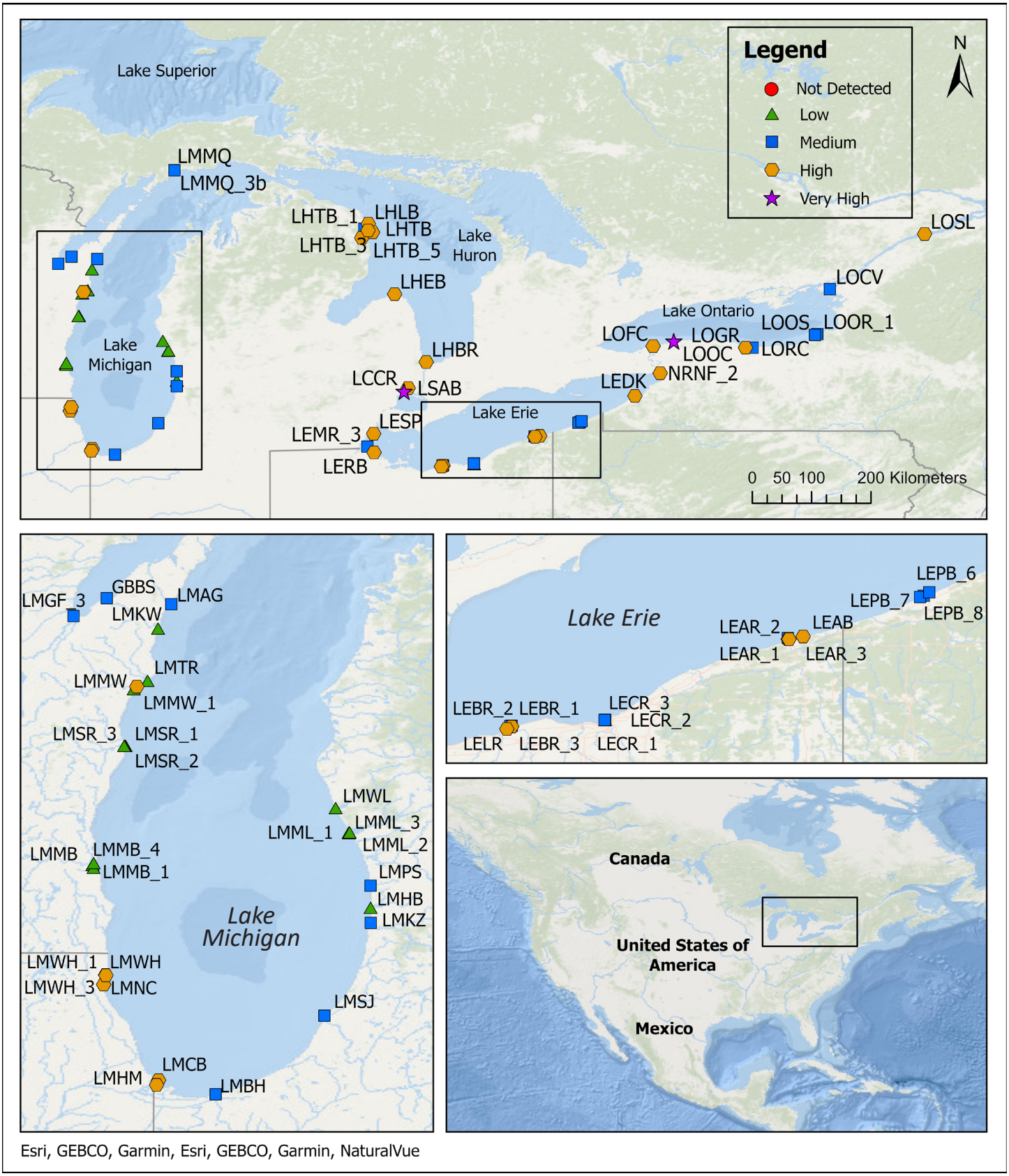


Figure 88. Map of sites in the Great Lakes Basin highlighting locations with low to high degrees of contamination in dreissenid mussel tissue in 2022, respective to one another in this study (Table A2).

Summary

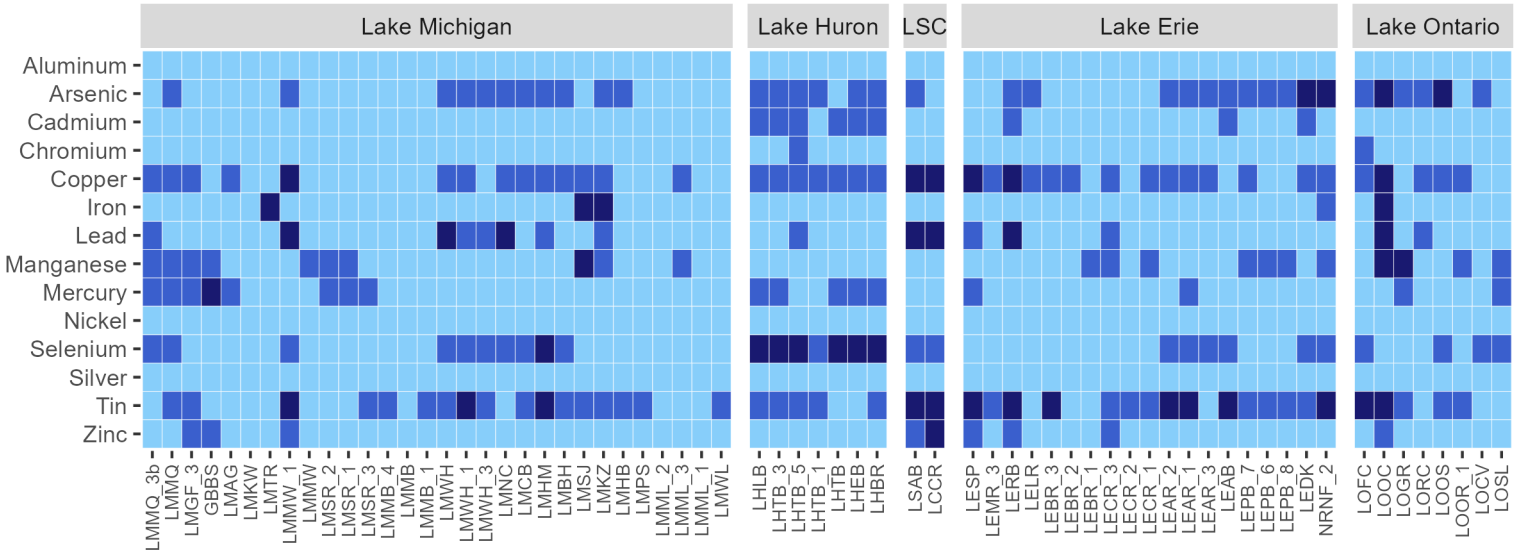


Figure 89. Cluster analysis heat map showing the distribution and magnitude of 2022 concentrations detected in the Great Lakes Basin in dreissenid mussel tissue, respective to all historic MWP contamination analyzed in Dreissena *species. Colors represent contamination magnitude as follows: low ( ), medium ( ), high ( ), and not detected ( ).*

Table 4. Summary of site-based trend analysis for trace metals in dreissenid mussel tissue; D = decreasing trend, I = increasing trend, blank = no trend. Only long term, historic MWP sites successfully sampled in 2022 were analyzed for temporal trends.

Lake	Site	Al	As	Cd	Cr	Cu	Fe	Pb	Mn	Hg	Ni	Se	Ag	Sn	Zn
Lake Michigan	GBBS					I							I	I	
	LMMB			D	D		I				D				
	LMNC			D	D							D			
	LMCB			D	D					D	D	D			
	LMHM			D	D					D					D
	LMHB	D		D	D						D			I	
Lake Huron	LHTB							I							
	LHBR													I	
Lake St. Clair	LSAB			D		I									
Lake Erie	LESP	D	D	D							D	D			
	LERB	D		D				I		D		D	I	I	
	LELR	D	D	D							D				D
	LEAB	D	D				D		D		D	D			D
	LEDK		D									D			
Lake Ontario	LOOC			D							D				
	LORC		D	D							D	D			
	LOOS			D					D	D	D	D			
	LOCV		D	D				I				D		I	
Regional		**D**	**D**	**D**	**D**	**I**			**D**	**D**	**D**	**D**		**I**	**D**

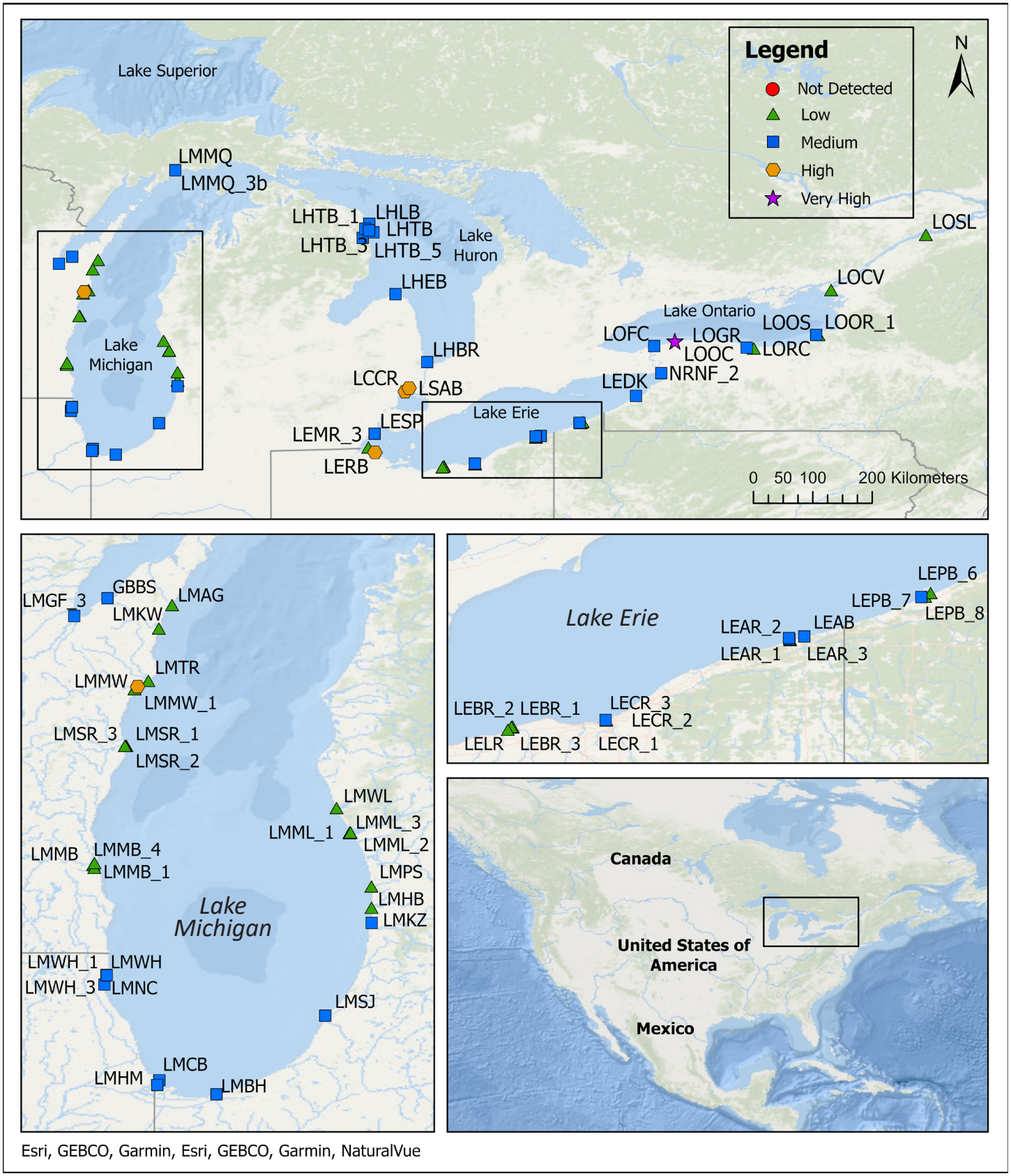


Figure 90. Map of sites in the Great Lakes Basin highlighting locations with low, medium, high, and very high degrees of contamination in dreissenid mussel tissue in 2022, respective to all historic MWP contamination analyzed in Dreissena *species (Table A3).*

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APPENDICES

Table A1. Percent dry values for tissue samples at each site collected in the Great Lakes Basin in 2022.

Site	% Dry of Tissue Samples*	Site	% Dry of Tissue Samples*
GBBS	7.94	LMKW	13.57
LCCR	9.31	LMKZ	17.24
LEAB	10.65	LMMB	12.27
LEAR_1	9.08	LMMB_1	15.17
LEAR_2	9.76	LMMB_4	14.84
LEAR_3	9.13	LMML_1	10.33
LEBR_1	8.96	LMML_2	9.86
LEBR_2	9.21	LMML_3	12.82
LEBR_3	9.92	LMMQ	12.71
LECR_1	11.02	LMMQ_3b	12.28
LECR_2	10.86	LMMW	14.47
LECR_3	11.88	LMMW_1	9.84
LEDK	7.89	LMNC	10.94
LELR	8.57	LMPS	13.96
LEMR_3	10.20	LMSJ	13.54
LEPB_6	7.80	LMSR_1	16.54
LEPB_7	8.40	LMSR_2	17.36
LEPB_8	8.50	LMSR_3	17.21
LERB	10.56	LMTR	14.53
LESP	15.11	LMWH	12.00
LHBR	11.83	LMWH_1	10.65
LHEB	9.56	LMWH_3	11.48
LHLB	11.68	LMWL	13.64
LHTB	10.14	LOCV	7.55
LHTB_1	9.56	LOFC	9.04
LHTB_3	10.09	LOGR	10.90
LHTB_5	11.05	LOOC	12.33
LMAG	14.78	LOOR_1	13.99
LMBH	10.61	LOOS	8.75
LMCB	11.97	LORC	8.34
LMGF_3	10.88	LOSL	11.74
LMHB	14.56	LSAB	11.81
LMHM	8.90	NRNF_2	7.77

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

Appendices

Table A2. 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 compound (0 = not detected, 1 = low, 2 = medium, 3 = high). The second section of the table is the calculation conducted to normalize the compound cluster sums by number of compounds 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).

Site	Al	As	Cd	Cr	Cu	Fe	Pb	Mn	Hg	Ni	Se	Ag	Sn	Zn	Cluster Sum	# Classes Analyzed	Normalized Cluster	Overall Cluster
GBBS	1	1	1	1	1	1	1	2	3	2	1	1	1	2	19	14	45.24	2
LCCR	1	2	2	1	3	1	3	1	2	3	2	2	3	3	29	14	69.05	4
LEAB	1	2	3	1	1	1	1	1	1	2	2	1	2	1	20	14	47.62	3
LEAR_1	1	3	2	1	1	1	1	1	2	2	2	1	2	1	21	14	50.00	3
LEAR_2	1	2	2	1	1	1	1	1	1	2	2	1	2	1	19	14	45.24	2
LEAR_3	1	3	3	1	1	1	1	1	1	2	2	1	1	1	20	14	47.62	3
LEBR_1	2	2	2	1	1	1	1	2	1	3	2	1	1	1	21	14	50.00	3
LEBR_2	1	2	2	1	1	1	1	1	1	2	1	1	1	1	17	14	40.48	2
LEBR_3	1	2	2	2	1	1	1	1	1	2	1	1	2	1	19	14	45.24	2
LECR_1	1	2	1	1	1	1	1	2	1	2	1	1	1	1	17	14	40.48	2
LECR_2	1	2	1	1	1	1	1	1	1	2	1	1	1	1	16	14	38.10	1
LECR_3	1	2	1	1	1	1	1	2	1	2	2	1	1	2	19	14	45.24	2
LEDK	2	3	3	1	1	1	1	1	1	2	2	1	1	1	21	14	50.00	3
LELR	1	2	3	2	1	1	1	1	1	3	1	1	1	1	20	14	47.62	3
LEMR_3	1	2	2	1	1	1	1	1	1	2	1	1	1	1	17	14	40.48	2
LEPB_6	1	2	2	1	1	1	1	1	1	2	1	1	1	1	17	14	40.48	2
LEPB_7	1	3	2	1	1	1	1	2	1	2	1	1	1	1	19	14	45.24	2
LEPB_8	1	2	2	2	1	1	1	2	1	2	1	1	1	1	19	14	45.24	2
LERB	2	2	3	2	2	1	2	1	1	3	1	2	2	2	26	14	61.90	3
LESP	1	2	2	2	2	1	2	1	2	3	1	1	2	2	24	14	57.14	3
LHBR	1	3	3	2	1	1	1	1	2	2	3	1	1	1	23	14	54.76	3
LHEB	1	2	3	2	1	1	1	1	3	3	3	1	1	1	24	14	57.14	3
LHLB	1	3	3	2	1	1	1	1	2	1	3	1	1	1	22	14	52.38	3
LHTB	1	2	3	2	1	1	1	1	2	1	3	1	1	1	21	14	50.00	3
LHTB_1	1	2	2	2	1	1	1	1	1	1	3	1	1	1	19	14	45.24	2
LHTB_3	1	3	3	2	1	1	1	1	2	1	3	1	1	1	22	14	52.38	3
LHTB_5	1	3	3	3	1	1	1	1	2	2	3	1	1	1	24	14	57.14	3
LMAG	2	1	1	1	1	1	1	1	2	1	2	1	1	1	17	14	40.48	2
LMBH	2	3	1	2	1	1	1	1	1	1	2	1	1	1	19	14	45.24	2
LMCB	2	3	2	2	1	1	1	1	1	1	3	2	1	1	22	14	52.38	3
LMGF_3	2	1	1	1	1	1	1	2	2	1	1	1	1	2	18	14	42.86	2
LMHB	1	2	1	1	1	1	1	1	1	1	1	1	1	1	15	14	35.71	1
LMHM	2	3	1	2	1	1	1	1	1	1	3	2	2	1	22	14	52.38	3

Table A2 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 compound (0 = not detected, 1 = low, 2 = medium, 3 = high). The second section of the table is the calculation conducted to normalize the compound cluster sums by number of compounds 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).

Site	Al	As	Cd	Cr	Cu	Fe	Pb	Mn	Hg	Ni	Se	Ag	Sn	Zn	Cluster Sum	# Classes Analyzed	Normalized Cluster	Overall Cluster
LMKW	2	1	1	1	1	1	1	1	1	1	1	1	1	1	15	14	35.71	1
LMKZ	1	2	1	1	1	3	1	2	1	1	1	1	1	1	18	14	42.86	2
LMMB	1	2	1	1	1	1	1	1	1	1	1	1	1	1	15	14	35.71	1
LMMB_1	1	2	1	1	1	1	1	1	1	1	1	1	1	1	15	14	35.71	1
LMMB_4	1	2	1	1	1	1	1	1	1	1	1	1	1	1	15	14	35.71	1
LMML_1	1	2	1	1	1	1	1	1	1	1	1	1	1	1	15	14	35.71	1
LMML_2	1	2	1	1	1	1	1	1	2	1	1	1	1	1	16	14	38.10	1
LMML_3	1	2	1	1	1	1	1	2	1	1	1	1	1	1	16	14	38.10	1
LMMQ	1	2	1	2	1	1	1	2	2	1	2	1	1	1	19	14	45.24	2
LMMQ_3b	1	2	1	1	1	1	1	2	3	1	2	1	1	1	19	14	45.24	2
LMMW	2	1	1	1	1	1	1	2	1	1	1	1	1	1	16	14	38.10	1
LMMW_1	1	3	1	1	2	1	2	1	1	1	2	1	2	2	21	14	50.00	3
LMNC	2	3	2	2	1	1	2	1	1	1	2	2	1	1	22	14	52.38	3
LMPS	1	2	2	1	1	1	1	1	1	1	2	1	1	1	17	14	40.48	2
LMSJ	1	2	1	1	1	3	1	3	1	1	1	1	1	1	19	14	45.24	2
LMSR_1	1	1	1	1	1	1	1	2	2	1	1	1	1	1	16	14	38.10	1
LMSR_2	1	1	1	1	1	1	1	1	3	1	1	1	1	1	16	14	38.10	1
LMSR_3	1	1	1	1	1	1	1	1	2	1	1	1	1	1	15	14	35.71	1
LMTR	2	1	1	1	1	2	1	1	1	1	1	1	1	1	16	14	38.10	1
LMWH	2	3	2	2	1	1	2	1	1	1	2	2	1	1	22	14	52.38	3
LMWH_1	2	3	2	2	1	1	1	1	1	2	2	2	2	1	23	14	54.76	3
LMWH_3	2	3	2	1	1	1	1	1	1	1	2	2	1	1	20	14	47.62	3
LMWL	1	1	1	1	1	1	1	1	2	1	1	1	1	1	15	14	35.71	1
LOCV	1	3	2	1	1	1	1	1	1	2	2	1	1	1	19	14	45.24	2
LOFC	2	3	2	3	1	1	1	1	1	1	2	1	2	1	22	14	52.38	3
LOGR	2	2	1	1	1	1	1	3	2	2	1	3	1	1	22	14	52.38	3
LOOC	3	3	2	2	2	2	2	3	1	2	1	2	2	2	29	14	69.05	4
LOOR_1	1	2	1	1	1	1	1	2	2	1	1	1	1	1	17	14	40.48	2
LOOS	1	3	2	1	1	1	1	1	1	1	2	1	1	1	18	14	42.86	2
LORC	2	3	2	1	1	1	1	1	1	1	1	2	1	1	19	14	45.24	2
LOSL	2	2	1	1	1	1	1	2	2	2	2	2	1	1	21	14	50.00	3
LSAB	2	2	1	1	2	1	2	1	1	2	2	2	2	2	23	14	54.76	3
NRNF_2	2	3	2	2	1	2	1	2	1	2	2	1	2	1	24	14	57.14	3

Appendices

Table A3. 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 compound (0 = not detected, 1 = low, 2 = medium, 3 = high). The second section of the table is the calculation conducted to normalize the compound cluster sums by number of compounds 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).

Site	Al	As	Cd	Cr	Cu	Fe	Pb	Mn	Hg	Ni	Se	Ag	Sn	Zn	Cluster Sum	# Classes Analyzed	Normalized Cluster	Overall Cluster
GBBS	1	1	1	1	1	1	1	2	3	1	1	1	1	2	18	14	42.86	2
LCCR	1	1	1	1	3	1	3	1	1	1	2	1	3	3	23	14	54.76	3
LEAB	1	2	2	1	1	1	1	1	1	1	2	1	3	1	19	14	45.24	2
LEAR_1	1	2	1	1	2	1	1	1	2	1	2	1	3	1	20	14	47.62	2
LEAR_2	1	2	1	1	2	1	1	1	1	1	2	1	3	1	19	14	45.24	2
LEAR_3	1	2	1	1	2	1	1	1	1	1	2	1	1	1	17	14	40.48	1
LEBR_1	1	1	1	1	1	1	1	2	1	1	1	1	1	1	15	14	35.71	1
LEBR_2	1	1	1	1	2	1	1	1	1	1	1	1	1	1	15	14	35.71	1
LEBR_3	1	1	1	1	2	1	1	1	1	1	1	1	3	1	17	14	40.48	1
LECR_1	1	1	1	1	2	1	1	2	1	1	1	1	2	1	17	14	40.48	1
LECR_2	1	1	1	1	1	1	1	1	1	1	1	1	2	1	15	14	35.71	1
LECR_3	1	1	1	1	2	1	2	2	1	1	1	1	2	2	19	14	45.24	2
LEDK	1	3	2	1	2	1	1	1	1	1	2	1	2	1	20	14	47.62	2
LELR	1	2	1	1	2	1	1	1	1	1	1	1	1	1	16	14	38.10	1
LEMR_3	1	1	1	1	2	1	1	1	1	1	1	1	2	1	16	14	38.10	1
LEPB_6	1	2	1	1	1	1	1	2	1	1	1	1	2	1	17	14	40.48	1
LEPB_7	1	2	1	1	2	1	1	2	1	1	1	1	2	1	18	14	42.86	2
LEPB_8	1	2	1	1	1	1	1	2	1	1	1	1	2	1	17	14	40.48	1
LERB	1	2	2	1	3	1	3	1	1	1	1	1	3	2	23	14	54.76	3
LESP	1	1	1	1	3	1	2	1	2	1	1	1	3	2	21	14	50.00	2
LHBR	1	2	2	1	2	1	1	1	2	1	3	1	2	1	21	14	50.00	2
LHEB	1	2	2	1	2	1	1	1	2	1	3	1	1	1	20	14	47.62	2
LHLB	1	2	2	1	2	1	1	1	2	1	3	1	2	1	21	14	50.00	2
LHTB	1	1	2	1	2	1	1	1	2	1	3	1	1	1	19	14	45.24	2
LHTB_1	1	2	1	1	2	1	1	1	1	1	2	1	2	1	18	14	42.86	2
LHTB_3	1	2	2	1	2	1	1	1	2	1	3	1	2	1	21	14	50.00	2
LHTB_5	1	2	2	2	2	1	2	1	1	1	3	1	2	1	22	14	52.38	2
LMAG	1	1	1	1	2	1	1	1	2	1	1	1	1	1	16	14	38.10	1
LMBH	1	2	1	1	2	1	1	1	1	1	2	1	2	1	18	14	42.86	2
LMCB	1	2	1	1	2	1	1	1	1	1	2	1	2	1	18	14	42.86	2
LMGF_3	1	1	1	1	2	1	1	2	2	1	1	1	2	2	19	14	45.24	2
LMHB	1	2	1	1	1	1	1	1	1	1	1	1	2	1	16	14	38.10	1
LMHM	1	2	1	1	2	1	2	1	1	1	3	1	3	1	21	14	50.00	2

Table A3 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 compound (0 = not detected, 1 = low, 2 = medium, 3 = high). The second section of the table is the calculation conducted to normalize the compound cluster sums by number of compounds 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).

Site	Al	As	Cd	Cr	Cu	Fe	Pb	Mn	Hg	Ni	Se	Ag	Sn	Zn	Cluster Sum	# Classes Analyzed	Normalized Cluster	Overall Cluster
LMKW	1	1	1	1	1	1	1	1	1	1	1	1	1	1	14	14	33.33	1
LMKZ	1	2	1	1	2	3	2	2	1	1	1	1	2	1	21	14	50.00	2
LMMB	1	1	1	1	1	1	1	1	1	1	1	1	1	1	14	14	33.33	1
LMMB_1	1	1	1	1	1	1	1	1	1	1	1	1	2	1	15	14	35.71	1
LMMB_4	1	1	1	1	1	1	1	1	1	1	1	1	2	1	15	14	35.71	1
LMML_1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	14	14	33.33	1
LMML_2	1	1	1	1	1	1	1	1	1	1	1	1	1	1	14	14	33.33	1
LMML_3	1	1	1	1	2	1	1	2	1	1	1	1	1	1	16	14	38.10	1
LMMQ	1	2	1	1	2	1	1	2	2	1	2	1	2	1	20	14	47.62	2
LMMQ_3b	1	1	1	1	2	1	2	2	2	1	2	1	1	1	19	14	45.24	2
LMMW	1	1	1	1	1	1	1	2	1	1	1	1	1	1	15	14	35.71	1
LMMW_1	1	2	1	1	3	1	3	1	1	1	2	1	3	2	23	14	54.76	3
LMNC	1	2	1	1	2	1	3	1	1	1	2	1	1	1	19	14	45.24	2
LMPS	1	1	1	1	1	1	1	1	1	1	1	1	2	1	15	14	35.71	1
LMSJ	1	1	1	1	2	3	1	3	1	1	1	1	2	1	20	14	47.62	2
LMSR_1	1	1	1	1	1	1	1	2	2	1	1	1	1	1	16	14	38.10	1
LMSR_2	1	1	1	1	1	1	1	2	2	1	1	1	1	1	16	14	38.10	1
LMSR_3	1	1	1	1	1	1	1	1	2	1	1	1	2	1	16	14	38.10	1
LMTR	1	1	1	1	1	3	1	1	1	1	1	1	1	1	16	14	38.10	1
LMWH	1	2	1	1	2	1	3	1	1	1	2	1	2	1	20	14	47.62	2
LMWH_1	1	2	1	1	2	1	2	1	1	1	2	1	3	1	20	14	47.62	2
LMWH_3	1	2	1	1	1	1	2	1	1	1	2	1	2	1	18	14	42.86	2
LMWL	1	1	1	1	1	1	1	1	1	1	1	1	2	1	15	14	35.71	1
LOCV	1	2	1	1	1	1	1	1	1	1	2	1	1	1	16	14	38.10	1
LOFC	1	2	1	2	2	1	1	1	1	1	2	1	3	1	20	14	47.62	2
LOGR	1	2	1	1	1	1	1	3	2	1	1	1	2	1	19	14	45.24	2
LOOC	1	3	1	1	3	3	3	3	1	1	1	1	3	2	27	14	64.29	4
LOOR_1	1	1	1	1	2	1	1	2	1	1	1	1	2	1	17	14	40.48	1
LOOS	1	3	1	1	2	1	1	1	1	1	2	1	2	1	19	14	45.24	2
LORC	1	2	1	1	2	1	2	1	1	1	1	1	1	1	17	14	40.48	1
LOSL	1	1	1	1	1	1	1	2	2	1	2	1	1	1	17	14	40.48	1
LSAB	1	2	1	1	3	1	3	1	1	1	2	1	3	2	23	14	54.76	3
NRNF_2	1	3	1	1	2	2	1	2	1	1	2	1	3	1	22	14	52.38	2

Table A4. Results of statistically significant Spearman's Rank tests on site specific temporal trends in dreissenid mussel tissue for sites sampled in the Great Lakes Basin in 2022. I = Increasing trend, D = Decreasing trend.

Metal	Site	p value	rho value	Trend	Metal	Site	p value	rho value	Trend
Al	LMHB	0.02	-0.68	D	Pb	LOCV	0.02	0.73	I
Al	LESP	0.04	-0.63	D	Mn	LEAB	0.00	-0.89	D
Al	LERB	0.01	-0.76	D	Mn	LOOS	0.04	-0.63	D
Al	LELR	0.04	-0.64	D	Hg	LMCB	0.00	-0.93	D
Al	LEAB	0.04	-0.62	D	Hg	LMHM	0.02	-0.79	D
As	LESP	0.00	-0.87	D	Hg	LERB	0.04	-0.66	D
As	LELR	0.00	-0.87	D	Hg	LOOS	0.00	-0.91	D
As	LEAB	0.00	-0.94	D	Ni	LMMB	0.01	-0.69	D
As	LEDK	0.01	-0.76	D	Ni	LMCB	0.01	-0.81	D
As	LORC	0.04	-0.63	D	Ni	LMHB	0.02	-0.68	D
As	LOCV	0.03	-0.67	D	Ni	LESP	0.04	-0.62	D
Cd	LMMB	0.00	-0.71	D	Ni	LELR	0.05	-0.61	D
Cd	LMNC	0.01	-0.75	D	Ni	LEAB	0.05	-0.61	D
Cd	LMCB	0.00	-0.90	D	Ni	LOOC	0.02	-0.69	D
Cd	LMHM	0.01	-0.81	D	Ni	LORC	0.00	-0.82	D
Cd	LMHB	0.00	-0.78	D	Ni	LOOS	0.00	-0.80	D
Cd	LSAB	0.00	-1.00	D	Se	LMNC	0.00	-0.75	D
Cd	LESP	0.00	-0.87	D	Se	LMCB	0.03	-0.76	D
Cd	LERB	0.00	-0.89	D	Se	LESP	0.00	-0.85	D
Cd	LELR	0.01	-0.76	D	Se	LERB	0.04	-0.65	D
Cd	LOOC	0.00	-0.80	D	Se	LEAB	0.03	-0.66	D
Cd	LORC	0.00	-0.82	D	Se	LEDK	0.01	-0.74	D
Cd	LOOS	0.01	-0.72	D	Se	LORC	0.00	-0.84	D
Cd	LOCV	0.00	-0.82	D	Se	LOOS	0.02	-0.70	D
Cr	LMMB	0.04	-0.56	D	Se	LOCV	0.00	-0.88	D
Cr	LMNC	0.02	-0.64	D	Ag	GBBS	0.04	0.62	I
Cr	LMCB	0.01	-0.83	D	Ag	LERB	0.00	0.82	I
Cr	LMHM	0.01	-0.81	D	Sn	GBBS	0.02	0.69	I
Cr	LMHB	0.00	-0.81	D	Sn	LMHB	0.01	0.68	I
Cu	GBBS	0.00	0.91	I	Sn	LHBR	0.00	0.80	I
Cu	LSAB	0.00	1.00	I	Sn	LERB	0.01	0.80	I
Fe	LMMB	0.04	0.56	I	Sn	LOCV	0.04	0.66	I
Fe	LEAB	0.00	-0.80	D	Zn	LMHM	0.03	-0.76	D
Pb	LHTB	0.04	0.70	I	Zn	LELR	0.00	-0.81	D
Pb	LERB	0.05	0.64	I	Zn	LEAB	0.03	-0.65	D



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