HTTPS://DOI.ORG/10.25923/2N6C-MB39

NATIONAL STATUS AND TRENDS, MUSSEL WATCH PROGRAM

A 2017 Assessment of Metals in the Gulf of Mexico

December 2022

NOAA TECHNICAL MEMORANDUM NOS NCCOS 314

NOAA NCCOS Monitoring and Assessment Branch

Citation

Dampier, J. Middleton, Apeti, D.A., Rider, M. and Reed, L. 2022. A 2017 Assessment of Metals in the Gulf of Mexico. NOAA Technical Memorandum NOS NCCOS 314. Silver Spring, MD. 50 pp. DOI 10.25923/2n6c-mb39.

Disclaimers

This report has been reviewed and approved for publication according to the NOAA's Scientific Integrity Policy and Fundamental Research Communications (FRC) framework, and the National Ocean Service (NOS) process for FRC review. The opinions, findings, conclusions, and recommendations expressed in this report are those of the authors, and they do not necessarily reflect those of NOAA. Any use of trade, firm, or product names is for descriptive purposes only and does not imply endorsement by the U.S. Government.

This NOAA Experiential Research and Training Opportunities (NERTO) internship was made possible by the National Oceanic and Atmospheric Administration (NOAA), Office of Education Educational Partnership Program award (NA16SEC4810009). The contents of this report are solely the responsibility of the award recipient and do not necessarily represent the official views of the U.S. Department of Commerce, National Oceanic and Atmospheric Administration. Any opinions, findings, conclusions, or recommendations expressed in this report are those of the author(s) and do not necessarily reflect the view of the U.S. Department of Commerce, National Oceanic and Atmospheric Administration.

Front page images credit: NOAA

Back page image credit: NOAA

NATIONAL STATUS AND TRENDS, MUSSEL WATCH PROGRAM
A 2017 Assessment of Metals in the Gulf of Mexico
December 2022

Authors:

Jeanna Middleton Dampier[1], Dennis A. Apeti[2], Mary M. Rider[2,3], and LouAnn Reed[4]

[1] Jackson State University, Jackson, MS

[2] NOAA National Ocean Service, National Centers for Coastal Ocean Science, Stressor Detection and Impacts Division, Silver Spring, MD

[3] Consolidated Safety Services, Inc., Fairfax, VA, under contract to NOAA

[4] NOAA National Ocean Service, National Centers for Coastal Ocean Science, Stressor Detection and Impacts Division, Charleston, SC

NOAA Technical Memorandum NOS NCCOS 314

United States Department of Commerce	National Oceanic and Atmospheric Administration	National Ocean Service
Gina M. Raimondo Secretary	Richard W. Spinrad Under Secretary	Nicole LeBoeuf Assistant Administrator

Executive Summary

In 1986, the National Mussel Watch Program (MWP), 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 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. Currently, five regions (Northeast, Southeast, Gulf of Mexico, West Coast, & Great Lakes) are sampled on a five-year rotation, with one region being sampled each year. The focus of this Gulf of Mexico technical report is to discuss the general distribution of trace metal contaminants in bivalve tissues collected from 39 sites across five states (Texas, Louisiana, Mississippi, Alabama, and Florida) in the northern Gulf of Mexico on the US coast. This report contains the analysis of both the most recent (2017) data as well as data previously collected by the MWP.

Of the 8 metals reported here, none displayed regional trends based on Spearman-Rank analyses, possibly due to the innate variability of the data. However, a combination of regional 3-point moving averages, site-based trend analyses and comparisons to the historic national MWP data set helps to elucidate the overall condition of these metals in the Gulf of Mexico region. Most of the metals, including copper, lead, mercury, tin and zinc, do not show any indication of increasing or decreasing on a regional scale since 1986. Notable concentrations for these metals are site specific and do not appear to fall outside of the normal variations detected by the MWP. Comparatively, cadmium concentrations appear to have decreased in the region since 1986 and concentrations remain low in 2017 compared to the historic national MWP data. Conversely, arsenic and nickel concentrations appear to be increasing regionally. Arsenic concentrations remain relatively low with the exception of one historically high site in Tampa Bay and a handful of other sites in Florida whose site concentrations have been increasing over time. Nickel concentrations have seemingly been increasing over time at both the regional scale and at many individual sites. Furthermore, many sites in the Gulf of Mexico now have nickel concentrations that compare to the highest nickel concentrations detected nationally for *C. virginica* by the MWP.

The authors do not attempt to offer many direct explanations for the trends and variations of the data presented in this report, instead leaving that analysis to the managers and stakeholders that have a better understanding of the local dynamics and potential sources. The aim of this report is to contextualize recent regional monitoring data in both time and space and to provide a perspective that may not be attainable at the local level. By doing so, we hope to support the allocation of resources and the ongoing studies and efforts necessary to manage coastal chemical contaminants nationwide.

Table of Contents

1.0 INTRODUCTION

In 1986, the National Mussel Watch Program (MWP), 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, among other activities, to initiate a continuous monitoring program. 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 found along all the United States coastlines, including Alaska, the Great Lakes, Hawaii, and in territories such as Puerto Rico.

A fundamental challenge faced by any long-term environmental monitoring program is how (or whether) to evolve in response to changing technologies, societal factors and environmental conditions. In 2013, due to budgetary constraints, the National Centers for Coastal Ocean Science (NCCOS) undertook the task of re-designing the MWP, moving from a nationwide yearly monitoring approach targeting every MWP station, to the rotating regional monitoring model that is currently employed. The five regions (Northeast, Southeast, Gulf of Mexico, West Coast, & Great Lakes) are sampled on a five-year rotation, with one region being sampled each year. The regional approach allows the program to improve its presence in coastal communities by increasing interaction and collaboration 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 and nearly 600 chemical contaminants. The list of chemical contaminants includes metals, legacy organic compounds and chemicals of emerging concern (CECs) (Figure 1). The MWP provides unique data that are vital to evaluating the health of the Nation's estuarine

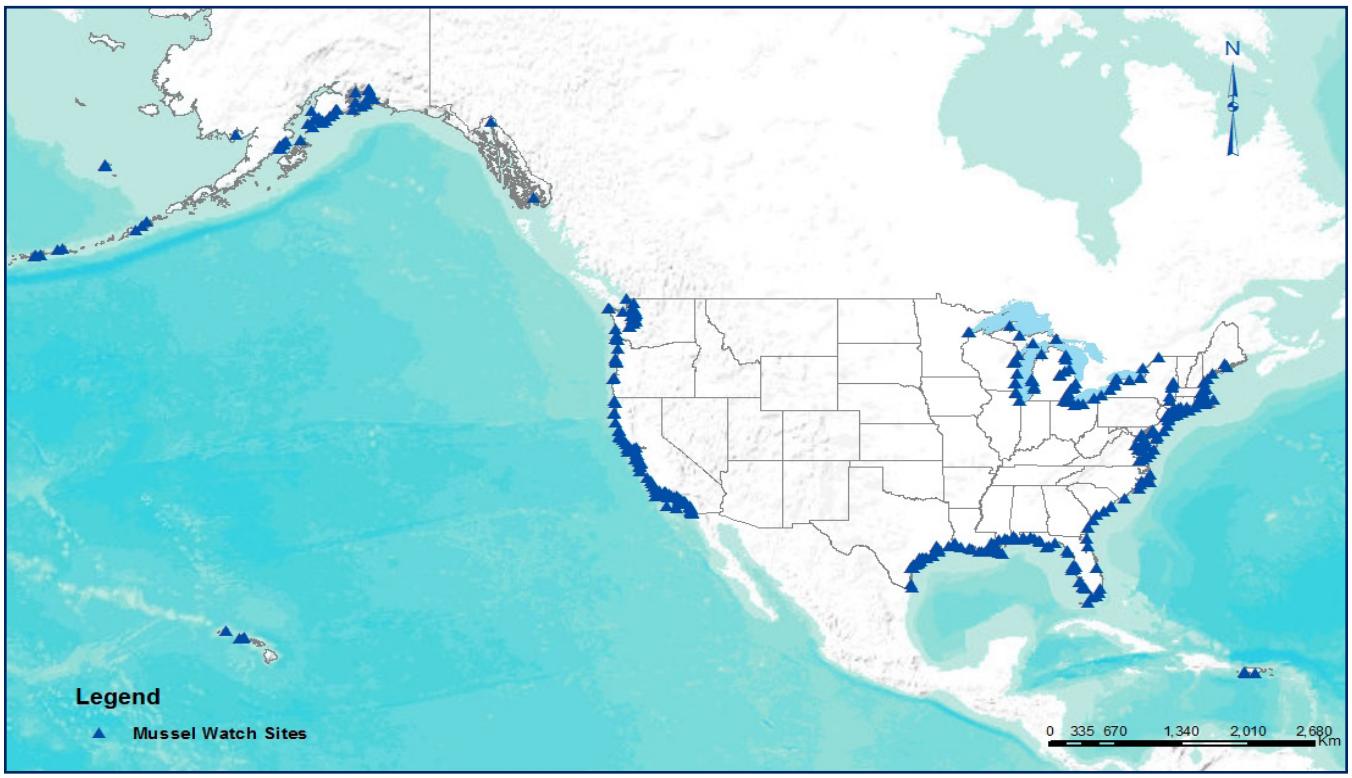


Figure 1. National Mussel Watch sites.

and coastal waters, particularly describing the levels of chemical contamination. The MWP dataset allows for temporal and spatial evaluation of regional and national changes in chemical distribution and concentration. The program's long-term data collection supports the assessment of potential impacts of unforeseen events such as oil spills and hurricanes, as well as evaluating the effectiveness of regulations that ban or limit toxic chemical usage s or support legislation such as the Clean Air and Clean Water Acts.

In 2017, the MWP performed their regional sampling along the Gulf of Mexico. The focus of this Gulf of Mexico report is to discuss the general distribution of trace metal contaminants in bivalve tissues collected from 39 sites across five states (Texas, Louisiana, Mississippi, Alabama, and Florida) in the northern Gulf of Mexico on the US coast.

2.0 METHODS

2.1 Sampling Design

Mussels and oysters are widely distributed along the coast, minimizing the issues inherent when comparing data from markedly different and mobile species. This wide-ranging distribution and the fact that adult bivalves are fixed to a local substrate makes bivalves a good integrator of contamination within a given area (Berner et al., 1976; Farrington et al., 1980; Farrington, 1983; and Tripp and Farrington, 1984). They are good surrogates for monitoring environmental quality because contaminant levels in their tissue respond to changes in ambient environmental factors and these organisms generally accumulate chemicals with little metabolic transformation (Roesijadi et al., 1984; Sericano, 1993). Historically, Mussel Watch sites were selected to represent large coastal areas that can be used to construct a nationwide assessment without the interferences or bias that can be attributed to known site specific contaminant input. Sites selected for monitoring are generally 10 to 100 km apart along the U.S. coastline, including the Great Lakes, Puerto Rico, Alaska, and Hawaii. Where possible, sites were selected to coincide with historical mussel and oyster monitoring locations from other programs, such as the U.S. EPA's Mussel Watch sites that were sampled from 1976 to 1978 (Goldberg et al., 1983), and to complement sites sampled through state programs, such as the California Mussel Watch Program (Martin, 1985). Because one single species of mussel or oyster is not common to all coastal regions, a variety of species are collected to gain a national perspective. A target species is identified for each site based on abundance and ease of collection. Mussels (*Mytilus* species) are collected from the North Atlantic and Pacific coasts, oysters (*Crassostrea virginica*) from the mid-Atlantic (Delaware Bay) southward and along the Gulf Coast, and zebra mussels (Dreissena species), an invasive species, are collected from sites in the Great Lakes. Despite the number of sites for a coastline as large as that of the U.S., relatively few species are required to determine a national contaminant perspective. For organic contaminants it is possible to compare across all sites because Mussel Watch species have a similar ability to bioaccumulate contaminants. For trace metals there are clear differences in bioaccumulation abilities between coastal mussels and oysters. Oysters have a greater affinity for zinc, copper and silver, while mussels are better able to accumulate lead and chromium (Kimbrough et al., 2008).

The oysters in this report were collected by hand or dredged from intertidal to shallow subtidal zones, brushed clean, packed in iced containers, and shipped to analytical laboratories within two days of collection. Sample collection protocols are described in detail in Apeti et al. (2012). Sample preparation, extraction techniques and analytical methods used by the Mussel Watch Program are available in Kimbrough and Lauenstein (2006). The Mussel Watch Program uses a performance-based quality assurance (QA) process to ensure data quality. This effort has been in operation since 1986 and is designed to document sampling protocols, analytical procedures and laboratory performance.

Methods

This Gulf of Mexico report contains the analysis of both the most recent (2017) as well as previously generated data for 39 sites across five states in the Gulf of Mexico on the US coast. In the 2017 assessment, sixty sites were visited for potential oyster retrieval, however, 21 of the sites had insufficient oyster population for metals analysis (Figure 2, Table 1).

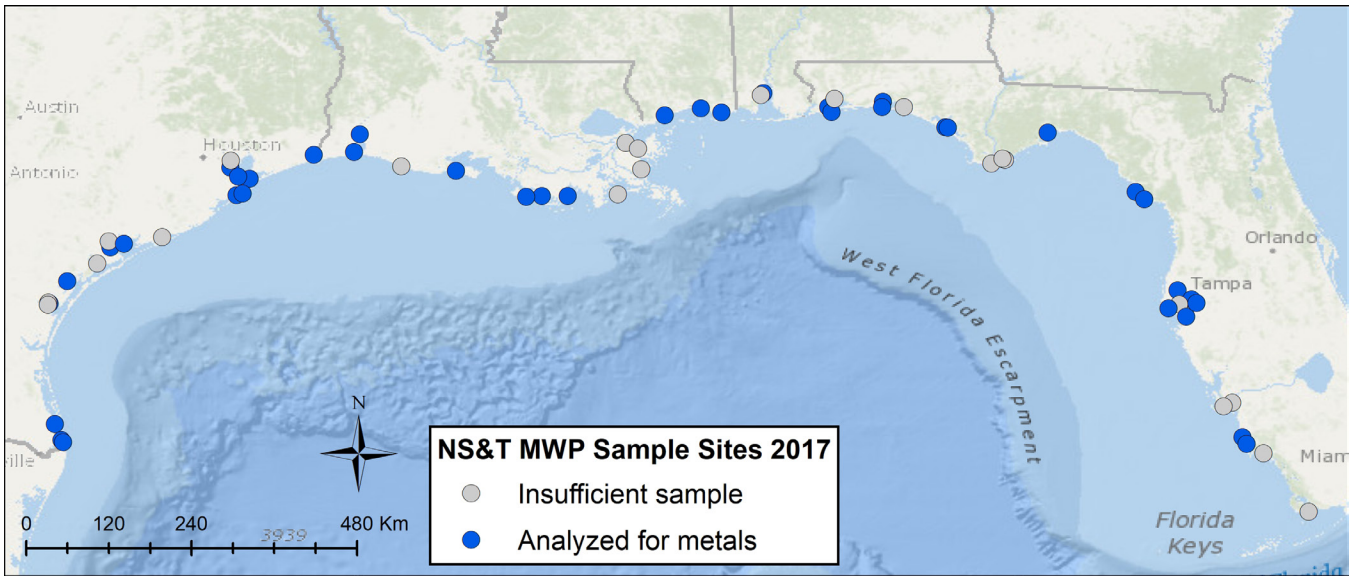


Figure 2. Mussel Watch sites visited in the Gulf of Mexico in 2017. 21 of the 60 sites visited had insufficient oyster populations for metals analysis.

Table 1. Description of Mussel Watch sites selected for the 2017 survey. 21 of the 60 sites visited had insufficient oyster population for metals analysis. "•" signifies the site was not analyzed for metals in 2017. TX = Texas, LA = Louisiana, MS = Mississippi, AL = Alabama, FL = Florida

	MWP Site	State	General Location	Specific Location	Latitude	Longitude	Sample Date
	LMSB	TX	Lower Laguna Madre	South Bay	26.04272	-97.18147	9/14/2017
	LMPI	TX	Lower Laguna Madre	Port Isabel	26.07367	-97.19927	9/14/2017
	LMAC	TX	Lower Laguna Madre	Arroyo Colorado	26.28010	-97.28827	9/14/2017
•	CCBH	TX	Corpus Christi	Boat Harbor	27.83649	-97.37970	9/15/2017
•	CCDC	TX	Corpus Christi	Nueces Bay, Doyle Cove	27.86183	-97.37262	9/15/2017
	CCNB	TX	Corpus Christi	Nueces Bay	27.85158	-97.35961	9/13/2017
	CBCR	TX	Copano Bay	Copano Reef	28.14251	-97.12864	9/28/2017
•	SAMP	TX	San Antonio Bay	Mosquito Point	28.37028	-96.73611	9/27/2017
•	MBLR	TX	Matagorda Bay	Lavaca River Mouth	28.66032	-96.58430	9/26/2017
	MBGP	TX	Matagorda Bay	Gallinipper Point	28.58397	-96.56463	9/26/2017
	MBCB	TX	Matagorda Bay	Carancahua Bay	28.63152	-96.38497	9/27/2017
•	MBEM	TX	Matagorda Bay	East Matagorda	28.71417	-95.88637	9/27/2017
•	GBSC	TX	Galveston Bay	Ship Channel	29.71314	-94.99254	11/6/2017
	GBCR	TX	Galveston Bay	Confederate Reef	29.26454	-94.91530	9/16/2017
	GBOB	TX	Galveston Bay	Offatts Bayou	29.28479	-94.83608	9/16/2017
	GBTD	TX	Galveston Bay	Todd's Dump	29.50259	-94.89604	9/17/2017
	GBYC	TX	Galveston Bay	Yacht Club	29.62096	-94.99579	9/17/2017

Methods

	GBHR	TX	Galveston Bay	Hanna Reef	29.47841	-94.74537	9/16/2017
	SLBB	LA	Sabine Lake	Blue Buck Point	29.78893	-93.90686	10/7/2017
	CLSJ	LA	Calcasieu Lake	St. Johns Island	29.82696	-93.38447	10/7/2017
	CLLC	LA	Calcasieu Lake	Lake Charles	30.05867	-93.30750	10/8/2017
•	JHJH	LA	Joseph Harbor Bayou	Joseph Harbor Bayou	29.63916	-92.76839	10/13/2017
	VBSP	LA	Vermilion Bay	Southwest Pass	29.58057	-92.05366	10/8/2017
	ABOB	LA	Atchafalaya Bay	Oyster Bayou	29.24027	-91.13314	10/12/2017
	CLCL	LA	Caillou Lake	Caillou Lake	29.25407	-90.92947	10/12/2017
	TBLB	LA	Terrebonne Bay	Lake Barre	29.25334	-90.59284	10/12/2017
•	BBMB	LA	Barataria Bay	Middle Bank	29.27667	-89.94200	10/11/2017
•	LBGO	LA	Lake Borgne	Gulf Outlet	29.94672	-89.83689	10/9/2017
•	LBMP	LA	Lake Borgne	Malheureux Point	29.86671	-89.67652	10/9/2017
•	BSBG	LA	Breton Sound	Bay Gardene	29.60077	-89.63283	10/9/2017
	MSPC	MS	Mississippi Sound	Pass Christian	30.30271	-89.32790	10/10/2017
	MSBB	MS	Mississippi Sound	Biloxi Bay	30.39293	-88.85794	10/10/2017
	MSPB	MS	Mississippi Sound	Pascagoula Bay	30.34083	-88.58809	10/10/2017
•	MBHI	AL	Mobile Bay	Hollingers Is. Chan.	30.56607	-88.07257	11/4/2017
	MBDR	AL	Mobile Bay	Dog River	30.59090	-88.04227	11/4/2017
	PBPH	FL	Pensacola Bay	Public Harbor	30.41367	-87.19133	11/5/2017
	PBSP	FL	Pensacola Bay	Sabine Point	30.34642	-87.15248	11/5/2017
•	PBIB	FL	Pensacola Bay	Indian Bayou	30.51677	-87.11145	11/4/2017
	CBJB	FL	Choctawhatchee Bay	Joe's Bayou	30.41038	-86.49095	11/5/2017
	CBPP	FL	Choctawhatchee Bay	Postil Point	30.48267	-86.47923	11/5/2017
•	CBSR	FL	Choctawhatchee Bay	Off Santa Rosa	30.41082	-86.20790	11/5/2017
	PCMP	FL	Panama City	Municipal Pier	30.15068	-85.66325	11/6/2017
	SAWB	FL	St. Andrew Bay	Watson Bayou	30.14182	-85.63490	11/6/2017
•	APDB	FL	Apalachicola Bay	Dry Bar	29.67577	-85.06227	11/6/2017
•	APCP	FL	Apalachicola Bay	Cat Point Bar	29.72392	-84.88654	11/6/2017
	AESP	FL	Apalachee Bay	Spring Creek	30.07753	-84.32909	11/7/2017
	SRWP	FL	Suwannee River	West Pass	29.30493	-83.18273	11/7/2017
	CKBP	FL	Cedar Key	Black Point	29.20700	-83.06907	11/8/2017
	TBNP	FL	Tampa Bay	Navarez Park	27.78713	-82.75398	11/9/2017
	TBOT	FL	Tampa Bay	Old Tampa Bay	28.02457	-82.63240	11/8/2017
•	TBPB	FL	Tampa Bay	Papys Bayou	27.84438	-82.61115	11/8/2017
	TBKA	FL	Tampa Bay	Peter O. Knight Airport	27.90870	-82.45378	11/8/2017
	TBHB	FL	Tampa Bay	Hillsborough Bay	27.86017	-82.38662	11/13/2017
	TBCB	FL	Tampa Bay	Cockroach Bay	27.67917	-82.52023	11/13/2017
•	CBBI	FL	Charlotte Harbor	Bird Island	26.51390	-82.03485	11/9/2017
•	CBFM	FL	Charlotte Harbor	Fort Meyers	26.55887	-81.92340	11/9/2017
	NBNB	FL	Naples Bay	Naples Bay	26.11187	-81.78480	11/10/2017
	RBHC	FL	Rookery Bay	Henderson Creek	26.02628	-81.73547	11/10/2017
•	EVFU	FL	Everglades	Faka Union Bay	25.90153	-81.51255	11/11/2017
•	FBFO	FL	Florida Bay	Flamingo	25.14110	-80.92317	11/12/2017

2.2 Study Area

The Gulf of Mexico region along the United States includes marine waters along the Southern United States and encompasses coastal habitats in Texas, Louisiana, Mississippi, Alabama, and Florida. When compared to other regional estuarine groupings, the average Gulf of Mexico estuary is distinguished by shallower water depth with shorter estuarine residence times, includes more wetland acreage per open water area, greater fisheries yield per area wetland, smaller tidal range, and higher sedimentation rates (Turner, 2001). Some Gulf of Mexico estuaries have flora and fauna not usually found in most other U.S. estuaries (e.g., manatees and mangroves) (Zimmerman et al., 2002). The large wetland-estuarine systems along the northern coast of the Gulf of Mexico are the result of continuous transport and deposition of riverine and marine sediments in an area with a low to moderate wave energy and low tidal range (generally less than 1.0 m) (Ellis and Smith, 2021; Mata et al., 2011).

The Gulf Coast is made of many inlets, bays, and lagoons. The coast is intersected by numerous rivers, the largest of which is the Mississippi River. Much of the land along the Gulf Coast is, or was, marshland. Forming a crescent along the Gulf Coast is the Gulf Coastal Plain, which reaches from Southern Texas to the western Florida Panhandle, while the western portions of the Gulf Coast are made up of many barrier islands and peninsulas, including the 130-mile (210 km) Padre Island along the Texas coast. These landforms protect the numerous bays and inlets providing as a barrier to oncoming waves but also serve to entrain sediments from upland areas. The central part of the Gulf Coast, from eastern Texas through Louisiana, consists primarily of marshland. The eastern part of the Gulf Coast, predominantly Florida, is dotted with many bays and inlets.

The Gulf Coast climate is considered a humid subtropical habitat, although the southern-most area of Florida does resemble a tropical climate. This region is vulnerable to extreme weather events including hurricanes and severe thunderstorms. These events help define the summer months where precipitation is greatest. July or August are commonly the wettest months of the year due to the combination of frequent summer thunderstorms, and tropical weather systems (tropical depressions, tropical storms and hurricanes), Tornadoes are infrequent along this coast but do occur; however, they occur more frequently in inland portions of Gulf Coast states.

The coastal areas along the Gulf of Mexico Coast are a major center of economic activity. The marshlands along the Louisiana and Texas provide breeding grounds and nurseries for marine life that drive the fishing and shrimping industries. The Apalachicola, Grand Bay, and Weeks Bay NERRs are of particular interest because of their economic and ecologic importance in the northern Gulf of Mexico (Passeri et al. 2016). Each estuary has its own unique morphology and hydrodynamic influences. Apalachicola is a wide, shallow estuary located within the Florida Panhandle. It is the second largest watershed system in the northern Gulf of Mexico surpassed only by the Mobile River basin (Isphording, 1985). The Apalachicola River discharges into East Bay through a delta and distributary system nearly three km wide. The estuary is sheltered from the Gulf of Mexico by a chain of barrier islands. Apalachicola is an ecologically and economically significant estuary that contains oyster reefs, seagrass beds, and salt marshes. Oysters, shrimp, blue crab, and finfish are the most harvested species with a value over $134 million in economic impact annually. In addition, Apalachicola Bay provides approximately 90% of Florida's oyster harvest and 10% of the total U.S. harvest (FDEP, 2013)

The regional economy is also dominated by industries related to energy, petrochemical and tourism sectors. The large cities of the region are (from west to east) Brownsville, Corpus Christi, Houston, Galveston, Beaumont, Lake Charles, Lafayette, Baton Rouge, New Orleans, Gulfport, Biloxi, Mobile, Pensacola, St. Petersburg, Tampa, and increasingly, Sarasota. All of these cities are considered as

metropolitan areas and most contain large ports (https://worldpopulationreview.com/state-rankings/gulf-states). The Port of South Louisiana (Metropolitan New Orleans in Laplace) and the Port of Houston are two of the ten busiest ports in the world by cargo volume. As of 2004, seven of the top ten busiest ports in the U.S. are on the Gulf Coast (https://www.iwr.usace.army.mil/).

The discovery of oil and gas deposits along the coast and offshore, combined with easy access to shipping, have made the Gulf of Mexico the heart of the U.S. petrochemical industry. There are approximately 4,000 oil platforms found in this region. This region also features other important industries including aerospace and biomedical research sectors, as well as established industries such as agriculture. For these reasons, the vibrant Gulf of Mexico coastal economy and population can impact coastal habitat quality.

2.3 Data Analysis

This report contains the analysis of the most recent MWP sampling data in the Gulf of Mexico (2017) and compares them to existing NS&T MWP data (1986-2012). Data management and analysis were conducted using a combination of R version 3.4.4 (R Core Team, 2013), Microsoft Excel (2016), JMP® version 12 (SAS Institute Inc., Cary, NC, 1989-2019) and ArcGIS (ESRI, 2011).

Concentrations were blank corrected by subtracting the method blank from the sample concentrations to correct for signal originating from the reagent or solvents. Concentration values for individual contaminants that were below the method detection limit (MDL) were qualified as undetected and were assigned a value of zero. This only applied to tin, where all but one site were below the MDL.

Individual site concentrations from 2017 were compared to boxplots and the median of their historic site concentrations. The 39 sites sampled in 2017 were analyzed for site-based trends and a regional trend based on yearly medians using Spearman Rank Correlations. Significance was determined as p-value <0.05.

To bring further perspective to the 2017 data, it was compared to previously existing MWP data. The NS&T data used for comparison comprise oyster tissue from the species *Crassostrea virginica* sample concentrations collected by the national MWP along the Southeast and Gulf coasts since the initiation of the program (1565 individual samples across 129 sites collected over 30 years) (Figure 3). The data were clustered into three groups using the Ward Hierarchical Cluster Analysis. The 2017 data were then compared to these groups to see how it related to the lowest, middle and highest concentrations ever detected by the MWP.

3.0 CONTAMINANTS

3.1 Background Information

The Mussel Watch Program monitors nearly 600 contaminants including metals, organic compounds and contaminants of emerging concern (CECs). Of the 22 metals analyzed by the MWP, a subset of eight metals have been selected for this report. There are three principal reasons for this, 1) the availability of historical MWP data for the metals, 2) several of these elements are considered to be abundant “earth metals” and 3) the current state of science and associated methods are less certain of guaranteeing accurate and precise quantitation of several metals. For example, aluminum (Al), iron (Fe), silicon (Si) and manganese (Mn) are all abundant earth metals. As such, the overriding signal for these chemicals tends to be a direct correlation to local earth crustal composition. Chromium (Cr), antimony (Sb), silver (Ag) and thallium (Tl) can be counted among those difficult to quantify.

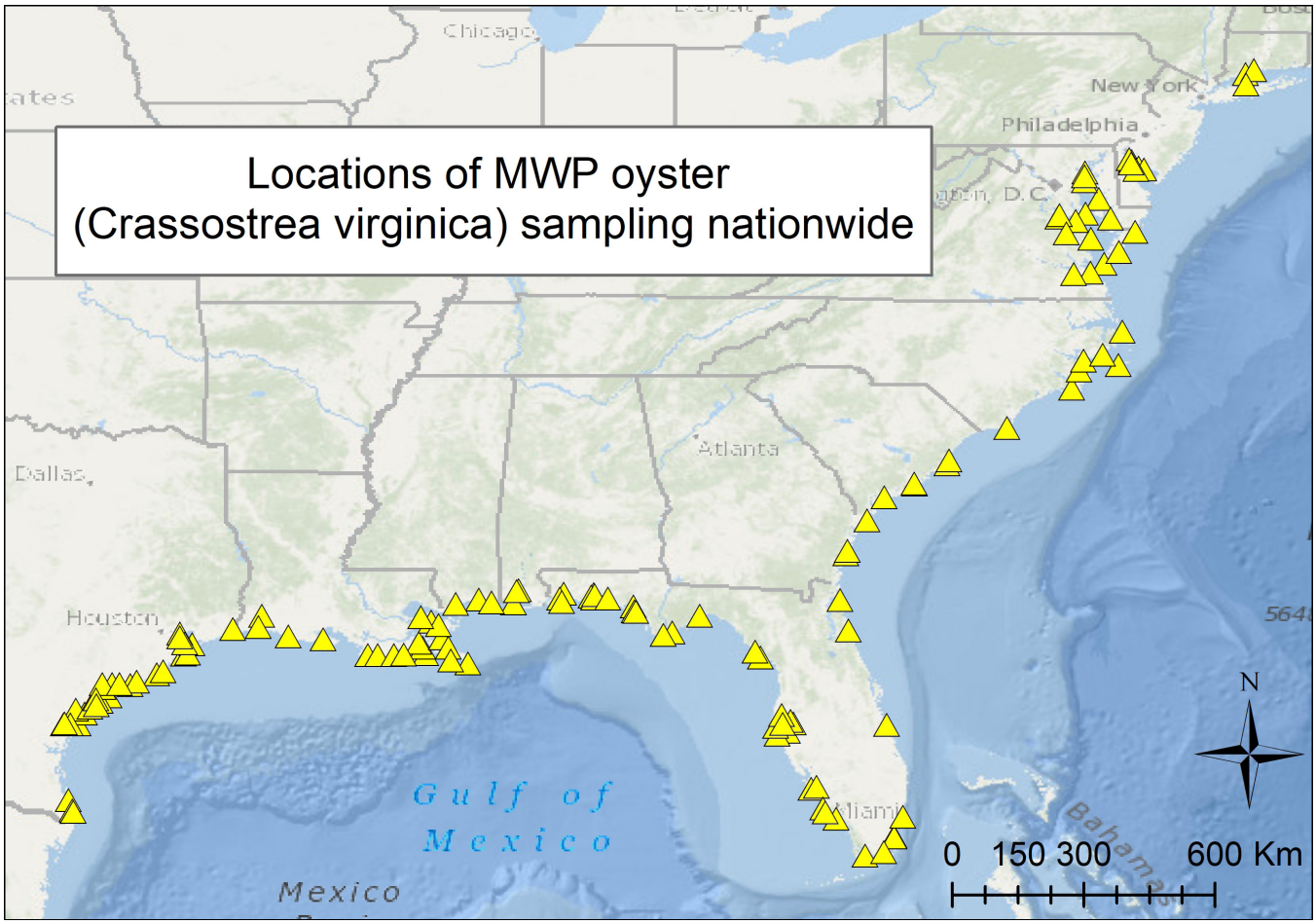


Figure 3. The 129 nationwide Mussel Watch Program sites where *Crassostrea virginica* have been collected.

Moreover, thallium is generally found in such low concentrations that our ability to detect its mere presence is restricted. The eight metals presented in this report can be found in Table 2.

3.1.1 Environmental Sources and Transport

Metals occur naturally in the environment, but human use (anthropogenic) of metal products, particularly since the industrial age, has resulted in excessive releases. Chemical contaminants enter the environment through point or non-point sources. Point source pollution, such as industrial and municipal effluents from a pipe or smokestack, are more easily regulated. In contrast, pollution from non-point sources are diffuse releases of chemicals to the environment such as runoff from agricultural and urban lawns and volatilization of chemicals from land or water to the atmosphere. As a result, non-point source pollution is difficult to regulate. Anthropogenic sources of metals include fossil fuel and waste burning, mining and ore processing, chemical production, and agriculture. These sources are largely responsible for the elevated environmental concentrations observed in coastal waters. Atmosphere releases can occur from both point and nonpoint anthropogenic sources such as smokestack emission, motor vehicle exhaust, and volatilization of pesticides from soil and plants, as well as natural sources such as volcanic eruptions, and forest fires.

Table 2. Metals currently monitored by the Mussel Watch Program and presented in this report.

SYMBOL	ELEMENT
As	Arsenic
Cd	Cadmium
Cu	Copper
Pb	Lead
Hg	Mercury
Ni	Nickel
Sn	Tin
Zn	Zinc

Once released, a chemical will interact with its environment based upon its unique chemical and physical properties, and the prevailing environmental conditions. Atmospheric transport, in contrast to other forms of chemical transport, results in diffuse regional, intercontinental and global distribution of contaminants, especially for persistent compounds that degrade slowly. Wide dispersion results in ambient levels being found globally. The “grasshopper effect” (global distillation) is a type of atmospheric transport whereby volatile chemicals released to the environment in lower (warmer) latitudes volatilize from land and surface waters and are transported in the atmosphere and redeposited in higher (cooler) latitudes. The process is repeated in “hops” and leads to a net gain in concentration at higher latitudes where these chemicals remain trapped. Through this process, chemicals originating from warmer climates can settle in Arctic environments, far from where they were used or released by human activities.

Transport of metals to coastal and estuarine water occurs primarily from runoff and atmospheric deposition. Point and non-point sources of pollution to streams, rivers and coastal waters have left a legacy of pollution in some areas from industrial discharges, along with agricultural and urban runoff. Contaminants that enter water may become more reactive, attach to suspended particles, settle to the bottom or be taken up by organisms. Resuspension of sediments can reintroduce contaminants to the overlying water column, thereby making sediments both a source and a sink for contaminants. In addition, sediment accumulation is also associated with permanent storage of contaminants.

Some chemicals have been identified as harmful and these chemicals are often regulated by policy or mandate. Over time, one would expect a net decrease in the regulated contaminant (parent compound) and potentially a net increase in transformation or degradation products. These processes can result in dilution and/or concentration of chemicals in specific environmental media, such as water, sediment or biota. Fate and transport processes are briefly summarized here. For a more detailed discussion, see Manahan (2005).

3.1.2 Bioaccumulation and Toxicity

An organism’s behavior and physiology, coupled with a chemical contaminant’s physico-chemical properties and bioavailability, determine which compounds are taken up by an organism and the associated biological effects. Some chemicals may be toxic to an organism while others may simply accumulate in tissue without harm. Metals tend to accumulate in selected tissues such as liver, kidney or bone, while organic contaminants usually accumulate in fat tissues. By the process of biomagnification through trophic level transfer, predators, particularly those at the top of the food chain (including humans), can be exposed to large amounts of contaminants that are accumulated in tissue of their prey. Mussels and oysters accumulate contaminants across their gills and by ingestion of particles. For some metals, like arsenic and cadmium, mussels and oysters do not regulate concentrations in their tissue, but instead respond to changes in their immediate environment.

Metals exist in the environment in several forms and each form may result in varying toxicity. Copper, for example, has the ability to cycle between an oxidized state, Cu(II), and reduced state, Cu(I), and is used by cuproenzymes involved in redox reactions (ASTDR, 2004; Harvey and McArdie, 2008; Stern, 2010). It is this property of copper that also makes it potentially toxic because the transitions between Cu(II) and Cu(I) can result in the generation of superoxide and hydroxyl radicals (ASTDR, 2004; Harvey and McArdie, 2008; Stern, 2010). The analytical methods used by the Mussel Watch Program do not distinguish between these various forms, but instead report values as total metal (aggregation of all species of a metal).

3.2 Arsenic (As)

3.2.1 Chemical Description

Arsenic is a toxic metal that is found in the environment at high levels as a result of natural sources and industrial production. Products that contain arsenic include preserved wood, semiconductors, pesticides, defoliants, pigments, antifouling paints, and veterinary medicines. In the recent past, as much as 90% of arsenic in industrial production was used for wood preservation (ATSDR, 2007a). Atmospheric sources of arsenic include smelting, fossil fuel combustion, power generation, and pesticide application. 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. Less harmful organic forms, such as arsenobetaine, predominate in seafood (Edmonds and Francesconi, 1977, 1988, 1993; Phillips, 1990; US FDA, 1993a). 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 US FDA (US 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 to coastal and estuarine water occurs primarily from river runoff and atmospheric deposition. The major source responsible for apparent elevated levels of arsenic in the Nation is natural crustal rock (Welch et al. 1988). This is important because it affects concentrations on the regional level. Concentrations of arsenic exceeding the current US EPA drinking water standard (10 parts per billion) (US EPA, 1979) have been documented in Interior Alaska, Seward and Kenai peninsulas, Mat-Su Valley, and Anchorage (Athey et al., 2018).

3.2.2 Arsenic Results

<u>2017 Arsenic Data Statistics</u>
- Concentration range: 3.39 – 41.01 µg/dry g
- Mean concentration: 11.52 ± 7.45 (SD) µg/dry g
- Maximum concentration: 41.01 µg/dry g (Tampa Bay Navaez Park – TBNP)

<u>Summary of Arsenic:</u>
There was a total of 12 sites that showed a significant trend of arsenic concentrations over time, with eight of the sites showing an increase over the course of MWP monitoring, and the remaining four displaying a decrease. The increasing trendline over time for arsenic concentration was at the following sites: Galveston Bay Offatts Bayou - GBOB ($p < 0.001$, rho = 0.76), Galveston Bay Yacht Club - GBYC ($p = 0.034$, rho = 0.53), Pensacola Bay Sabine Point - PBSP($p = 0.025$, rho = 0.73), Choctawhatchee Bay Joe's Bayou - CBJB ($p = 0.002$, rho = 0.74), Choctawhatchee Bay Postil Point – CBPP ($p = 0.032$, rho = 0.51), Panama City Municipal Pier – PCMP ($p = 0.013$, rho = 0.72), St. Andrew Bay Watson Bayou – SAWB ($p = 0.005$, rho = 0.67), and Tampa Bay Cockroach Bay – TBCB ($p = 0.001$, rho = 0.76). The following four sites displayed a decreasing trendline over time for arsenic concentration: Mississippi Sound Pascagoula Bay – MSPB ($p = 0.027$, rho = -0.53), Cedar Key Black Point – CKBP ($p = 0.046$, rho = -0.48), Naples Bay Naples Bay – NBNB ($p = 0.015$, rho = -0.58), and Rookery Bay Henderson Creek – RBHC ($p = 0.012$, rho = -0.59) (Figure 5, Appendix 2).

When compared to the historic MWP arsenic concentrations, 65% of the 2017 sites were above their historic MWP medians. There is was no significant regional trend in arsenic concentrations over time (Figure 6, Appendix 3). Tampa Bay Navaez Park – TBNP arsenic concentration for 2017 fell within the highest historic MWP National cluster range (32.60 – 123.00 ug/dry g) (Figure 4). This is among the highest values ever detected for arsenic by the MWP and could suggest a particular point or non-point source of contamination, possibly from industrial or agricultural pollution.

National MWP Comparison

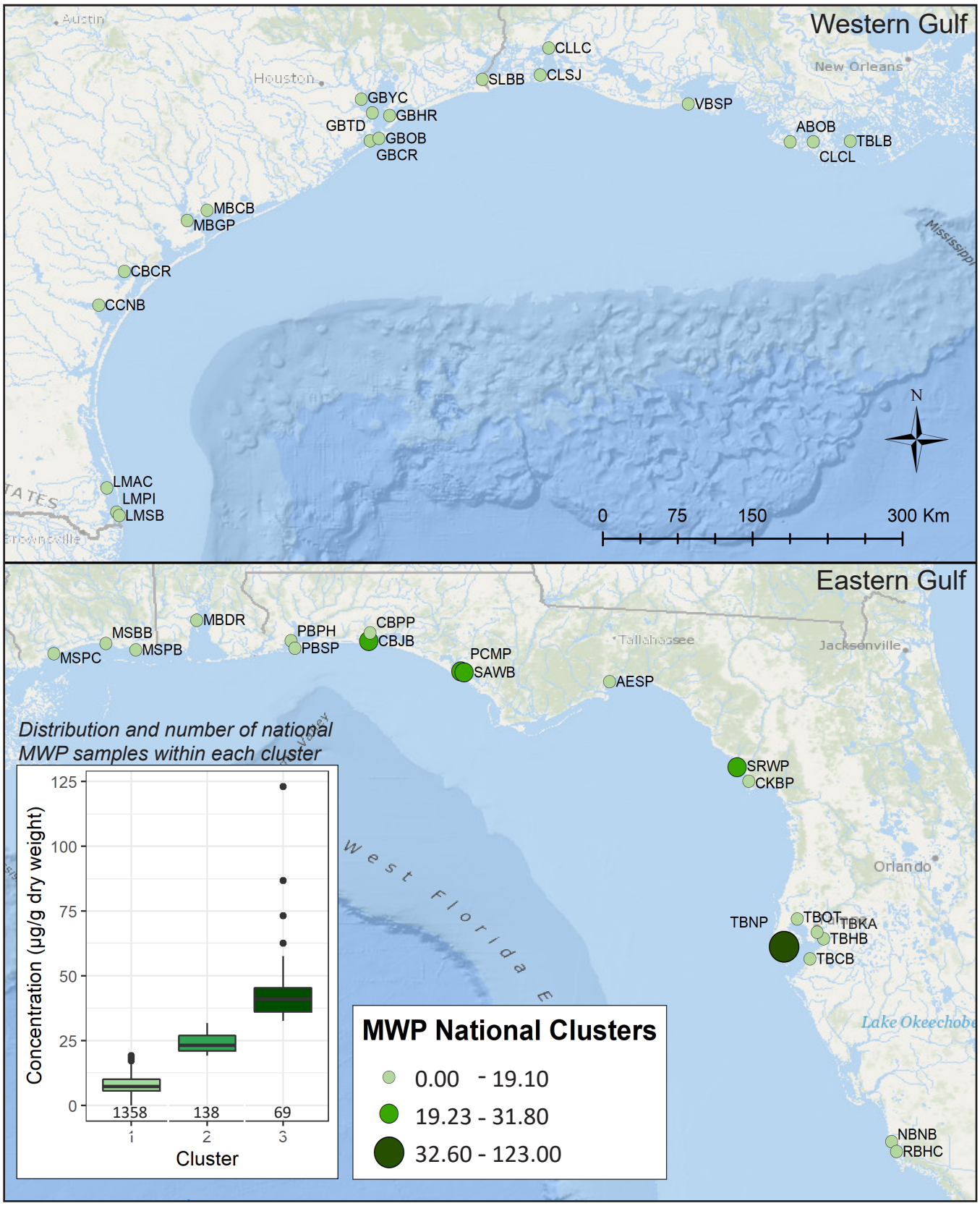


Figure 4. 2017 arsenic concentrations compared to the historic national MWP *Crassostrea virginica* arsenic concentrations (µg/dry g) (129 national sites sampled between 1986-2012 for a total of 1565 samples).

HISTORICAL SITE DATA

65% of sites were above their historic medians

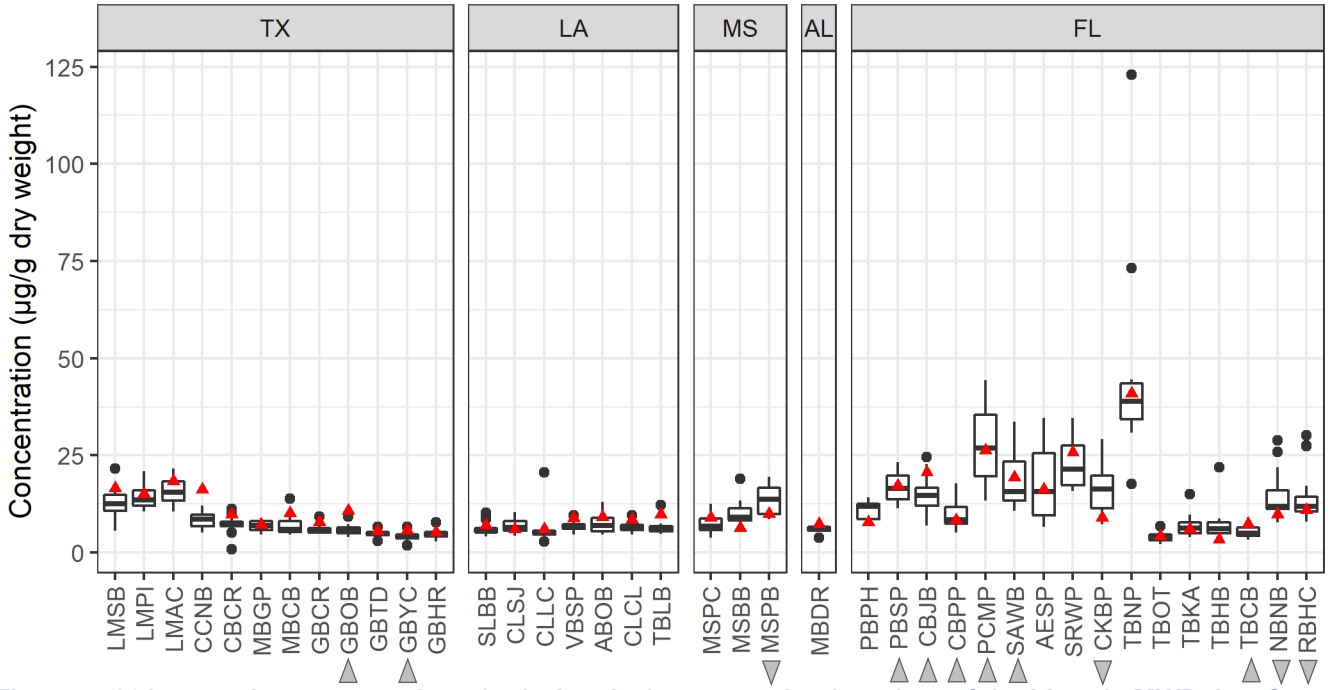


Figure 5. 2017 arsenic concentrations (red triangles) compared to boxplots of the historic MWP data for each site (1986-2012) and the results of the trend analysis showing signifcant increasing or decreasing trends (grey triangles).

REGIONAL TREND ANALYSIS

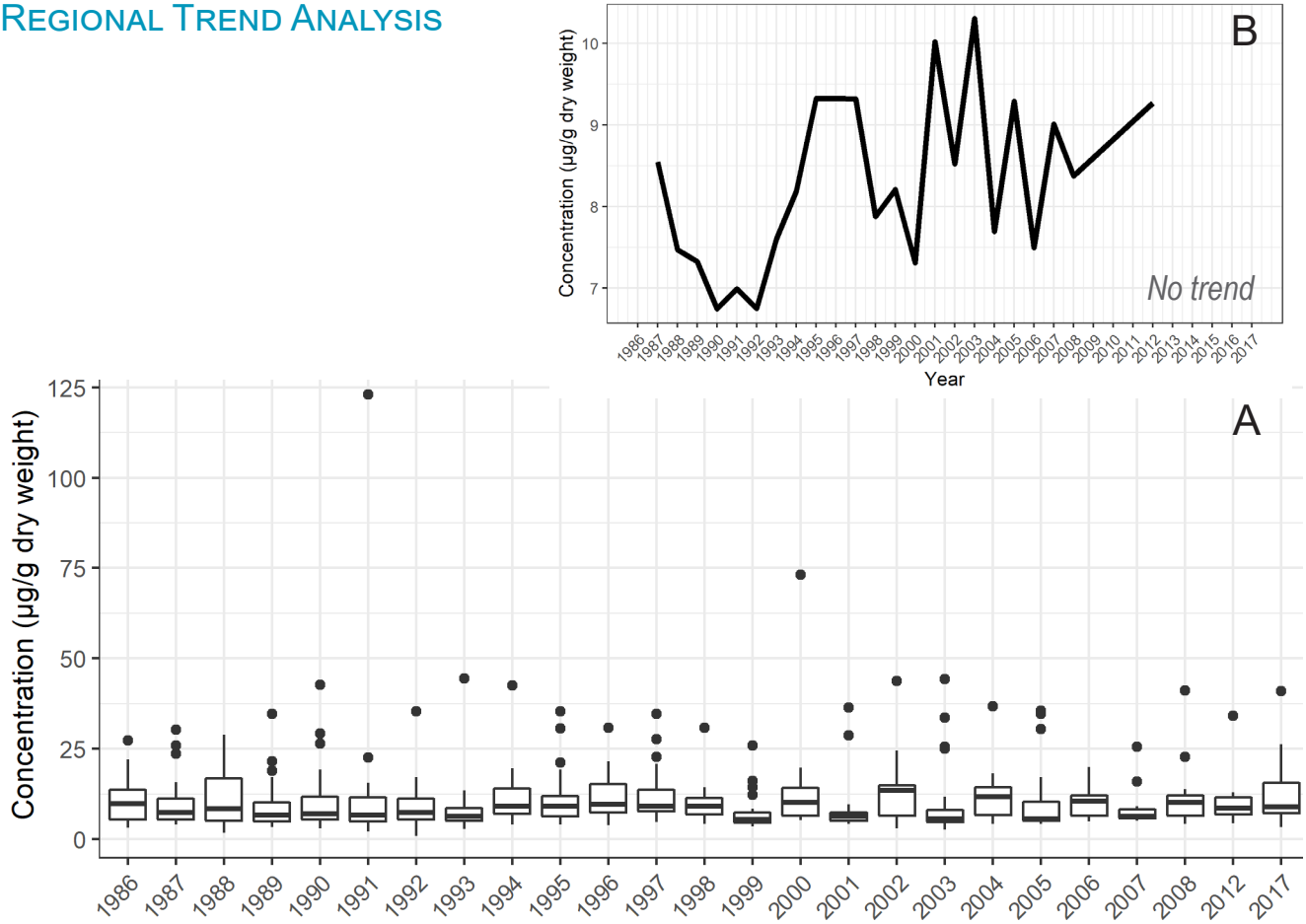


Figure 6. (A) Boxplots representing the historic arsenic concentrations of the 39 site analyzed in this report and (B) the three-point moving average of the yearly median concentrations.

3.3 Cadmium (Cd)

3.3.1 Chemical Description

Cadmium occurs naturally in the Earth's crust as complex oxides and sulfides in ores but is not regarded as an essential element for human life. Environmental contamination of cadmium in coastal and estuarine environments can be linked to both natural and non-point anthropogenic sources (Roesijadi, 1984). 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, 1999a; US FDA 1993b). In addition to abundant industrial applications, other products that contain cadmium include batteries, color pigment, plastics, and phosphate fertilizers. As a result of fossil fuel burning, erosion, and biological activities, cadmium becomes airborne and is transported by atmospheric processes. Land-based runoff and ocean upwelling are the main conveyors of cadmium into coastal environments. Respiration and food represent the two major exposure pathways for humans to cadmium. Exposure to high levels occurs primarily as a result of occupational exposure. Cadmium is toxic to fish, especially salmonoid species and juveniles, and chronic exposure can result in reductions in growth. Safety guidance levels for cadmium in fish and shellfish are no longer listed by the US FDA (US FDA, 2011).

3.3.2 Cadmium Results

<u>2017 Cadmium Data Statistics</u>
- Concentration range: 0.58 - 10.57 µg/dry g
- Mean concentration: 3.27 ± 2.39 (SD) µg/dry g
- Maximum concentration: 10.57 µg/dry g (Sabine Lake Blue Buck Point - SLBB)

<u>Summary of Cadmium:</u>
There was a total of seven sites that showed a significant decreasing trend of cadmium concentrations over the course of MWP monitoring. Lower Laguna Madre South Bay – LMSB ($p = 0.002$, rho = -0.68), Galveston Bay Confederate Reef – GBCR ($p = 0.049$, rho = -0.5), Galveston Bay Todd's Dump – GBTD ($p = 0.022$, rho = -0.56), Atchafalaya Bay Oyster Bayou – ABOB ($p = 0.004$, rho = -0.68), Suwannee River West Pass – SRWP ($p = 0.037$, rho = -0.9), Cedar Key Black Point – CKBP ($p < 0.001$, rho = -0.83), and Naples Bay Naples Bay – NBNB ($p = 0.005$, rho = -0.65) display a decreasing trend (Figure 8, Appendix 2). The 2017 cadmium concentration for all seven sites fall within the lowest historic MWP national cluster range (0.00 – 4.40 ug/dry g) (Figure 7).

There were no regional trends for cadmium (Figure 9, Appendix 3). The presence of cadmium at every site suggests that natural sources are driving concentrations with some more localized influences potentially affecting a small number of sites, such as Galveston Bay Confederate Reef - CBCR, thereby creating the minimal observed site and regional variability.

When compared to the historic MWP cadmium concentrations, 36% of the 2017 sites were above their historic MWP medians (Figure 8).

National MWP Comparison

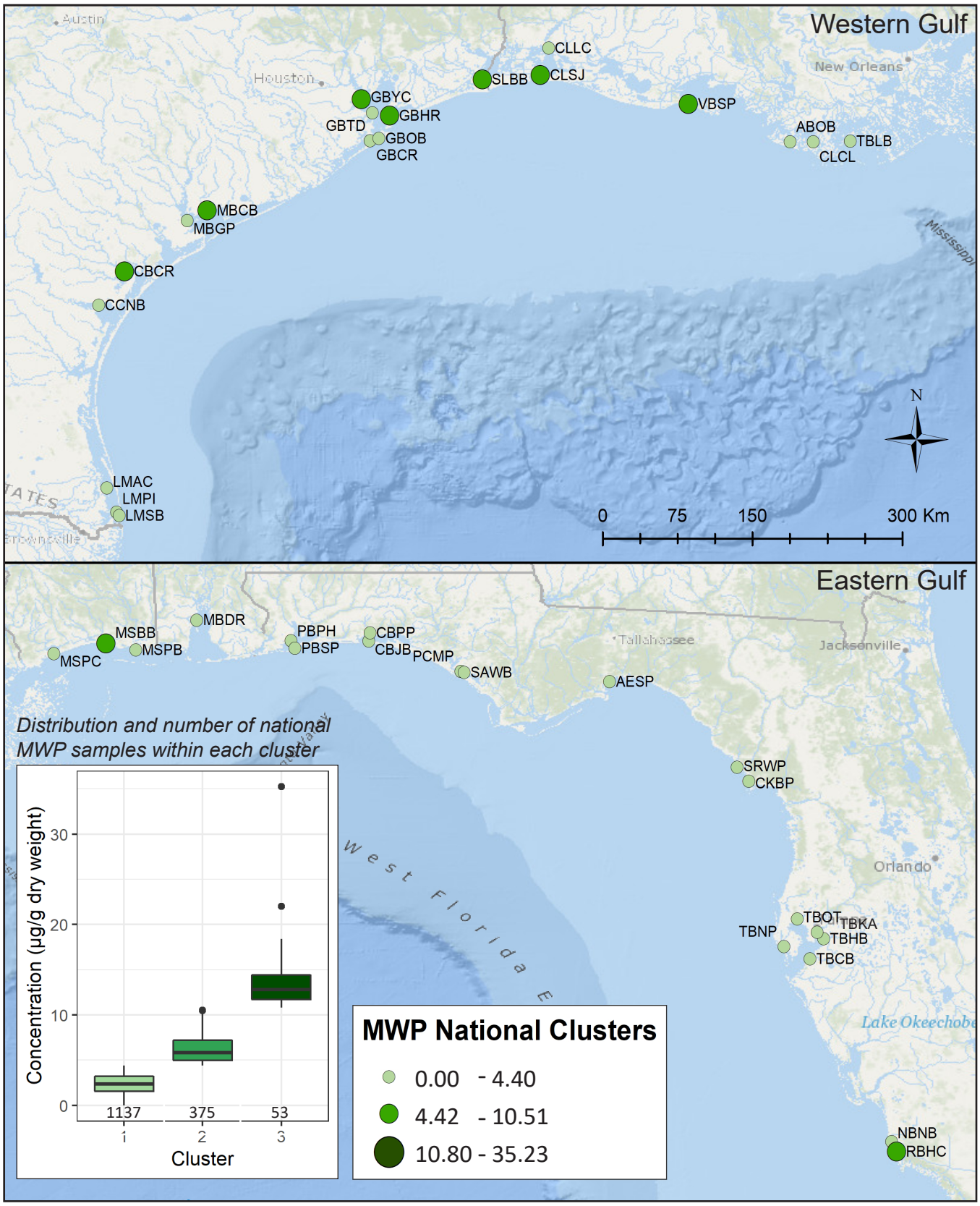


Figure 7. 2017 cadmium concentrations compared to the historic national MWP *Crassostrea virginica* cadmium concentrations (µg/dry g) (129 national sites sampled between 1986-2012 for a total of 1565 samples).

HISTORICAL SITE DATA

36% of sites were above their historic medians

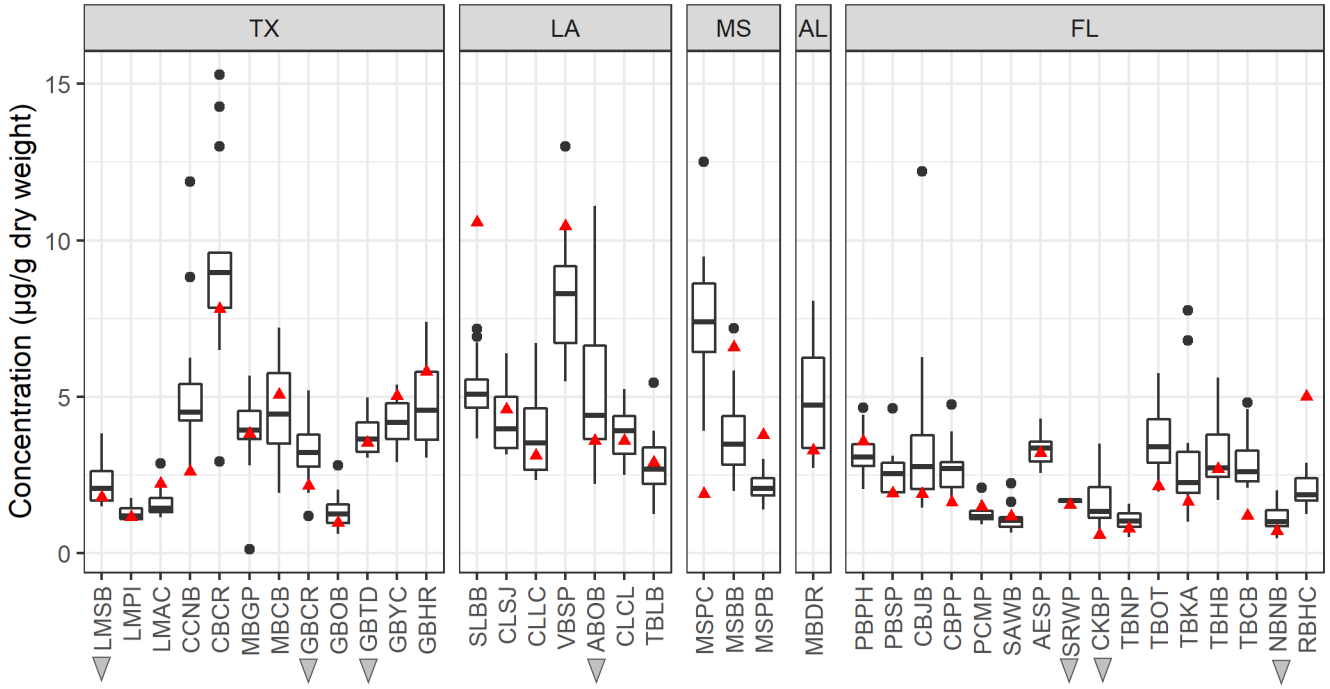


Figure 8. 2017 cadmium concentrations (red triangles) compared to boxplots of the historic MWP data for each site (1986-2012) and the results of the trend analysis showing signifcant increasing or decreasing trends (grey triangles).

REGIONAL TREND ANALYSIS

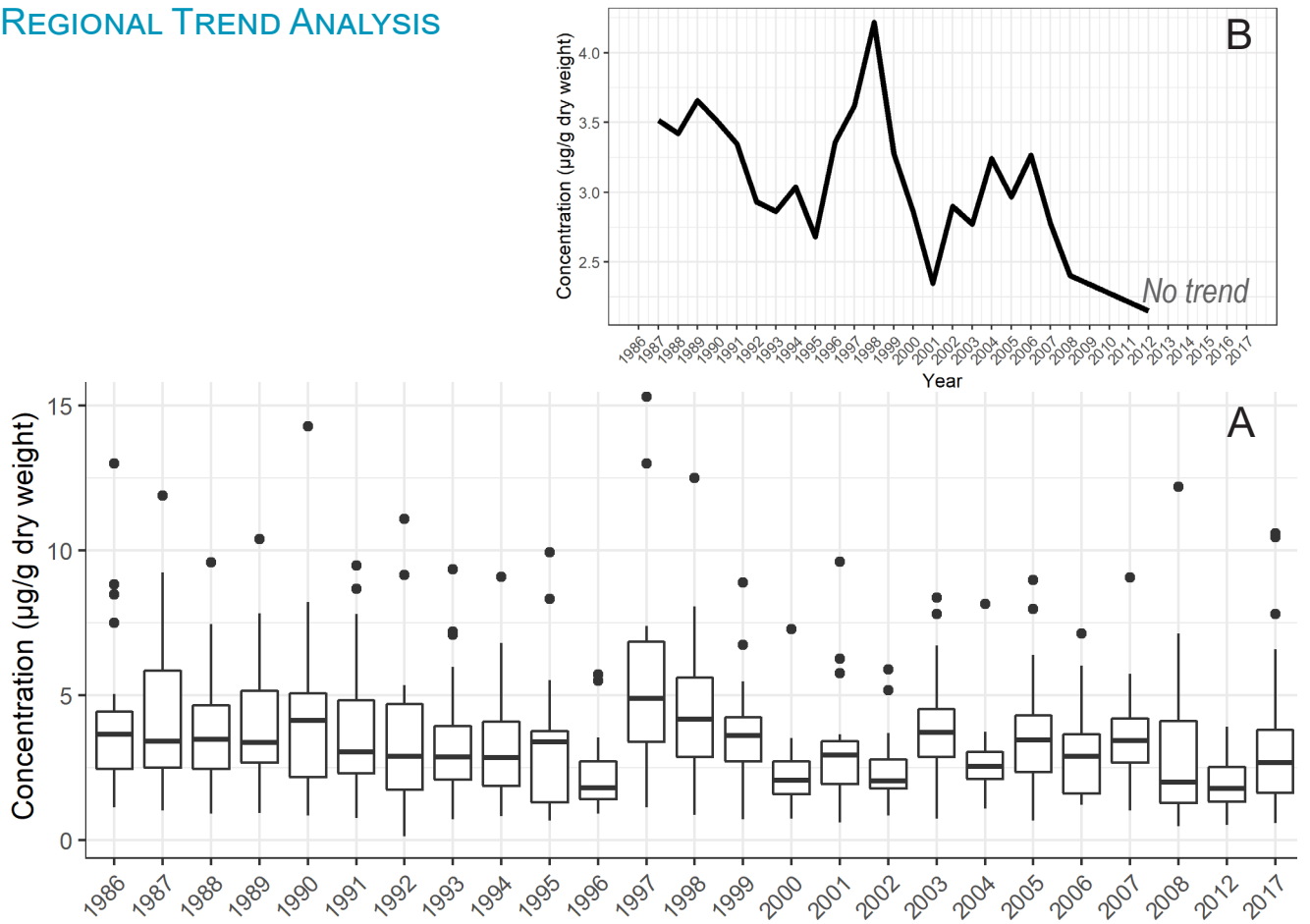


Figure 9. (A) Boxplots representing the historic cadmium concentrations of the 39 site analyzed in this report and (B) the three-point moving average of the yearly median concentrations.

3.4 Copper (Cu)

3.4.1 Chemical Description

Copper is a naturally occurring element that is ubiquitous in the environment. Trace amounts of copper are an essential nutrient for plants and animals. Anthropogenic sources include mining, manufacturing, agriculture, sewage sludge, antifouling paint, fungicides, wood preservatives, and vehicle brake pads. The US 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 paint (US EPA, 1979). Copper can be toxic to aquatic organisms; juvenile fishes and invertebrates are much more sensitive to copper than adults (Mohammed, 2013). Although copper is not highly toxic to humans, chronic effects of copper as a result of prolonged exposure to large doses, can cause damage to the digestive tract and eye irritation (ATSDR, 2004). Excessive exposure to copper has been linked to cellular damage leading to Wilson's disease in humans (Tchounwou et. al., 2008; Stern, 2010). There is no recommended US FDA safety level for copper in fish and fish products. 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 a significant source of copper to coastal waters.

3.4.2 Cadmium Results

<u>2017 Copper Data Statistics</u>
- Concentration range: 25.93 – 1302.78 µg/dry g
- Mean concentration: 342.60 ± 307.85 (SD) µg/dry g
- Maximum concentration: 1302.78 µg/dry g (Vermilion Bay Southwest Pass -VBSP)

<u>Summary of Copper:</u>
There was a total of seven sites that showed a significant increasing trend of copper concentrations over the course of MWP monitoring. Galveston Bay Offatts Bayou – GBOB ($p = 0.050$, $rho = 0.52$), Mississippi Sound Biloxi Bay – MSBB ($p = 0.020$, $rho = 0.60$), Choctawhatchee Bay Joe's Bayou – CBJB ($p = 0.012$, $rho = 0.63$), St. Andrew Bay Watson Bayou – SAWB ($p = 0.044$, $rho = 0.51$), Tampa Bay Navaez Park – TBNP ($p = 0.004$, $rho = 0.70$), Tampa Bay Old Tampa Bay – TBOT ($p = 0.017$, $rho = 0.60$), Tampa Bay Peter O. Knight Airport – TBKA, Tampa Bay Hillsborough Bay – TBHB ($p = 0.004$, $rho = 0.71$), and Naples Bay Naples Bay – NBNB ($p < 0.001$, $rho = 0.74$) all display an increasing trend. Only one site, Atchafalaya Bay Oyster Bayou – ABOB ($p = 0.015$, $rho = -0.60$) displayed a significant decreasing trend for copper concentration over time (Figure 11, Appendix 2).

Copper concentrations showed greater variability within and between sites than arsenic or cadmium. However, like arsenic, the 2017 copper concentration for four of the seven sites (Sabine Lake Blue Buck Point -SLBB, Vermilion Bay Southwest Pass -VBSP, St. Andrew Bay Watson Bayou – SAWB, and Rookery Bay Henderson Creek – RBHC) fall within the highest historic MWP national cluster range (694.00 – 2179.67 ug/dry g) (Figure 10). There were no regional-based trends in copper over time (Figure 12, Appendix 3). When compared to the historic MWP copper concentrations, 79% of the 2017 sites were above their historic MWP medians (Figure 11). There are no US FDA guideline for copper in shellfish. Many of the sites that were sampled in 2017 had increases in their copper concentrations as compared to the historic copper concentrations. Since copper continues to be used in both marine and terrestrial industries, it would be prudent to continue to monitor these concentrations over time.

National MWP Comparison

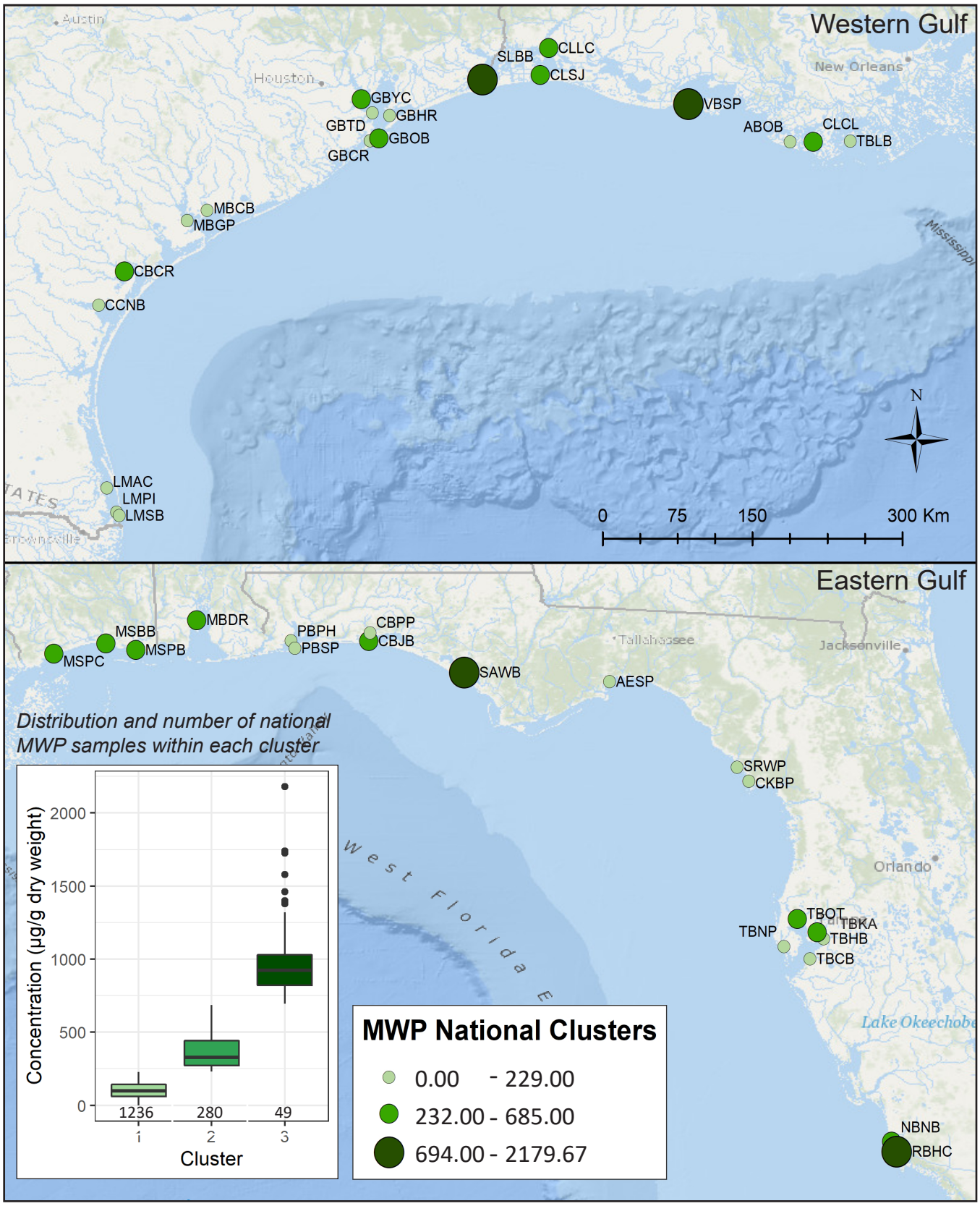


Figure 10. 2017 copper concentrations compared to the historic national MWP *Crassostrea virginica* copper concentrations (µg/dry g) (129 national sites sampled between 1986-2012 for a total of 1565 samples).

HISTORICAL SITE DATA

79% of sites were above their historic medians

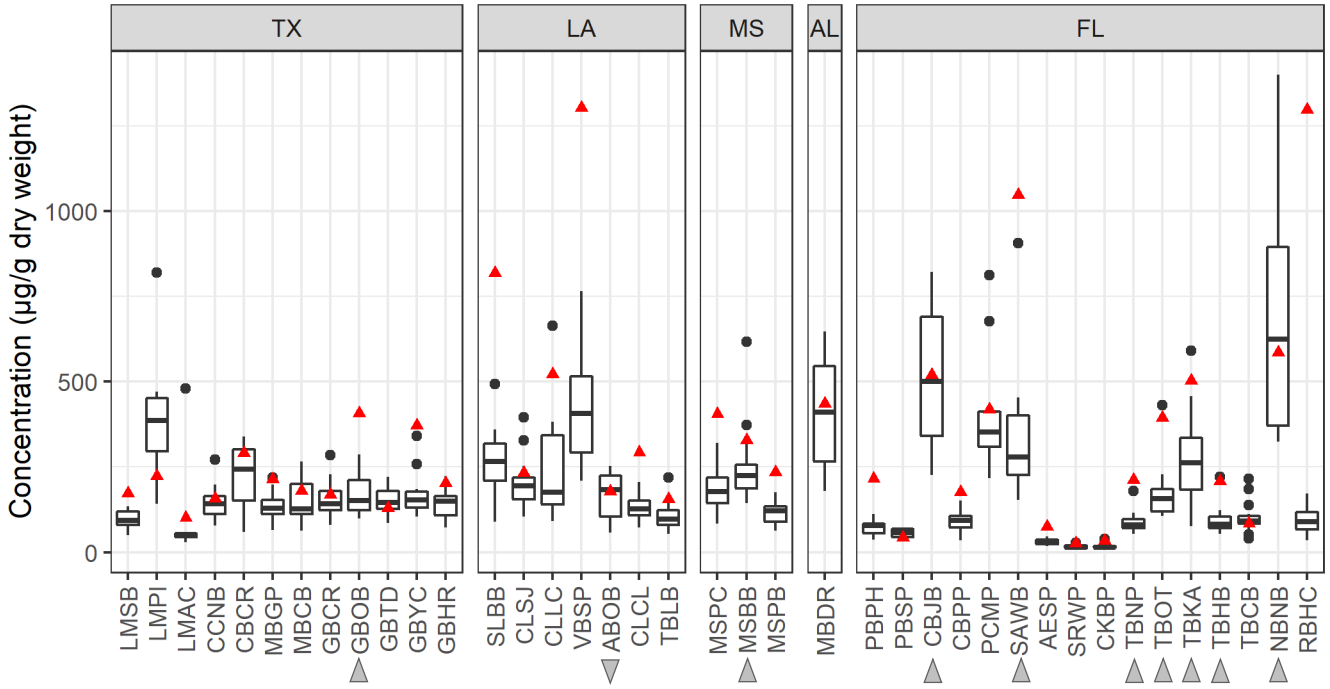


Figure 11. 2017 copper concentrations (red triangles) compared to boxplots of the historic MWP data for each site (1986-2012) and the results of the trend analysis showing signifcant increasing or decreasing trends (grey triangles).

REGIONAL TREND ANALYSIS

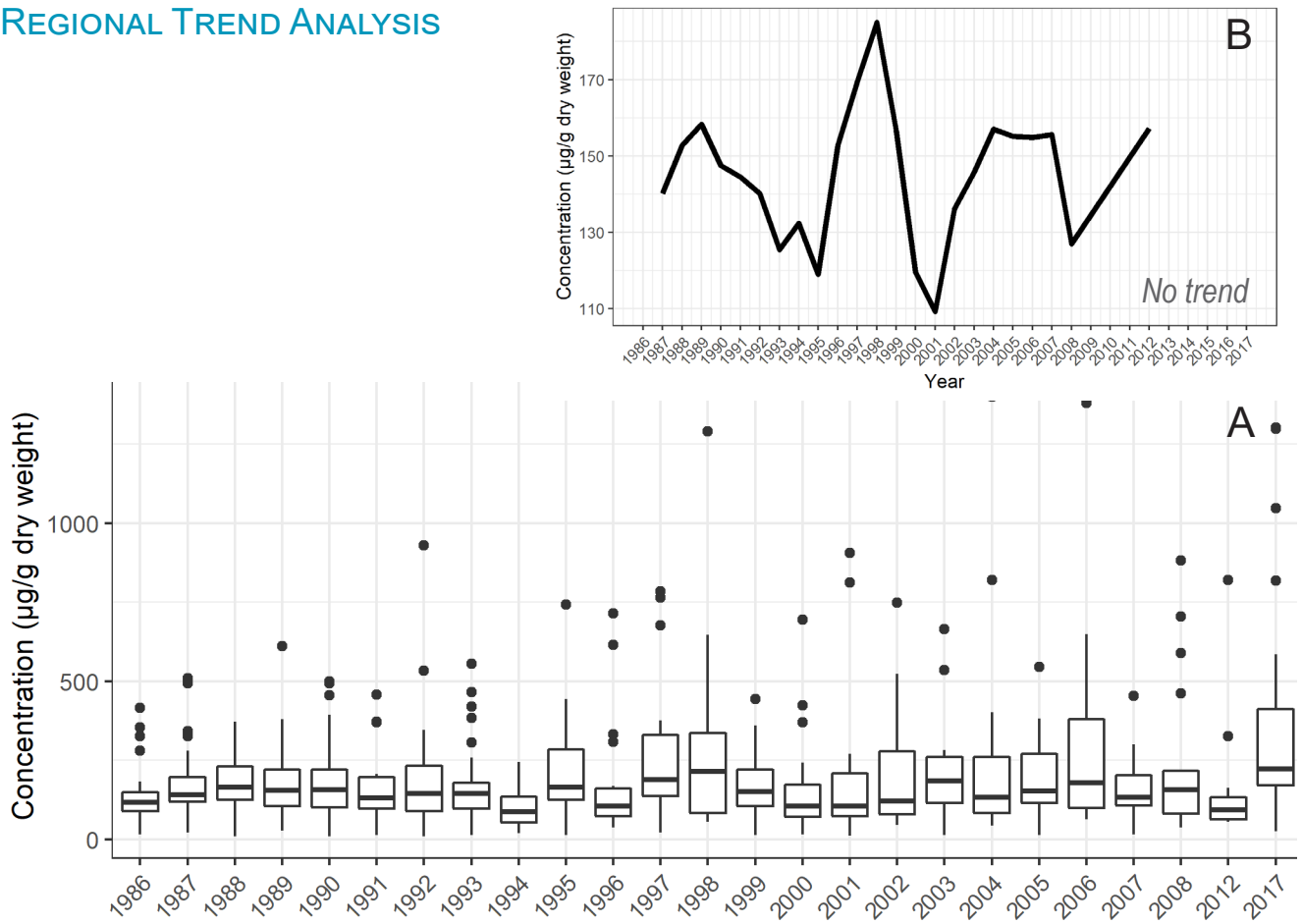


Figure 12. (A) Boxplots representing the historic copper concentrations of the 39 site analyzed in this report and (B) the three-point moving average of the yearly median concentrations.

3.5 Lead (Pb)

3.5.1 Chemical Description

Lead is a ubiquitous metal that occurs naturally in the Earth's crust. Loadings of lead into coastal waters are primarily linked with 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 is not a biologically required micronutrient 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, which results in decreased performance and inhibits typical mental developmental 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 US FDA (US FDA, 2011).

3.5.2 Lead Results

<u>2017 Lead Data Statistics</u>
- Concentration range: 0.32 – 11.16 µg/dry g
- Mean concentration: 1.65 ± 1.83 (SD) µg/dry g
- Maximum concentration: 11.16 µg/dry g (Vermilion Bay Southwest Pass - VBSP)

<u>Summary of Lead:</u>
There was a total of two sites that showed a significant increasing trend of lead concentrations over course of MWP monitoring: Corpus Christi Nueces Bay – CCNB ($p < 0.001$, rho = 0.81), and Galveston Bay Confederate Reef – GBCR ($p = 0.035$, rho = 0.53). Only one site Choctawhatchee Bay Joe's Bayou – CBJB ($p = 0.010$, rho = -0.65) displayed a decreasing trend in lead concentration over time (Figure 14, Appendix 2).

Lead concentrations showed less site-based variability over time than arsenic, cadmium, and copper concentrations. Vermilion Bay Southwest Pass (VBSP) which had the highest individual year concentrations for copper, also had the highest individual year concentration of lead (11.16 µg/dry g, 2017). Furthermore, when compared to the historic MWP lead concentrations, VBSP along with Calcasieu Lake, Lake Charles (CLLC) fell within the highest concentration range (3.73 – 28.90 µg/dry g) (Figure 13). There were no regional trends in lead concentrations over time (Figure 15, Appendix 3). When compared to the historic MWP lead concentrations, 90% of the 2017 sites were above their historic MWP medians (Figure 14).

National MWP Comparison

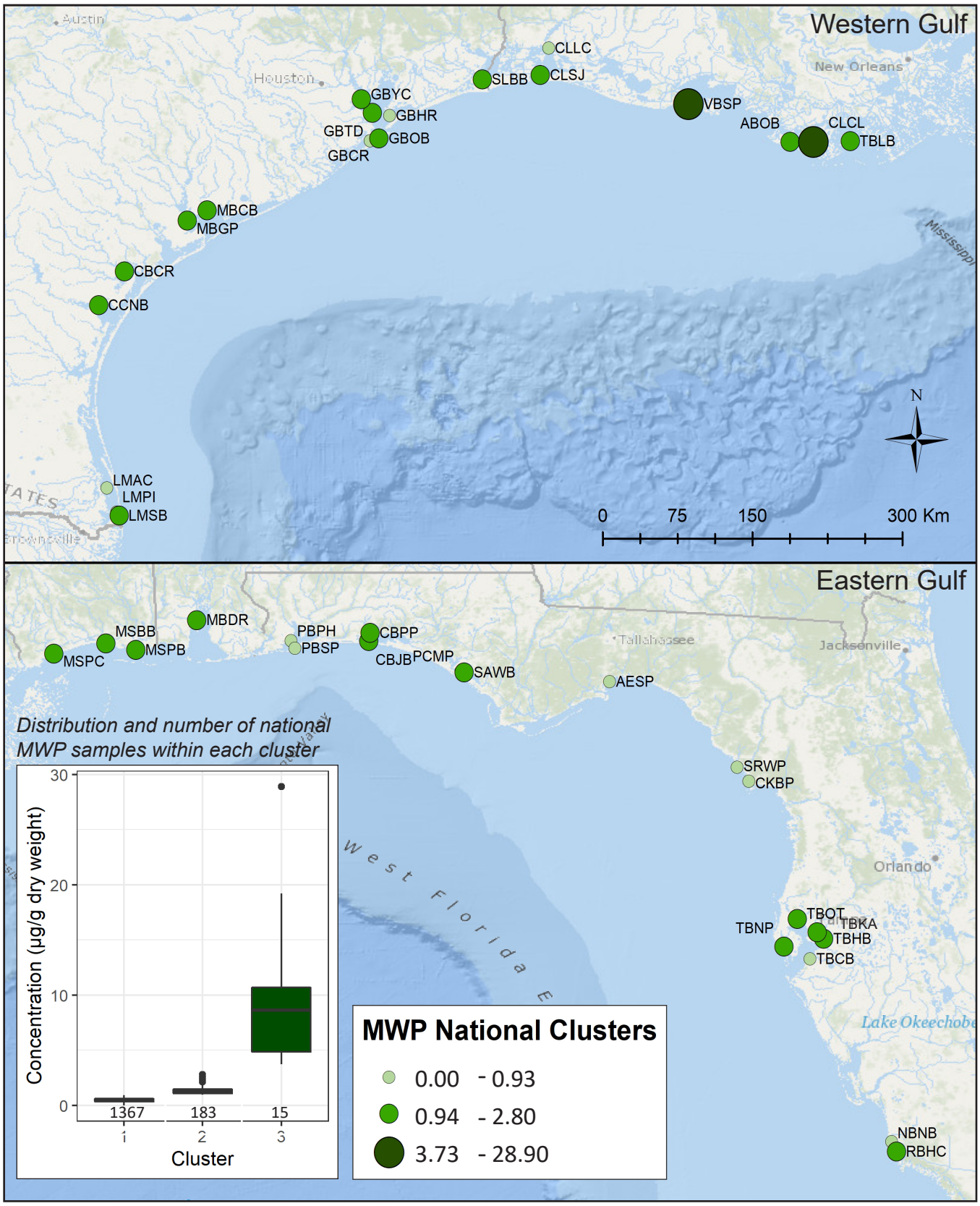


Figure 13. 2017 lead concentrations compared to the historic national MWP *Crassostrea virginica* lead concentrations (µg/dry g) (129 national sites sampled between 1986-2012 for a total of 1565 samples).

HISTORICAL SITE DATA

90% of sites were above their historic medians

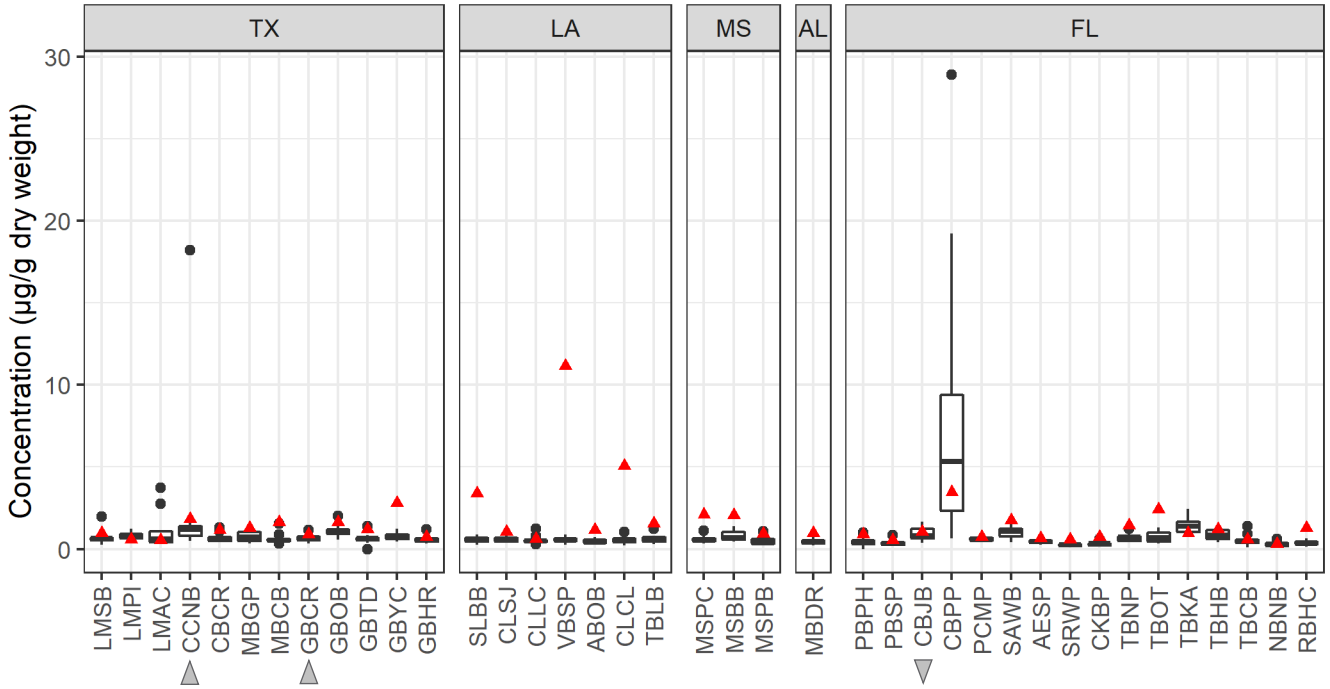


Figure 14. 2017 lead concentrations (red triangles) compared to boxplots of the historic MWP data for each site (1986-2012) and the results of the trend analysis showing signifcant increasing or decreasing trends (grey triangles).

REGIONAL TREND ANALYSIS

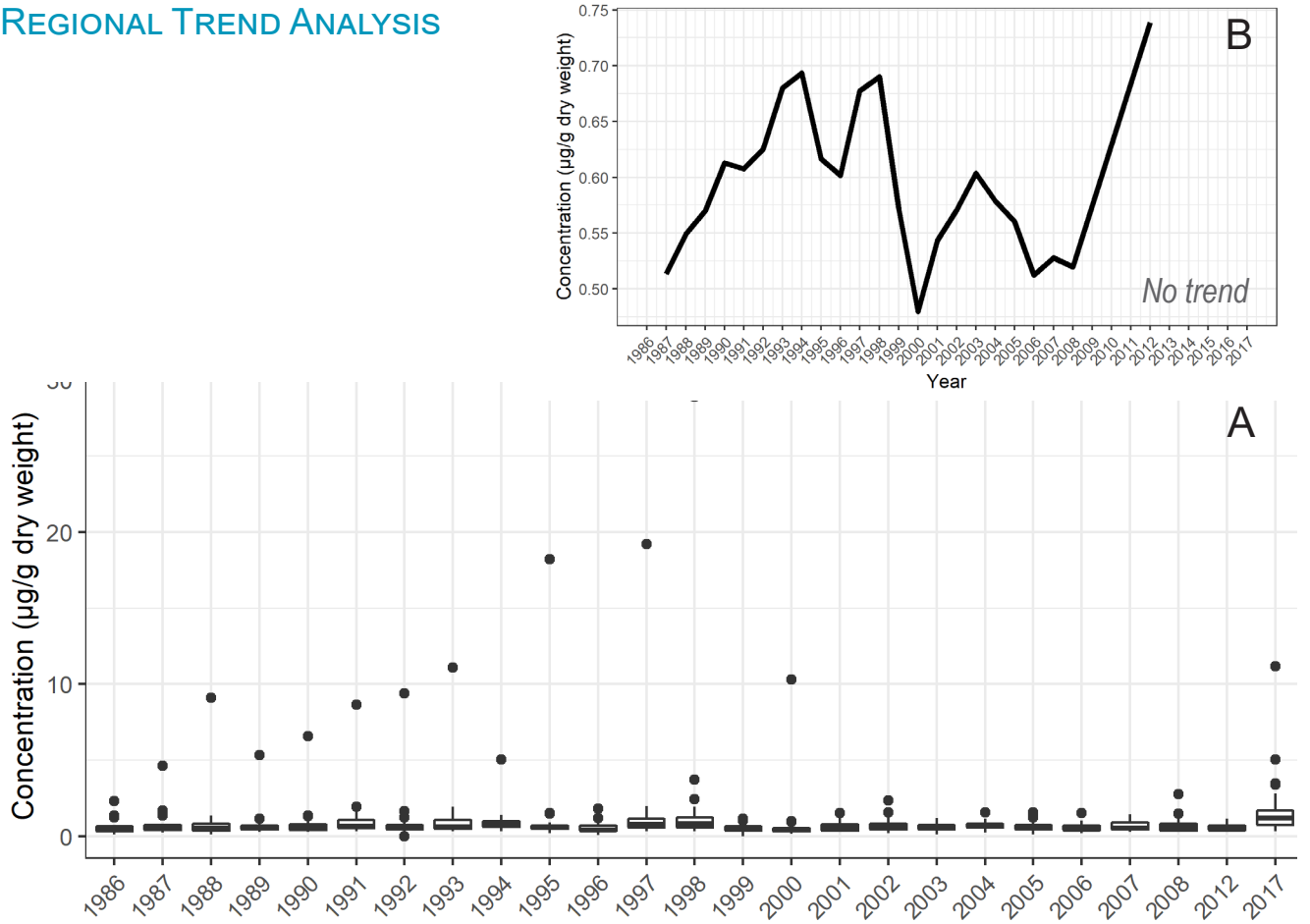


Figure 15. (A) Boxplots representing the historic lead concentrations of the 39 site analyzed in this report and (B) the three-point moving average of the yearly median concentrations.

3.6 Mercury (Hg)

3.6.1 Chemical Description

Mercury is a highly toxic, non-essential trace metal that occurs naturally. Elevated levels occur as a result of human activity (ATSDR, 1999b). In the US, coal fired-electric turbines, municipal and medical waste incinerators, mining, landfills and sewage sludge are the primary emitters of mercury into the air. 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 US 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 animal and humans (US FDA, 2011). The Mussel Watch Program 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 US FDA and US EPA (US EPA, 1979; US FDA, 2011). In the environment, mercury may change forms between elemental, inorganic, and organic. Natural sinks, such as sediment and soil, represent the largest source of mercury to the environment. 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 (NADP, 2020).

3.6.2 Mercury Results

<u>2017 Mercury Data Statistics</u>
- Concentration range: 0.06 – 0.87 µg/dry g
- Mean concentration: 0.17 ± 0.14 (SD) µg/dry g
- Maximum concentration: 0.87 µg/dry g (Rookery Bay Henderson Creek – RBHC)

<u>Summary of Mercury:</u>
Like arsenic, there were more sites displaying a significant decreasing trend of mercury concentrations over the course of MWP monitoring than there were sites displaying an increasing trend. The following six sites showed a decrease in trend for mercury concentration: Galveston Bay Confederate Reef – GBCR ($p = 0.023$, rho = -0.58), Matagorda Bay Carancahua Bay – MBCB ($p = 0.007$, rho = -0.73), Choctawhatchee Bay Joe's Bayou – CBJB ($p < 0.001$, rho = -0.82), Tampa Bay Navaez Park – TBNP ($p = 0.007$, rho = -0.66), Tampa Bay Old Tampa Bay – TBOT ($p = 0.011$, rho = -0.64), and Tampa Bay Cockroach Bay – TBCB ($p = 0.013$, rho = -0.59). While Lower Laguna Madre Port Isabel – LMPI ($p = 0.030$, rho = 0.65) and Calcasieu Lake St. Johns Island – CLSJ both show an increasing trend for mercury concentration over time (Figure 17, Appendix 2).

Unlike the 2017 arsenic, cadmium, copper, and lead concentrations, mercury has ten sites that fell within the highest concentration cluster when compared to the historic MWP mercury concentration (0.17 – 1.55 µg/dry g) (Figure 16). The ten sites are listed as follows: Matagorda Bay Gallinipper Point – MBGP, Sabine Lake Blue Buck Point – SLBB, Calcasieu Lake St. Johns Island – CLSJ, Calcasieu Lake, Lake Charles – CLLC, Mississippi Sound Pass Christian – MSPC, Pensacola Bay Public Harbor – PBPH, Pensacola Bay Sabine Point – PBSP, Choctawhatchee Bay Postil Point – CBPP, Tampa Bay Old Tampa Bay – TBOT, and Tampa Bay Cockroach Bay – TBCB.

There were no regional trends for mercury concentration over time (Figure 18, Appendix 3). When compared to the historic MWP mercury concentrations, 77% of the 2017 sites were above their historic MWP medians (Figure 17).

National MWP Comparison

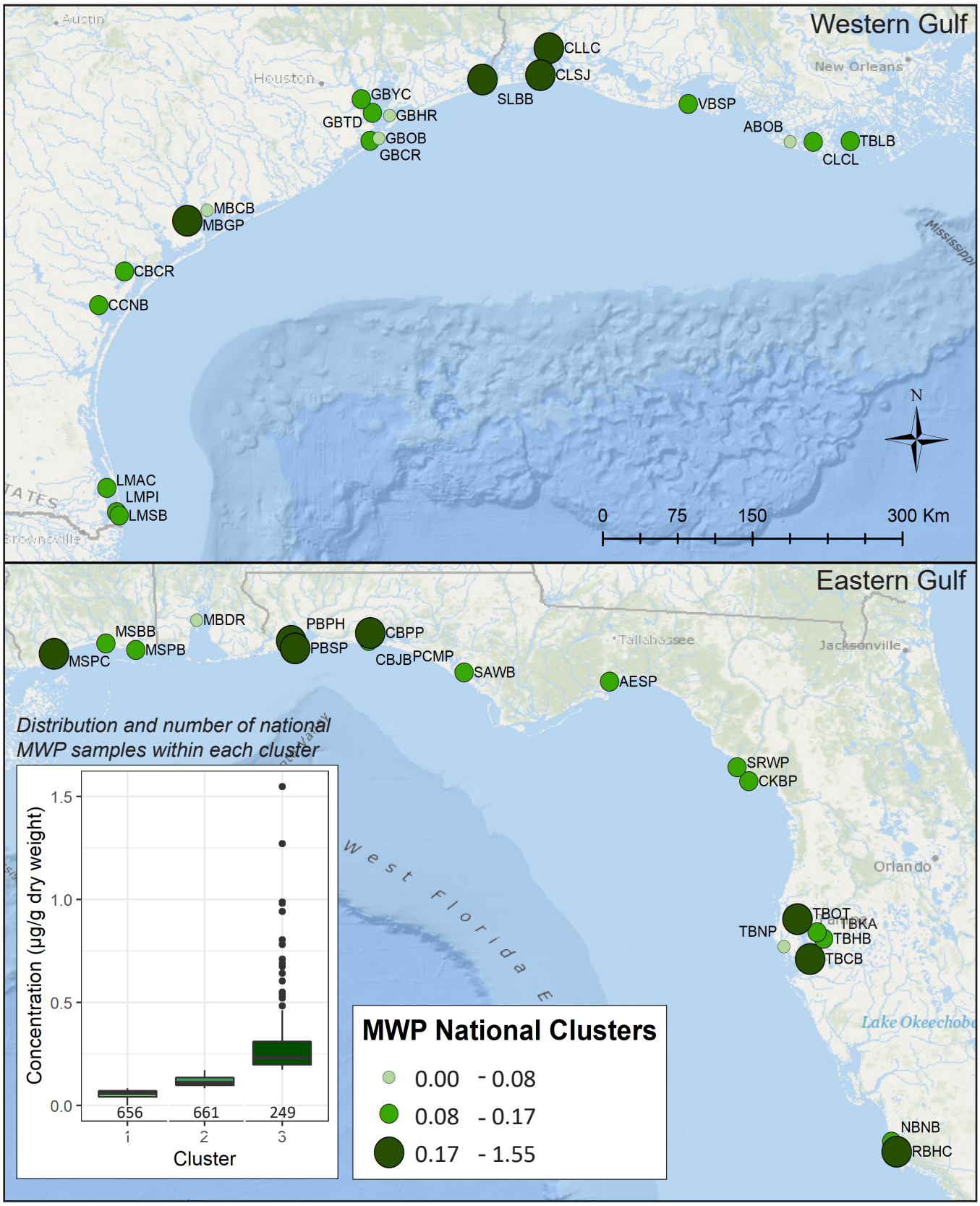


Figure 16. 2017 mercury concentrations compared to the historic national MWP *Crassostrea virginica* mercury concentrations (µg/dry g) (129 national sites sampled between 1986-2012 for a total of 1566 samples).

HISTORICAL SITE DATA

77% of sites were above their historic medians

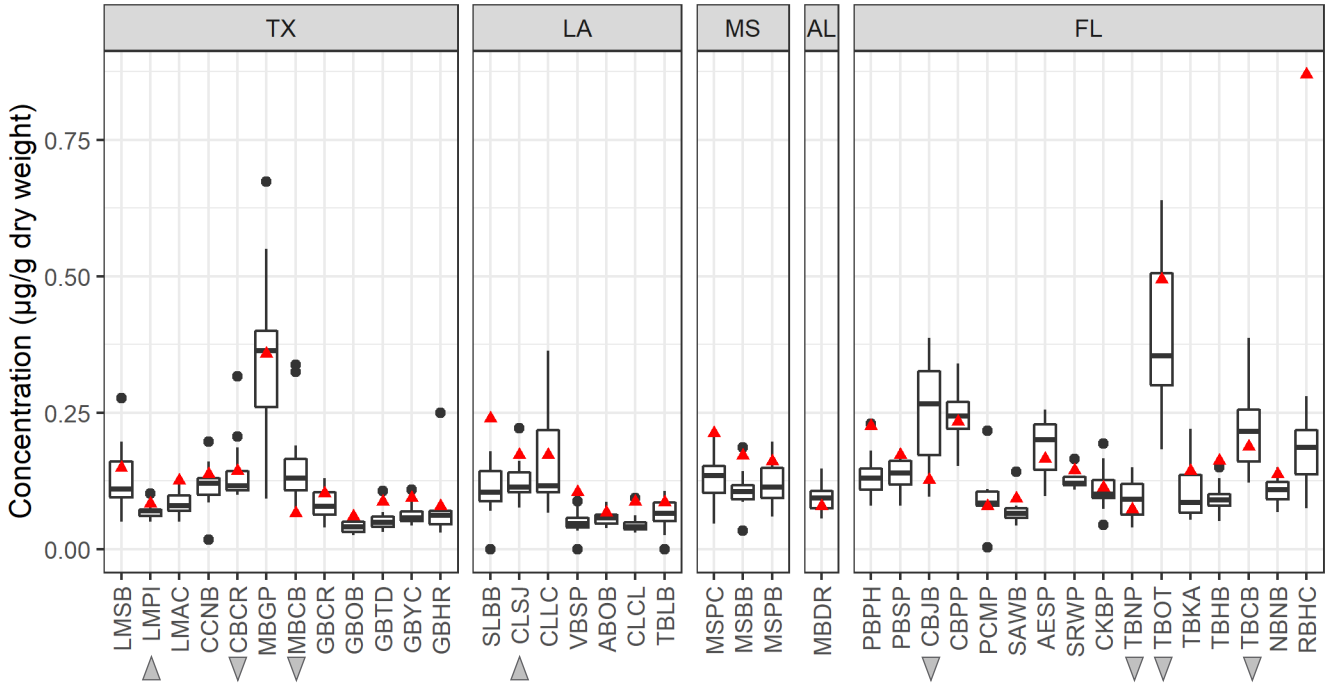


Figure 17. 2017 mercury concentrations (red triangles) compared to boxplots of the historic MWP data for each site (1986-2012) and the results of the trend analysis showing signifcant increasing or decreasing trends (grey triangles).

REGIONAL TREND ANALYSIS

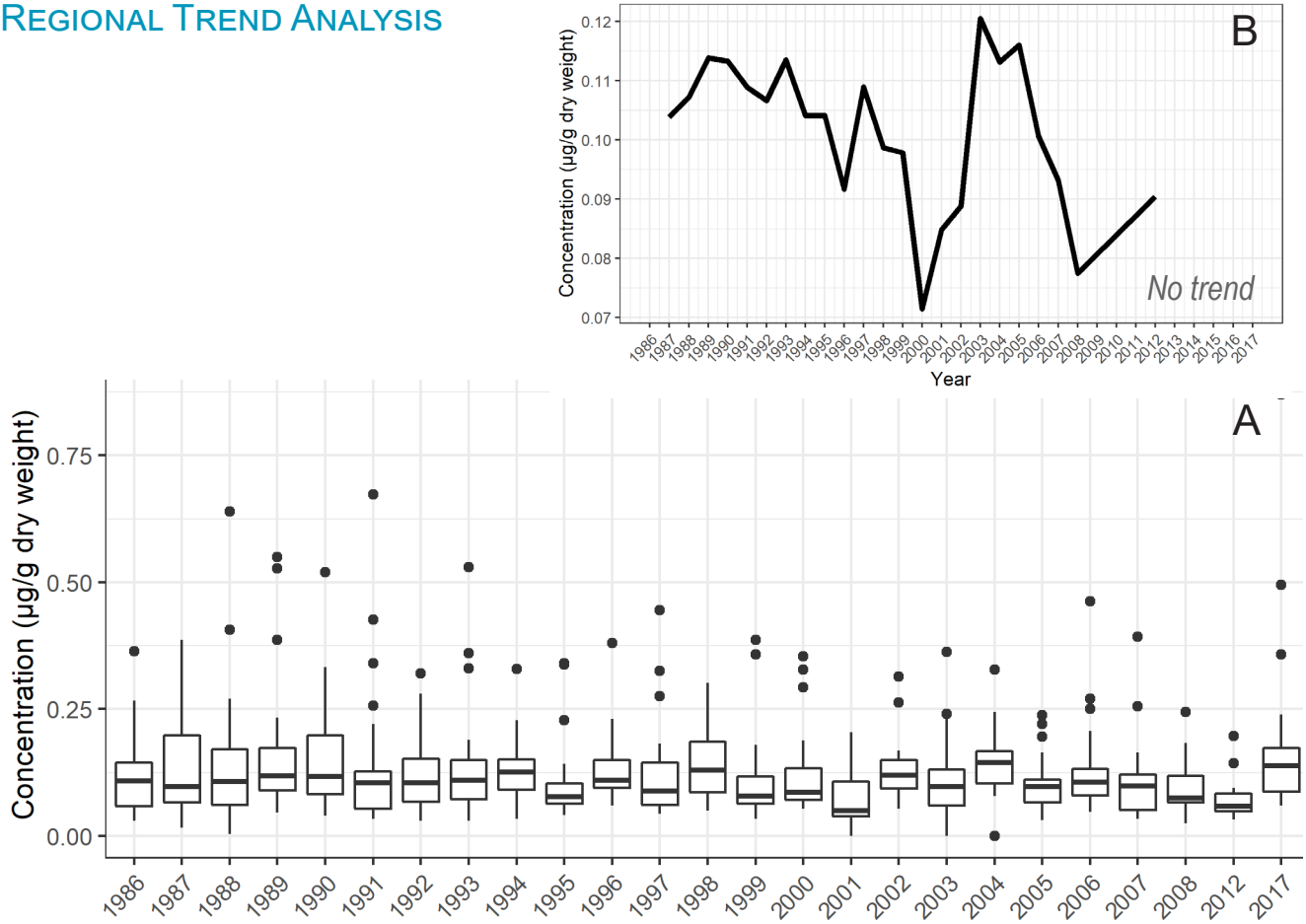


Figure 18. (A) Boxplots representing the historic mercury concentrations of the 39 site analyzed in this report and (B) the three-point moving average of the yearly median concentrations.

3.7 Nickel (Ni)

3.7.1 Chemical Description

Nickel is a naturally occurring, biologically essential trace element that is widely distributed in the environment. It exists in alloy form in combination with other metals and as a soluble element. Nickel is found in stainless steel, nickel-cadmium batteries, pigments, computers, wire, coinage and is used for electroplating (ATSDR, 2005a). 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. River and stream input of nickel are the largest sources for oceans and coastal waters. Atmospheric sources are usually not significant, except in the Great Lakes where the 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 Ni is derived from municipal and industrial wastewaters and thus is likely to be complexed with organic matter (Sweet et al., 1998). 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). Food is the major source of human exposure to nickel (ATSDR, 2005a). Exposure to large doses of nickel can cause serious health effects, such as bronchitis, while long-term exposure can result in cancer. There is no evidence that nickel biomagnifies in the food chain (McGeer et al., 2003; Suedel et al.,1994). Safety guidance levels for nickel in fish and shellfish are no longer listed by the US FDA (US FDA, 2011).

3.7.2 Nickel Results

2017 Nickel Data Statistics
- Concentration range: 1.34 – 16.02 µg/dry g
- Mean concentration: 4.48 ± 3.17 (SD) µg/dry g
- Maximum concentration: 16.02 µg/dry g (Matagorda Bay Gallinipper Point – MBGP)

Summary of Nickel:
Unlike cadmium, there were no sites that display a decreasing trend of nickel concentrations over the course of MWP monitoring. Nickel concentration for the following 12 sites show a significant increase in concentration over time: Lower Laguna Madre South Bay – LMSB ($p = 0.0397$, $rho = 0.4884$), Lower Laguna Madre Port Isabel – LMPI ($p = 0.0021$, $rho = 0.8182$), Matagorda Bay Carancahua Bay – MBCB ($p = 0.0386$, $rho = 0.6014$), Galveston Bay Confederate Reef – GBCR ($p = 0.0134$, $rho = 0.6029$), Galveston Bay Todd's Dump – GBTD ($p = 0.0254$, $rho = 0.5395$), Galveston Bay Yacht Club – GBYC ($p = 0.0134$, $rho = 0.6029$), Galveston Bay Hanna Reef – GBHR ($p = 0.0010$, $rho = 0.7432$), Calcasieu Lake Lake Charles – CLLC ($p = 0.0232$, $rho = 0.5466$), Pensacola Bay Public Harbor – PBPH ($p = 0.0396$, $rho = 0.5357$), Tampa Bay Navaez Park – TBNP ($p = 0.0198$, $rho = 0.5929$), Tampa Bay Peter O. Knight Airport – TBKA ($p = 0.0097$, $rho = 0.6429$), Tampa Bay Cockroach Bay – TBCB ($p = 0.0008$, $rho = 0.7328$) (Figure 20, Appendix 2).

Nickel concentrations in 2017 when compared to the historic MWP nickel concentration resulted in 19 out of 39 sites falling within the highest concentration cluster (3.52 – 12.57 µg/dry g) (Figure 19). The 19 sites are as follows: Copano Bay Copano Reef – CBCR, Matagorda Bay Gallinipper Point – MBGP, Matagorda Bay Carancahua Bay – MBCB, Galveston Bay Confederate Reef – GBCR, Galveston Bay Offatts Bayou – GBOB, Galveston Bay Hanna Reef – GBHR, Galveston Bay Yacht Club – GBYC, Sabine Lake Blue Buck Point – SLBB, Calcasieu Lake St. Johns Island – CLSJ, Calcasieu Lake Lake Charles – CLLC, Vermilion Bay Southwest Pass - VBSP, Atchafalaya Bay Oyster Bayou – ABOB, Caillou Lake Caillou Lake – CLCL, Terrebonne Bay Lake Barre – TBLB, Mississippi Sound Biloxi Bay – MSBB, Mississippi Sound Pascagoula Bay – MSPB, Mobile Bay Dog River – MBDR, St. Andrew Bay Watson Bayou – SAWB, and Tampa Bay Navaez Park – TBNP.

There was so significant regional trend calculated for nickel over time. However, the data, while variable, suggest that nickel concentrations in tissue have been slowly increasing over time, but more data and further analysis are needed to confirm this trend (Figure 21, Appendix 3).

All the sites in 2017 (100%) are above their historic means (Figure 20).

National MWP Comparison

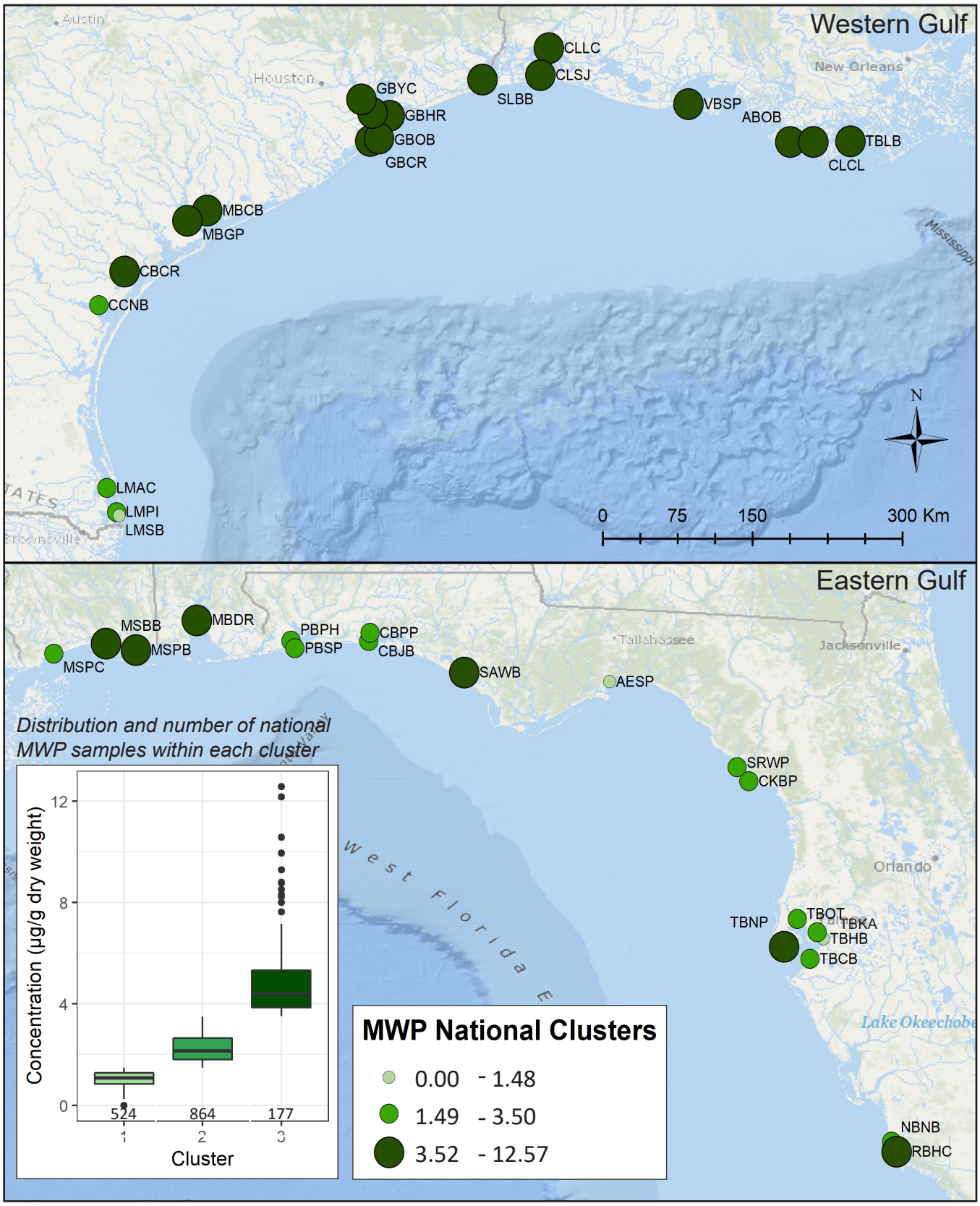


Figure 19. 2017 nickel concentrations compared to the historic national MWP *Crassostrea virginica* nickel concentrations (µg/dry g) (129 national sites sampled between 1986-2012 for a total of 1565 samples).

HISTORICAL SITE DATA

100% of sites were above their historic medians

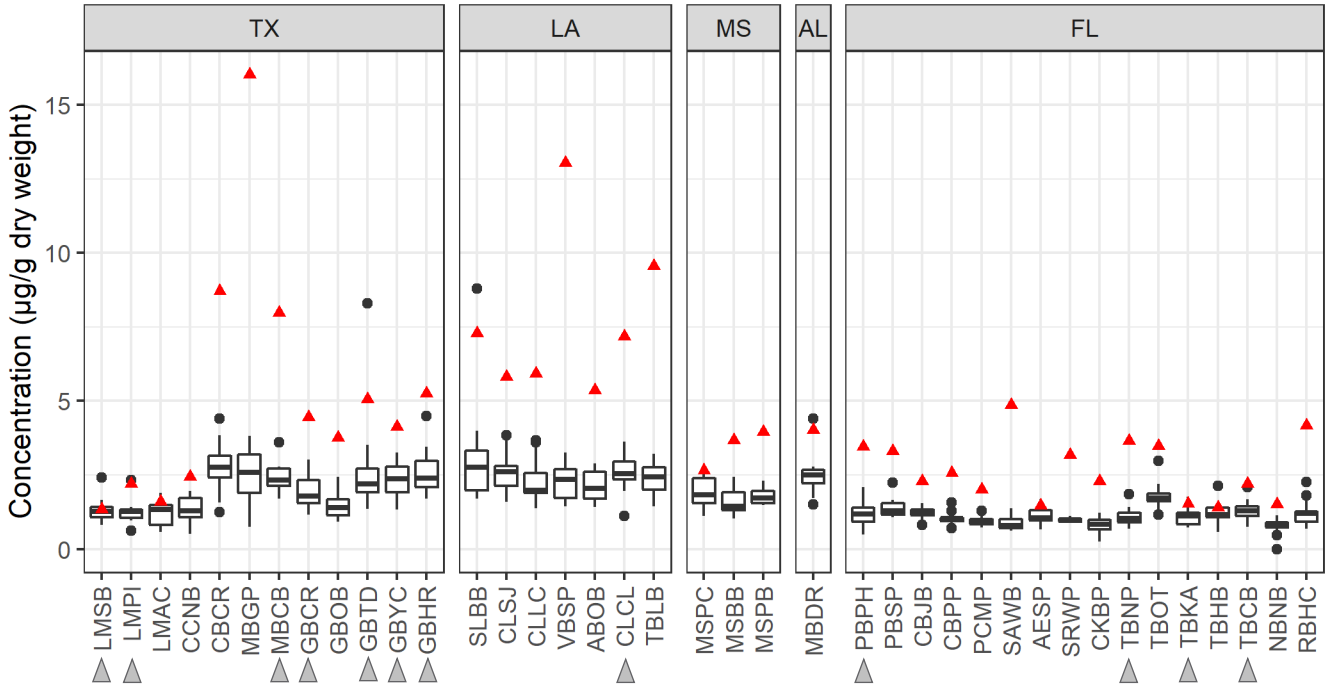


Figure 20. 2017 nickel concentrations (red triangles) compared to boxplots of the historic MWP data for each site (1986-2012) and the results of the trend analysis showing signifcant increasing or decreasing trends (grey triangles).

REGIONAL TREND ANALYSIS

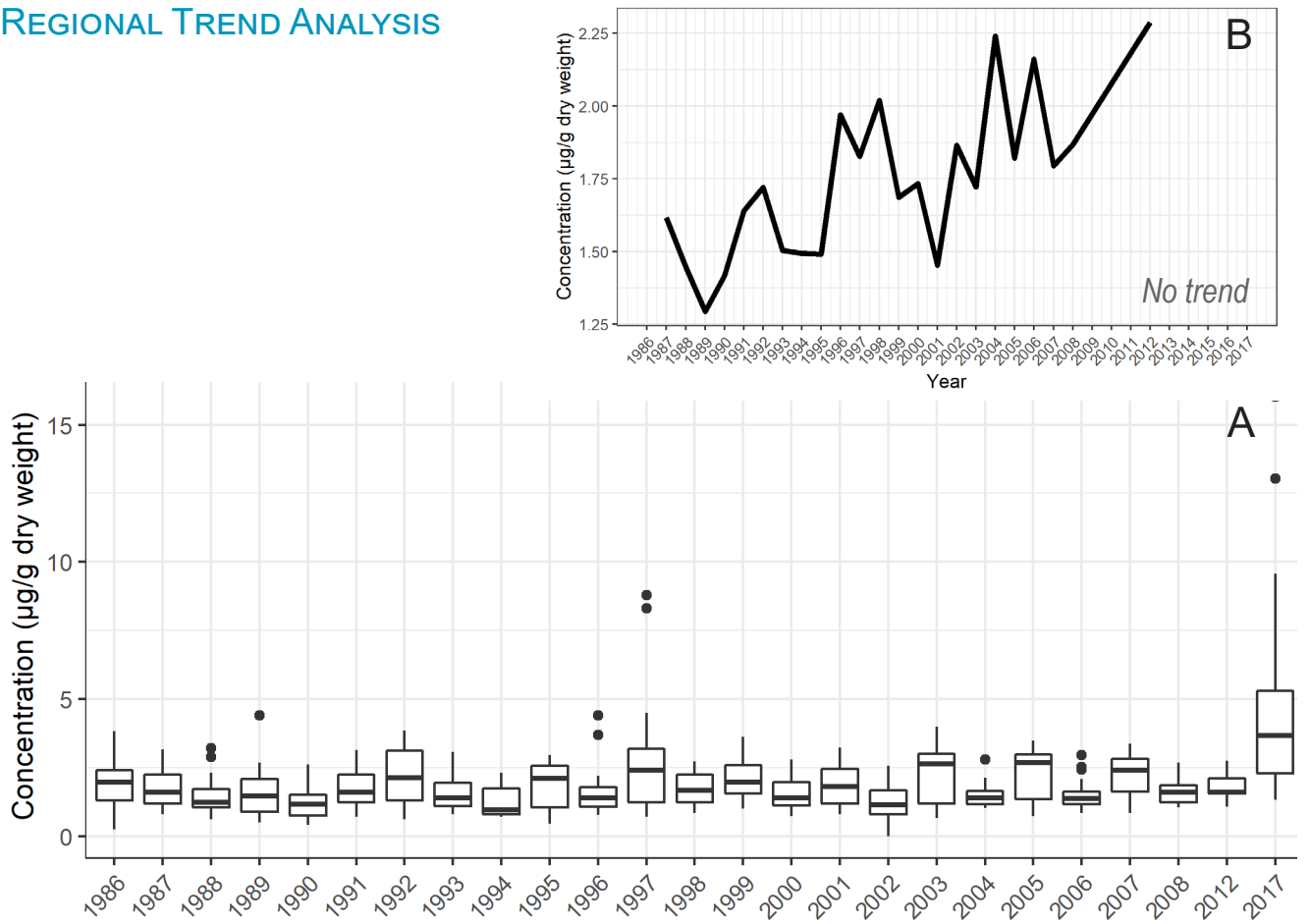


Figure 21. (A) Boxplots representing the historic nickel concentrations of the 39 site analyzed in this report and (B) the three-point moving average of the yearly median concentrations.

3.8 Tin (Sn)

3.8.1 Chemical Description

Tin sources in coastal water and soil include manufacturing and processing facilities, as well as the degradation of legacy organotins (Tributyltin). Tin also occurs in trace amounts in natural waters. Concentrations in unpolluted waters and the atmosphere are often near analytical detection limits. Tin has not been mined in the U.S. since 1993 (USGS, 2008); however, Canadian tin mining occurs in the Great Lakes Region. 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 US 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 (Hadjispyou et al., 1998). Tin is regarded as being 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).

3.8.2 Tin Results

2017 Tin Data Statistics
- Concentration range: 0.00 – 0.12 µg/dry g
- Mean concentration: 0.00 ± 0.02 (SD) µg/dry g
- Maximum concentration: 0.12 µg/dry g (Naples Bay Naples Bay - NBNB)

Summary of Tin:
2017 tissue tin concentrations display minimal variability both temporally and spatially with a concentration range of 0.00 – 0.12 µg/dry g (Appendix 1). There was a total of five sites that showed a significant decreasing trend of tin concentrations over the course of MWP monitoring, Galveston Bay Offatts Bayou – GBOB, Choctawhatchee Bay Joe's Bayou – CBJB, Panama City Municipal Pier – PCMP, Tampa Bay Peter O. Knight Airport – TBKA, and Naples Bay Naples Bay – NBNB (Figure 23, Appendix 2).

All 39 sites were below the historic mean concentration for tin, and all 2017 tin concentrations fell in the lowest cluster when compared to the historic MWP tin concentrations (Figure, 23, Figure 22).

There were no regional trends for tin concentrations over time (Figure 24, Appendix 3).

National MWP Comparison

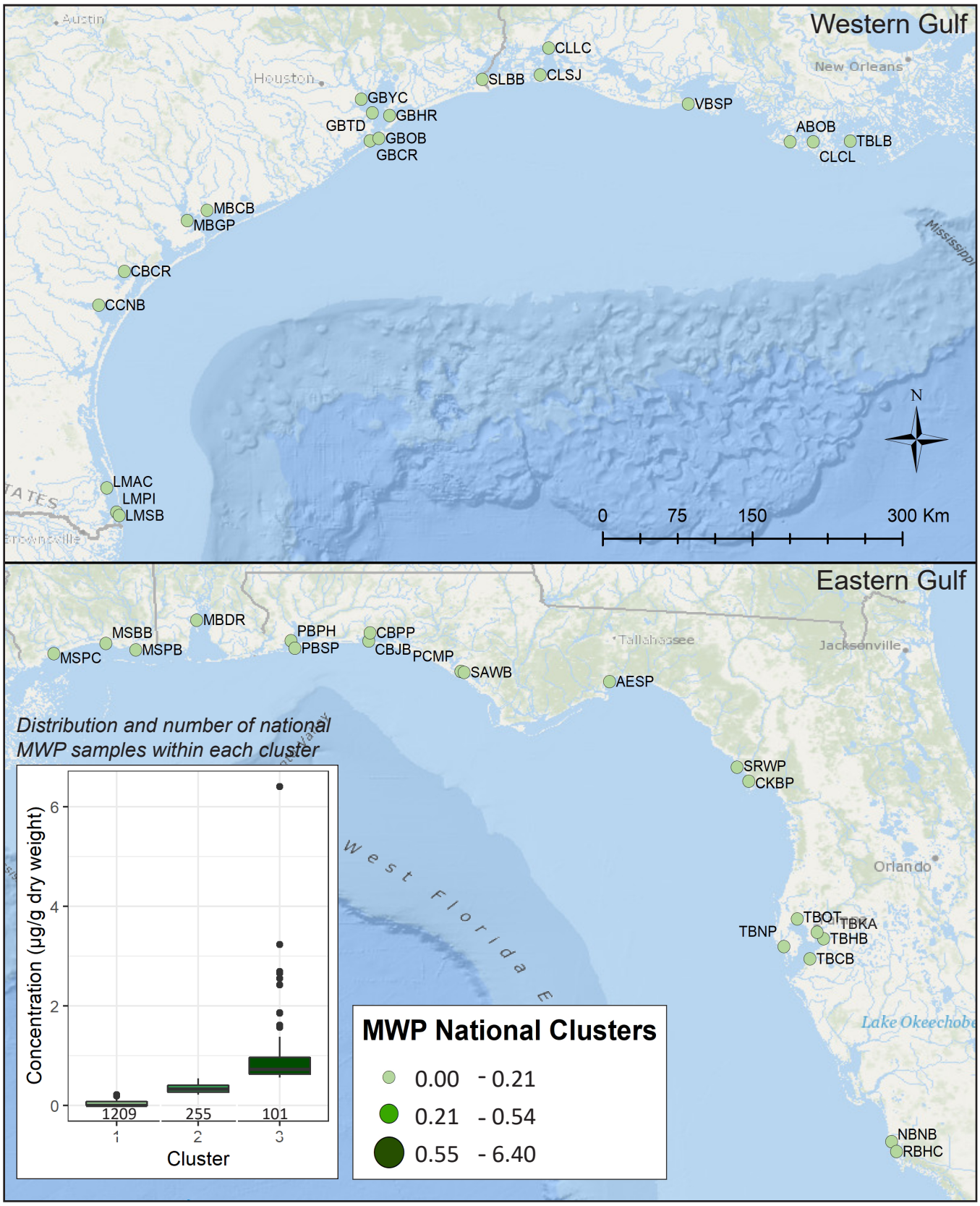


Figure 22. 2017 tin concentrations compared to the historic national MWP *Crassostrea virginica* tin concentrations (µg/dry g) (129 national sites sampled between 1986-2012 for a total of 1565 samples).

HISTORICAL SITE DATA

0% of sites were above their historic medians

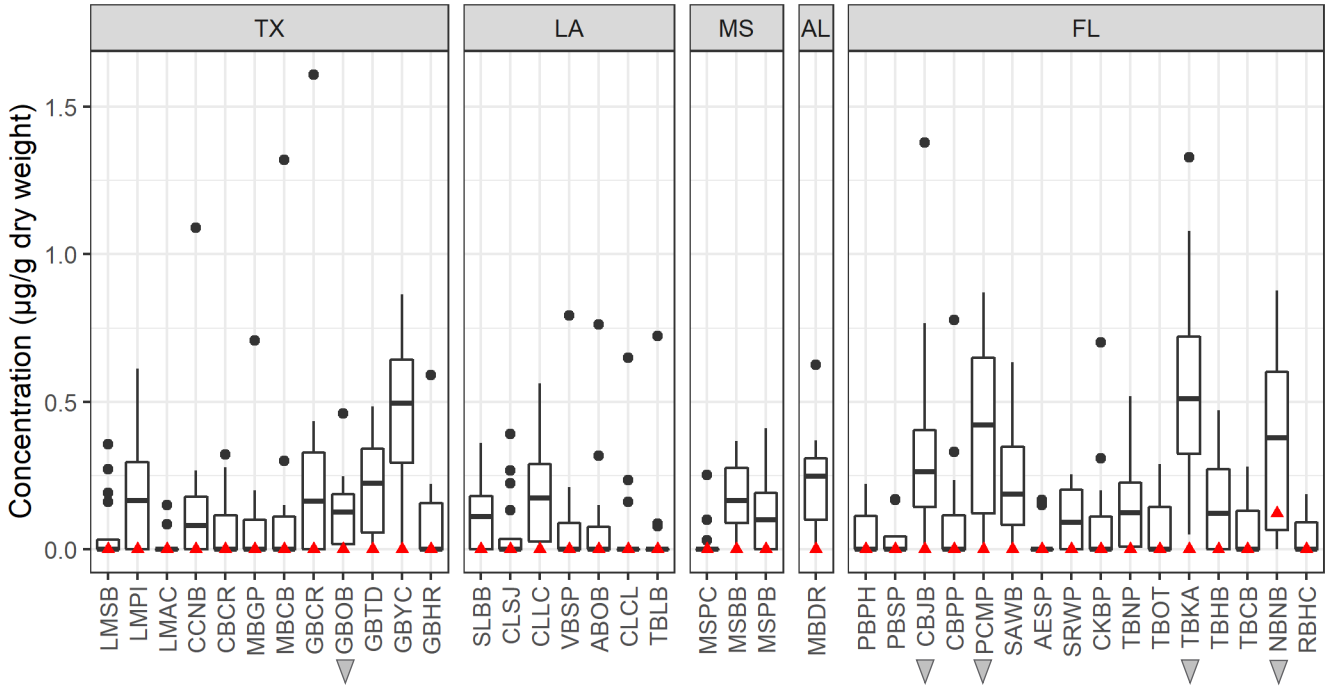


Figure 23. 2017 tin concentrations (red triangles) compared to boxplots of the historic MWP data for each site (1986-2012) and the results of the trend analysis showing signifcant increasing or decreasing trends (grey triangles).

REGIONAL TREND ANALYSIS

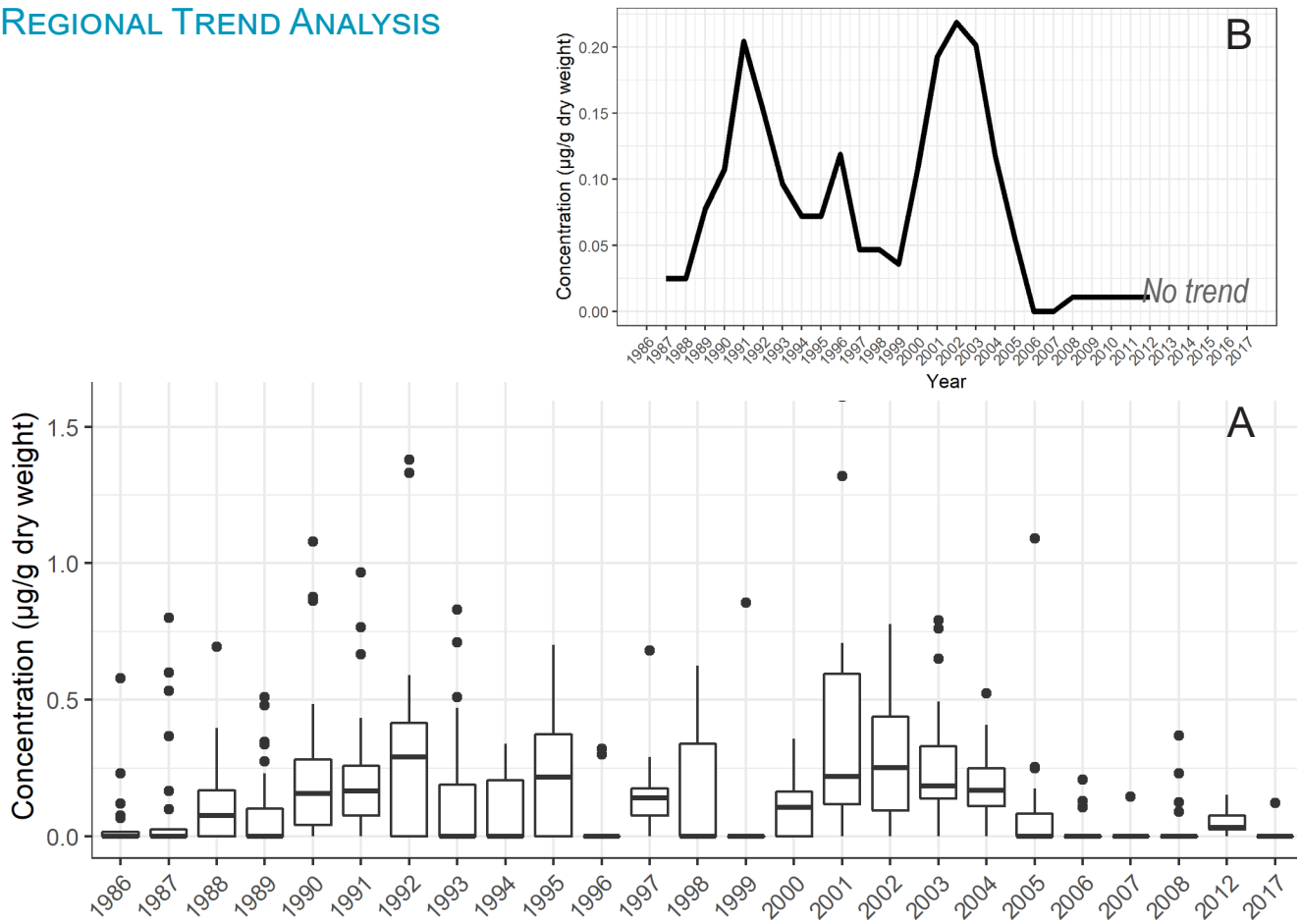


Figure 24. (A) Boxplots representing the historic tin concentrations of the 39 site analyzed in this report and (B) the three-point moving average of the yearly median concentrations.

3.9 Zinc (Zn)

3.9.1 Chemical Description

Zinc releases from anthropogenic sources are greater than those from natural sources (ATSDR, 2005c). The primary anthropogenic sources of zinc in the environment (air, water, soil) are related to mining and metallurgic operations involving zinc and use of commercial products containing zinc, including tire wear particles and brake pads. The most important sources of anthropogenic zinc in soil come from discharges of smelter slags and wastes, mine tailings, coal and bottom fly ash, and the use of commercial products such as fertilizers and wood preservatives that contain zinc (ATSDR, 2005c). The 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, and wire and wire rope). Canada is one of the largest producers and exporters of zinc. The United States is the largest customer for Canadian refined zinc, and the automobile industry is the largest user of galvanized steel. 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 (US EPA, 1979). In 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, while remaining as the free ion at lower pHs. As a result of natural and anthropogenic activities, zinc is found in all environmental compartments (air, water, soil, and biota).

Zinc is an essential nutrient. 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 US FDA recommended safety level for zinc in fish and fish products.

3.9.2 Zinc Results

<u>2017 Zinc Data Statistics</u>
- Concentration range: 309.93 – 21007.75 µg/dry g
- Mean concentration: 4407.96 ± 3941.40 (SD) µg/dry g
- Maximum concentration: 21007.75 µg/dry g (Rookery Bay Henderson Creek - RBHC)

<u>Summary of Zinc:</u>
Zinc concentrations vary both temporally and spatially with a concentration range of 309.93 – 21007.75 µg/dry g (Appendix 1). There was one site with a decreasing trend of zinc concentrations over the course of MWP monitoring, Mobile Bay Dog River – MBDR ($p = 0.042$, rho = -0.68), and 3 sites with increasing trends of zinc concentratios; Lower Laguna Madre Arroyo Colorado – LMAC ($p = 0.021$, rho = 0.68), Galveston Bay Offatts Bayou – GBOB ($p = 0.001$, rho = 0.76), and Naples Bay Naples Bay – NBNB ($p = 0.018$, rho = 0.56) (Figure 26, Appendix 2).

2017 zinc concentrations were widely distributed among the three historic MWP national clusters. Zinc concentrations in 2017 when compared to the historic MWP zinc concentration resulted in four sites falling within the highest concentration cluster (8000.00 – 26796.33 µg/dry g); Galveston Bay Yacht Club – GBYC, Galveston Bay Offatts Bayou – GBOB, Tampa Bay Old Tampa Bay – TBOT, and Rookery Bay Henderson Creek – RBHC (Figure 25). There were no regional trends for tin concentrations over time (Figure 27, Appendix 3). When compared to the historic MWP zinc concentrations, 74% of the 2017 sites were above their historic MWP medians (Figure 26).

NATIONAL MWP COMPARISON

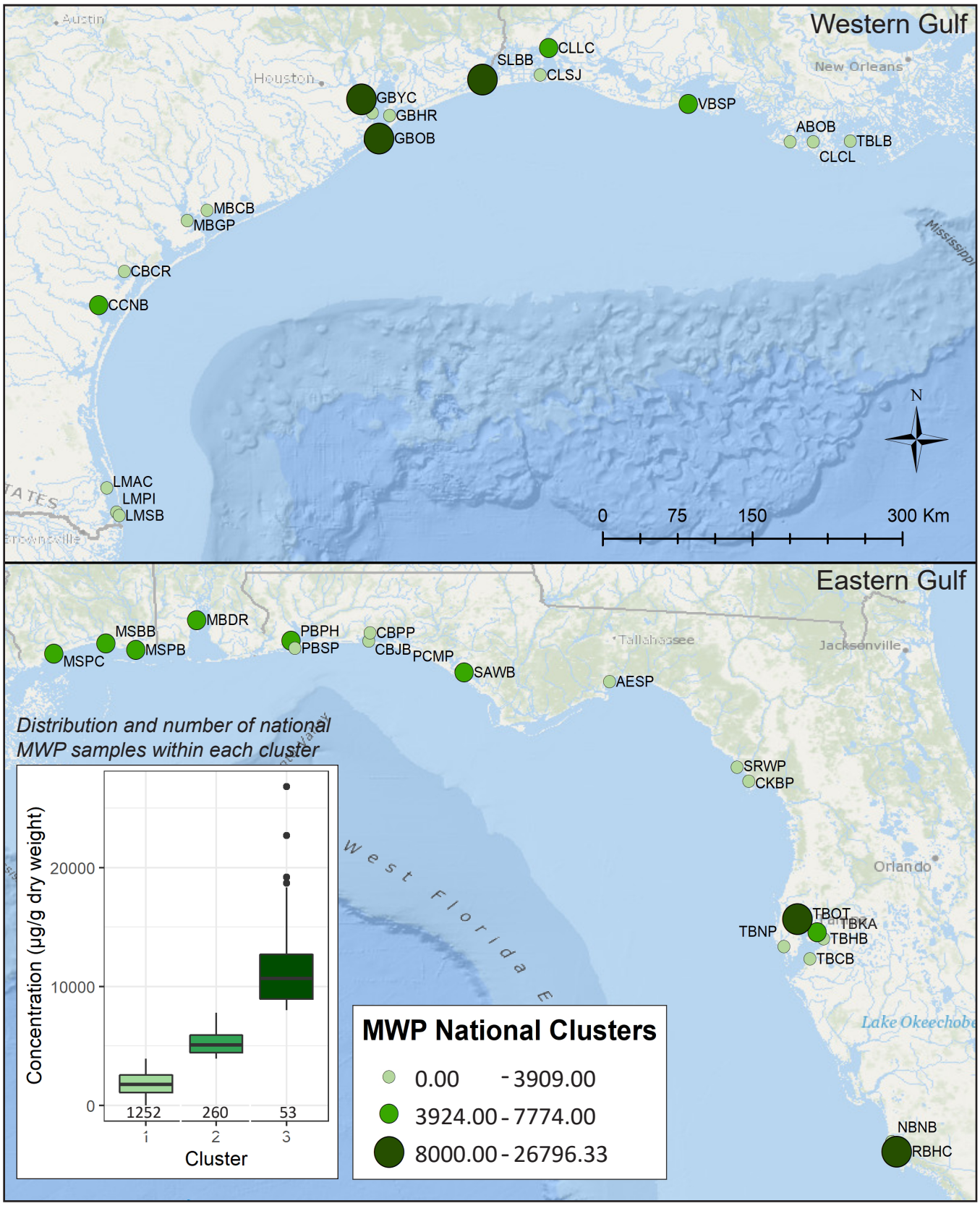


Figure 25. 2017 zinc concentrations compared to the historic national MWP *Crassostrea virginica* zinc concentrations (µg/dry g) (129 national sites sampled between 1986-2012 for a total of 1565 samples).

HISTORICAL SITE DATA

74% of sites were above their historic medians

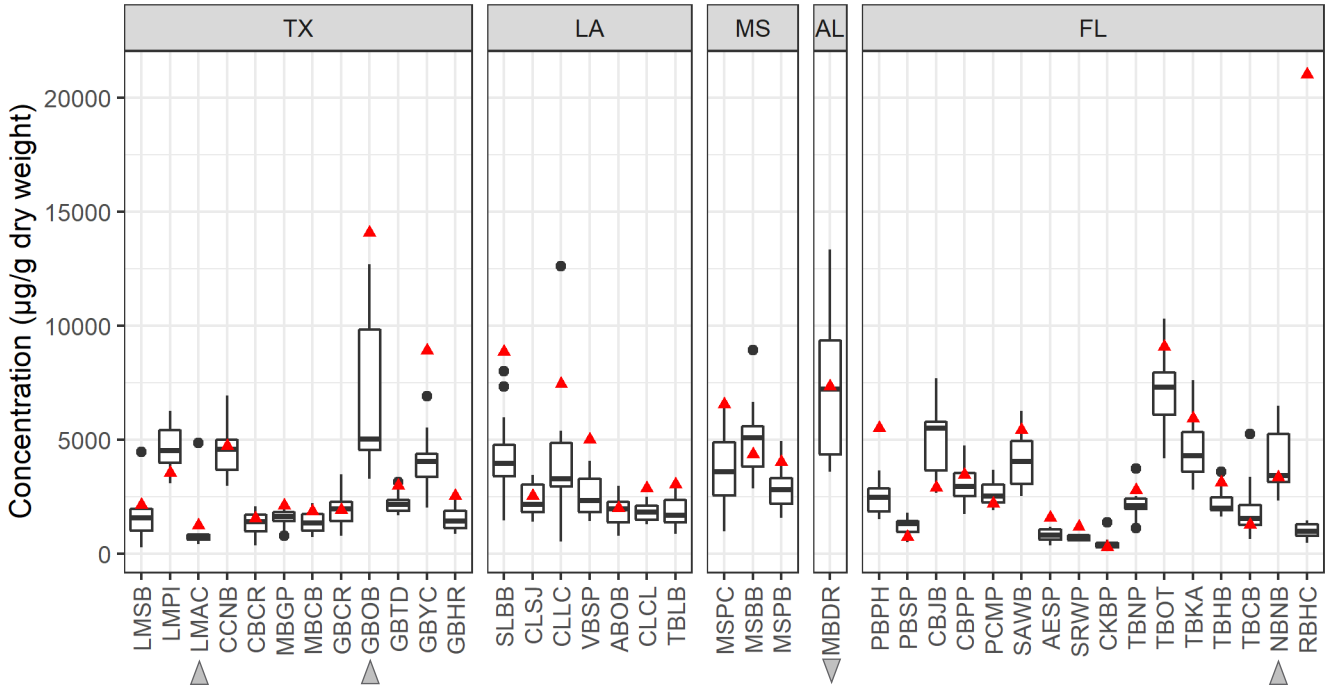


Figure 26. 2017 zinc concentrations (red triangles) compared to boxplots of the historic MWP data for each site (1986-2012) and the results of the trend analysis showing signifcant increasing or decreasing trends (grey triangles).

REGIONAL TREND ANALYSIS

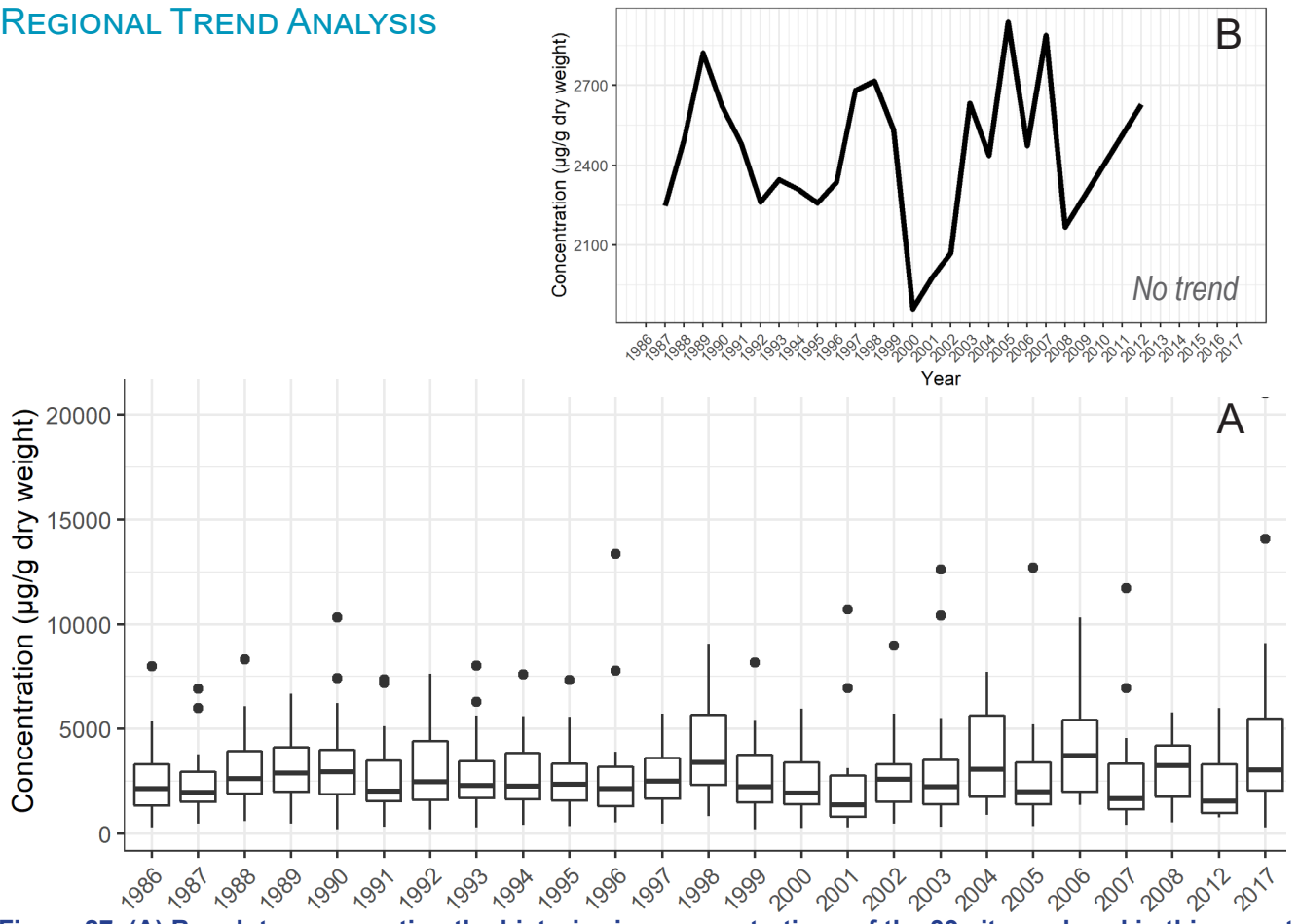


Figure 27. (A) Boxplots representing the historic zinc concentrations of the 39 site analyzed in this report and (B) the three-point moving average of the yearly median concentrations.

4.0 SUMMARY

Even though there are no significant regional trends for the eight metals analyzed, Table 3 displays the site-specific trends that are notable across the Gulf of Mexico (GOM) region. While cadmium, nickel and tin showed uniform site-based trends across the region (increasing or decreasing), all other contaminants showed variation depending on the site, highlighting the importance of local influences on many of these metal concentrations. Figure 28 summarizes the comparison of the 2017 data to the historical national MWP data clusters, highlighting the metals and sites for which multiple relatively high concentrations were detected. Nickel and mercury were the two metals with the most 2017 concentrations falling within the high national MWP clusters (21 and 10 sites, respectively). Sites SLBB and RBHC each had four metals whose 2017 concentrations fell within the high national MWP clusters. VBSP had high concentrations for three metals, and all of the other sites had two or fewer. A brief overview of the notable findings for each metal can be found below.

Arsenic concentrations showed both increasing and decreasing site specific temporal trends throughout the GOM, however, many of the sites with increasing trends in arsenic concentrations are situated along the Florida Panhandle. Additionally, the sites along the northwestern coastline of Florida, from Pensacola Bay to Tampa Bay, had the highest ranges of historic arsenic concentrations in the Gulf of Mexico and several of their 2017 arsenic concentrations fall within the medium or high national MWP clusters (Figure 28). Historically, TBNP has had two of the three highest arsenic concentration (125 µg/dry g and 75 µg/dry g) that MWP has ever detected.

Although there is no significant Spearman rank correlation for cadmium concentrations for the regional trend, it does appear that the rolling average for the regional and site-based trend is decreasing. There are no cadmium concentrations within the high MWP cluster for 2017, and 64% of the sites were below the historic medians. Seven of the thirty-nine sites displayed a decreasing trend for cadmium concentration with none increasing. Similar to findings in Apeti, et. al. (2009), there is a significant inverse correlation between concentration of cadmium in *C. virginica* and the salinity concentrations in the GOM in 2017 (Appendix 4). In estuaries, the salinity gradient is one of the main factors

Table 3. Summary of the site-based trend analyses; D = decreasing trend, I = increasing trend.

State	Site	Arsenic (As)	Cadmium (Cd)	Copper (Cu)	Lead (Pb)	Mercury (Hg)	Nickel (Ni)	Tin (Sn)	Zinc (Zn)
TX	LMSB		D				I		
TX	LMPI					I	I		
TX	LMAC								I
TX	CCNB				I				
TX	CBCR					D			
TX	MBGP								
TX	MBCB					D	I		
TX	GBCR		D		I		I		
TX	GBOB	I		I				D	I
TX	GBTD		D				I		
TX	GBYC	I					I		
TX	GBHR						I		
LA	SLBB								
LA	CLSJ					I			
LA	CLLC								
LA	VBSP								
LA	ABOB		D	D					
LA	CLCL						I		
LA	TBLB								
MS	MSPC								
MS	MSBB			I					
MS	MSPB	D							
AL	MBDR								D
FL	PBPH						I		
FL	PBSP	I							
FL	CBJB	I		I	D	D		D	
FL	CBPP	I							
FL	PCMP	I						D	
FL	SAWB	I		I					
FL	AESP								
FL	SRWP		D						
FL	CKBP	D	D						
FL	TBNP			I		D	I		
FL	TBOT			I		D			
FL	TBKA			I			I	D	
FL	TBHB			I					
FL	TBCB	I				D	I		
FL	NBNB	D	D	I				D	I
FL	RBHC	D							

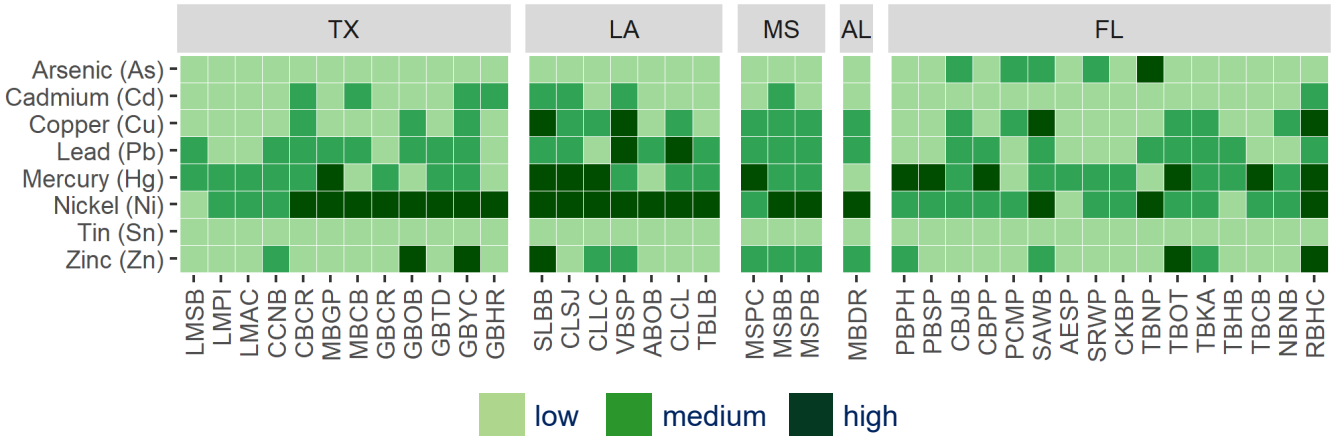


Figure 28. Heatmap summarizing the distribution of the 2017 metal tissue concentrations in comparison to the MPW national data clusters.

controlling Cd bioaccumulation from the dissolved phase. It has been shown that in coastal habitats, the rates of Cd uptake by mollusks decrease along an increasing salinity gradient in estuaries (Rainbow, 1995; Riedel et al., 1998; De Wolf et al., 2004). Salinity in the waters of the GOM is increasing due to a decrease in fresh water entering the waters as a result of several decades of river diversions (Das et al., 2012). This increase in salinity could lead to the possible decrease in cadmium uptake by *Crassostrea virginica*.

Even though the rolling average show no significant regional trend for copper, there were nine individual sites showing an increasing trend for copper concentration and only site where it was decreasing. In 2017, 79% of the sites were above their historic MWP medians for Cu concentrations. There are two sites in Louisiana, SLBB and VBSP, that had noticeably higher 2017 concentrations than their historic concentration ranges for copper. Likewise, Florida had two sites, RBHC and SAWB, that showed the same trend.

Across the region, there were no notable spatial patterns in lead concentrations as well as no notable regional trend; however, there is some site specific spatial and temporal variability. In 2017, VBSP had one of the highest concentrations ever measured in the region despite having a historically low range of lead concentrations. Site CBPP has had the highest range of lead concentrations in the region historically; although, the 2017 concentrations was below the sites historic median.

Mercury, unlike lead, displayed a wide array of spatial variability across the region. Despite many sites with 2017 concentrations falling within the historical high MWP cluster, there are few sites that stand out. Sites TBOT and MBGP have the highest historical MWP Hg concentrations ranges in the region, and their 2017 Hg concentrations fall within their usual range. Site RBHC historically had moderate Hg concentrations, but in 2017 it had the highest concentration of Hg measured to date in the region. Hopefully, future sampling will assist in determining if this is an anomaly or representative of an upward trend at the site (RBHC).

At all sites, the 2017 nickel concentrations were above their historic national MWP medians. There is spatial variability in the region for Ni concentrations, with sites from Copano Bay, TX through Alabama displaying the highest historic concentration ranges. Additionally, there were 12 sites across the region with site-based increasing trends. In general, free ionic metal concentrations are reduced in high salinity environments in comparison to low salinity or freshwater because of the increased presence of complexing anions. For nickel in sea water, the two of importance are SO_4^{2-} and Cl^-

(Sadiq, 1989). In addition, at higher salinities, there should be increased competition with metal ions by protective cations such as Na+, Mg2+ and Ca2+ for binding to sites at the biotic ligand (Paquin et al., 2000, Janssen et al., 2003). Therefore, salinity is thought to act protectively against the toxicity of many metals, including Ni (Eisler, 1998). In the case of nickel, it would be expected that, with the increase in salinity in the Gulf of Mexico, a decrease in nickel in the region would be notable (Appendix 4). However, this is not the case in the region.

All the 2017 tin tissue concentrations were in the lowest historical MWP cluster. Five sites displayed a decreasing trend and there were no increasing site-based trends detected. Since 2005, the concentrations of Sn in the region have been extremely low.

Similar to the mercury results, in 2017 site RBHC had the highest concentration of zinc ever detected in the region. As stated earlier, future sampling will assist in determining if this is an anomaly for both Hg and Zn concentrations or representative of an upward trend at site RBHC for both metals. Five of the sites in the region fell within the highest national MWP cluster for Zn concentrations. There was no regional trend and only 4 site-based trends for Zn (three increasing and two decreasing).

The aim of this report is to contextualize recent regional monitoring metals data in both time and space and to provide a perspective that may not be attainable at the local level. By doing so, we hope to support the allocation of resources and the ongoing studies and efforts necessary to manage coastal chemical contaminants nationwide.

References

Apeti, D. A., Lauenstein, G. G., & Riedel, G. F., 2009. Cadmium distribution in coastal sediments and mollusks of the US. Marine Pollution Bulletin, 58(7), 1016–1024.

Apeti, D.A., Johnson, W.E., Kimbrough, K.L. and Lauenstein, G.G., 2012. National Status and Trends Mussel Watch Program: Field Methods 2012 Update. NOAA National Centers for Coastal Ocean Science, Center for Coastal Monitoring and Assessment. NOAA NCCOS Technical Memorandum 134. Silver Spring, MD.

ATSDR (Agency for Toxic Substances and Disease Registry), 1999a. Toxicological Profile for Cadmium. Atlanta, GA: U.S. Department of Health and Human Services, Public Health Service.

ATSDR (Agency for Toxic Substances and Disease Registry), 1999b. Toxicological Profile for Mercury. Atlanta, GA: U.S. Department of Health and Human Services, Public Health Service.

ATSDR (Agency for Toxic Substances and Disease Registry), 2004. Toxicological Profile for Copper. Atlanta, GA: U.S. Department of Health and Human Services, Public Health Service.

ATSDR (Agency for Toxic Substances and Disease Registry), 2005a. Toxicological Profile for Nickel. Atlanta, GA: U.S. Department of Health and Human Services, Public Health Service.

ATSDR (Agency for Toxic Substances and Disease Registry), 2007a. Toxicological Profile for Arsenic. Atlanta, GA: U.S. Department of Health and Human Services, Public Health Service.

ATSDR (Agency for Toxic Substances and Disease Registry), 2007b. Toxicological Profile for Lead. Atlanta, GA: U.S. Department of Health and Human Services, Public Health Service.

ATSDR (Agency for Toxic Substances and Disease Registry), 2005b. Toxicological Profile for Tin. Atlanta, GA: U.S. Department of Health and Human Services, Public Health Service.

ATSDR (Agency for Toxic Substances and Disease Registry). 2005c. Toxicological Profile for Zinc. Atlanta, GA: U.S. Department of Health and Human Services, Public Health Service.

Athey, J.E., Daanen, R.D. and Hendricks, K.A., 2018. Naturally occurring arsenic in Alaska groundwater: Alaska Division of Geological & Geophysical Surveys Information Circular 69, 2 p. http://doi.org/10.14509/30060

Berner, L.H., McGowan, J., Martin, J.H., and Teal, J., 1976. Sampling marine organisms. In: Strategies for Marine Pollution Monitoring, E. D. Goldberg, (ed.). John Wiley & Sons, NY. pp. 269-73.

Das, A., Justic, D., Inoue, M., Hoda, A., Huang, H. and Park, D., 2012. Impacts of Mississippi River diversions on salinity gradients in a deltaic Louisiana estuary: Ecological and management implications. Estuarine, Coastal and Shelf Science, 111, pp.17-26.

De Wolf, H., Backeljau, T. and Blust, R., 2004. Sensitivity to cadmium along a salinity gradient in populations of the periwinkle, Littorina littorea, using time-to-death analysis. Aquatic Toxicology, 66(3), pp.241-253.

Denier van der Gon, H.A.C., Hulskotte, J.H.J., Visschedijk, A.J.H. and Schaap, M., 2007. A revised estimate of copper emissions from road transport in UNECE Europe and its impact on predicted copper concentrations. Atmospheric Environment 41(38):8697-8710.

Edmonds, J.S. and Francesconi, K.A., 1977. Methylated arsenic from marine fauna. Nature 265:436.

Edmonds, J.S. and Francesconi, K.A., 1988. The origin of arsenobetaine in marine animals. Applied Organometallic Chemistry 2:297-302.

References

Edmonds, J.S. and Francesconi, K.A., 1993. Arsenic in seafood: human health aspects and regulations. Marine Pollution Bulletin 26:665-674.

Ellis, A.M. and Smith, C.G., 2021. Emerging dominance of Paratrochammina simplissima (Cushman and McCulloch) in the northern Gulf of Mexico following hydrologic and geomorphic changes. Estuarine, Coastal and Shelf Science, 255, p.107312.

Eisler, R., Nickel Hazards to Fish, Wildlife, and Invertebrates: A Synoptic Review. 1998–0001. Biological Science Report United States Geological Survey, Biological Resources Division, Washington, D.C (1998).

Farrington, J. W. 1983. Bivalves as sentinels of coastal chemical pollution: The Mussel (and oyster) Watch. Oceanus 26(2):18-29.

Farrington, J. W., J. Albaiges, K. A. Burns, B. P. Dunn, P. Eaton, J. L. Laseter, P. L. Parker, and S. Wise. 1980. Fossil fuels. In: The International Mussel Watch. National Research Council. National Academy of Sciences - Office of Publications, Washington, D.C. pp. 7-77.

Florida Department of Environmental Protection (2013), Apalachicola National Estuarine Research Reserve management plan, June 2013, Rep., Fla. Dep. of Environ. Prot., Tallahassee, Fla.

Gerritse, R.G., R. Vriesema, J.W. Daleberg, and H.P. de Roos, 1982. Effect of sewage sludge on trace element mobility in soils. Journal of Environmental Quality 11:359-364.

Goldberg, E.D., Koide, M., Hodge, V., Flegal, A.R., Martin, J., 1983. US Mussel Watch: 1977–1978 results on trace metals and radionuclides. Estuarine, Coastal and Shelf Sciences 16, 69–93.

Goyer, R.A., 1986. Toxic Effects of Metals. In: Toxicology the Basic Science of Poisons. Third Edition. Edited by Klaassen, C. D., Amdur, M.O. and Doull, J. by Macmillan Publishing Company. New York, NY.

Hadjispyrou, S.A., A. Anagnostopoulos, K. Nicholson, M.K. Nimfopoulos, and K.M. Michailidis. 1998. Correlation of the methylating capacity of river and marine sediments to their organic sediment index. Environmental Geochemistry and Health 20(1):19-27.

Harvey, L.J. and McArdle, H.J., 2008. Biomarkers of copper status: a brief update. British Journal of Nutrition, 99(S3), pp. S10-S13.

IARC (International Agency for Research on Cancer), 2006. Inorganic and organic lead compounds 10-17 February 2004. Vol. 87.

Isphording, W. C. (1985), Sedimentological investigation of the Apalachicola Bay, Florida estuarine system: prepared for the Mobile District, Corps of Engineers, Rep., Univ. of Ala., Tuscaloosa, Ala.

Janssen, C.R., Heijerick, D.G., De Schamphelaere, K.A.C., Allen, H.E., Environmental risk assessment of metals: tools for incorporating bioavailability, Environ. Int., 28 (2003), pp. 793-800

Kimbrough, K.L. and Lauenstein, G.G., 2006. Trace Metal Analytical Methods of the National Status and Trends Program: 2000–2006. US Dept. Comm., NOAA Tech. Memo. 29, NOS NCCOS, Silver Spring, Maryland.

Kimbrough, K.L., Johnson, W.E., Lauenstein, G.G., Christensen, J.D. and Apeti, D.A., 2008. An Assessment of Two Decades of Contaminant Monitoring in the Nation's Coastal Zone. Silver Spring, MD. NOAA Technical Memorandum NOS NCCOS 74. 105 pp.

References

Manahan, S.E., 2005. Environmental Chemistry. 8th Edition. CRC Press: Boca Raton, Florida. 783 pp.

Mance, G. 1987. Pollution Threat of Heavy Metals in Aquatic Environments. Elsevier Science New York, NY.

Martin, M. 1985. State Mussel Watch: Toxics surveillance in California. Marine Pollution Bulletin 16(4):140-146.

Mata, D.I., Moreno-Casasola, P., Madero-Vega, C., Castillo-Campos, G. and Warner, B.G., 2011. Floristic composition and soil characteristics of tropical freshwater forested wetlands of Veracruz on the coastal plain of the Gulf of Mexico. Forest Ecology and Management, 262(8), pp.1514-1531.

McGeer, J.C., Brix, K.V., Skeaff, J.M., DeForest, D.K., Brigham, S.I., Adams, W.J. and Green, A., 2003. Inverse relationship between bioconcentration factor and exposure concentration for metals: implications for hazard assessment of metals in the aquatic environment. Environmental Toxicology and Chemistry 22(5):1017-1037.

Mirenda R.J., 1986. Acute toxicity and accumulation of zinc in the crayfish Orconectes virilis (Hagen). Bulletin of Environmental Contamination and Toxicology 37(1):387-394.

Mohammed, A., 2013. Why are early life stages of aquatic organisms more sensitive to toxicants than adults? New insights into toxicity and drug testing, 49-62.

NADP (National Atmospheric Deposition Program) (NRSP-3), 2000. NADP Program Office, Wisconsin State Laboratory of Hygiene, 465 Henry Mall, Madison, WI 53706.

Nriagu J.O., Lawson G, Wong H.K., and Cheam, V.,1995. Dissolved trace metals in lakes Superior, Erie, and Ontario. Environmental science & technology. Dec 27;30(1):178-87.

Paquin, P.R., Santore, R.C., Wu, K.B., Kavvadas, C.D. and Di Toro, D.M., 2000. The biotic ligand model: a model of the acute toxicity of metals to aquatic life. Environmental Science & Policy, 3, pp.175-182.

Passeri, D.L., Hagen, S.C., Plant, N.G., Bilskie, M.V., Medeiros, S.C. and Alizad, K., 2016. Tidal hydrodynamics under future sea level rise and coastal morphology in the Northern Gulf of Mexico. Earth's Future, 4(5), pp.159-176.

Phillips, D.J.H., 1990. Arsenic in aquatic organisms: a review, emphasizing chemical speciation. Aquatic Toxicology 16(3):151-186.

R Core Team, 2013. R: A language and environment for statistical computing. Vienna, Austria: R Foundation for Statistical Computing. Retrieved from http://www.Rproject.org/.

Rainbow, P.S., 1995. Physiology, physicochemistry and metal uptake: a crustacean perspective. Marine Pollution Bulletin 31 (1–3), 55–59.

Riedel, G.F., Abbe, G.R., Sanders, J.G., 1998. Trace metals concentrations in oysters from the Patuxent River, Maryland: temporal and spatial variation. Estuaries 21, 423–434.

Roesijadi, G., Young, J.S., Drum, A.S. and Gurtisen, J.M., 1984. Behavior of trace metals in Mytilus edulis during a reciprocal transplant field experiment. Marine Ecology Progress Series 18:155-70.

Sadiq, M., Nickel sorption and speciation in marine environment, Hydrobiologia, 176/177 (1989), pp. 225-232.

References

Seidel, S.L., V.F. Hodge, and E.D. Goldberg. 1980. Tin as an environmental pollutant. Thalassia Jugoslavica 16:209-223.

Sericano, J.L. 1993. The American Oyster (*Crassostrea virginica*) as a Bioindicator of Trace Organic Contamination. Doctoral dissertation, Texas A&M University, College Station, TX. 242 pp.

Sokolova, I.M., Ringwood, A.H. and Johnson, C., 2005. Tissue-specific accumulation of cadmium in subcellular compartments of eastern oysters *Crassostrea virginica* Gmelin (Bivalvia: Ostreidae). Aquatic Toxicology 74(3):218-228.

Stern, B.R., 2010. Essentiality and toxicity in copper health risk assessment: overview, update, and regulatory considerations. Journal of Toxicology and Environmental Health, Part A, 73(2-3), pp.114-127.

Suedel, B.C., Boraczek, J.A., Peddicord, R.K., Clifford, P.A. and Dillon, T.M., 1994. Trophic transfer and biomagnification potential of contaminants in aquatic ecosystems. Reviews of environmental contamination and toxicology, pp.21-89.

Sweet, C.W., Weiss, A. and Vermette, S.J., 1998. Atmospheric deposition of trace metals at three sites near the Great Lakes. Water, Air, and Soil Pollution, 103(1), pp.423-439.

Tchounwou, P.B., Yedjou, C.G., Patlolla, A.K. and Sutton, D.J., 2012. Heavy metal toxicity and the environment. Molecular, clinical and environmental toxicology, pp.133-164.

Tchounwou P.B., Newsome C., Williams J., and Glass K., 2008. Copper-induced cytotoxicity and transcriptional activation of stress genes in human liver carcinoma (HepG2) cells. In Metal ions in biology and medicine: proceedings of the... International Symposium on Metal Ions in Biology and Medicine held...= Les ions metalliques en biologie et en medecine:... Symposium international sur les ions metalliques. (Vol. 10, p. 285). NIH Public Access.

Thompson, S.E., Burton, C.A., Quinn, D.J. and Ng, Y.C., 1972. Concentration factors of chemical elements in edible aquatic organisms. Livermore, CA: Lawrence Livermore Laboratory, Biomedical Division, University of California. 77 pp.

Tripp, B.W. and Farrington, J.W., 1984. Using sentinel organisms to monitor chemical changes in the coastal zone: progress or paralysis. Submitted to the Coastal Society, 9th Annual Conference, October 1984, Atlantic City, NJ. Woods Hole Oceanographic Institution Contribution No. 5830.

Turner, R.E., 2001. Of manatees, mangroves, and the Mississippi River: Is there an estuarine signature for the Gulf of Mexico? Estuaries, 24(2), pp.139-150.

US EPA (United States Environmental Protection Agency), 1979. Water-related environmental fate of 129 priority pollutants. Washington, DC: U.S. Environmental Protection Agency, Office of Water Planning and Standards. EPA440479029a

US FDA (United States Food and Drug Administration), 1993a. Guidance Document for Arsenic in Shellfish. U.S. Department of Health and Human Services, Public Health Service, Office of Seafood (HFS-416), 200 C Street, SW, Washington, D.C.

US FDA (United States Food and Drug Administration), 1993b. Guidance Document for Cadmium in Shellfish. U.S. Department of Health and Human Services, Public Health Service, Office of Seafood (HFS-416), 200 C Street, SW, Washington, D.C. 20204.

References

US FDA (United States Food and Drug Administration), 2011. Fish and Fishery Products Hazards and Controls Guide. 4th Ed. Department of Health and Human Services, Public Health Service, Center for Food Safety and Applied Nutrition, Office of Seafood, Washington, DC.

USGS (United States Geological Survey), 2008. Minerals Yearbook: Volume I. Metals and Minerals. Reston, Virginia.

Welch, A.H., Lico, M.S. and Hughes, J.L., 1988. Arsenic in ground water of the western United States. Groundwater, 26(3), pp.333-347.

WHO (World Health Organization) 1980. Tin and organotin compounds: A preliminary review. Environmental Heath criteria 15. Geneva, Switzerland. 22 Apr. 2008.

Zimmerman, R.J., Minello, T.J. and Rozas, L.P., 2002. Salt marsh linkages to productivity of penaeid shrimps and blue crabs in the northern Gulf of Mexico. In Concepts and controversies in tidal marsh ecology (pp. 293-314). Springer, Dordrecht.

Appendices

Appendix 1. 2017 metal tissue concentrations (µg/g dry weight) above MDL measured in oyster tissue in the Gulf of Mexico.

Site	Arsenic (As)	Cadmium (Cd)	Copper (Cu)	Lead (Pb)	Mercury (Hg)	Nickel (Ni)	Tin (Sn)	Zinc (Zn)
Minimum MDL	3.39	0.58	25.93	0.32	0.06	1.34	0.00	309.93
ABOB	9.10	3.59	178.30	1.18	0.07	5.35	0.00	2014.62
AESP	16.22	3.20	74.07	0.65	0.17	1.46	0.00	1587.47
CBCR	9.74	7.81	290.97	1.19	0.14	8.71	0.00	1572.81
CBJB	20.70	1.89	519.56	1.06	0.13	2.29	0.00	2900.15
CBPP	8.38	1.63	175.42	3.47	0.23	2.57	0.00	3459.40
CCNB	16.17	2.61	158.03	1.82	0.14	2.43	0.00	4747.10
CKBP	8.99	0.58	33.83	0.75	0.11	2.30	0.00	309.93
CLCL	8.51	3.60	292.30	5.07	0.09	7.18	0.00	2892.35
CLLC	6.05	3.12	522.38	0.64	0.17	5.93	0.00	7463.69
CLSJ	6.03	4.59	233.00	1.06	0.17	5.83	0.00	2553.06
GBCR	7.80	2.16	168.12	0.91	0.10	4.45	0.00	1929.74
GBHR	5.18	5.80	202.57	0.73	0.08	5.25	0.00	2536.75
GBOB	10.74	0.97	406.28	1.64	0.06	3.75	0.00	14068.28
GBTD	5.50	3.53	129.57	1.23	0.09	5.05	0.00	3004.21
GBYC	5.63	5.01	371.06	2.82	0.09	4.12	0.00	8907.03
LMAC	18.33	2.21	100.80	0.54	0.13	1.61	0.00	1247.41
LMPI	15.16	1.16	222.64	0.58	0.08	2.21	0.00	3552.33
LMSB	16.51	1.79	172.15	0.99	0.15	1.34	0.00	2160.88
MBCB	10.16	5.06	179.74	1.63	0.07	7.99	0.00	1874.39
MBDR	7.27	3.29	434.46	0.97	0.08	4.03	0.00	7338.08
MBGP	7.24	3.81	214.52	1.30	0.36	16.02	0.00	2131.13
MSBB	6.37	6.58	328.62	2.07	0.17	3.67	0.00	4367.39
MSPB	9.91	3.78	234.84	0.93	0.16	3.96	0.00	4038.66
MSPC	8.98	1.90	405.57	2.09	0.21	2.66	0.00	6546.59
NBNB	9.73	0.70	585.05	0.32	0.14	1.52	0.12	3349.79
PBPH	7.83	3.57	214.84	0.88	0.23	3.45	0.00	5532.89
PBSP	17.20	1.92	42.91	0.54	0.17	3.30	0.00	757.61
PCMP	26.35	1.48	418.15	0.75	0.08	2.02	0.00	2204.19
RBHC	10.98	5.00	1297.63	1.28	0.87	4.18	0.00	21007.75
SAWB	19.35	1.18	1048.09	1.76	0.09	4.86	0.00	5432.85
SLBB	7.04	10.57	818.63	3.39	0.24	7.28	0.00	8867.74
SRWP	25.86	1.55	25.93	0.58	0.14	3.18	0.00	1212.63
TBCB	7.38	1.20	83.46	0.59	0.19	2.21	0.00	1292.20
TBHB	3.39	2.68	209.00	1.19	0.16	1.42	0.00	3143.72
TBKA	5.95	1.65	503.73	0.98	0.14	1.54	0.00	5945.87
TBLB	9.74	2.89	156.15	1.56	0.09	9.57	0.00	3059.85
TBNP	41.01	0.79	212.38	1.42	0.07	3.66	0.00	2805.68
TBOT	4.18	2.14	393.71	2.42	0.49	3.49	0.00	9088.40
VBSP	8.84	10.44	1302.78	11.16	0.10	13.05	0.00	5005.88

Appendices

Appendix 2. Statistical results from Spearman Rank Correlations on site-based trends.

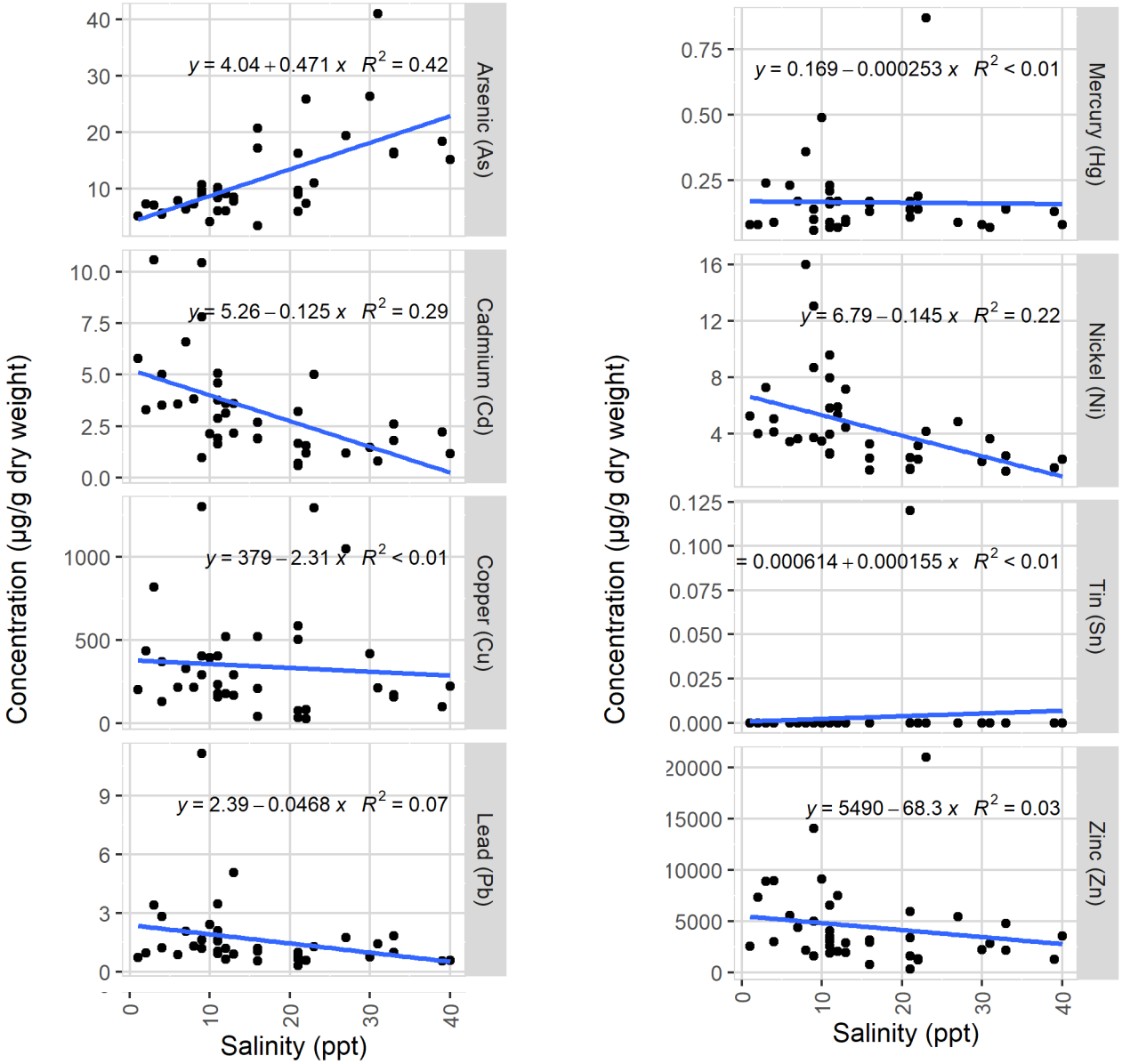
Site	Arsenic (As)		Cadmium (Cd)		Copper (Cu)		Lead (Pb)		Mercury (Hg)		Nickel (Ni)		Tin (Sn)		Zinc (Zn)	
Values	*p*	*rho*	*p*	*rho*	*p*	*rho*	*p*	*rho*	*p*	*rho*	*p*	*rho*	*p*	*rho*	*p*	*rho*
ABOB	0.125	-0.40	0.004	-0.68	0.015	-0.60	0.564	-0.16	0.191	0.34	0.557	0.16	0.516	0.18	0.072	-0.46
AESP	0.994	0.00	0.958	0.02	0.418	0.24	0.605	-0.15	0.620	0.15	0.164	0.39	0.811	-0.07	0.605	0.15
CBCR	1.000	0.00	0.413	-0.23	0.612	0.14	0.095	0.45	0.023	-0.58	0.355	0.26	0.556	-0.17	0.960	-0.01
CBJB	0.002	0.74	0.860	0.05	0.012	0.63	0.009	-0.65	<0.001	-0.82	0.689	0.11	0.027	-0.57	0.376	0.25
CBPP	0.032	0.51	0.278	-0.27	0.197	0.32	0.219	-0.30	0.791	-0.07	0.269	0.28	0.641	-0.12	0.358	-0.23
CCNB	0.594	0.14	0.070	-0.46	0.837	-0.06	<0.001	0.81	0.557	-0.16	0.116	0.41	0.872	0.04	0.333	0.26
CKBP	0.046	-0.48	<0.001	-0.83	0.657	-0.11	0.964	0.01	0.668	-0.11	0.521	0.16	0.839	-0.05	0.463	-0.18
CLCL	0.751	-0.08	0.348	-0.24	0.363	0.24	0.184	0.34	0.178	0.34	0.023	0.55	0.757	0.08	0.866	0.04
CLLC	0.562	0.16	0.571	0.16	0.355	0.26	0.550	0.17	0.095	0.45	0.091	0.45	0.747	-0.09	0.074	0.48
CLSJ	0.072	-0.45	0.400	-0.22	0.955	-0.01	0.158	0.36	0.034	0.51	0.061	0.46	0.970	0.01	0.673	0.11
GBCR	0.249	0.31	0.049	-0.50	0.922	-0.03	0.035	0.53	0.765	0.08	0.013	0.60	0.804	-0.07	0.240	-0.31
GBHR	0.753	0.09	0.144	-0.38	0.617	-0.14	0.163	0.37	0.957	0.01	<0.001	0.74	0.784	0.07	0.345	-0.25
GBOB	<0.001	0.76	0.079	-0.47	0.048	0.52	0.483	0.20	0.334	-0.27	0.666	-0.12	0.014	-0.62	0.001	0.76
GBTD	0.548	0.16	0.020	-0.56	0.715	-0.10	0.794	0.07	0.833	-0.06	0.025	0.54	0.562	-0.15	0.205	0.32
GBYC	0.034	0.53	0.105	-0.42	0.240	0.31	0.158	0.37	0.905	0.03	0.013	0.60	0.931	-0.02	0.966	-0.01
LMAC	0.201	-0.42	0.679	0.14	0.264	0.37	0.223	0.40	0.492	-0.23	0.051	0.60	0.693	-0.13	0.021	0.68
LMPI	0.199	-0.42	0.137	-0.48	0.170	0.45	0.272	-0.36	0.030	0.65	0.002	0.82	0.157	-0.46	0.958	0.02
LMSB	0.349	-0.23	0.002	-0.68	0.990	0.00	0.073	0.43	0.069	-0.44	0.040	0.49	0.865	0.04	0.570	0.14
MBCB	0.587	-0.17	0.175	-0.42	0.106	-0.49	0.085	0.52	0.007	-0.73	0.039	0.60	0.165	-0.43	0.379	-0.28
MBDR	0.244	0.43	0.058	-0.65	0.170	-0.50	0.898	0.05	0.139	-0.53	0.244	0.43	0.539	-0.24	0.042	-0.68
MBGP	0.110	-0.41	0.520	-0.17	0.295	0.28	0.158	0.37	0.147	-0.38	0.172	0.36	0.718	-0.10	0.305	0.27
MSBB	0.108	-0.43	0.840	-0.06	0.020	0.59	0.940	-0.02	0.643	-0.13	0.909	0.03	0.101	-0.44	0.201	0.35
MSPB	0.023	-0.53	0.108	-0.39	0.505	-0.17	0.791	0.07	0.074	-0.43	0.748	0.08	0.888	-0.04	0.097	-0.40
MSPC	0.852	-0.05	0.105	-0.41	0.708	0.10	0.232	0.31	0.413	0.21	0.178	0.34	0.431	0.20	0.374	0.23
NBNB	0.015	-0.58	0.005	-0.65	<0.001	0.74	0.896	-0.03	0.286	-0.27	0.058	0.47	<0.001	-0.74	0.018	0.56
PBPH	0.884	0.04	0.737	-0.09	0.850	-0.05	0.087	0.46	0.894	0.04	0.040	0.54	0.250	-0.32	0.576	0.16
PBSP	0.025	0.73	0.488	-0.27	0.798	-0.10	0.308	0.38	0.798	0.10	0.668	0.17	0.725	-0.14	0.170	-0.50
PCMP	0.013	0.72	0.937	0.03	0.853	0.06	0.077	-0.55	0.937	0.03	0.332	0.32	0.037	-0.63	0.051	-0.60
RBHC	0.012	-0.59	0.619	0.13	0.808	0.06	0.224	0.31	0.371	-0.23	0.058	0.47	0.638	-0.12	0.794	0.07
SAWB	0.004	0.67	0.154	0.37	0.044	0.51	0.192	-0.34	0.074	0.46	0.310	0.27	0.465	-0.20	0.478	-0.19
SLBB	0.933	-0.02	0.162	0.36	0.236	0.30	0.580	-0.14	0.445	0.20	0.068	0.45	0.862	-0.05	0.680	0.11
SRWP	0.747	0.20	0.037	-0.90	0.624	0.30	0.624	0.30	0.747	0.20	0.505	0.40	0.718	-0.22	0.624	0.30
TBCB	<0.001	0.75	0.101	-0.41	0.063	-0.46	0.439	0.20	0.013	-0.59	<0.001	0.73	0.241	0.30	0.758	-0.08
TBHB	0.982	-0.01	0.366	0.26	0.004	0.71	0.110	-0.45	0.487	-0.20	0.197	0.37	0.586	-0.16	0.692	0.12
TBKA	0.081	0.46	0.771	0.08	0.050	0.51	0.226	-0.33	0.198	-0.35	0.010	0.64	0.003	-0.71	0.830	0.06
TBLB	0.715	0.10	0.343	-0.25	0.772	0.08	0.837	-0.05	0.139	-0.37	0.248	0.30	0.475	-0.19	0.837	-0.05
TBNP	0.344	-0.26	0.187	-0.36	0.004	0.70	0.899	-0.04	0.007	-0.66	0.020	0.59	0.587	-0.15	0.398	0.24
TBOT	0.079	0.47	0.550	-0.17	0.017	0.60	0.869	-0.05	0.011	-0.64	0.081	0.46	0.361	-0.25	0.550	-0.17
VBSP	0.963	-0.01	0.837	-0.05	0.660	0.12	0.593	0.14	0.574	0.15	0.081	0.44	0.533	0.16	0.680	0.11

APPENDICES

Appendix 3. Statistical results from Spearman Rank Correlations on regional trends.

Metals	*p-value*	*rho*
Arsenic (As)	0.537	0.13
Cadmium (Cd)	0.052	-0.39
Copper (Cu)	0.666	0.09
Lead (Pb)	0.968	-0.01
Mercury (Hg)	0.385	-0.18
Nickel (Ni)	0.248	0.24
Tin (Sn)	0.825	-0.05
Zinc (Zn)	0.870	0.03

Appendix 4. 2017 metal tissue concentrations (µg/g dry weight) compared to site water salinity (ppt) concentrations.



U.S. Department of Commerce

Gina M. Raimondo, *Secretary*

National Oceanic and Atmospheric Administration

Richard W. Spinrad, *Under Secretary for Oceans and Atmosphere*

National Ocean Service

Nicole LeBoeuf, *Assistant Administrator for Ocean Service and Coastal Zone Management*

The mission of the National Centers for Coastal Ocean Science is to provide managers with scientific information and tools needed to balance society's environmental, social and economic goals. For more information, visit: http://www.coastalscience.noaa.gov/