

NATIONAL STATUS AND TRENDS, MUSSEL WATCH PROGRAM A 2017 Assessment of Legacy Organic Contaminants in the Gulf of Mexico



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Reviewers:

1. Dr. Erik Davenport
2. Dr. Tony Pait
3. Dr. Ed Wirth

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NATIONAL STATUS AND TRENDS, MUSSEL WATCH PROGRAM

A 2017 Assessment of Legacy Organic Contaminants in the Gulf of Mexico

January 2024

Authors:

Lauren M. Swam^{1,2}, Mary M. Rider^{1,2}, Dennis A. Apeti¹

¹ NOAA National Ocean Service, National Centers for Coastal Ocean Science,
Stressor Detection and Impacts Division, Silver Spring, MD

² Consolidated Safety Services, Inc., Fairfax, VA, under contract to NOAA



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United States Department
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Ocean Service

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Nicole LeBoeuf
Assistant Administrator

EXECUTIVE SUMMARY

In 2017, the National Oceanic and Atmospheric Administration (NOAA) Mussel Watch Program (MWP) conducted an assessment of the presence, distribution, and concentrations of legacy organic contaminants in Eastern oysters (*Crassostrea virginica*) from the Gulf of Mexico's coastal waters. To accomplish this, the MWP utilized a sentinel-based monitoring approach by collecting and analyzing bivalves as surrogates for coastal water pollution. Mussels and oysters are sessile organisms that filter and accumulate particles from water; therefore, measuring contaminant levels in their tissue is a good indicator of local chemical contamination. The oyster samples were measured for 9 legacy compound groups, including butyltins, chlordanes, chlorobenzenes, dichlorodiphenyltrichloroethane (DDTs), dieldrins, endosulfans, hexachlorocyclohexane (HCHs), polycyclic aromatic hydrocarbons (PAHs), and polychlorinated biphenyls (PCBs) in addition to 2 individual contaminants, chlorpyrifos and mirex. Oyster tissue samples from 44 monitoring sites across the Gulf Coasts of Florida, Alabama, Mississippi, Louisiana, and Texas were analyzed in this study.

The results indicated that legacy organic contaminants are present at varying concentrations in waters of the Gulf of Mexico. There were no sites within this analysis that were devoid of chemical contamination, emphasizing the ubiquity of these contaminants in coastal waters. The accumulation of legacy organic contaminants in organisms are often analyte and location-dependent. In this study, results indicated that the presence and concentration of a specific contaminant are heavily influenced by its chemistry, sources, fate, and transport. This information is useful to coastal resource managers because the long-term nature and geographic spread of the dataset can compliment data from other regional monitoring programs.

Broadly, the MWP provides unique data vital to evaluating the health of the Nation's coasts through temporal and spatial evaluation of chemical contamination. Studies such as this not only provide needed data and information for the MWP but also address contamination data gaps that are relevant to coastal managers as they develop long-term policies to protect ecosystem services provided by the coastal environment within the Gulf of Mexico.

KEY FINDINGS

- Legacy organic contaminants in oyster tissue were detected at low concentrations compared to historical national MWP data
- The only significantly increasing site-specific trend over time was Total Chlorobenzenes at site TBLB (Terrebonne Bay Lake Barre)
- All legacy organic contaminant compound groups showed significantly decreasing regional trends at historic MWP sites except for Total Chlorobenzenes, which did not exhibit a significant temporal trend
- Total Dieldrins, Total Endosulfans, and Mirex were not detected at any sites analyzed in the Gulf of Mexico in 2017

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COMMONLY USED ACRONYMS

AL	Alabama
DDT	dichlorodiphenyltrichloroethane
dw	dry weight
EPA	Environmental Protection Agency
FL	Florida
g	gram
HCH	hexachlorocyclohexane
LA	Louisiana
MDL	method detection limit
MS	Mississippi
MWP	Mussel Watch Program
NCCOS	National Centers for Coastal Ocean Science
ng	nanogram
NOAA	National Oceanic and Atmospheric Administration
NS&T	National Status and Trends
PAH	polycyclic aromatic hydrocarbon
PCB	polychlorinated biphenyls
SD	standard deviation
TX	Texas
µg	microgram
US	United States
ww	wet weight

Introduction

1.0 HISTORY OF MUSSEL WATCH PROGRAM

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

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

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

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



Figure 1. National Mussel Watch sites.

2.0 INTRODUCTION

The MWP has long-term monitoring sites spanning the Gulf of Mexico coast; a subset of these were analyzed in this study (Figure 2). The Gulf of Mexico is an expansive waterbody that receives waters from more than 150 rivers, including the Mississippi, Ohio, and Missouri Rivers, and runoff from 31 of the 50 states (Kim et al., 1999; Mitsch et al., 2001). The presence of many barrier islands and peninsulas, including the 130-mile (210 km) Padre Island along the Texas coast, and the many inlets, bays, and lagoons throughout the region cause this coastline to be very complex. These landforms and expansive marshland protect the numerous bays and inlets by acting as a barrier to oncoming waves, but they also serve to entrain sediments from upland areas. 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 climate is considered a humid subtropical habitat, and as such, the region is vulnerable to extreme weather events including hurricanes and severe thunderstorms. The physiography, climate, and hydrology in the Gulf of Mexico provide natural conditions that support a rich and abundant diversity of plant and animal communities in the basin. The Gulf of Mexico region is highly productive both ecologically and economically. The marshlands along the Louisiana and Texas coasts provide breeding grounds and nurseries for marine life that drive the fishing and shrimping industries. Many estuaries along the coast also contain 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. Apalachicola Bay alone provides approximately 90% of Florida's oyster harvest and 10% of the total U.S. harvest of those species (FDEP, 2013). The regional economy is also dominated by industries related to the energy, petrochemical, and tourism sectors. 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 United States (U.S.) petrochemical industry. This region also features other important industries including aerospace and biomedical research sectors, as well as established agricultural industries. The water quality that sustains this high productivity has been affected by a combination of natural and mainly anthropogenic factors such as growing urbanization, industries, and agriculture (Kim et al., 1999; LaMourie et al., 2023; Sunkara et al., 2023). Chemical contaminants in the Gulf of Mexico may be caused by nonpoint sources such as river input and long-range atmospheric deposition (Vazquez-Botello et al., 2004). Significant point sources of toxic hydrocarbon related contaminants in the Gulf region are seepage from its abundant oil reserves and oil spills such as Deepwater Horizon in 2010 (Apeti et al., 2013).

Coastal chemical pollution along the Gulf of Mexico coast of the U.S. has been assessed and monitored by the National Oceanic and Atmospheric Administration (NOAA) National Status and Trends Program (NS&T) for resource and ecosystem management and protection since 1986 (Kimbrough et al., 2008; Battista et al., 2006). Statewide water quality monitoring efforts conducted by the Texas Parks and Wildlife Department, the Louisiana Department of Wildlife and Fisheries, the Mississippi Department of Marine Resources, the Alabama Department of Marine Resources, and the Florida Department of Environmental Protection, among others, occur frequently in the region. However, the NS&T Program is one of the few, perhaps only, continuous monitoring programs for chemical contaminants in the region. These studies have provided relevant data and information to coastal managers and the scientific community with a primary focus on legacy organic contaminants. These legacy pollutants are routinely monitored and regulated and include trace elements ("trace metals") and persistent organic pollutants such as butyltins, chlordanes, chlorobenzenes, dichlorodiphenyltrichloroethane (DDTs), dieldrins, endosulfans, hexachlorocyclohexane (HCHs), mirex, polycyclic aromatic hydrocarbons (PAHs), and polychlorinated biphenyls (PCBs).

In 2017, the MWP conducted a comprehensive assessment of the magnitude and distribution of legacy organic contaminants along the US coastline in the Gulf of Mexico (Figure 2). The study was designed within the framework of the MWP regional monitoring approach, which balances short-term flexibility in study design against the cost of broad contaminant surveys. The objectives of study were to 1) assess the magnitude and distribution of persistent organic contaminants historically assessed by the MWP; 2) compare coastal contamination in the Gulf of Mexico in 2017 to previous MWP studies in the same and other regions; and 3) make data electronically available to coastal resource managers in the Gulf of Mexico region.

3.0 METHODS

3.1 Study Area and Sampling Design

The MWP has 85 long-term monitoring sites in coastal waters in the Gulf of Mexico. Monitoring sites were historically selected in locations with abundant bivalve populations to allow for repetitive sampling and to convey information about the degree of chemical contamination in the general area over time. The sites were not randomly selected nor designed to target specific pollution sources.

Sample collection at these sites was conducted by TDI Brooks International following standard protocols utilized by the MWP (Apeti et al., 2012) in primarily September - November 2017. Oyster samples (*Crassostrea virginica*) were collected via hand picking, oyster tongs, or oyster dredging from 44 sites (Figure 2; Table 1). Although 61 sites were identified for collection throughout the Gulf of Mexico, only 44 sites yielded sufficient oysters to allow for analysis (Figure 2; Table 1). Out of the 61 identified sites, APEB was the only site not attempted for sampling in 2017 based on the recent and realized absence of oysters throughout the area. Analyses of butyltins (4 compounds) were only conducted for 41 sites due to limited quantity of oyster tissue available for laboratory analysis.

3.2 Analytical Methods

Chemical analyses in oyster tissue followed procedures routinely used in the NOAA NS&T Program. Detailed descriptions of analytical methods for legacy organic contaminants analyzed in this study can be found in Kimbrough et al., (2007). Laboratory results were subjected to regular NS&T performance-based quality assessment and quality control procedures for data accuracy and precision. Legacy organic contaminant data is presented in nanograms of contaminant per gram of dry tissue (ng/g dw) with the exception of butyltins which are reported as ng Sn/g dw, as they are organotin compounds. The laboratory at which analyses were conducted has changed over time; however, inter-laboratory calibrations have been conducted with low concern. In the MWP, legacy organic contaminants were analyzed by the Geochemical and Environmental Research Group or the Department of Oceanography at Texas A&M University in College Station, TX from 1986 - 2000 and have been analyzed by TDI-Brooks Int. since 2000. The data in this report will be archived and publicly available at NOAA's National Centers for Environmental Information (NCEI) (<https://www.ncei.noaa.gov/>). For all contaminant classes, a background summary ("Chemical Description") and analysis summary ("Results Summary") can be found within this document.

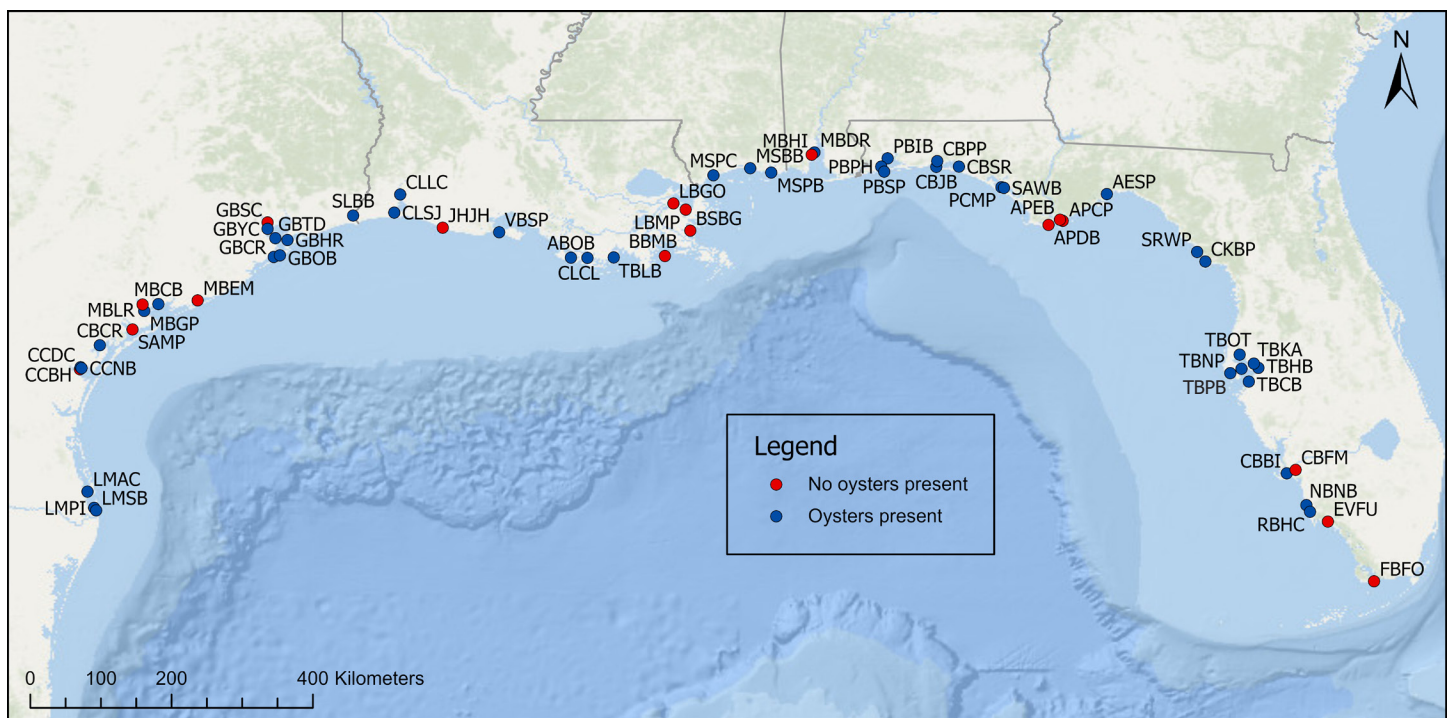


Figure 2. Map of MWP sites in the Gulf of Mexico region sampled in 2017 and their respective collection status. Esri, GEBCO, DeLorme, NaturalVue

3.3 Data Analysis

Data management and analysis were conducted using a combination of R version 4.1.2 (R Core Team, 2013), Microsoft Excel (2016), ArcGIS (ESRI, 2011), and JMP12 Software (JMP, 2022).

Concentrations of all compounds were blank corrected and any values below the method detection limit (MDL) were categorized as undetected and were assigned a value of 0. The MDL is defined as the lowest concentration able to be detected by the analytical instrument or method. Legacy organic compounds are reported as the arithmetic sum of all the individual compounds or congeners within a contaminant group. For historical analysis in this report, these sums were the total of a subset of the compounds tested in 2017. Historic analysis dates back as early as possible while still including the same subset of compounds over time. Data for each contaminant group from sites in 2017 were statistically analyzed at site and regional scales using a Spearman's Rank test. Spearman's rank correlation was used to evaluate whether concentrations co-varied predictably as a function of time (Zar, 1998), and therefore imply a significant increasing or decreasing temporal trend. The Spearman's rank correlation procedure is a nonparametric technique that is free of assumptions about concentrations being normally distributed with a common variance about sites (Kimbrough et al., 2008). The variables used for the Spearman's test were year and concentration sums. Regional temporal trends were also depicted using a three-point moving average of the yearly mean for all sites combined.

To provide context of the magnitude of concentrations detected in 2017, contaminant concentrations in tissue were evaluated against the US FDA maximum permissible action levels for molluscan shellfish consumption for human health protection (FDA, 2011) and the US EPA Screening Values (SVs) (EPA, 2000) for recreational fishers. The US FDA Action and Tolerance Levels (Table A2) represent concentration limits at which the US FDA will take legal action to remove shellfish from the market to protect human health (FDA, 2011). The US EPA SVs (Table A2) were developed to provide guidance to state, local, regional and tribal environmental health officials for their contaminant monitoring programs and for issuing fish and shellfish consumption advisories (EPA, 2000). The SVs represent a threshold concentration of concern for a chemical contaminant in fish and shellfish tissue for a critical toxic or a carcinogenic effect in humans. In cases where there were both carcinogenic and non-carcinogenic SVs available, the SV for the carcinogenic effects was used. Values higher than the SVs provide an indication of where more intensive site-specific monitoring and/or evaluation of human health risks should be conducted (EPA, 2000). The US EPA and US FDA guideline values are reported on wet tissue weights basis, and where available for comparison, concentrations of chemical contaminants in oysters from the Gulf of Mexico were converted to wet weights using the percent dry fraction (Table A1).

Additionally, contamination levels detected at sites in 2017 were compared to national long-term NOAA NS&T monitoring data. The NS&T data used for comparison comprises oyster tissue sample concentrations collected by the national MWP since the initiation of the program in 1986. This comparison was done using a multivariate cluster analysis (the Ward method) on the sums of concentrations within each contaminant class for legacy organic compounds. This analysis clusters contaminant concentrations into significantly different groups such that values contained within a group are more like each other than any other value of a different group. The categories derived from the clusters were not representative measurements that have exceeded any regulatory thresholds; rather, they denoted concentrations that were significantly higher than the preceding category. For each contaminant class, sites were clustered into three groups to represent high contamination (value=3), medium contamination (value=2), and low contamination (value=1) relative to all data collected within the MWP. If the resulting high (value = 3) cluster had fewer than 10 data points, those points were temporarily removed as "outliers" (due to the large sample size of this dataset), the data were re-clustered, and the outliers were added back in as part of cluster 3 before resulting boxplots were created. Clustering results were reported in a map on the Summary page of each contaminant class. Any instances of "not detected" contaminants were differentiated from low contamination (value=1) on the map.

To provide further context, cluster analysis results were summarized for all classes per site assessed in this study in two heatmaps showing (1) cluster results of the 2017 study sites based on relative concentrations found within the sites assessed in this study and (2) cluster results of the 2017 study sites based on national long-term NOAA NS&T monitoring data. The clustering process for comparing 2017 oyster tissue contaminant data only to sites analyzed within the 2017 Gulf of Mexico study was similar to the process described for the historic MWP clustering. The contaminant sums were similarly clustered into high, medium, and low contamination, but "outliers" (as described previously) were not removed to preserve results due to the smaller sample size. Then, cluster values of each contaminant class for each site were summed and were again clustered using the Ward Method to generate five groups of sites with statistically different degrees of overall contamination within this study (not detected, low, medium, high, and very high) and were presented in a map. This clustering was conducted with respect to sites sampled in 2017 and to all historic MWP contamination analyzed in oysters. Total summed cluster values for the historic MWP dataset were normalized by the maximum value possible at that site (i.e., $(\text{sum cluster values})/(\# \text{ chemical classes analyzed } * 3) \times 100$) (Table A4; Table A5).

Methods

Table 1. Mussel Watch sites selected for 2017 Gulf of Mexico survey. ● signifies the site was analyzed for legacy organic contaminants in 2017. 61 sites were attempted to be sampled, 44 sites yielded oysters to be analyzed. FL - Florida, AL - Alabama, MS - Mississippi, LA - Louisiana, TX - Texas.

Site	State	General Location	Specific Location	Latitude	Longitude	Tissue Sampled?
ABOB	LA	Atchafalaya Bay	Oyster Bayou	29.25550	-91.13617	●
AESP	FL	Apalachee Bay	Spring Creek	30.06333	-84.32200	●
APCP	FL	Apalachicola Bay	Cat Point Bar	29.72417	-84.88417	
APDB	FL	Apalachicola Bay	Dry Bar	29.67250	-85.06567	
APEB	FL	Apalachicola Bay	East Bay	29.73830	-84.91850	
BBMB	LA	Barataria Bay	Middle Bank	29.27667	-89.94200	
BSBG	LA	Breton Sound	Bay Gardene	29.59800	-89.62083	
CBBI	FL	Charlotte Harbor	Bird Island	26.51433	-82.03450	●
CBCR	TX	Copano Bay	Copano Reef	28.14200	-97.12800	●
CBFM	FL	Charlotte Harbor	Fort Meyers	26.55833	-81.92283	
CBJB	FL	Choctawhatchee Bay	Joe's Bayou	30.41083	-86.49083	●
CBPP	FL	Choctawhatchee Bay	Postil Point	30.48233	-86.47933	●
CBSR	FL	Choctawhatchee Bay	Off Santa Rosa	30.41200	-86.20367	●
CCBH	TX	Corpus Christi	Boat Harbor	27.83617	-97.38017	
CCDC	TX	Corpus Christi	Doyle City	27.86183	-97.37262	●
CCNB	TX	Corpus Christi	Nueces Bay	27.85217	-97.35983	●
CKBP	FL	Cedar Key	Black Point	29.20667	-83.06950	●
CLCL	LA	Caillou Lake	Caillou Lake	29.25317	-90.92667	●
CLLC	LA	Calcasieu Lake	Lake Charles	30.05867	-93.30750	●
CLSJ	LA	Calcasieu Lake	St. Johns Island	29.82900	-93.38400	●
EVFU	FL	Everglades	Faka Union Bay	25.90233	-81.51233	
FBFO	FL	Florida Bay	Flamingo	25.14117	-80.92367	
GBCR	TX	Galveston Bay	Confederate Reef	29.26333	-94.91633	●
GBHR	TX	Galveston Bay	Hanna Reef	29.48033	-94.74183	●
GBOB	TX	Galveston Bay	Offatts Bayou	29.28400	-94.83633	●
GBSC	TX	Galveston Bay	Ship Channel	29.70450	-94.99300	
GBTD	TX	Galveston Bay	Todd's Dump	29.50300	-94.89600	●
GBYC	TX	Galveston Bay	Yacht Club	29.62200	-94.99583	●
JHJH	LA	Joseph Harbor Bayou	Joseph Harbor Bayou	29.63683	-92.76683	
LBGO	LA	Lake Borgne	Gulf Outlet	29.94483	-89.83533	
LBMP	LA	Lake Borgne	Malheureux Point	29.86700	-89.67850	

Methods

Table 1 cont. Mussel Watch sites selected for 2017 Gulf of Mexico survey. ● signifies the site was analyzed for legacy organic contaminants in 2017. 61 sites were attempted to be sampled, 44 sites yielded oysters to be analyzed. FL - Florida, AL - Alabama, MS - Mississippi, LA - Louisiana, TX - Texas.

Site	State	General Location	Specific Location	Latitude	Longitude	Tissue Sampled?
LMAC	TX	Lower Laguna Madre	Arroyo Colorado	26.28250	-97.28533	●
LMPI	TX	Lower Laguna Madre	Port Isabel	26.07483	-97.19950	●
LMSB	TX	Lower Laguna Madre	South Bay	26.04317	-97.17600	●
MBCB	TX	Matagorda Bay	Carancahua Bay	28.66500	-96.38300	●
MBDR	AL	Mobile Bay	Dog River	30.59167	-88.03983	●
MBEM	TX	Matagorda Bay	East Matagorda	28.71117	-95.88333	
MBGP	TX	Matagorda Bay	Gallinipper Point	28.57883	-96.56300	●
MBHI	AL	Mobile Bay	Hollingers Is. Chan.	30.56333	-88.07500	
MBLR	TX	Matagorda Bay	Lavaca River Mouth	28.66033	-96.58450	
MSBB	MS	Mississippi Sound	Biloxi Bay	30.39250	-88.85750	●
MSPB	MS	Mississippi Sound	Pascagoula Bay	30.33600	-88.58917	●
MSPC	MS	Mississippi Sound	Pass Christian	30.30233	-89.32717	●
NBNB	FL	Naples Bay	Naples Bay	26.11183	-81.78517	●
PBIB	FL	Pensacola Bay	Indian Bayou	30.51667	-87.11167	●
PBPH	FL	Pensacola Bay	Public Harbor	30.41367	-87.19133	●
PBSP	FL	Pensacola Bay	Sabine Point	30.34983	-87.15467	●
PCMP	FL	Panama City	Municipal Pier	30.15117	-85.66300	●
RBHC	FL	Rookery Bay	Henderson Creek	26.02700	-81.73883	●
SAMP	TX	San Antonio Bay	Mosquito Point	28.34400	-96.71233	
SAWB	FL	St. Andrew Bay	Watson Bayou	30.14250	-85.63217	●
SLBB	LA	Sabine Lake	Blue Buck Point	29.79083	-93.90633	●
SRWP	FL	Suwannee River	West Pass	29.32917	-83.17417	●
TBCB	FL	Tampa Bay	Cockroach Bay	27.68100	-82.51767	●
TBHB	FL	Tampa Bay	Hillsborough Bay	27.85483	-82.39467	●
TBKA	FL	Tampa Bay	Peter O. Knight Airport	27.90967	-82.45383	●
TBLB	LA	Terrebonne Bay	Lake Barre	29.25950	-90.59433	●
TBNP	FL	Tampa Bay	Navarez Park	27.78717	-82.75400	●
TBOT	FL	Tampa Bay	Old Tampa Bay	28.02367	-82.63283	●
TBPB	FL	Tampa Bay	Papys Bayou	27.84433	-82.61150	●
VBSP	LA	Vermilion Bay	Southwest Pass	29.57950	-92.05100	●

Results - Total Butyltins

4.0 RESULTS - TOTAL BUTYLTINS

4.1 Butyltins Chemical Description

For this document, Total Butyltins analyzed in 2017 is the sum of 4 organometallic compounds and Total Butyltins analyzed historically is the sum of 3 organometallic compounds (Table 2). The parent butyltin compound is tributyltin and the other butyltin compounds are its less toxic transformation products. Tributyltin has had a variety of uses ranging from a biocide in antifouling paints to a catalyst and an ingredient in glass coatings (Bennett, 1996; Birchenough et al., 2002). In the late 1960s, tributyltin was incorporated into an antifouling polymer paint system, quickly becoming one of the most effective paints used on boat hulls (Birchenough et al., 2002). Beginning in 1989, the use of tributyltin as an antifouling agent was banned in the US on non-aluminum vessels smaller than 25 meters in length (Gibbs and Bryan, 1996). However, the continued use of tributyltin on ships and other antifouling paint applications increased the ubiquity of the compound in aquatic environments. Thus, tributyltin and its metabolites continue to be detected in many components of coastal and marine ecosystems in the US.

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

In the aquatic environment, tributyltin is degraded by microorganisms and sunlight into monobutyltin and dibutyltin (Bennett, 1996). Experiments have shown that the half-life of tributyltin is on the order of days and degradation to monobutyltin takes approximately a month. However, in deeper anoxic sediments, the half-life of tributyltin appears to be on the order of 2-4 years or longer (Batley, 1996). Although banned decades ago, tributyltin and its metabolites continue to be detected in many components of the environment. While organotin compounds are readily bioaccumulated by aquatic organisms from water, there is no evidence for biomagnification up the food chain (WHO, 1980). Tributyltin is sparingly soluble in water and associates readily with suspended particles in the water column. Butyltins are persistent in the aquatic environment and accumulate in sediment; therefore, they will continue to affect the aquatic environment (Gibbs and Bryan, 1996; EPA, 2003). Releases of organotins to the atmosphere are not significant due to their low vapor pressure and rapid photodegradation.

Note: Total Butyltins concentration from sites TBKA in 1989 and PCMP in 1989 were removed from figures in the Historical Context section for better visualization due to their outlying concentration (1,883.00 and 1,824.00 ng/g Sn, respectively). This data point remained in the dataset for all statistical analyses.

Table 2. Butyltin compounds composing the "Total Butyltins" summation for the 2017 Gulf of Mexico survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2017	Analyzed in Historical Data
Monobutyltin	●	●
Dibutyltin	●	●
Tributyltin	●	●
Tetrabutyltin	●	

Results - Total Butyltins

4.2 Magnitude and Distribution of Butyltins in 2017

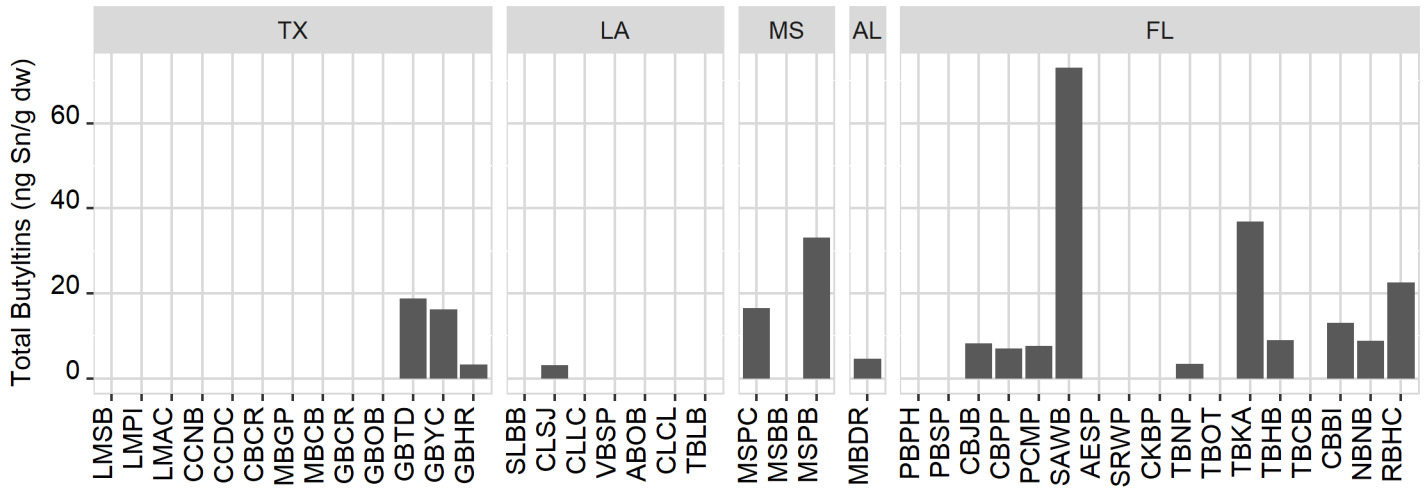


Figure 3. Bar graph showing magnitude of Total Butyltin concentration sums detected in oyster tissue in the Gulf of Mexico in 2017. Sites are listed geographically from west to east, following the coastline.

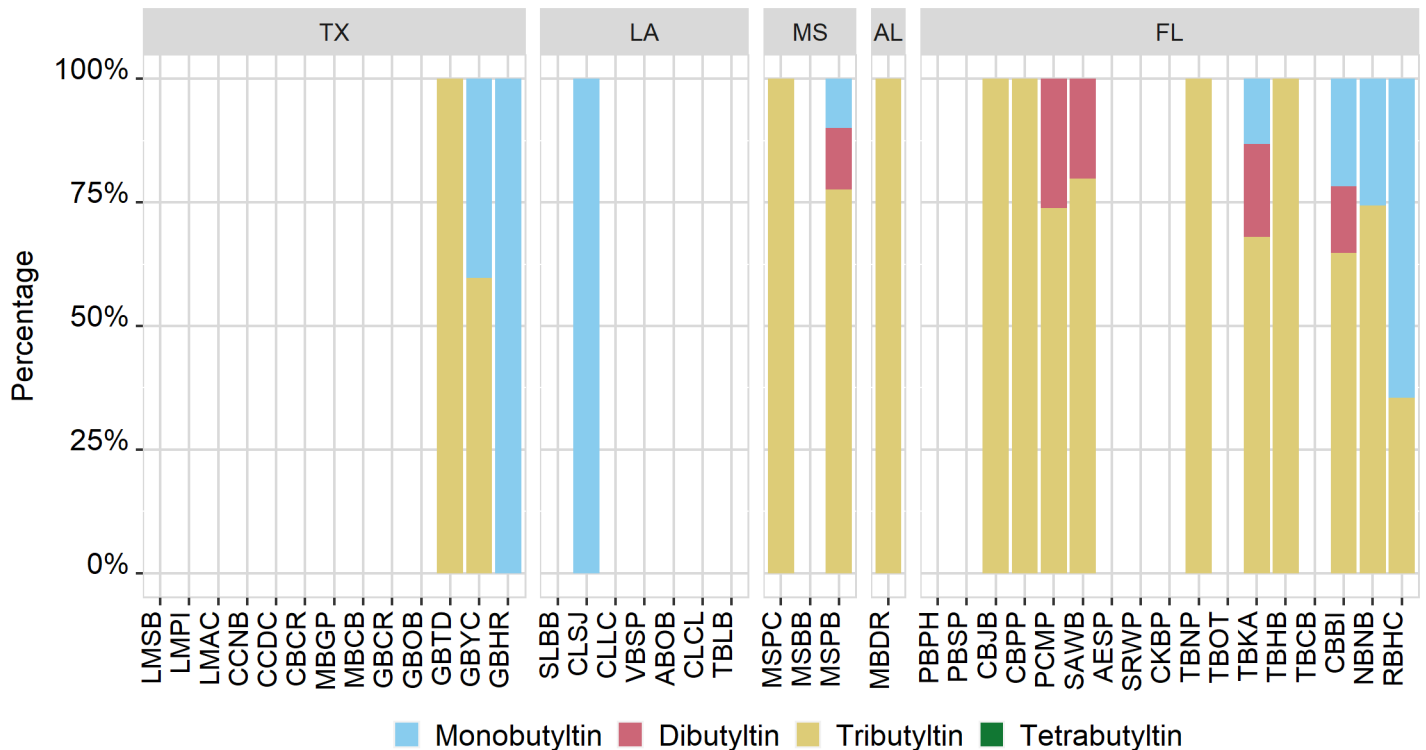


Figure 4. Percent composition of individual butyltin contaminants in Total Butyltin concentration sums per site in the Gulf of Mexico in 2017. Sites are listed geographically from west to east, following the coastline.

Results - Total Butyltins

4.3 Historical Context of Butyltins Magnitude and Distribution

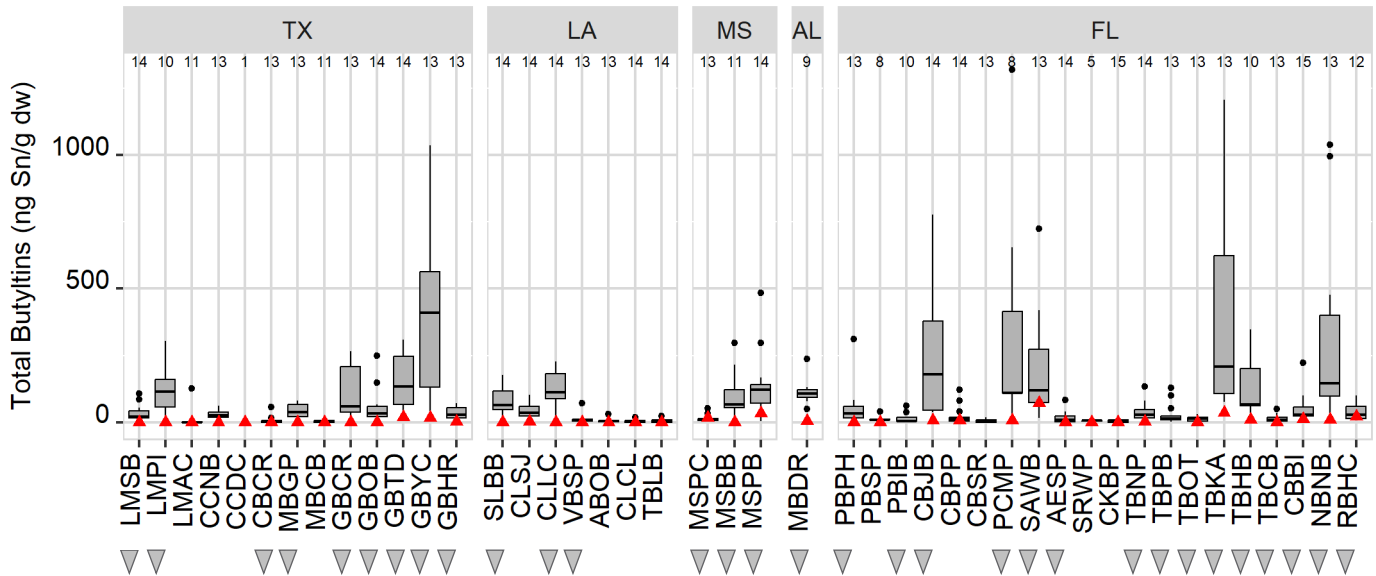


Figure 5. Total Butyltin concentration sums in 2017 (red triangles) compared to boxplots of the historic Total Butyltin concentration sums for each site (1989 - 2012) and results of the trend analysis showing significant increasing or decreasing contamination trends over time (gray triangles). Sites are listed geographically from west to east, following the coastline. The number of years each site has been sampled since 1989 is noted at the top of the plot for each site.

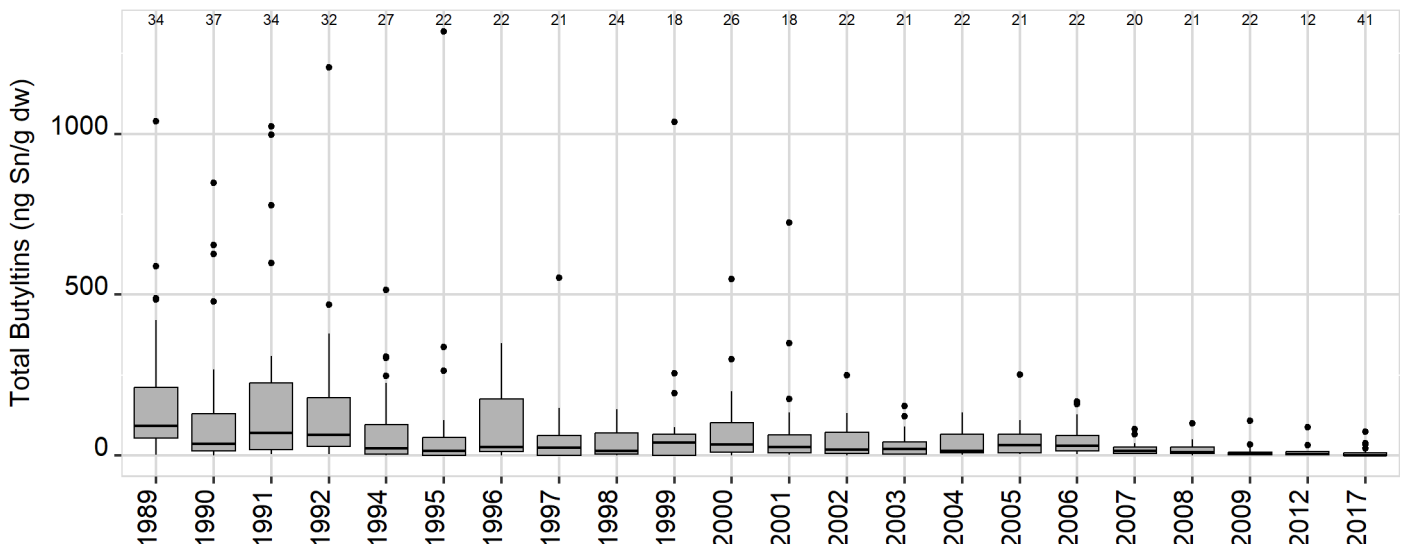


Figure 6. Boxplots representing the historic Total Butyltin concentration sums of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot for each year.

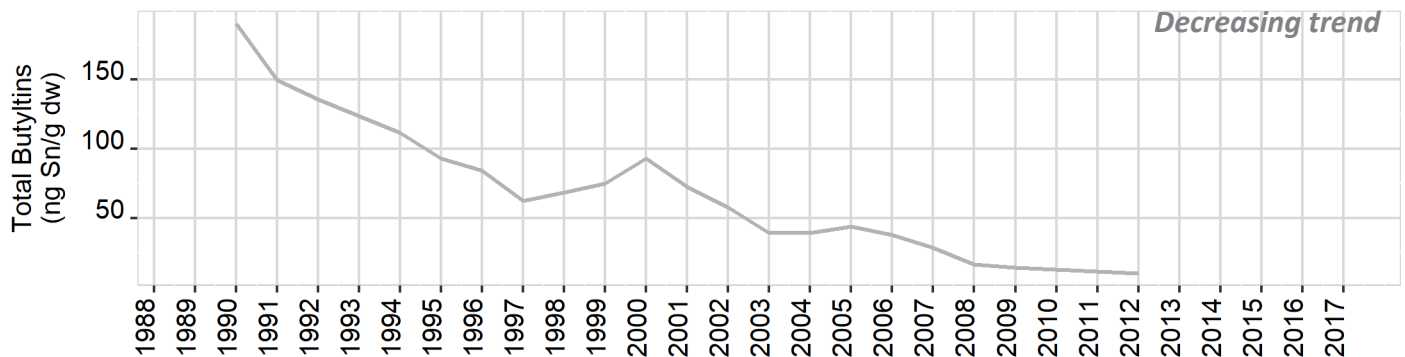


Figure 7. Three-point moving average of the yearly mean Total Butyltin concentration sums in the sites analyzed in this study.

Results - Total Butyltins

4.4 Butyltins Summary

2017 Results (sum of 4 compounds):

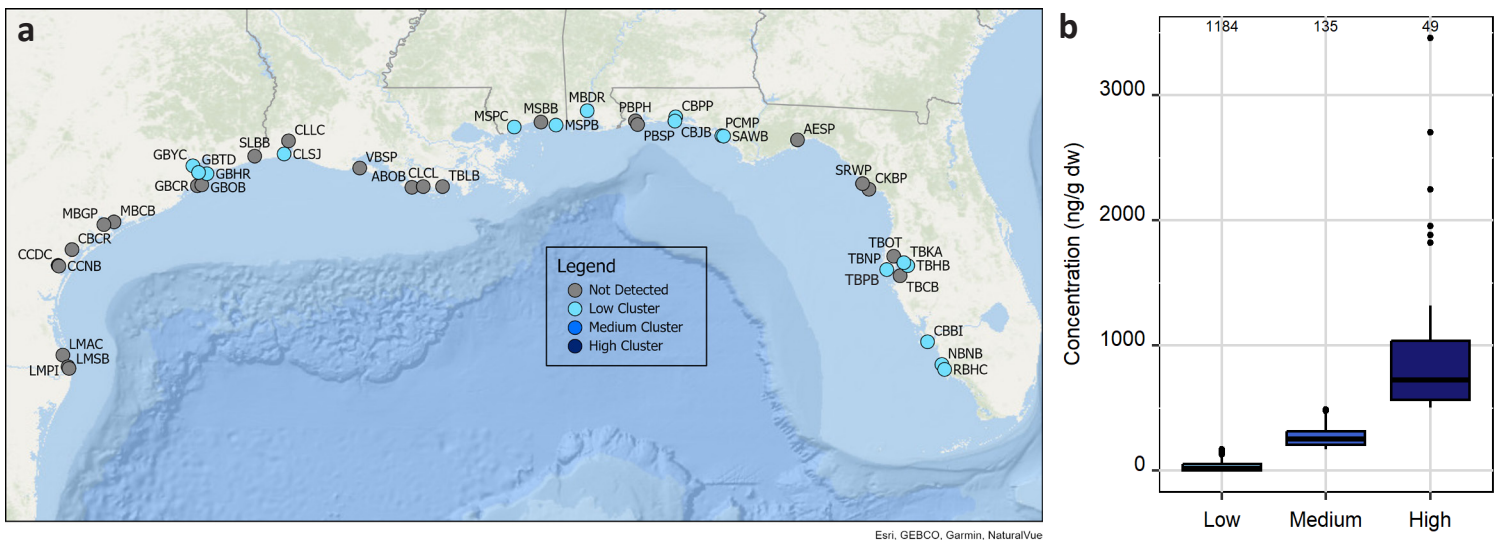
- Total Butyltins were detected at 41.5% of 41 sites analyzed
- Total Butyltins concentration sums stats (Figure 3; Figure 4):
 - Range: 3.08 – 73.06 ng/g Sn
 - Total Butyltins were not detected at 24 sites
 - Maximum concentration sum was detected at site SAWB (St. Andrew Bay, Watson Bayou)
 - Median: 0.00 ng/g Sn
 - Mean \pm SD: 6.96 \pm 13.93 ng/g Sn

Historical Context (sum of 3 compounds):

- 2.4% sites were above their historic median concentrations in 2017
- 31 sites showed decreasing temporal trends of Total Butyltin concentration sums at $\alpha = 0.05$ (Figure 5; Figure 6; Table A6)
- 0 sites showed increasing temporal trends of Total Butyltin concentration sums at $\alpha = 0.05$ (Figure 5; Figure 6; Table A6)
- There was a significant decreasing regional trend over time for sites in 2017 ($p < 0.001$, $\rho = -0.91$) (Figure 7)
- The percentage of sites in the Gulf of Mexico in 2017 grouped into each cluster based on historic data (Figure 8):
 - 100% of sites in low cluster (0.0 – 170.5 ng/g Sn)
 - 0% of sites in medium cluster (172.0 – 488.0 ng/g Sn)
 - 0% of sites in high cluster (502.3 – 3,456.7 ng/g Sn)

General Observations:

- Total Butyltin concentration sums were low or not detected throughout the Gulf of Mexico (Figure 8).
- The continued presence of tributyltin (the butyltin parent compound) suggests that there continues to be inputs into the environment, perhaps from resuspended sediments or the refurbishing of historic vessels, since tributyltin has been banned in the US for use on most vessels since 1989 (Figure 4).
- Total Butyltin concentration sums were well below the US EPA recommended screening values for both recreational and subsistence fishers (Table A2).



Results - Total Chlordanes

5.0 RESULTS - TOTAL CHLORDANES

5.1 Chlordanes Chemical Description

For this document, Total Chlordanes analyzed in 2017 is the sum of 7 compounds and Total Chlordanes analyzed historically is the sum of 4 compounds (Table 3). Chlordane belongs to a group of organic pesticides called cyclodienes. Technical chlordane, an insecticide, was used in the US from 1948-1983 for agricultural and urban settings to control insect pests. It was also the predominant insecticide for the control of subterranean termites. Agricultural uses were banned in 1983 and all uses were banned by 1988. These compounds are some of the most ubiquitous contaminants measured by the Mussel Watch Program.

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

Removal from both soil and water sources is primarily by volatilization and particle-bound runoff. In air, chlordane degrades as a result of photolysis and oxidation. Chlordane exists in the atmosphere primarily in the vapor-phase, but the particle-bound fraction is important for long-range transport. Chlordane is prevalent in the Arctic due to the grasshopper effect and distributed in the food web (Hargrave et al., 1992). Chlordane binds to dissolved organic matter, further facilitating its transport in natural waters.

Note: Total Chlordanes concentration from site CBPP in 1987 was removed from figures in the Historical Context section for better visualization due to its outlying concentration (287.88 ng/g). This data point remained in the dataset for all statistical analyses.

Table 3. Chlordane compounds composing the "Total Chlordanes" summation for the 2017 Gulf of Mexico survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2017	Analyzed in Historical Data
Alpha-Chlordane	●	●
Trans-Nonachlor	●	●
Gamma-Chlordane	●	
Cis-Nonachlor	●	
Heptachlor	●	●
Heptachlor-Epoxide	●	●
Oxychlordane	●	

Results - Total Chlordanes

5.2 Magnitude and Distribution of Chlordanes in 2017

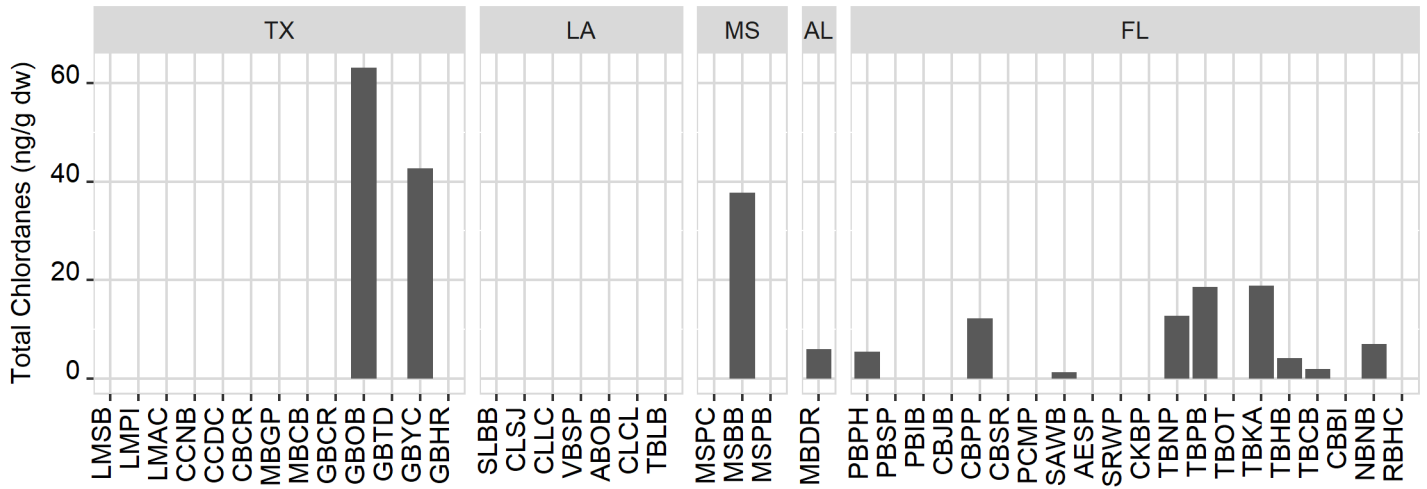


Figure 9. Bar graph showing magnitude of Total Chlordane concentration sums detected in oyster tissue in the Gulf of Mexico in 2017. Sites are listed geographically from west to east, following the coastline.

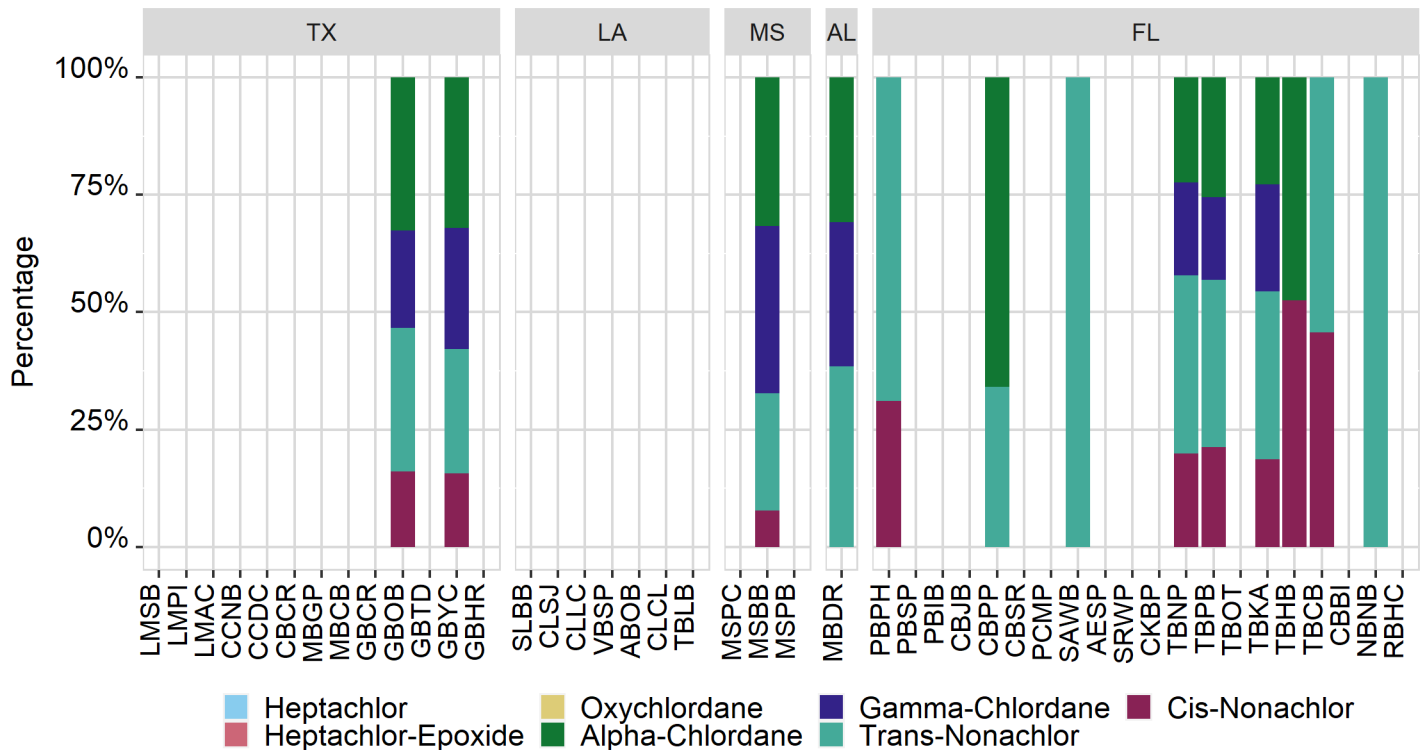


Figure 10. Percent composition of individual chlordane contaminants in Total Chlordane concentration sums per site in the Gulf of Mexico in 2017. Sites are listed geographically from west to east, following the coastline.

Results - Total Chlordanes

5.3 Historical Context of Chlordanes Magnitude and Distribution

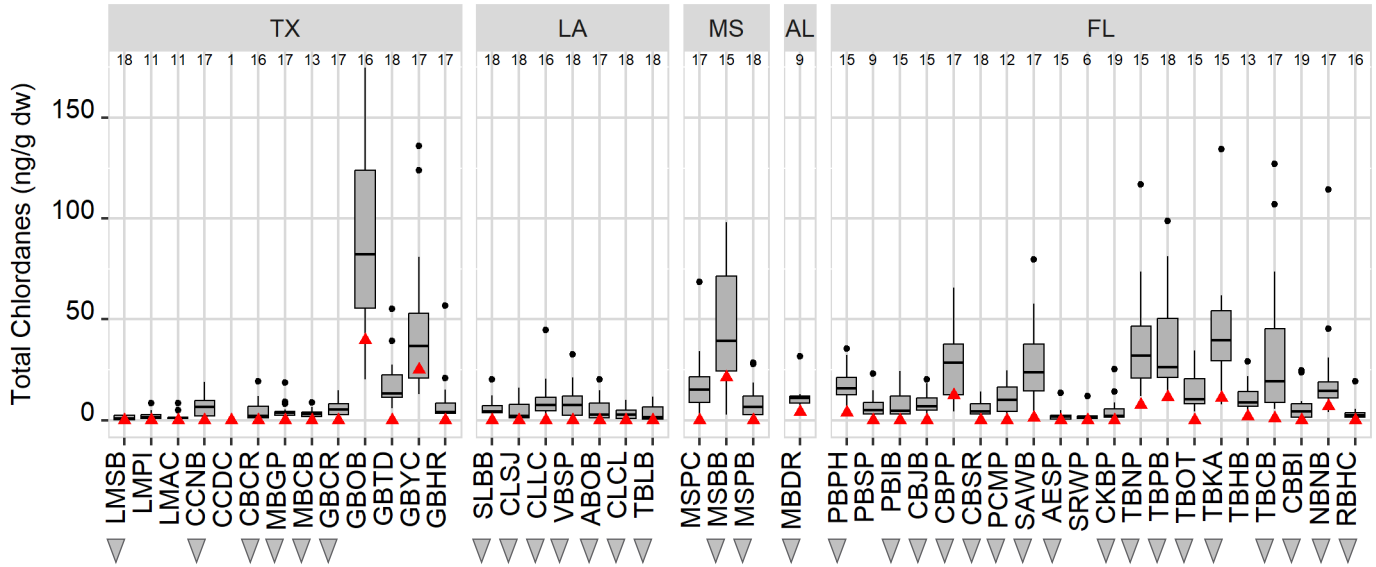


Figure 11. Total Chlordane concentration sums in 2017 (red triangles) compared to boxplots of the historic Total Chlordane concentration sums for each site (1986 - 2012) and results of the trend analysis showing significant increasing or decreasing contamination trends over time (gray triangles). Sites are listed geographically from west to east, following the coastline. The number of years each site has been sampled since 1986 is noted at the top of the plot for each site.

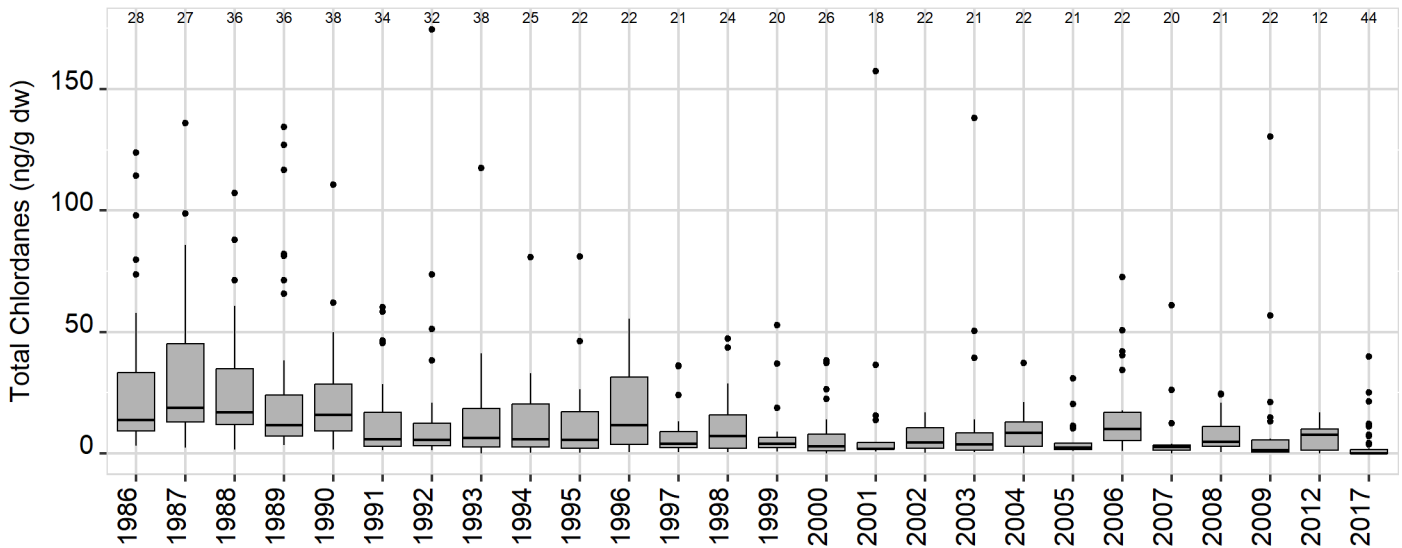


Figure 12. Boxplots representing the historic Total Chlordane concentration sums of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot for each year.

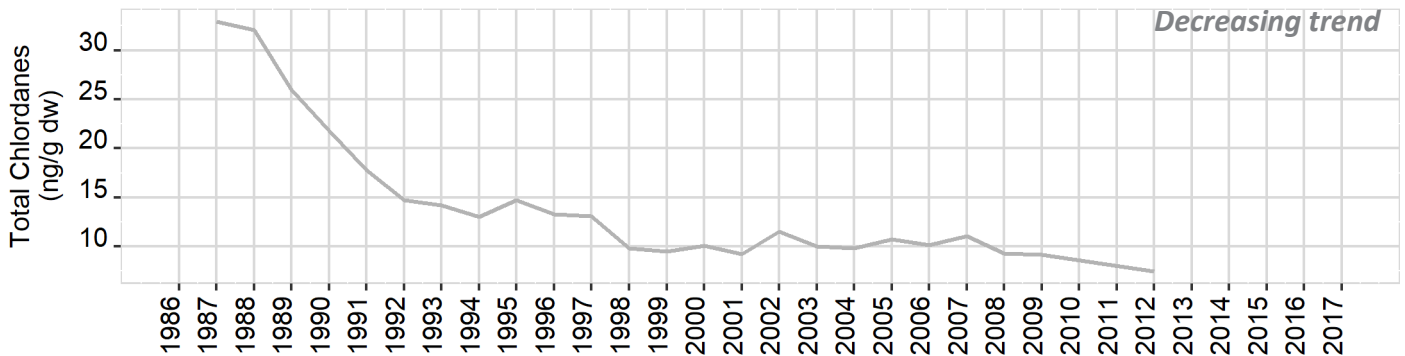


Figure 13. Three-point moving average of the yearly mean Total Chlordane concentration sums in the sites analyzed in this study.

Results - Total Chlordanes

5.4 Chlordanes Summary

2017 Results (sum of 7 compounds):

- Total Chlordanes were detected at 29.5% of 44 sites analyzed
- Total Chlordanes concentration sums stats (Figure 9; Figure 10):
 - Range: 1.24 – 63.15 ng/g
 - Total Chlordanes were not detected at 31 sites
 - Maximum concentration sum was detected at site GBOB (Galveston Bay, Offatts Bayou)
 - Median: 0.00 ng/g
 - Mean \pm SD: 5.27 \pm 12.91 ng/g

Historical Context (sum of 4 compounds):

- 0% sites were above their historic median concentrations in 2017
- 33 sites showed decreasing temporal trends of Total Chlordane concentration sums at $\alpha = 0.05$ (Figure 11; Figure 12; Table A6)
- 0 sites showed increasing temporal trends of Total Chlordane concentration sums at $\alpha = 0.05$ (Figure 11; Figure 12; Table A6)
- There was a significant decreasing regional trend over time for sites in 2017 ($p < 0.001$, $\rho = -0.78$) (Figure 13)
- The percentage of sites in the Gulf of Mexico in 2017 grouped into each cluster based on historic data (Figure 14):
 - 100% of sites in low cluster (0.0 – 43.5 ng/g)
 - 0% of sites in medium cluster (44.9 – 91.0 ng/g)
 - 0% of sites in high cluster (98.0 – 287.9 ng/g)

General Observations:

- Total Chlordane concentration sums were low or not detected throughout the Gulf of Mexico (Figure 14).
- Total Chlordane concentration sums were well below the US EPA recommended screening values for both recreational and subsistence fishers (Table A2).

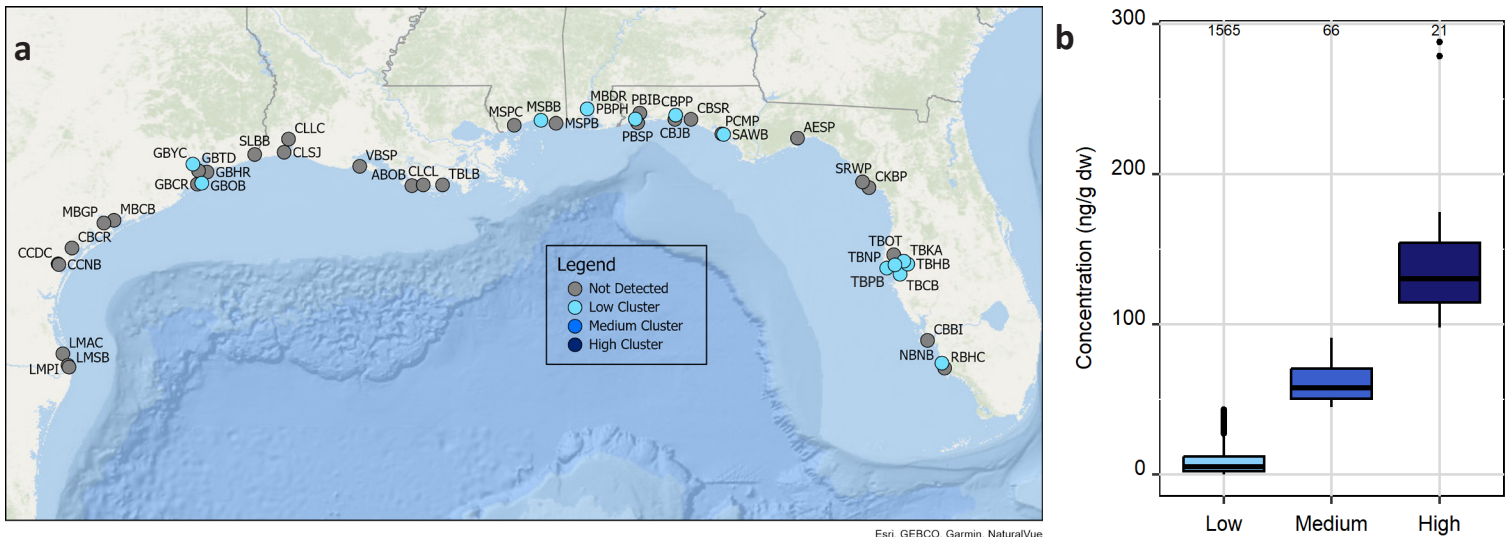


Figure 14. (a) Total Chlordane concentration sums in 2017 compared to the (b) historic national MWP *Crassostrea virginica* Total Chlordane concentration sums (ng/g dw). 129 national sites sampled between 1986 - 2017 for a total of 1652 samples.

Results - Total Chlorobenzenes

6.0 RESULTS - TOTAL CHLOROBENZENES

6.1 Chlorobenzenes Chemical Description

For this document, Total Chlorobenzenes analyzed in 2017 is the sum of 5 compounds and Total Chlorobenzenes analyzed historically is only 1 compound (Table 4). Chlorobenzenes belong to the family of organic halogen compounds and are widely used as degreasers, chemical intermediates and solvents for pesticide formulations, adhesives, paints, polishes, dyes and drugs. For example, pentachloroanisole comes from the biomethylation of pentachlorophenol, a chemical used as a general biocide, fungicide, bactericide, herbicide, molluscicide, algacide and insecticide by agriculture and other industries including textiles, paints, oil drilling and forestry (Canada, 2012). Although chlorobenzenes are not banned, their production has decreased by 60% since the peak in 1960 due primarily to regulations on DDT, in which it was used as part of the manufacturing process (ATSDR, 1990a).

There is inadequate evidence to classify chlorobenzenes as carcinogens, however, animal studies indicate that livers, kidneys and the central nervous system are affected by exposure to chlorobenzenes (ATSDR, 1990a).

Chlorobenzenes typically evaporate rapidly into the air or are broken down by bacteria and are not considered to build up in the food chain.

Note: Total Chlorobenzenes concentration from sites TBOT in 2006 and CLCL in 2007 were removed from figures in the Historical Context section for better visualization due to their outlying concentration (5.05 and 3.34 ng/g, respectively). These data points remained in the dataset for all statistical analyses.

Table 4. Chlorobenzene compounds composing the "Total Chlorobenzenes" summation for the 2017 Gulf of Mexico survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2017	Analyzed in Historical Data
1,2,3,4-Tetrachlorobenzene	●	
1,2,4,5-Tetrachlorobenzene	●	
Hexachlorobenzene	●	●
Pentachloroanisole	●	
Pentachlorobenzene	●	

Results - Total Chlorobenzenes

6.2 Magnitude and Distribution of Chlorobenzenes in 2017

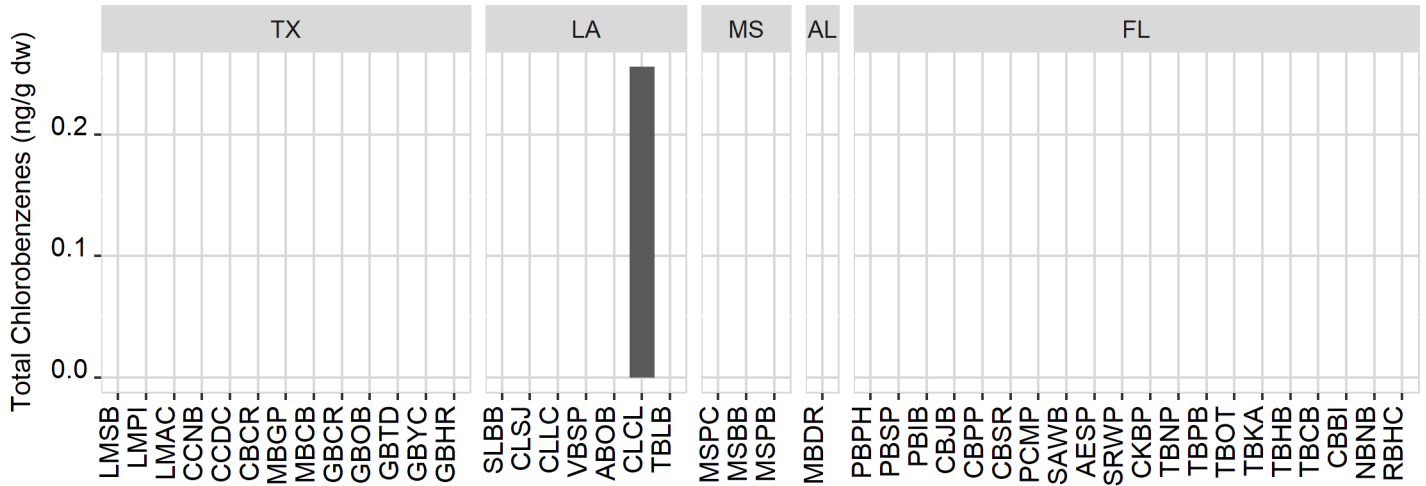


Figure 15. Bar graph showing magnitude of Total Chlorobenzene concentration sums detected in oyster tissue in the Gulf of Mexico in 2017. Sites are listed geographically from west to east, following the coastline.

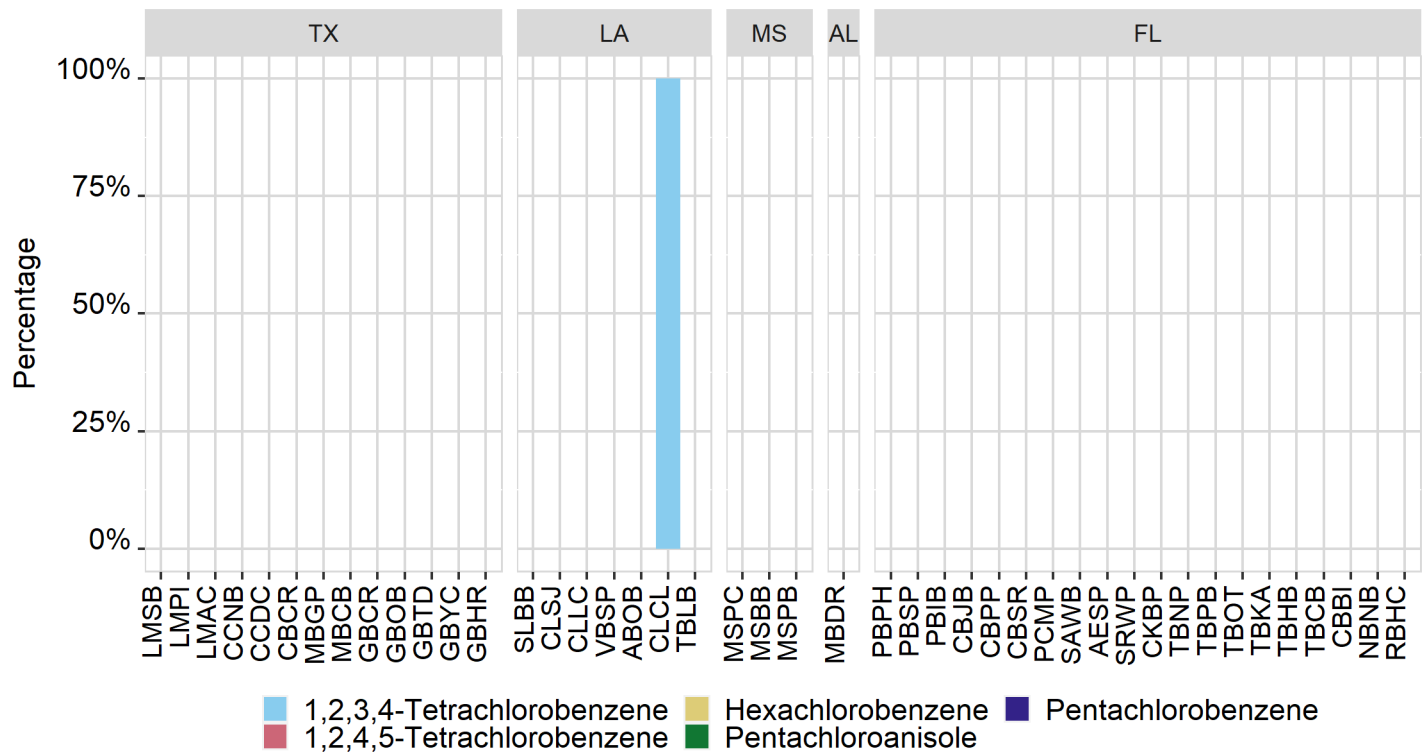


Figure 16. Percent composition of individual chlorobenzene contaminants in Total Chlorobenzene concentration sums per site in the Gulf of Mexico in 2017. Sites are listed geographically from west to east, following the coastline.

Results - Total Chlorobenzenes

6.3 Historical Context of Chlorobenzenes Magnitude and Distribution

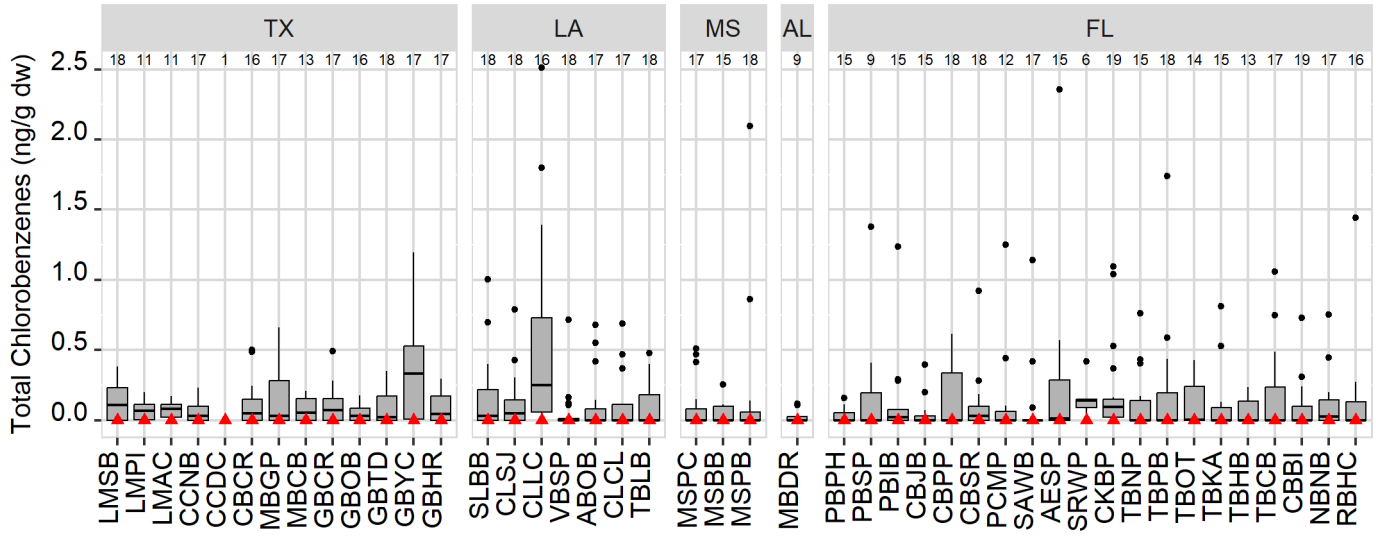


Figure 17. Total Chlorobenzene concentration sums in 2017 (red triangles) compared to boxplots of the historic Total Chlorobenzene concentration sums for each site (1986 - 2012) and results of the trend analysis showing significant increasing or decreasing contamination trends over time (gray triangles). Sites are listed geographically from west to east, following the coastline. The number of years each site has been sampled since 1986 is noted at the top of the plot for each site.

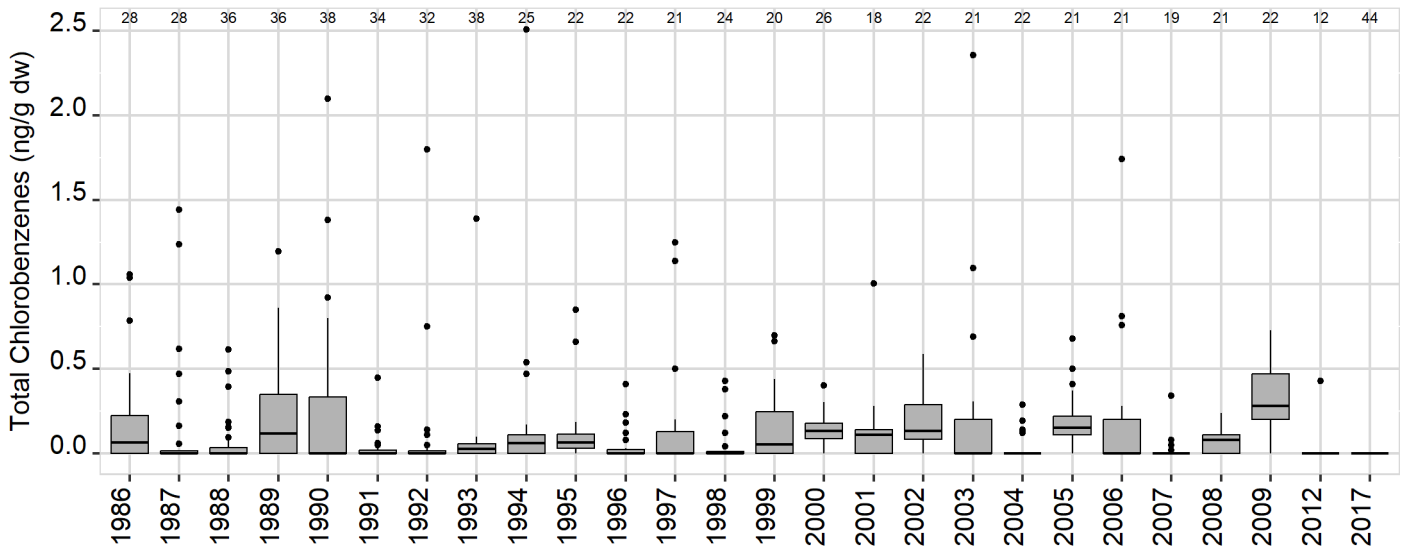


Figure 18. Boxplots representing the historic Total Chlorobenzene concentration sums of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot for each year.

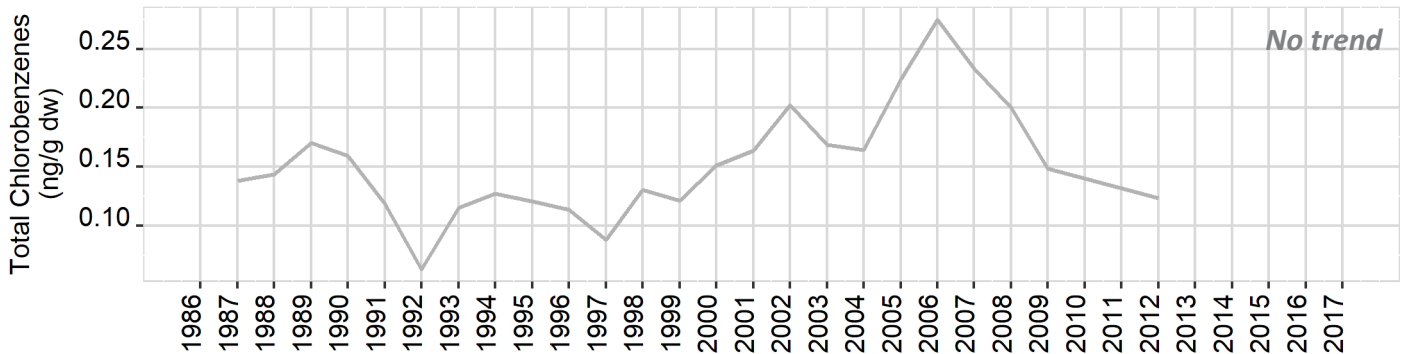


Figure 19. Three-point moving average of the yearly mean Total Chlorobenzene concentration sums in the sites analyzed in this study.

Results - Total Chlorobenzenes

6.4 Chlorobenzenes Summary

2017 Results (sum of 5 compounds):

- Total Chlorobenzenes were detected at 2.3% of 44 sites analyzed
- Total Chlorobenzenes concentration sums stats (Figure 15; Figure 16):
 - Range: 0.00 – 0.26 ng/g
 - Total Chlorobenzenes were not detected at 43 sites
 - Maximum concentration sum was detected at site CLCL (Caillou Lake, Caillou Lake)
 - Median: 0.00 ng/g
 - Mean \pm SD: 0.01 \pm 0.04 ng/g

Historical Context (1 compound):

- 0% sites were above their historic median concentrations in 2017
- 2 sites showed decreasing temporal trends of Total Chlorobenzene concentration sums at $\alpha = 0.05$ (Figure 17; Figure 18; Table A6)
- 1 site showed increasing temporal trends of Total Chlorobenzene concentration sums at $\alpha = 0.05$ (Figure 17; Figure 18; Table A6)
- There was no significant increasing or decreasing regional trend over time for sites in 2017 ($p = 0.95$, $\rho = -0.01$) (Figure 19)
- The percentage of sites in the Gulf of Mexico in 2017 grouped into each cluster based on historic data (Figure 20):
 - 100% of sites in low cluster (0.0 – 0.3 ng/g)
 - 0% of sites in medium cluster (0.3 – 1.5 ng/g)
 - 0% of sites in high cluster (1.6 – 6.1 ng/g)

General Observations:

- Only 1,2,3,4-Tetrachlorobenzene was detected at one site (CLCL) in the Gulf of Mexico, suggesting that widespread contamination of chlorobenzenes is unlikely in the region (Figure 20).
- The historic chlorobenzene compound hexachlorobenzene was not detected in 2017, meaning that the Total Chlorobenzene concentration sums were well below the US EPA recommended screening values for both recreational and subsistence fishers (Table A2).

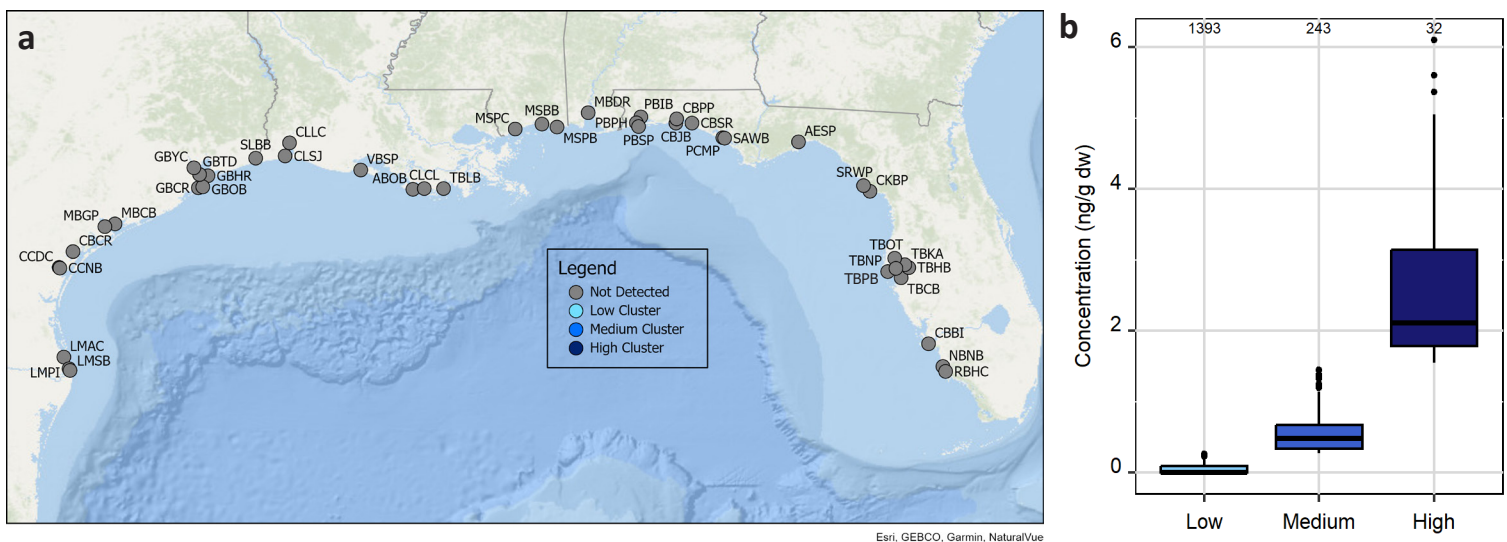


Figure 20. (a) Total Chlorobenzene concentration sums in 2017 compared to the (b) historic national MWP *Crassostrea virginica* Total Chlorobenzene concentration sums (ng/g dw). 129 national sites sampled between 1986 - 2017 for a total of 1668 samples.

Results - Chlorpyrifos

7.0 RESULTS - CHLORPYRIFOS

7.1 Chlorpyrifos Chemical Description

For this document, chlorpyrifos analyzed both in 2017 and historically is a single compound. Chlorpyrifos is a white, crystal-like solid with a strong odor that has been used as a pesticide in the United States since 1965 in both agricultural and non-agricultural settings (ATSDR, 1997; EPA, 2022). In 2000, there was a voluntary agreement to eliminate, phase out, and modify certain uses of chlorpyrifos, specifically private homeowner usage except for roaches, discontinuing uses on tomato plants and lowering the maximum residue level of grapes (EPA, 2022). As of 2022, chlorpyrifos is no longer used on food and animal feed crops, although it may still be used on non-fruit bearing trees (NPIC, 2023).

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

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

7.2 Magnitude and Distribution of Chlorpyrifos in 2017

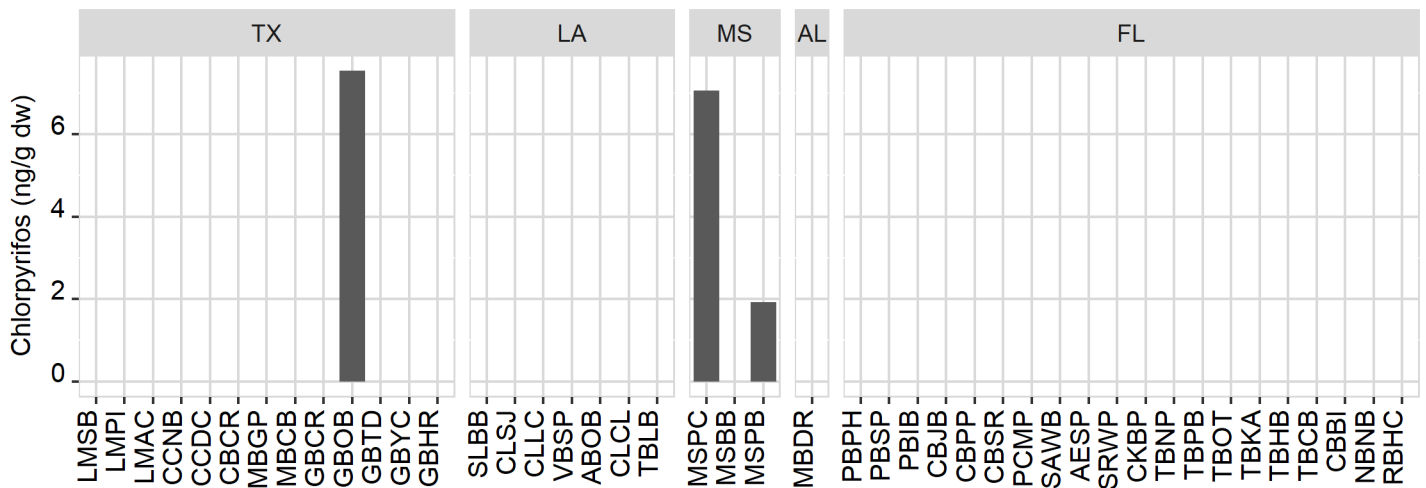


Figure 21. Bar graph showing magnitude of Chlorpyrifos concentrations detected in oyster tissue in the Gulf of Mexico in 2017. Sites are listed geographically from west to east, following the coastline.

Results - Chlorpyrifos

7.1 Historical Context of Chlorpyrifos Magnitude and Distribution

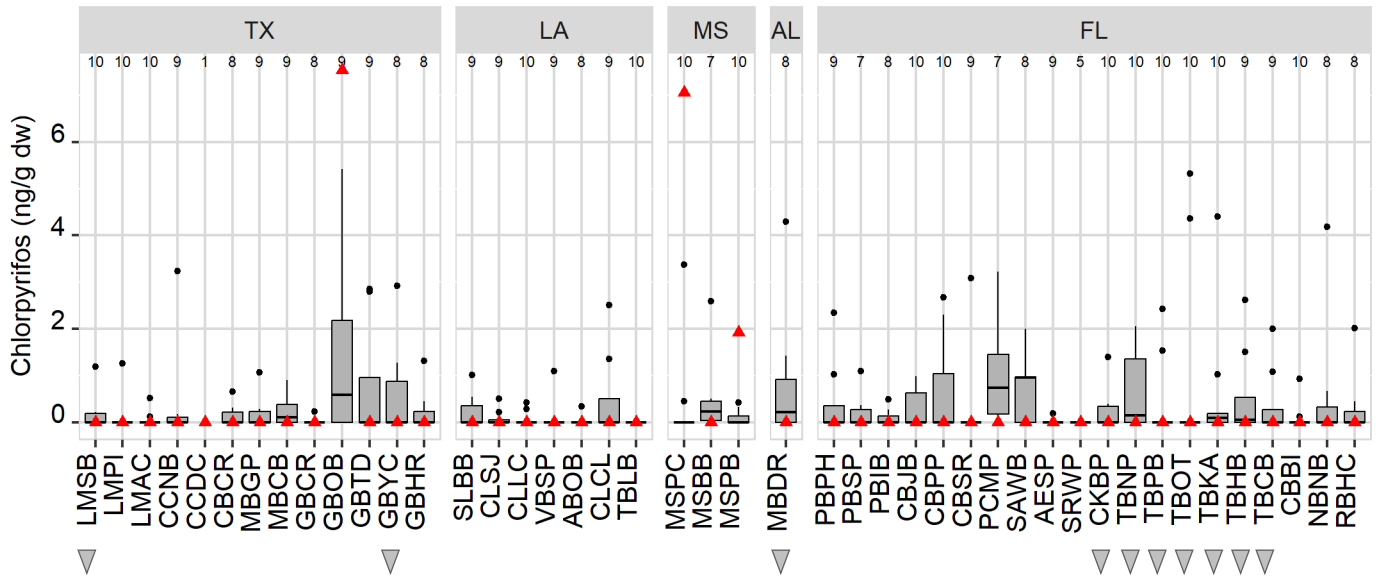


Figure 22. Chlorpyrifos concentrations in 2017 (red triangles) compared to boxplots of the historic Total Chlorobenzene concentrations for each site (1994 - 2012) and results of the trend analysis showing significant increasing or decreasing contamination trends over time (gray triangles). Sites are listed geographically from west to east, following the coastline. The number of years each site has been sampled since 1994 is noted at the top of the plot for each site.

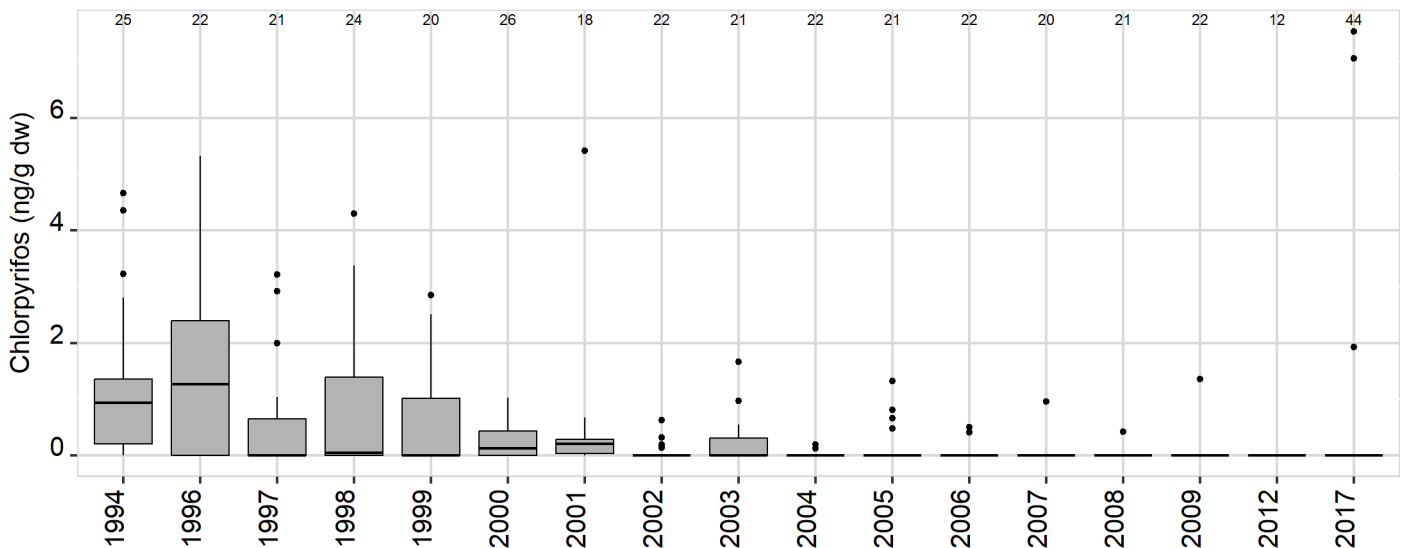


Figure 23. Boxplots representing the historic Chlorpyrifos concentrations of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot for each year.

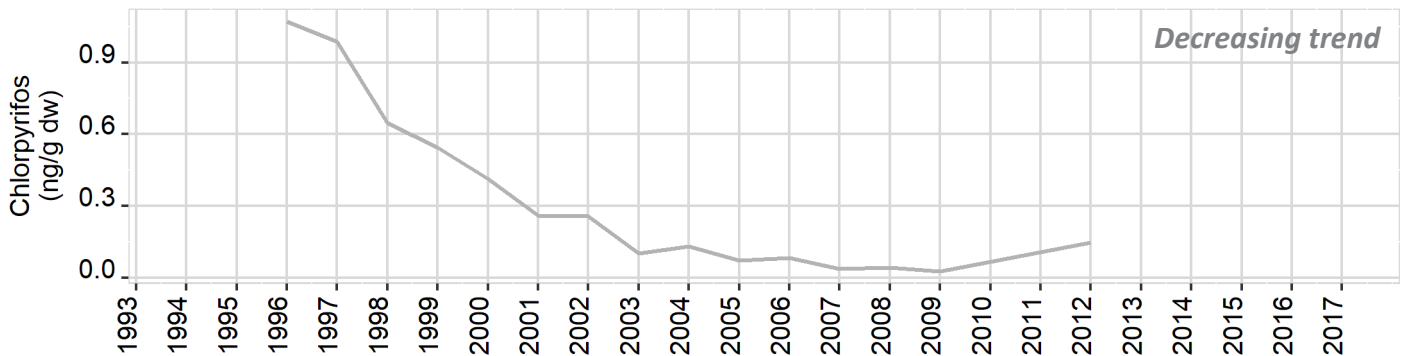


Figure 24. Three-point moving average of the yearly mean Chlorpyrifos concentrations in the sites analyzed in this study.

Results - Chlorpyrifos

7.4 Chlorpyrifos Summary

2017 Results (1 compound):

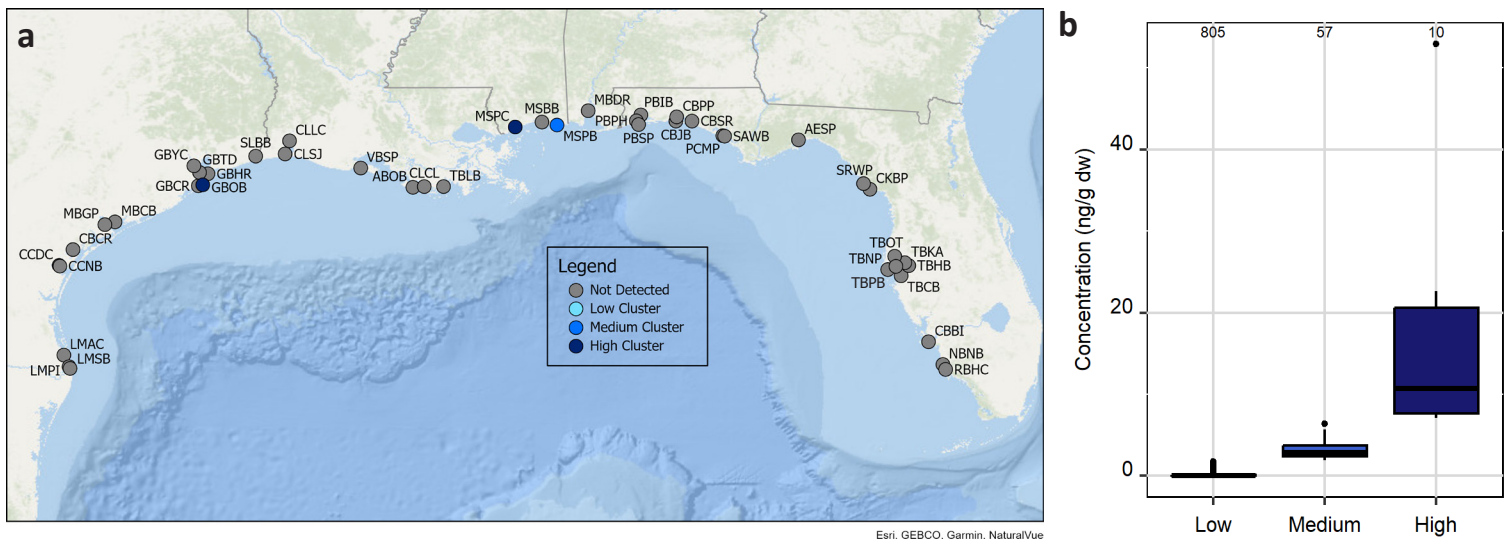
- Chlorpyrifos was detected at 6.8% of 44 sites analyzed
- Chlorpyrifos concentrations stats (Figure 21):
 - Range: 1.92 – 7.54 ng/g
 - Chlorpyrifos was not detected at 41 sites
 - Maximum concentration sum was detected at site GBOB (Galveston Bay, Offatts Bayou)
 - Median: 0.00 ng/g
 - Mean \pm SD: 0.38 \pm 1.56 ng/g

Historical Context (1 compound):

- 7.0% sites were above their historic median concentrations in 2017
- 10 sites showed decreasing temporal trends of Chlorpyrifos concentrations at $\alpha = 0.05$ (Figure 22; Figure 23; Table A6)
- 0 sites showed increasing temporal trends of Chlorpyrifos concentrations at $\alpha = 0.05$ (Figure 22; Figure 23; Table A6)
- There was a significant decreasing regional trend over time for sites in 2017 ($p < 0.001$, $\rho = -0.79$) (Figure 24)
- The percentage of sites in the Gulf of Mexico in 2017 grouped into each cluster based on historic data (Figure 25):
 - 93.2% of sites in low cluster (0.0 – 1.7 ng/g)
 - 2.3% of sites in medium cluster (1.9 – 6.4 ng/g)
 - 4.5% of sites in high cluster (7.1 – 52.9 ng/g)

General Observations:

- Chlorpyrifos was only detected at three sites within the Gulf of Mexico suggesting that widespread contamination of chlorpyrifos is unlikely in the region (Figure 25).
- The three sites where chlorpyrifos was detected, however, were clustered in the medium to high contamination clusters, possibly suggesting localized point sources of contamination (Figure 25).



Results - Total DDTs

8.0 RESULTS - TOTAL DDTs

8.1 DDTs Chemical Description

For this document, Total DDTs analyzed both in 2017 and historically are the sum of 6 compounds (Table 5). The six compounds within Total DDTs (dichlorodiphenyltrichloroethane) are comprised of ortho and para forms of DDT and its' transformation products DDE and DDD, the latter being the most predominant form found in the environment (ATSDR, 2022). Technical DDT, the insecticide, was composed of up to 14 compounds, of which 65-80% was the active ingredient, p,p'-DDT (4,4'-DDT). The next major component, o,p'-DDT (2,4'-DDT) (15-21%), is nearly inactive as an insecticide. DDT was used worldwide as an insecticide for agricultural pests and mosquito control. Its use in the United States was banned in 1972, but it is still used in some countries today. DDT was banned due to its environmental persistence, bioaccumulation, and toxicity to non-target organisms (ATSDR, 2022).

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

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

Table 5. DDT compounds composing the "Total DDTs" summation for the 2017 Gulf of Mexico survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2017	Analyzed in Historical Data
2,4'-DDD	●	●
4,4'-DDD	●	●
2,4'-DDE	●	●
4,4'-DDE	●	●
2,4'-DDT	●	●
4,4'-DDT	●	●

Results - Total DDTs

8.2 Magnitude and Distribution of DDTs in 2017

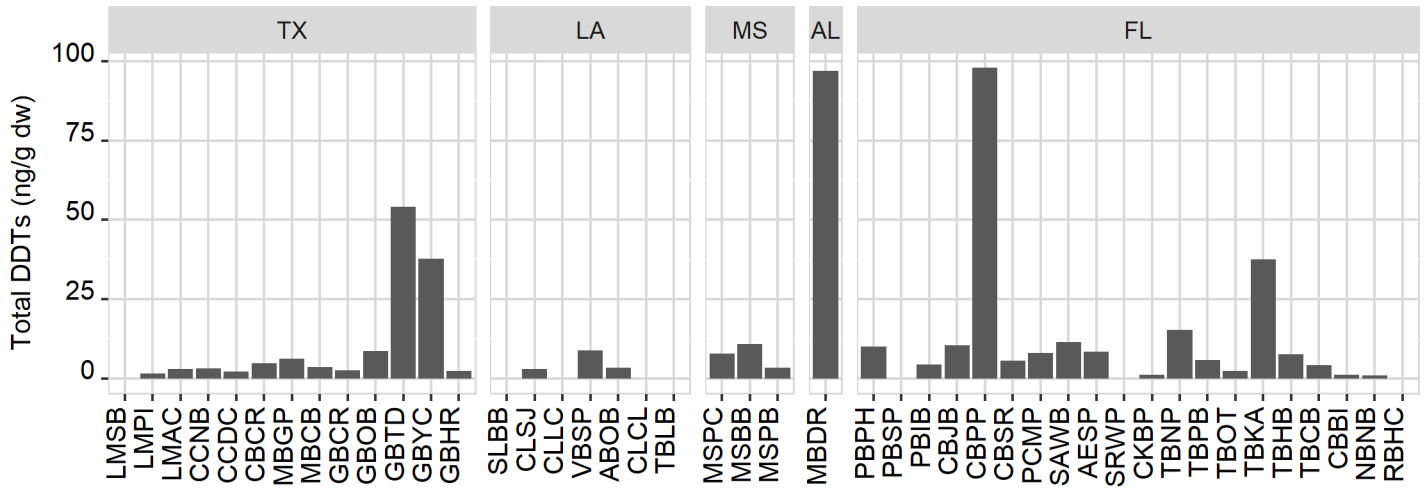


Figure 26. Bar graph showing magnitude of Total DDT concentration sums detected in oyster tissue in the Gulf of Mexico in 2017. Sites are listed geographically from west to east, following the coastline.

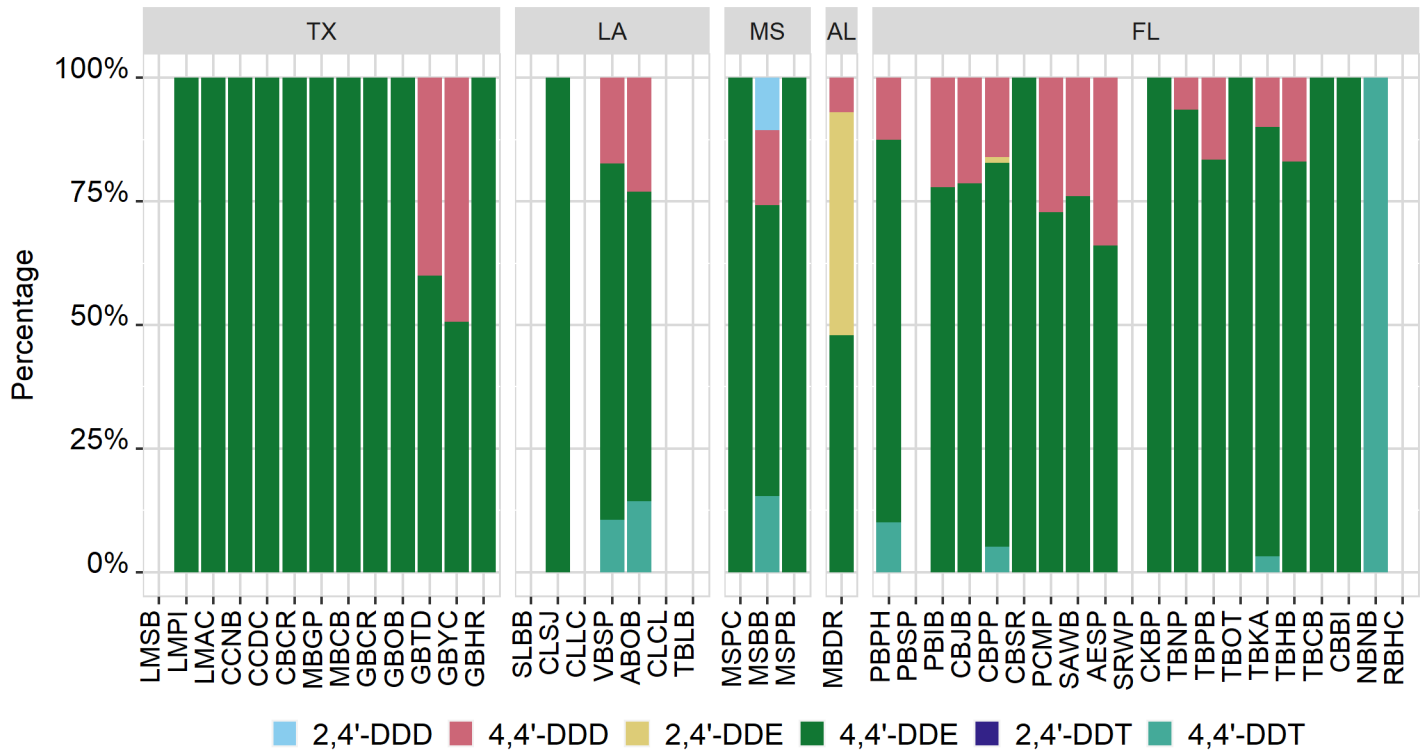


Figure 27. Percent composition of individual DDT contaminants in Total DDT concentration sums per site in the Gulf of Mexico in 2017. Sites are listed geographically from west to east, following the coastline.

Results - Total DDTs

8.3 Historical Context of DDTs Magnitude and Distribution

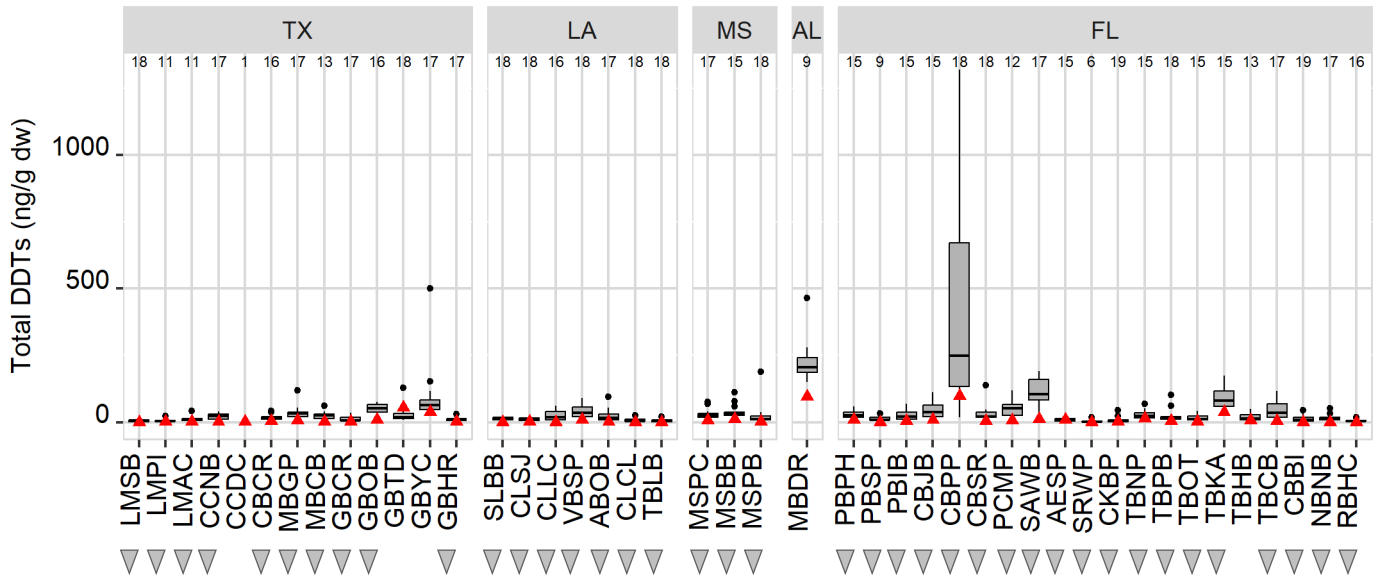


Figure 28. Total DDT concentration sums in 2017 (red triangles) compared to boxplots of the historic Total DDT concentration sums for each site (1986 - 2012) and results of the trend analysis showing significant increasing or decreasing contamination trends over time (gray triangles). Sites are listed geographically from west to east, following the coastline. The number of years each site has been sampled since 1986 is noted at the top of the plot for each site.

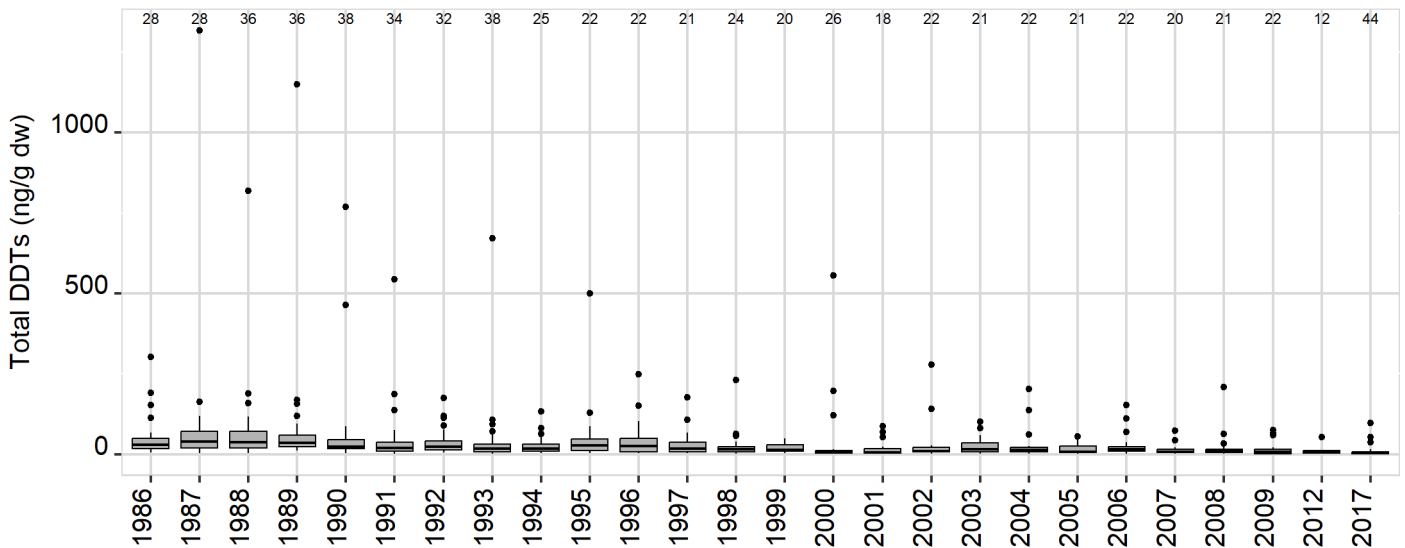


Figure 29. Boxplots representing the historic Total DDT concentration sums of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot for each year.

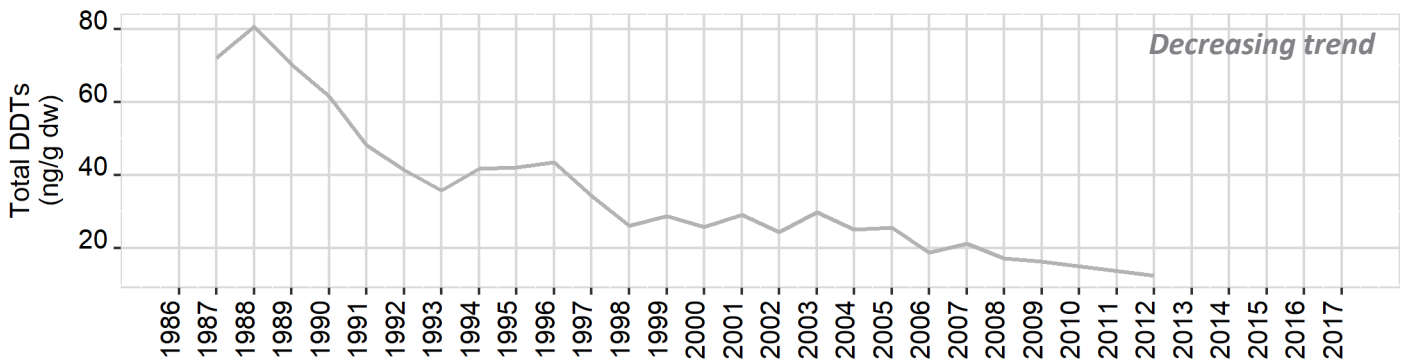


Figure 30. Three-point moving average of the yearly mean Total DDT concentration sums in the sites analyzed in this study.

Results - Total DDTs

8.4 DDTs Summary

2017 Results (sum of 6 compounds):

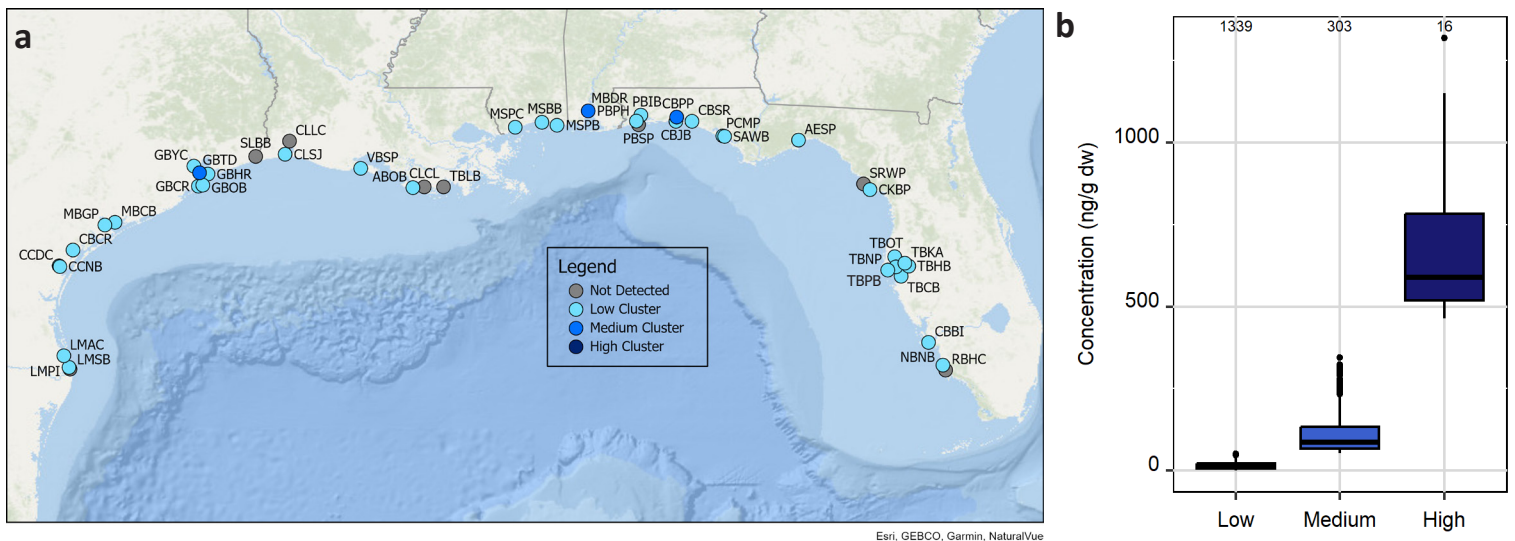
- Total DDTs were detected at 81.8% of 44 sites analyzed
- Total DDTs concentration sums stats (Figure 26; Figure 27):
 - Range: 0.85 – 97.94 ng/g
 - Total DDTs were not detected at 8 sites
 - Maximum concentration sum was detected at site CBPP (Choctawhatchee Bay, Postil Point)
 - Median: 3.82 ng/g
 - Mean \pm SD: 11.26 \pm 21.85 ng/g

Historical Context (sum of 6 compounds):

- 4.7% sites were above their historic median concentrations in 2017
- 39 sites showed decreasing temporal trends of Total DDT concentration sums at $\alpha = 0.05$ (Figure 28; Figure 29; Table A6)
- 0 sites showed increasing temporal trends of Total DDT concentration sums at $\alpha = 0.05$ (Figure 28; Figure 29; Table A6)
- There was a significant decreasing regional trend over time for sites in 2017 ($p < 0.001$, $\rho = -0.89$) (Figure 30)
- The percentage of sites in the Gulf of Mexico in 2017 grouped into each cluster based on historic data (Figure 31):
 - 93.2% of sites in low cluster (0.0 – 52.2 ng/g)
 - 6.8% of sites in medium cluster (52.7 – 344.1 ng/g)
 - 0.0% of sites in high cluster (464.4 – 1,317.9 ng/g)

General Observations:

- Total DDTs concentration sums were mostly low or not detected throughout the Gulf of Mexico (Figure 31).
- Total DDTs were clustered into the medium cluster at three sites within the Gulf of Mexico (MBDR, CBPP, and GBTD), possibly suggesting localized point sources of contamination (Figure 31).
- Total DDT concentration sums were well below the US EPA recommended screening values for both recreational and subsistence fishers (Table A2).



Results - Total Dieldrins

9.0 RESULTS - TOTAL DIELDRINS

9.1 Dieldrins Chemical Description

For this document, Total Dieldrins analyzed in 2017 is the sum of 3 compounds and Total Dieldrins analyzed historically is the sum of 2 compounds (Table 6). Dieldrins were widely used as insecticides in the 1960s for the control of termites around buildings and general crop protection from insects. In 1970, all uses of dieldrins were banned due to concern that they could cause severe aquatic environmental change and potential carcinogenicity (EPA, 1980). The ban was lifted in 1972 to allow limited use of dieldrins, primarily for termite control. All uses of dieldrins were finally banned in 1989 (EPA, 1990).

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

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

Note: Total Dieldrins concentration from sites MSPC in 2006 and GBOB in 2001 were removed from figures in the Historical Context section for better visualization due to their outlying concentration (127.74 and 59.56 ng/g, respectively). This data point remained in the dataset for all statistical analyses.

Table 6. Dieldrin compounds composing the "Total Dieldrins" summation for the 2017 Gulf of Mexico survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2017	Analyzed in Historical Data
Aldrin	●	●
Dieldrin	●	●
Endrin	●	

9.2 Magnitude and Distribution of Dieldrins in 2017

Dieldrins were not detected at any sites in the Gulf of Mexico in 2017.

Results - Total Dieldrins

9.3 Historical Context of Dieldrins Magnitude and Distribution

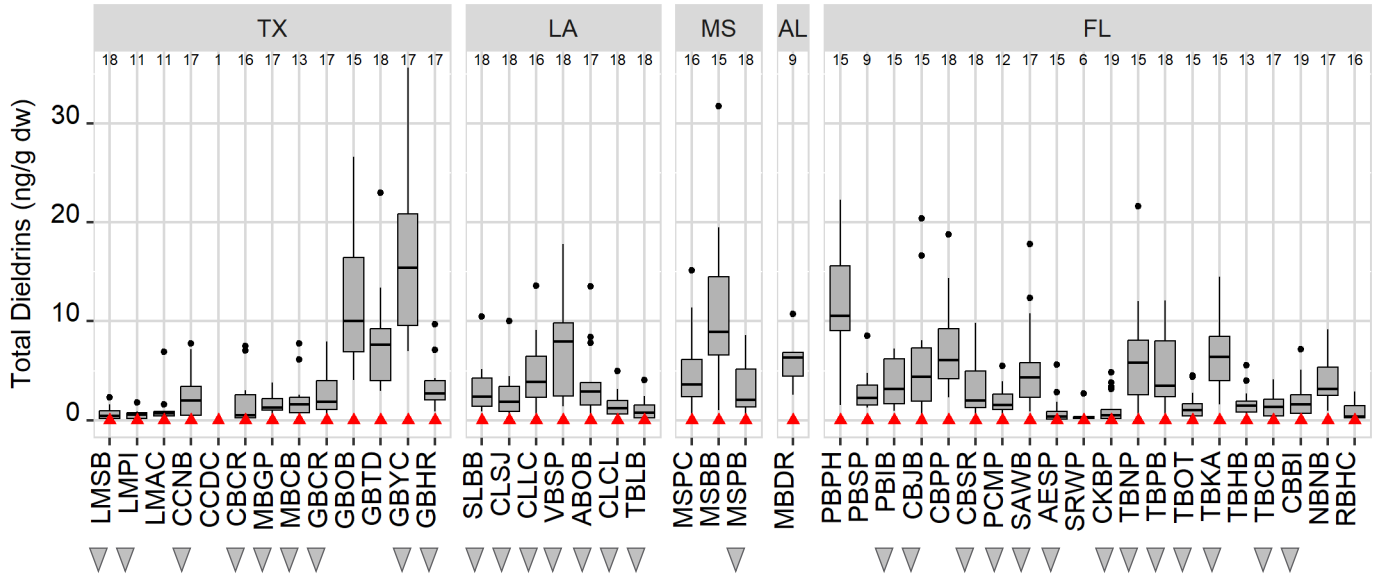


Figure 32. Total Dieldrin concentration sums in 2017 (red triangles) compared to boxplots of the historic Total Dieldrin concentration sums for each site (1986 - 2012) and results of the trend analysis showing significant increasing or decreasing contamination trends over time (gray triangles). Sites are listed geographically from west to east, following the coastline. The number of years each site has been sampled since 1986 is noted at the top of the plot for each site.

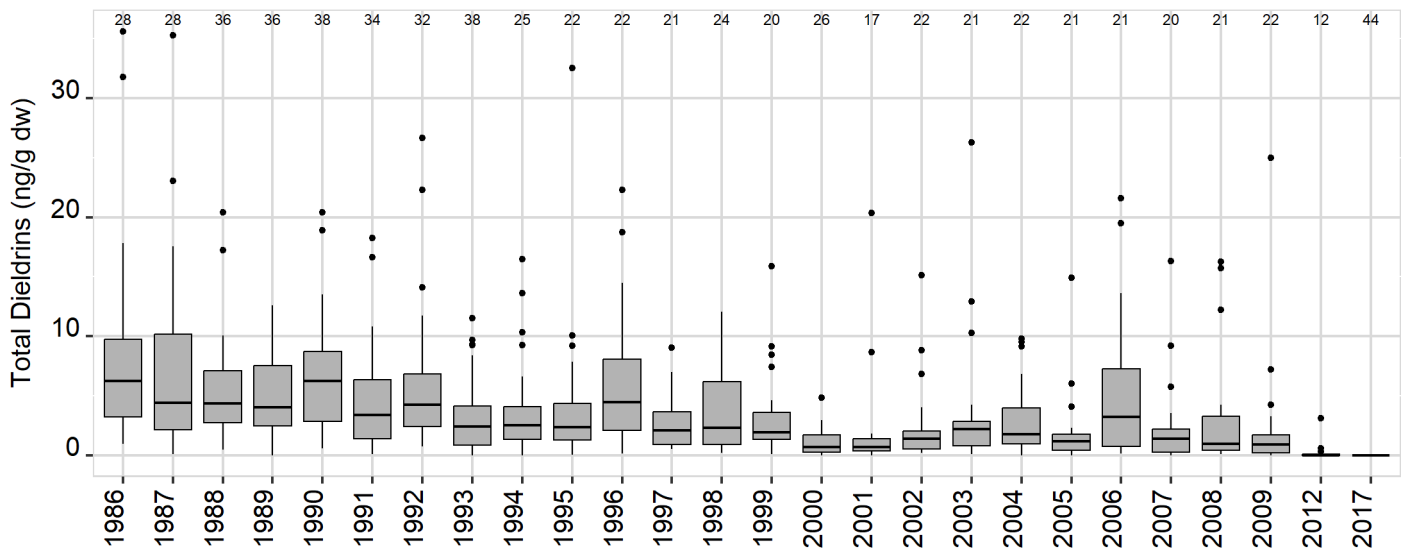


Figure 33. Boxplots representing the historic Total Dieldrin concentration sums of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot for each year.

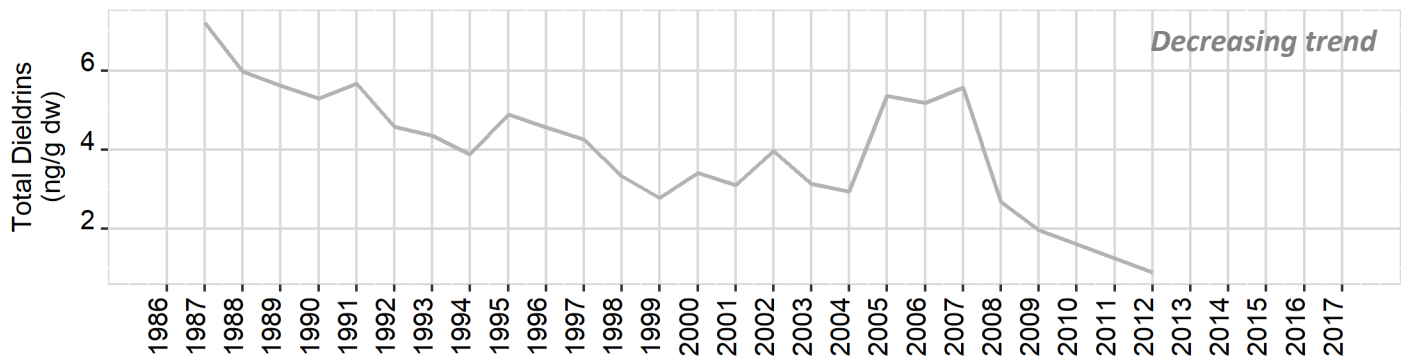


Figure 34. Three-point moving average of the yearly mean Total Dieldrin concentration sums in the sites analyzed in this study.

Results - Total Dieldrins

9.4 Dieldrins Summary

2017 Results (sum of 3 compounds):

- Dieldrins were not detected at any sites in the Gulf of Mexico in 2017.

Historical Context (sum of 2 compounds):

- 0.0% sites were above their historic median concentrations in 2017
- 30 sites showed decreasing temporal trends of Total Dieldrin concentration sums at $\alpha = 0.05$ (Figure 32; Figure 33; Table A6)
- 0 sites showed increasing temporal trends of Total Dieldrin concentration sums at $\alpha = 0.05$ (Figure 32; Figure 33; Table A6)
- There was a significant decreasing regional trend over time for sites in 2017 ($p < 0.001$, $\rho = -0.70$) (Figure 34)
- The percentage of sites in the Gulf of Mexico in 2017 grouped into each cluster based on historic data (Figure 35):
 - 100% of sites in low cluster (0.0 – 5.9 ng/g)
 - 0.0% of sites in medium cluster (5.9 – 20.4 ng/g)
 - 0.0% of sites in high cluster (21.3 – 127.7 ng/g)

General Observations:

- Dieldrins were not detected at any sites in the Gulf of Mexico in 2017 (Figure 35).

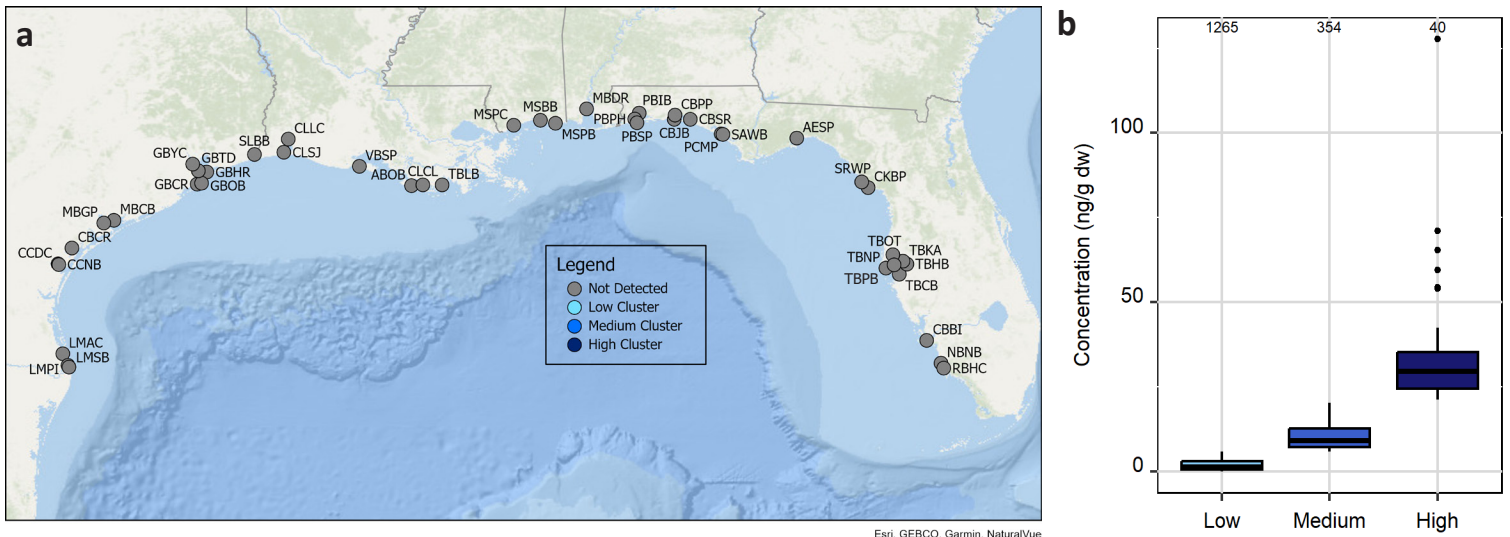


Figure 35. (a) Total Dieldrin concentration sums in 2017 compared to the (b) historic national MWP *Crassostrea virginica* Total Dieldrin concentration sums (ng/g dw). 129 national sites sampled between 1986 - 2017 for a total of 1659 samples.

Results - Total Endosulfans

10.0 RESULTS - TOTAL ENDOSULFANS

10.1 Endosulfans Chemical Description

For this document, Total Endosulfans analyzed in 2017 is the sum of 3 compounds and Total Endosulfans analyzed historically is the sum of 2 compounds (Table 7). Technical grade endosulfan is a mixture of two isomers (Endosulfan I and II) and Endosulfan sulfate is a product of oxidation and can be found in technical grade endosulfan. Endosulfan was a restricted-application pesticide, used to treat certain crops against aphids, beetles, leafhoppers, white flies, etc. (ATSDR, 2015). The use of endosulfans was restricted to certain crops before its phase-out by 2016 (ATSDR, 2015).

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

Endosulfan can be found in the environment in the atmosphere, soil, and water (ATSDR, 2015). Endosulfans can travel long distances by air and may be broken down by sunlight. In soil, endosulfans attach to soil particles, limiting its movement further. In water, Endosulfans change into the less toxic endosulfan diol, but endosulfan sulfate is more resistant to break down. Endosulfans can bioaccumulate in organisms living in contaminated water (ATSDR, 2015).

Note: Total Endosulfans concentration from site GBOB in 2001 was removed from figures in the Historical Context section for better visualization due to its outlying concentration (79.29 ng/g). This data point remained in the dataset for all statistical analyses.

Table 7. Endosulfan compounds composing the “Total Endosulfans” summation for the 2017 Gulf of Mexico survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2017	Analyzed in Historical Data
Endosulfan I	●	●
Endosulfan II	●	●
Endosulfan Sulfate	●	

10.2 Magnitude and Distribution of Endosulfans in 2017

Endosulfans were not detected at any sites in the Gulf of Mexico in 2017.

Results - Total Endosulfans

10.3 Historical Context of Endosulfans Magnitude and Distribution

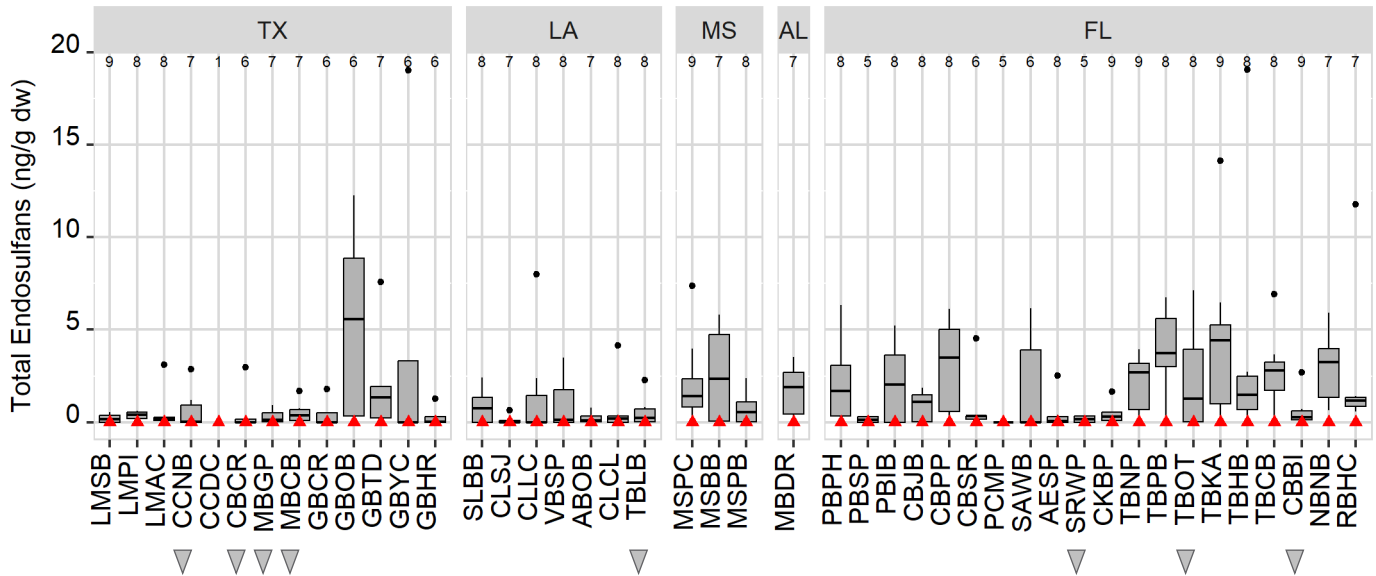


Figure 36. Total Endosulfan concentration sums in 2017 (red triangles) compared to boxplots of the historic Total Endosulfan concentration sums for each site (1994 - 2012) and results of the trend analysis showing significant increasing or decreasing contamination trends over time (gray triangles). Sites are listed geographically from west to east, following the coastline. The number of years each site has been sampled since 1994 is noted at the top of the plot for each site.

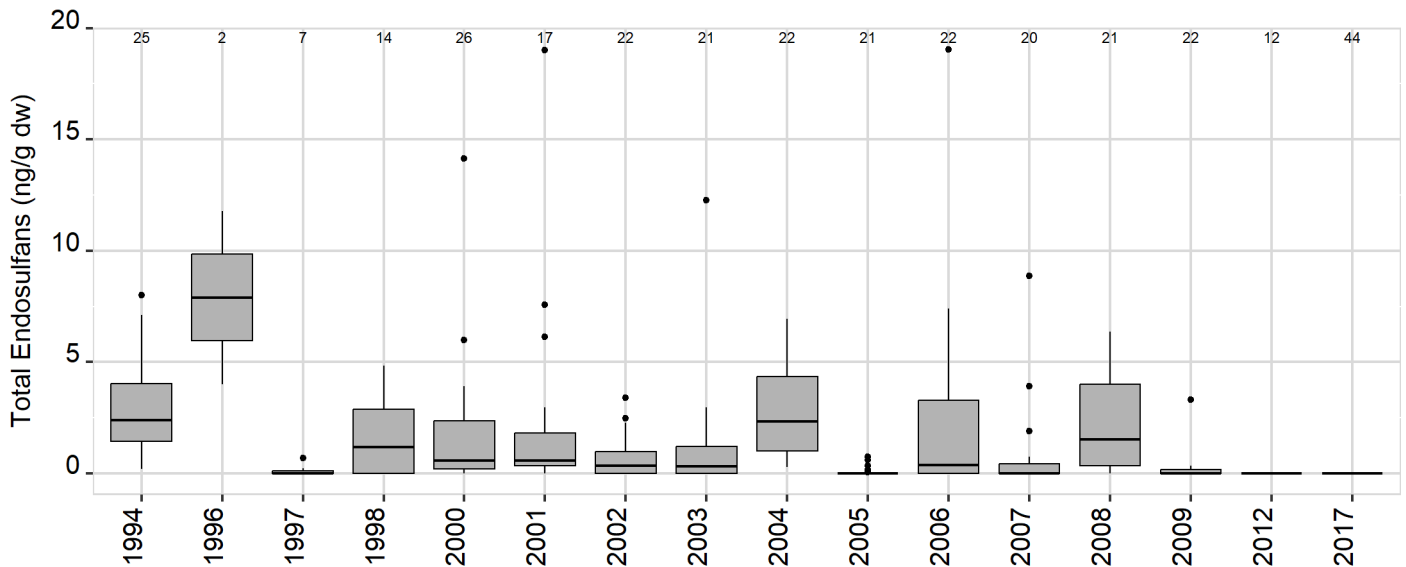


Figure 37. Boxplots representing the historic Total Endosulfan concentration sums of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot for each year.

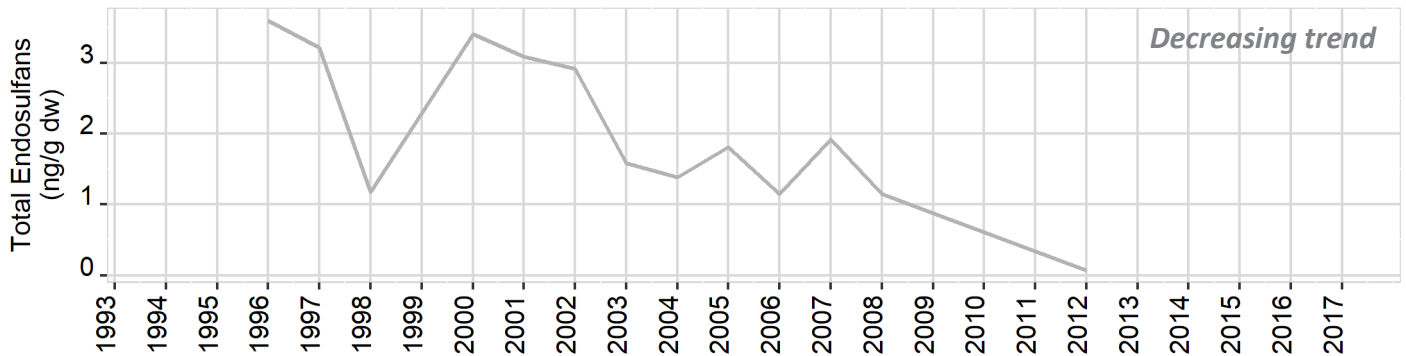


Figure 38. Three-point moving average of the yearly mean Total Endosulfan concentration sums in the sites analyzed in this study.

Results - Total Endosulfans

10.4 Endosulfans Summary

2017 Results (sum of 3 compounds):

- Endosulfans were not detected at any sites in the Gulf of Mexico in 2017.

Historical Context (sum of 2 compounds):

- 0.0% sites were above their historic median concentrations in 2017
- 8 sites showed decreasing temporal trends of Total Endosulfan concentration sums at $\alpha = 0.05$ (Figure 36; Figure 37; Table A6)
- 0 sites showed increasing temporal trends of Total Endosulfan concentration sums at $\alpha = 0.05$ (Figure 36; Figure 37; Table A6)
- There was a significant decreasing regional trend over time for sites in 2017 ($p = 0.029$, $\rho = -0.55$) (Figure 38)
- The percentage of sites in the Gulf of Mexico in 2017 grouped into each cluster based on historic data (Figure 39):
 - 100% of sites in low cluster (0.0 – 5.6 ng/g)
 - 0.0% of sites in medium cluster (5.8 – 14.1 ng/g)
 - 0.0% of sites in high cluster (18.9 – 134.0 ng/g)

General Observations:

- Endosulfans were not detected at any sites in the Gulf of Mexico in 2017 (Figure 39).

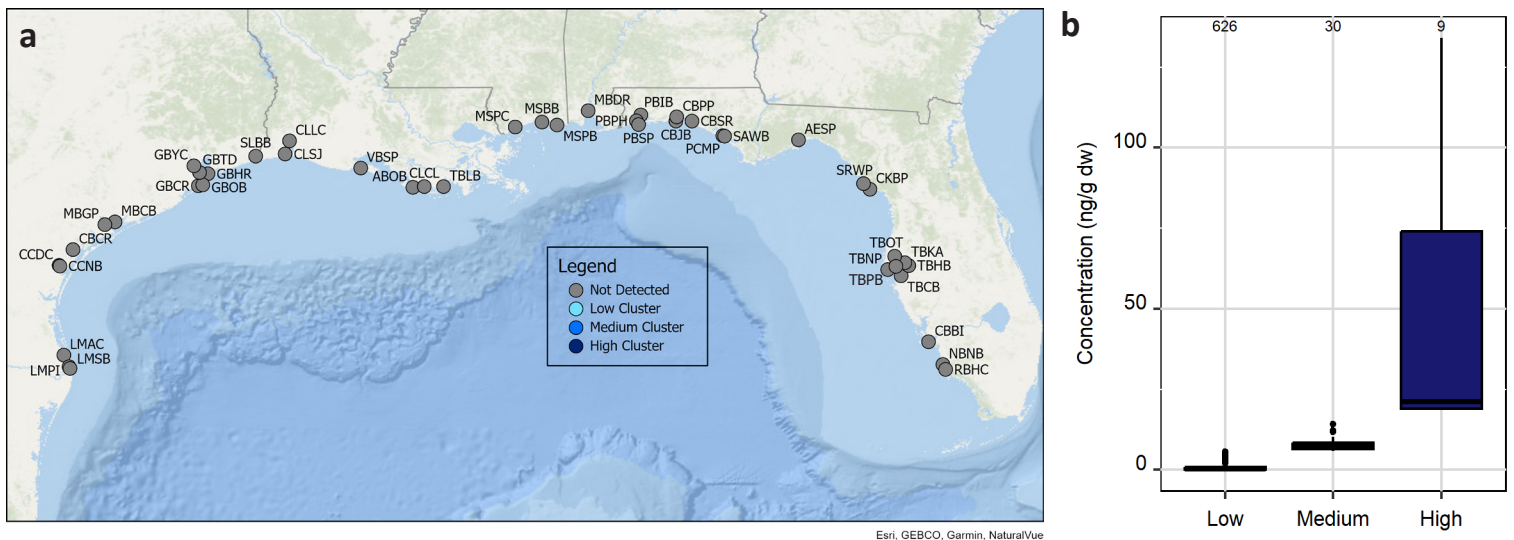


Figure 39. (a) Total Endosulfan concentration sums in 2017 compared to the (b) historic national MWP *Crassostrea virginica* Total Endosulfan concentration sums (ng/g dw). 117 national sites sampled between 1994 - 2017 for a total of 665 samples.

Results - Total HCHs

11.0 RESULTS - TOTAL HCHs

11.1 HCHs Chemical Description

For this document, Total HCHs analyzed in 2017 is the sum of 4 compounds and Total HCHs analyzed historically is only 1 compound (Table 8). Technical grade HCH (hexachlorocyclohexane) contains the alpha, beta, gamma, delta, and epsilon forms of HCH. Almost all of the insecticidal properties are found in gamma-HCH (lindane) which is used as an insecticide on fruit, vegetables and forest crops. It is also found in lotion, cream or shampoo as a prescription to treat head and body lice and scabies (ATSDR, 2005). In 2009, the Stockholm Convention on Persistent Organic Pollutants implemented an international ban on the use of lindane in agriculture but allowed a 5-year extension for its use in the treatment of head lice and scabies (UNEP, 2009). The US did not ratify the convention; however, the EPA requested the voluntary cancellation of the last agricultural uses of lindane in 2006 (EPA, 2006a).

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

In sediments and water, HCH can be broken down into less toxic substances by algae, bacteria, and fungi, but it is a slow process. HCH has been shown to accumulate in the fatty tissue of fish.

Note: Total HCHs concentration from site GBYC in 1995 was removed from figures in the Historical Context section for better visualization due to its outlying concentration (15.48 ng/g). This data point remained in the dataset for all statistical analyses.

Table 8. HCH compounds composing the “Total HCHs” summation for the 2017 Gulf of Mexico survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2017	Analyzed in Historical Data
Alpha-Hexachlorocyclohexane	●	
Beta-Hexachlorocyclohexane	●	
Gamma-Hexachlorocyclohexane	●	●
Delta-Hexachlorocyclohexane	●	

Results - Total HCHs

11.2 Magnitude and Distribution of HCHs in 2017

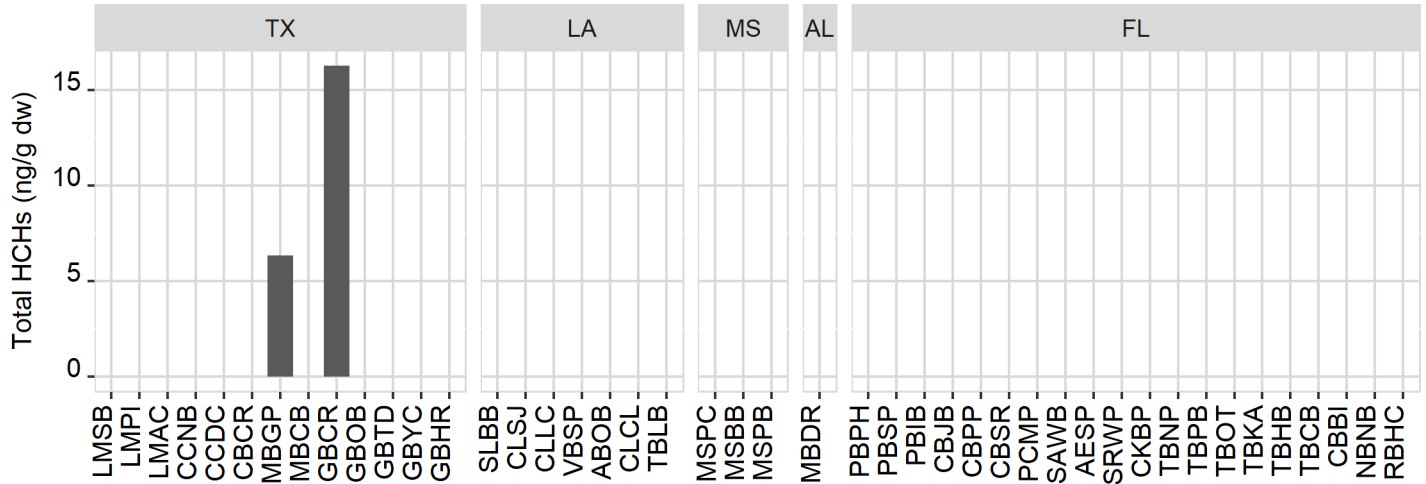


Figure 40. Bar graph showing magnitude of Total HCH concentration sums detected in oyster tissue in the Gulf of Mexico in 2017. Sites are listed geographically from west to east, following the coastline.

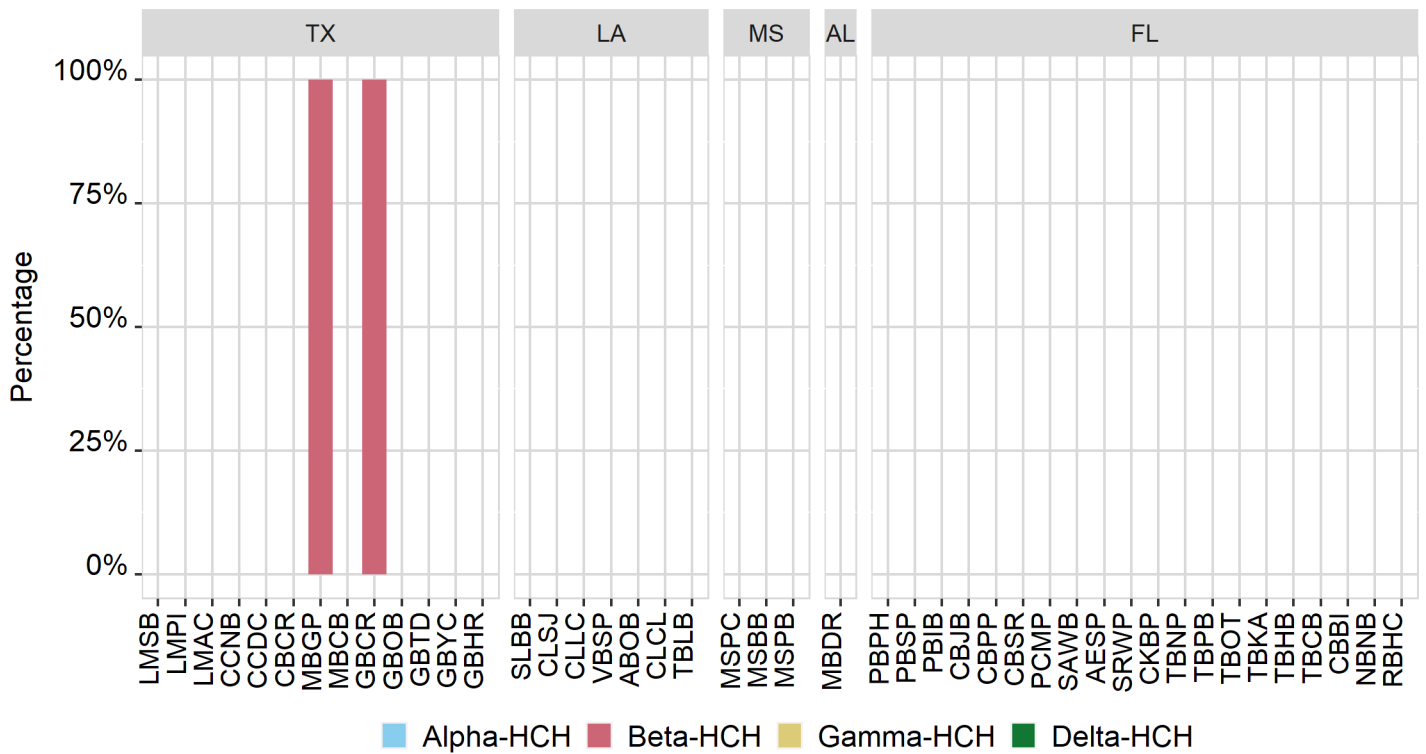


Figure 41. Percent composition of individual HCH contaminants in Total HCH concentration sums per site in the Gulf of Mexico in 2017. Sites are listed geographically from west to east, following the coastline.

Results - Total HCHs

11.3 Historical Context of HCHs Magnitude and Distribution

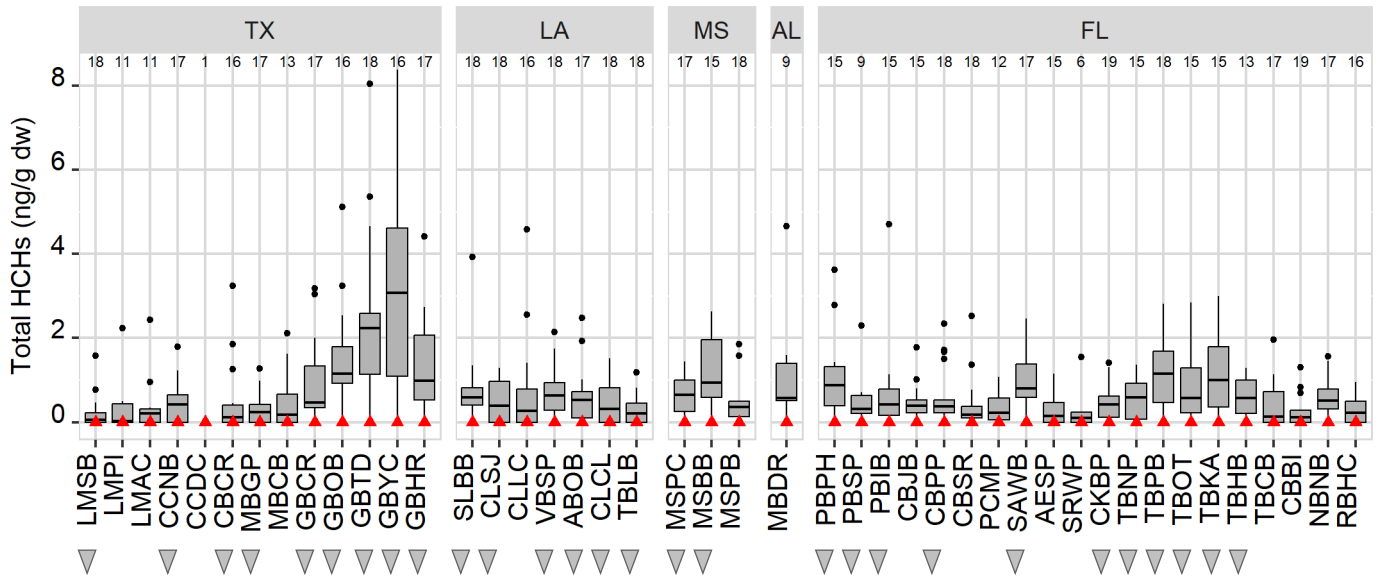


Figure 42. Total HCH concentration sums in 2017 (red triangles) compared to boxplots of the historic Total HCH concentration sums for each site (1986 - 2012) and results of the trend analysis showing significant increasing or decreasing contamination trends over time (gray triangles). Sites are listed geographically from west to east, following the coastline. The number of years each site has been sampled since 1986 is noted at the top of the plot for each site.

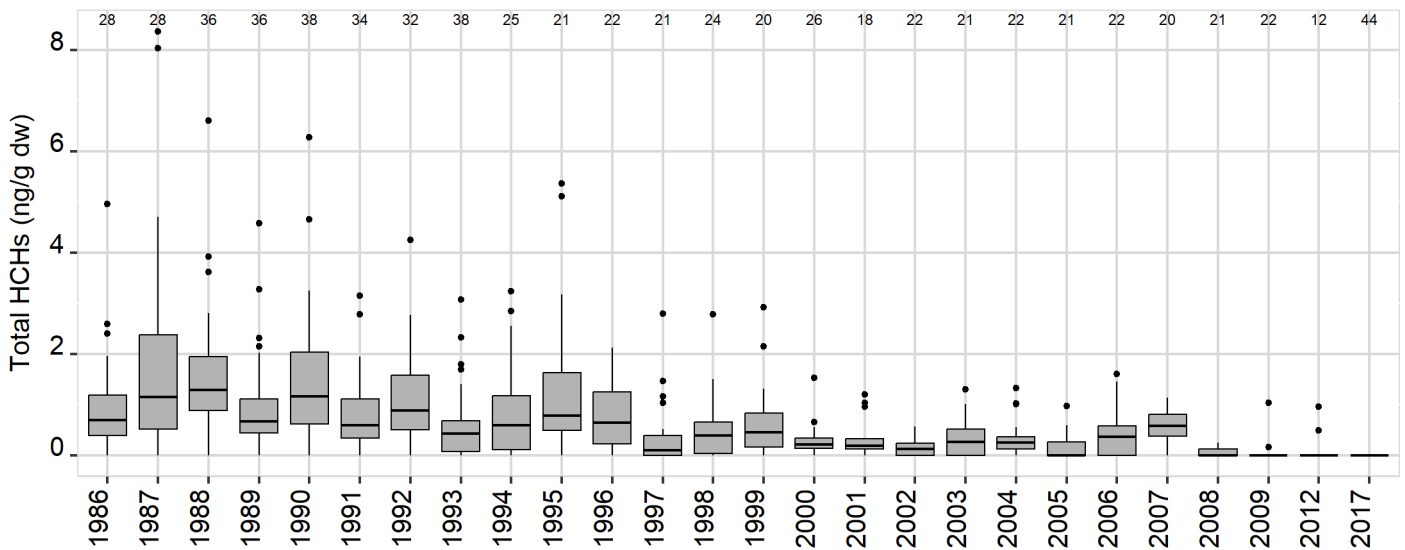


Figure 43. Boxplots representing the historic Total HCH concentration sums of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot for each year.

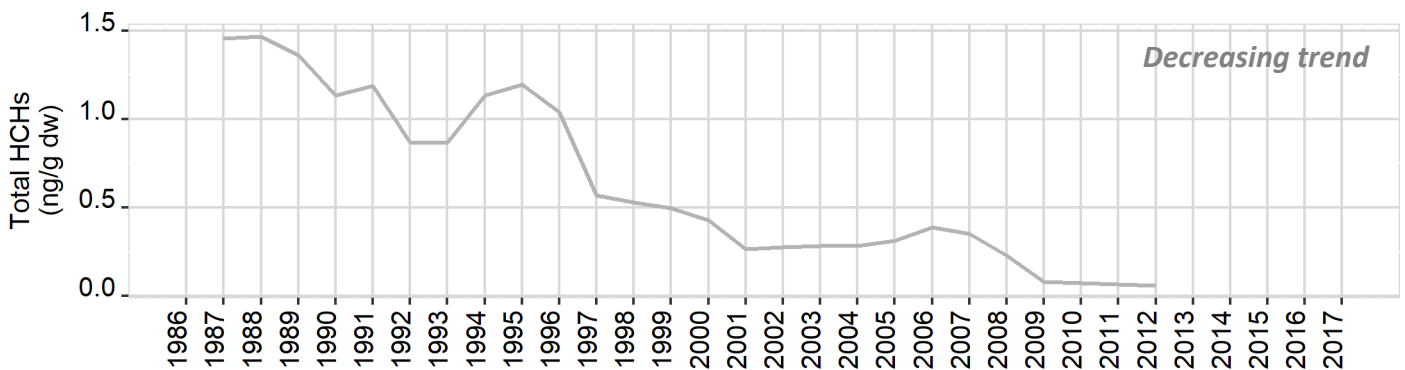


Figure 44. Three-point moving average of the yearly mean Total HCH concentration sums in the sites analyzed in this study.

Results - Total HCHs

11.4 HCHs Summary

2017 Results (sum of 4 compounds):

- Total HCHs were detected at 4.5% of 44 sites analyzed
- Total HCHs concentration sums stats (Figure 40; Figure 41):
 - Range: 6.33 – 16.29 ng/g
 - Total HCHs were not detected at 42 sites
 - Maximum concentration sum was detected at site GBCR (Galveston Bay, Confederate Reef)
 - Median: 0.00 ng/g
 - Mean \pm SD: 0.51 \pm 2.61 ng/g

Historical Context (1 compound):

- 0% sites were above their historic median concentrations in 2017
- 28 sites showed decreasing temporal trends of Total HCH concentration sums at $\alpha = 0.05$ (Figure 42; Figure 43; Table A6)
- 0 sites showed increasing temporal trends of Total HCH concentration sums at $\alpha = 0.05$ (Figure 42; Figure 43; Table A6)
- There was a significant decreasing regional trend over time for sites in 2017 ($p < 0.001$, $\rho = -0.88$) (Figure 44)
- The percentage of sites in the Gulf of Mexico in 2017 grouped into each cluster based on historic data (Figure 45):
 - 100% of sites in low cluster (0.0 – 0.9 ng/g)
 - 0% of sites in medium cluster (0.9 – 3.3 ng/g)
 - 0% of sites in high cluster (3.4 – 17.3 ng/g)

General Observations:

- Only beta-HCH was detected at two sites (MBGP and GBCR) in the Gulf of Mexico, suggesting that widespread contamination of HCHs is unlikely in the region (Figure 41).
- The historic HCH compound gamma-HCH was not detected in 2017, meaning that the Total HCH concentration sums were well below the US EPA recommended screening values for both recreational and subsistence fishers (Table A2).

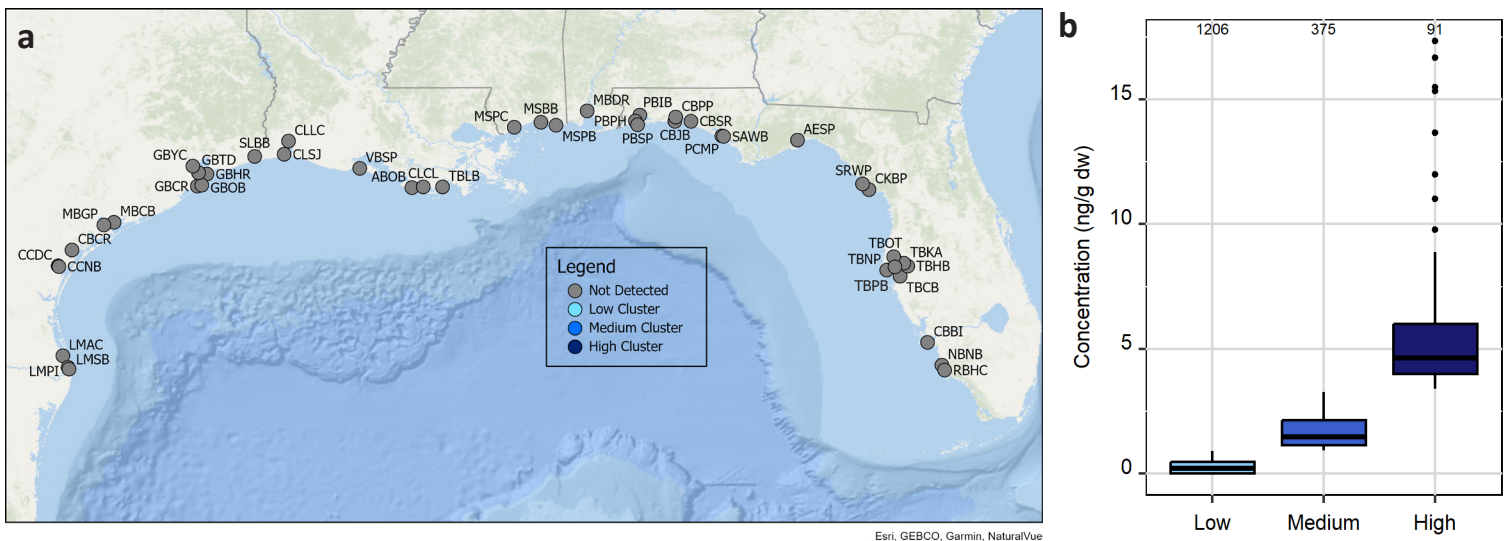


Figure 45. (a) Total HCH concentration sums in 2017 compared to the (b) historic national MWP *Crassostrea virginica* Total HCH concentration sums (ng/g dw). 129 national sites sampled between 1986 - 2017 for a total of 1672 samples.

12.0 RESULTS - MIREX

12.1 Mirex Chemical Description

For this document, Mirex analyzed both in 2017 and historically is a single compound. Mirex was commercially introduced in the United States in 1959 for use in pesticide formulations and as an industrial fire retardant (ATSDR, 2020). Mirex was used in products including rubber, plastic, paints, paper, and electrical goods. In the 1960s, mirex was commonly used to control fire ants in southern States. Mirex was banned for use in the United States in 1978, except for use on pineapples until stocks on hand were exhausted (ATSDR, 2020).

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

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

12.2 Magnitude and Distribution of Mirex in 2017

Mirex was not detected at any sites in the Gulf of Mexico in 2017.

Results - Mirex

12.3 Historical Context of Mirex Magnitude and Distribution

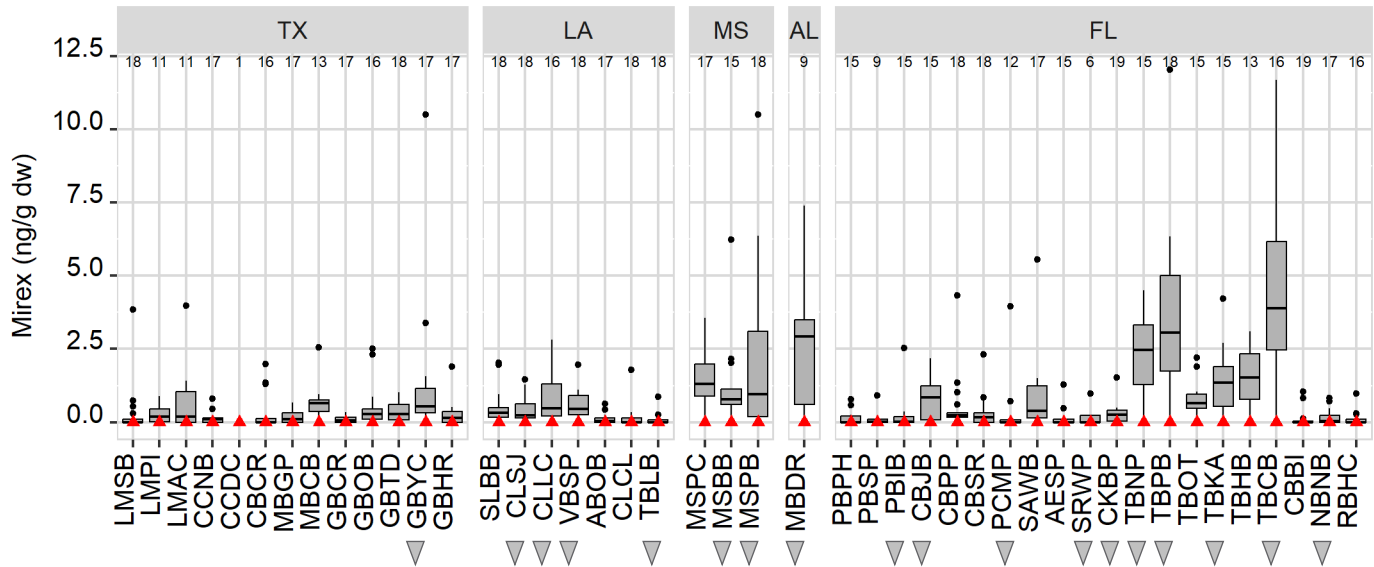


Figure 46. Mirex concentrations in 2017 (red triangles) compared to boxplots of the historic Mirex concentrations for each site (1986 - 2012) and results of the trend analysis showing significant increasing or decreasing contamination trends over time (gray triangles). Sites are listed geographically from west to east, following the coastline. The number of years each site has been sampled since 1986 is noted at the top of the plot for each site.

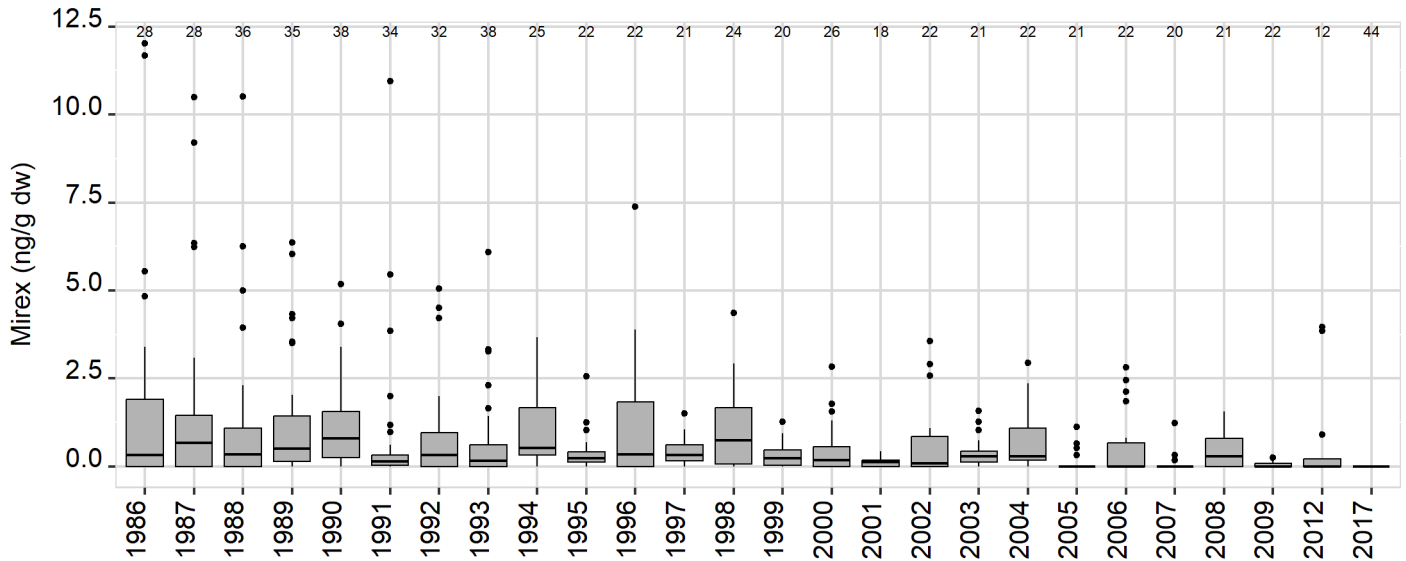


Figure 47. Boxplots representing the historic Mirex concentrations of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot for each year.

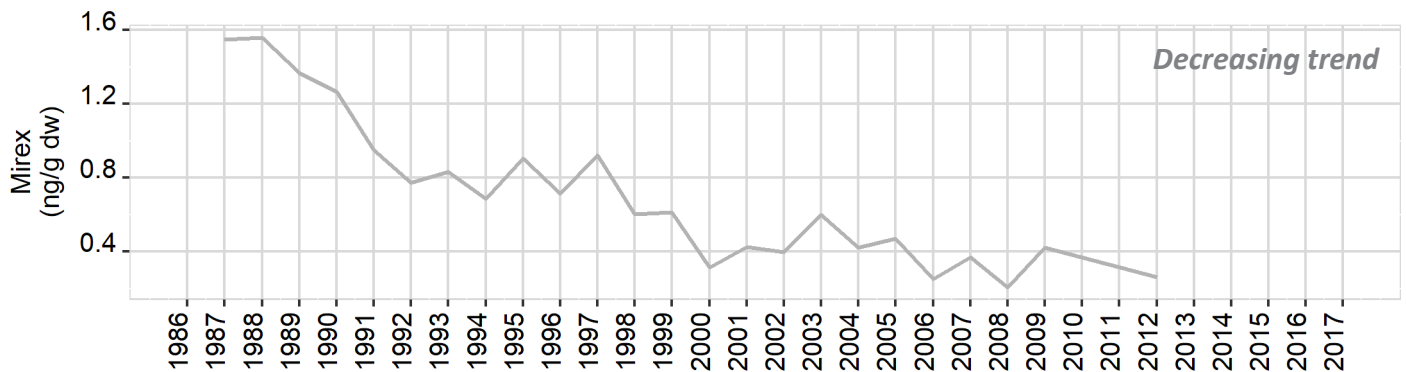


Figure 48. Three-point moving average of the yearly mean Mirex concentrations in the sites analyzed in this study.

Results - Mirex

12.4 Mirex Summary

2017 Results (1 compound):

- Mirex was not detected at any sites in the Gulf of Mexico in 2017.

Historical Context (1 compound):

- 0.0% sites were above their historic median concentrations in 2017
- 18 sites showed decreasing temporal trends of Mirex concentrations at $\alpha = 0.05$ (Figure 46; Figure 47; Table A6)
- 0 sites showed increasing temporal trends of Mirex concentrations at $\alpha = 0.05$ (Figure 46; Figure 47; Table A6)
- There was a significant decreasing regional trend over time for sites in 2017 ($p < 0.001$, $\rho = -0.75$) (Figure 48)
- The percentage of sites in the Gulf of Mexico in 2017 grouped into each cluster based on historic data (Figure 49):
 - 100% of sites in low cluster (0.0 – 2.9 ng/g)
 - 0.0% of sites in medium cluster (3.1 – 7.6 ng/g)
 - 0.0% of sites in high cluster (8.6 – 70.0 ng/g)

General Observations:

- Mirex was not detected at any sites in the Gulf of Mexico in 2017 (Figure 49).

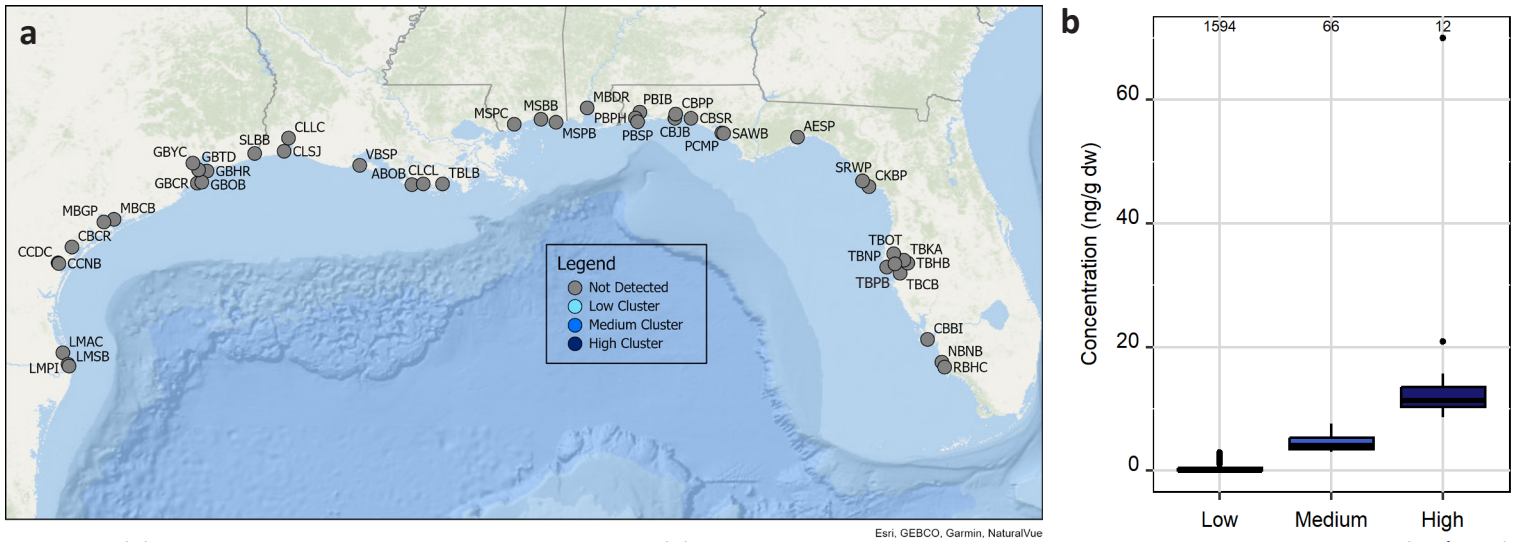


Figure 49. (a) Mirex concentrations in 2017 compared to the (b) historic national MWP *Crassostrea virginica* Mirex concentrations (ng/g dw). 129 national sites sampled between 1986 - 2017 for a total of 1672 samples.

Results - Total PAHs

13.0 RESULTS - TOTAL PAHs

13.1 PAHs Chemical Description

For this document, Total PAHs analyzed in 2017 is the sum of 64 compounds and Total PAHs analyzed historically is the sum of 39 compounds (Table 9). PAHs (polycyclic aromatic hydrocarbons) are formed from the fusing of benzene rings during the incomplete combustion of organic materials. PAHs are found in creosote, soot, petroleum, coal and tar, and are the only organic contaminants measured by the Mussel Watch Program that have natural sources (forest fires and volcanoes) in addition to anthropogenic sources (automobiles emissions, home heating, coal fired power plants). PAHs can also enter the aquatic environment by means of discharge from industrial and wastewater treatments plants (ATSDR, 1995) or can volatilize from an oil spill if they have a small molecular weight.

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

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

PAH50 is a contemporary assessment of PAHs that is commonly used to track environmental PAH concentrations. In addition to the 50 PAH chemicals included in PAH50, there are other PAHs that are influential in environmental pollutant assessment and management. For example, perylene is a PAH that is routinely used to help identify if the source of a given petroleum product or signature in the environment is a result of either petrogenic or pyrogenic (combustion) processes (Yang et al., 2022). The PAH50 subset was compared to the concentration sum of the full suite of PAHs analyzed in this study.

Note: Total PAHs concentration from site SAWB in 1991 was removed from figures in the Historical Context section for better visualization due to its outlying concentration (32,989.50 ng/g). This data point remained in the dataset for all

Table 9. PAH compounds composing the "Total PAHs" summation for the 2017 Gulf of Mexico survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2017	Analyzed in Historical Data
Acenaphthene	●	●
Acenaphthylene	●	●
Anthracene	●	●
Benz[a]anthracene	●	●
Benzo[a]fluoranthene	●	
Benzo[a]pyrene	●	●
Benzo[b]fluoranthene	●	●
Benzo[e]pyrene	●	●
Benzo[g,h,i]perylene	●	●
Benzo[k]fluoranthene	●	●
Benzothiophene	●	
Biphenyl	●	●
C1-Benzothiophene	●	
C1-Chrysenes	●	●
C1-Decalin	●	
C1-Dibenzo[a,h]anthracene	●	

Results - Total PAHs

Table 9 cont. PAH compounds composing the "Total PAHs" summation for the 2017 Gulf of Mexico survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2017	Analyzed in Historical Data
C1-Dibenzothiophenes	●	●
C1-Fluoranthenes_Pyrenes	●	●
C1-Fluorenes	●	●
C1-Naphthalenes	●	●
C1-Naphthobenzothiophene	●	
C1-Phenanthrenes_Anthracenes	●	●
C2-Benzothiophene	●	
C2-Chrysenes	●	●
C2-Decalin	●	
C2-Dibenzo[a,h]anthracene	●	
C2-Dibenzothiophenes	●	●
C2-Fluoranthenes_Pyrenes	●	
C2-Fluorenes	●	●
C2-Naphthalenes	●	●
C2-Naphthobenzothiophene	●	
C2-Phenanthrenes_Anthracenes	●	●
C3-Benzothiophene	●	
C3-Chrysenes	●	●
C3-Decalin	●	
C3-Dibenzo[a,h]anthracene	●	
C3-Dibenzothiophenes	●	●
C3-Fluoranthenes_Pyrenes	●	
C3-Fluorenes	●	●
C3-Naphthalenes	●	●

Table 9 cont. PAH compounds composing the "Total PAHs" summation for the 2017 Gulf of Mexico survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2017	Analyzed in Historical Data
C3-Naphthobenzothiophene	●	
C3-Phenanthrenes_Anthracenes	●	●
C4-Benzothiophenes	●	
C4-Chrysenes	●	●
C4-Decalin	●	
C4-Dibenzothiophenes	●	
C4-Fluoranthenes_Pyrenes	●	
C4-Naphthalenes	●	●
C4-Naphthobenzothiophenes	●	
C4-Phenanthrenes_Anthracenes	●	●
Carbazole	●	
Chrysene	●	●
Decalin	●	
Dibenzo[a,h]anthracene	●	●
Dibenzofuran	●	
Dibenzothiophene	●	●
Fluoranthene	●	●
Fluorene	●	●
Indeno[1,2,3-c,d]pyrene	●	●
Naphthalene	●	●
Naphthobenzothiophene	●	
Perylene	●	●
Phenanthrene	●	●
Pyrene	●	●

Results - Total PAHs

statistical analyses.

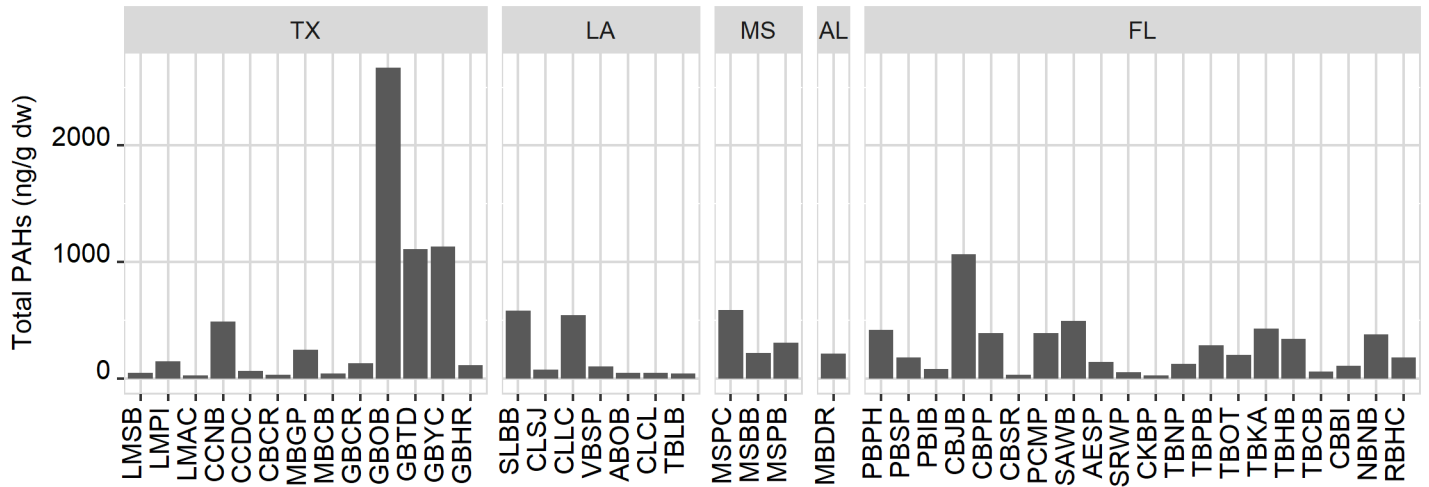


Figure 50. Bar graph showing magnitude of Total PAH concentration sums detected in oyster tissue in the Gulf of Mexico in 2017. Sites are listed geographically from west to east, following the coastline.

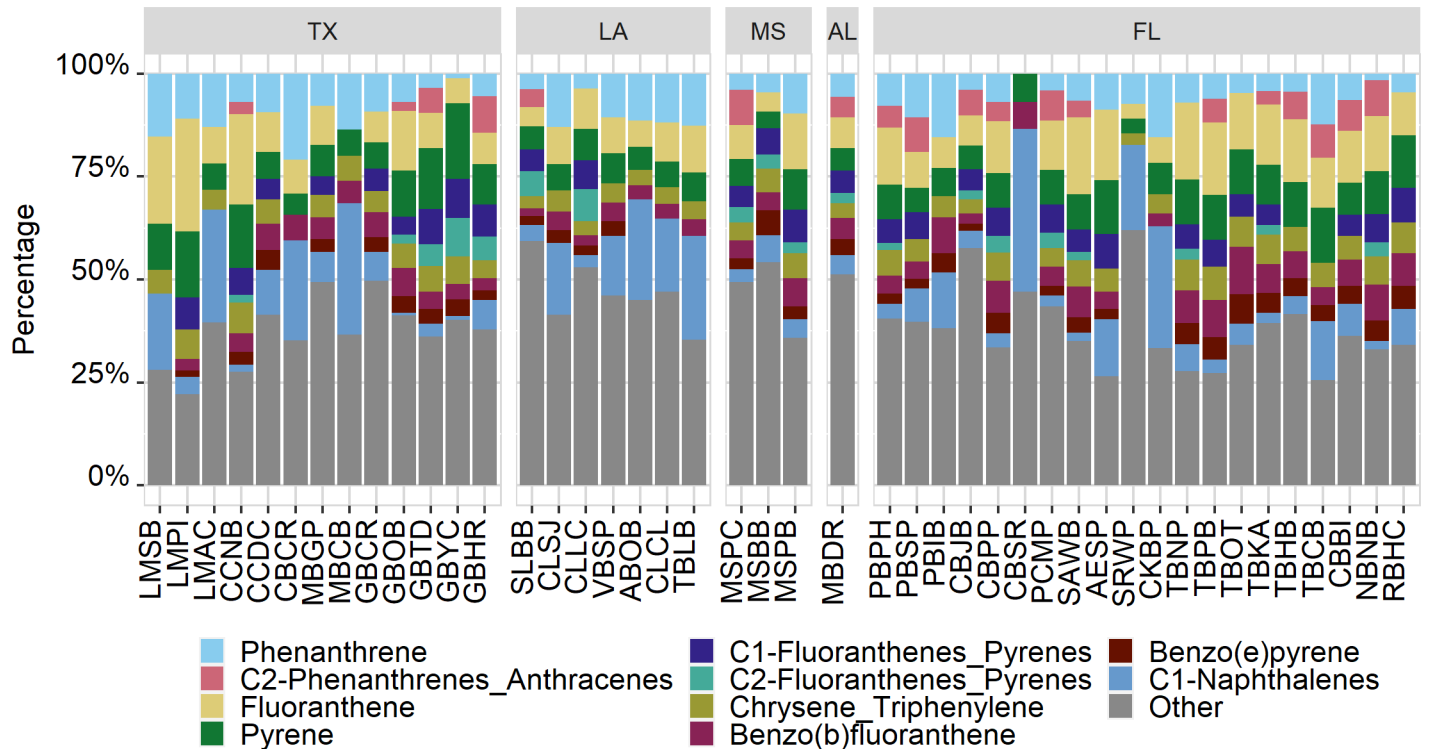


Figure 51. Percent composition of individual PAH contaminants in Total PAH concentration sums per site in the Gulf of Mexico in 2017. Sites are listed geographically from west to east, following the coastline. The 10 most prevalent PAH compounds were chosen to be included in this stacked boxplot, the remaining compounds were grouped together as "Other".

Results - Total PAHs

13.2 Magnitude and Distribution of PAHs in 2017

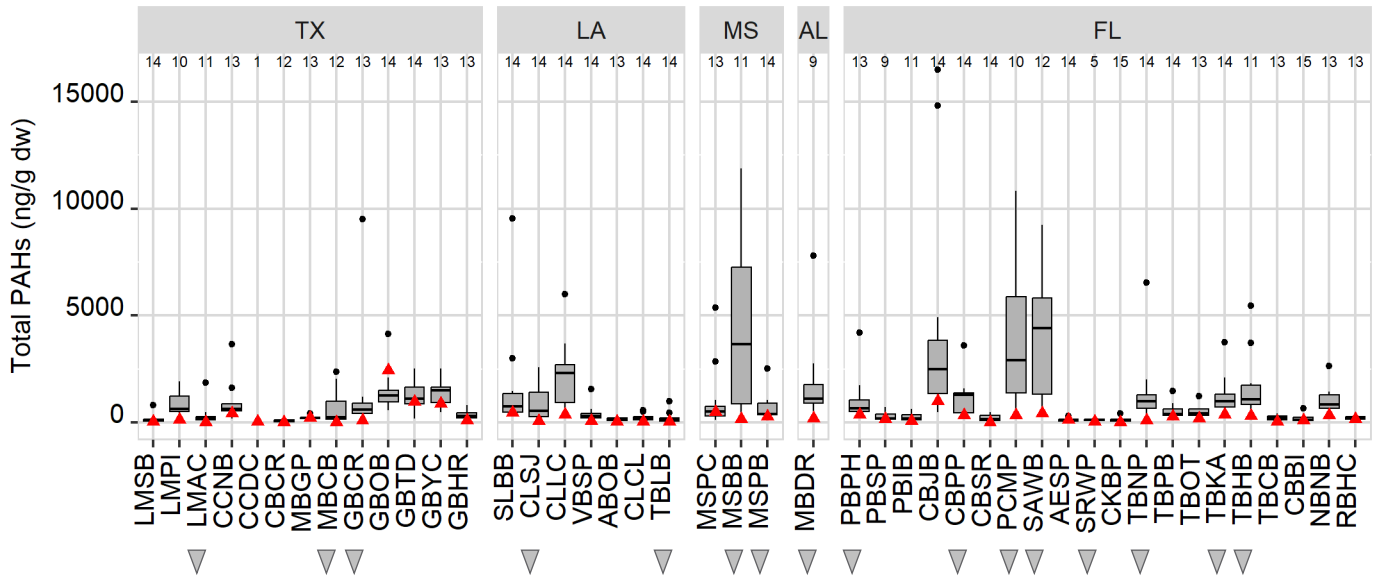


Figure 52. Total PAH concentration sums in 2017 (red triangles) compared to boxplots of the historic Total PAH concentration sums for each site (1990 - 2012) and results of the trend analysis showing significant increasing or decreasing contamination trends over time (gray triangles). Sites are listed geographically from west to east, following the coastline. The number of years each site has been sampled since 1990 is noted at the top of the plot for each site.

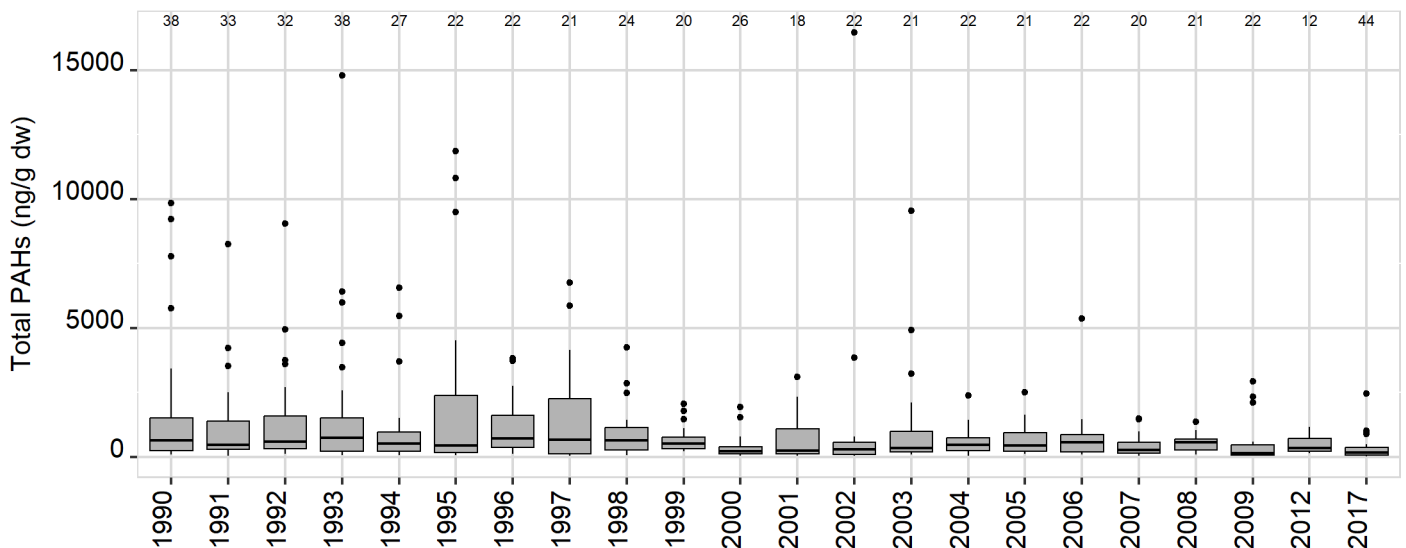


Figure 53. Boxplots representing the historic Total PAH concentration sums of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot for each year.

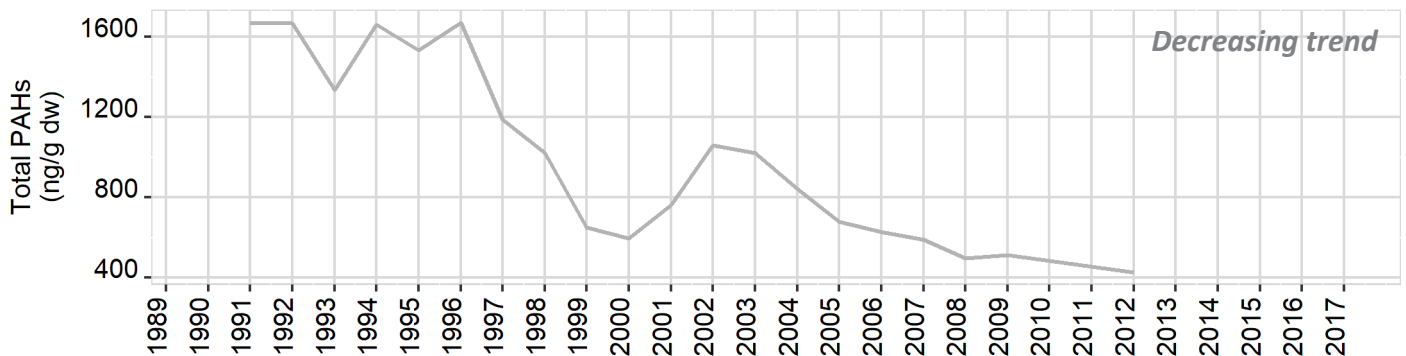


Figure 54. Three-point moving average of the yearly mean Total PAH concentration sums in the sites analyzed in this study.

Results - Total PAHs

13.3 Historical Context of PAHs Magnitude and Distribution

13.4 PAHs Summary

2017 Results (sum of 64 compounds):

- Total PAHs were detected at 100% of 44 sites analyzed
- Total PAHs concentration sums stats (Figure 50; Figure 51):
 - Range: 24.13 – 2,667.93 ng/g
 - Total PAHs were detected at all sites
 - Maximum concentration sum was detected at site GBOB (Galveston Bay, Offatts Bayou)
 - Median: 180.40 ng/g
 - Mean \pm SD: 326.47 \pm 458.44 ng/g

Historical Context (sum of 39 compounds):

- 7.0% sites were above their historic median concentrations in 2017
- 16 sites showed decreasing temporal trends of Total PAH concentration sums at $\alpha = 0.05$ (Figure 52; Figure 53; Table A6)
- 0 sites showed increasing temporal trends of Total PAH concentration sums at $\alpha = 0.05$ (Figure 52; Figure 53; Table A6)
- There was a significant decreasing regional trend over time for sites in 2017 ($p < 0.01$, $\rho = -0.83$) (Figure 54)
- The percentage of sites in the Gulf of Mexico in 2017 grouped into each cluster based on historic data (Figure 55):
 - 100% of sites in low cluster (19.5 – 3,236.1 ng/g)
 - 0% of sites in medium cluster (3,426.9 – 7,307.7 ng/g)
 - 0% of sites in high cluster (7,793.1 – 89,768.5 ng/g)

General Observations:

- Total PAH concentration sums were low throughout the Gulf of Mexico (Figure 55).
- Total PAH concentration sums at 10 sites exceeded the US EPA screening values for subsistence fishers and 1 site exceeded US EPA recommended screening values for recreational fishers (Table A2; Table A3).
- The concentrations of the 64 PAH compounds analyzed in 2017 were compared to the PAH50 totals from the same year. The only compound detected outside of the PAH50 was perylene. Across the region, perylene only accounted for 1.3% of the total PAHs detected (33.5 ng/g dw).

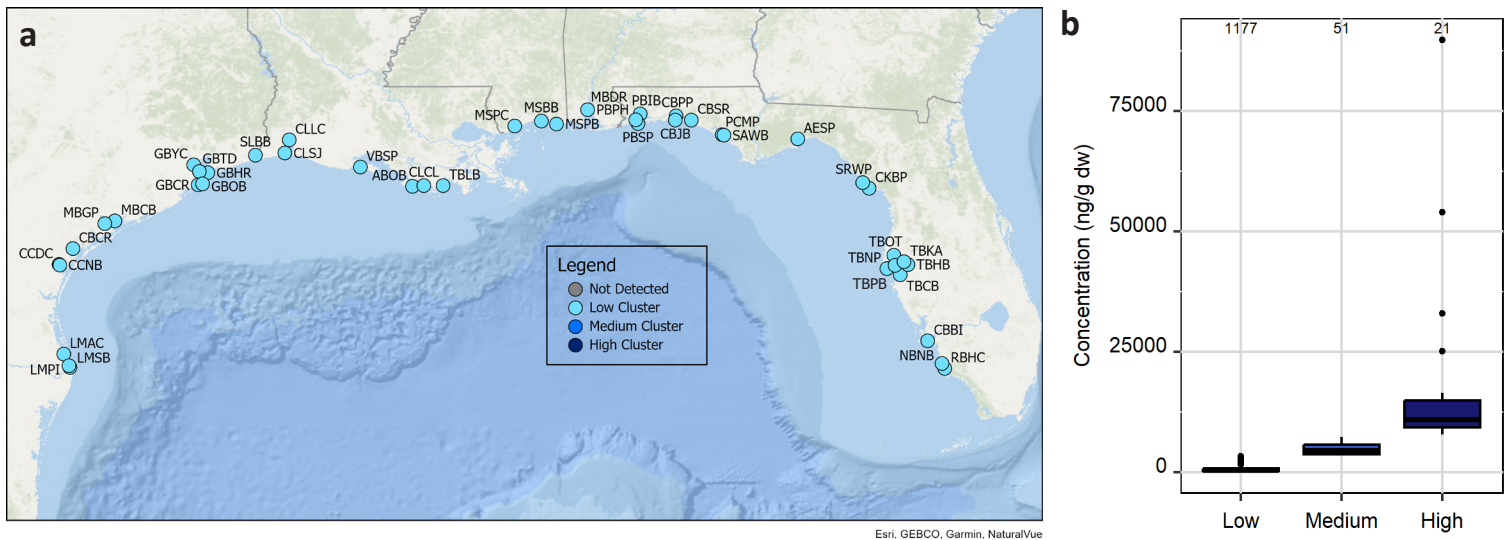


Figure 55. (a) Total PAH concentration sums in 2017 compared to the (b) historic national MWP *Crassostrea virginica* Total PAH concentration sums (ng/g dw). 123 national sites sampled between 1990 - 2017 for a total of 1249 samples.

Results - Total PCBs

14.0 RESULTS - TOTAL PCBs

14.1 PCBs Chemical Description

For this document, Total PCBs analyzed in 2017 is the sum of 156 compounds and Total PCBs analyzed historically is the sum of 18 compounds typically used to assess PCBs (Table 10). Polychlorinated Biphenyls (PCBs) are synthetic organic compounds that have been used in numerous applications including electrical transformers and capacitors, hydraulic and heat transfer fluids, pesticides and in paints (ATSDR, 2000). PCBs have a biphenyl ring structure (two benzene rings with a carbon to carbon bond) and a varying number (1-10) of chlorine atoms. There are 209 individual PCB compounds or congeners possible. PCBs were manufactured in the US between 1929 and 1977. In the US, a single manufacturer produced all PCBs and the commercial products were referred to as Aroclors, which are mixtures of PCB congeners. Approximately 65% of PCBs manufactured in the US were used in electrical applications (Eisler and Belisle, 1996). Other applications were lubricants, hydraulic fluids, paints, adhesives, plasticizers, and flame retardants (Kimbrough et al., 2008). Improper disposal and leakage are responsible for their original environmental introduction. Current pollution sources include volatilization and runoff from landfills, leaks from old electrical equipment, and dredging of contaminated sediments (WHO and IPCS, 1993).

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

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

As a note, combinations of which PCB compounds are grouped together in analysis change over time. The core 18 compounds that are used for most PCB comparisons were the basis for inclusion in historical analysis, and so any compound in the 2017 data that included one of those 18 compounds were included in the summary, even if the grouping included other compounds as well (i.e., PCB15 and PCB18 were analyzed separately in 2017 but were often grouped historically, so both were included in this historical analysis).

Table 10. PCB compounds composing the “Total PCBs” summation for the 2017 Gulf of Mexico survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2017	Analyzed in Historical Data
PCB1	●	
PCB2	●	
PCB3	●	
PCB4_10	●	
PCB6	●	
PCB7_9	●	
PCB8_5	●	●
PCB11	●	
PCB12	●	

Table 10 cont. PCB compounds composing the “Total PCBs” summation for the 2017 Gulf of Mexico survey. ● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2017	Analyzed in Historical Data
PCB13	●	
PCB14	●	
PCB15	●	●
PCB16_32	●	
PCB17	●	
PCB18	●	●
PCB19	●	
PCB21_20_33	●	
PCB22	●	

Results - Total PCBs

Table 10 cont. PCB compounds composing the "Total PCBs" summation for the 2017 Gulf of Mexico survey.

● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2017	Analyzed in Historical Data
PCB23	●	
PCB24	●	
PCB25	●	
PCB26	●	
PCB27	●	
PCB28_31	●	●
PCB29	●	
PCB30	●	
PCB34	●	
PCB35	●	
PCB36	●	
PCB37	●	
PCB38	●	
PCB39	●	
PCB40_57	●	
PCB42	●	
PCB43	●	
PCB44	●	●
PCB45	●	
PCB46_69_73	●	
PCB48_75_47	●	
PCB49	●	
PCB50	●	
PCB51	●	
PCB52	●	●
PCB53	●	
PCB54	●	
PCB55	●	
PCB56	●	
PCB58	●	
PCB59	●	
PCB60	●	
PCB61_74	●	
PCB62	●	
PCB63	●	

Table 10 cont. PCB compounds composing the "Total PCBs" summation for the 2017 Gulf of Mexico survey.

● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2017	Analyzed in Historical Data
PCB65	●	
PCB66_80	●	●
PCB67	●	
PCB68_41_64	●	
PCB71	●	
PCB72	●	
PCB76_70	●	
PCB77	●	
PCB78	●	
PCB79	●	
PCB81	●	
PCB82	●	
PCB85	●	
PCB88	●	
PCB89_113	●	
PCB91	●	
PCB92	●	
PCB94	●	
PCB96_103	●	
PCB97_125_86	●	
PCB99	●	
PCB100	●	
PCB101_84_90	●	●
PCB102_98	●	
PCB104	●	
PCB105_127	●	●
PCB106_107	●	
PCB109	●	
PCB110	●	
PCB111_115_87	●	
PCB112	●	
PCB114_122	●	
PCB116_117	●	
PCB118_108	●	●
PCB119	●	

Results - Total PCBs

Table 10 cont. PCB compounds composing the "Total PCBs" summation for the 2017 Gulf of Mexico survey.

● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2017	Analyzed in Historical Data
PCB120_83	●	
PCB121_93_95	●	
PCB123	●	
PCB124	●	
PCB126	●	
PCB128_167	●	●
PCB129	●	
PCB130	●	
PCB132	●	●
PCB134_133	●	
PCB135	●	
PCB136_154	●	
PCB137	●	
PCB138_164_163	●	●
PCB140	●	
PCB141	●	
PCB142_146_161	●	
PCB143	●	
PCB144	●	
PCB147	●	
PCB148_145	●	
PCB149_139	●	
PCB150	●	
PCB151	●	
PCB152	●	
PCB153_168	●	●
PCB155	●	
PCB156	●	
PCB157	●	
PCB159	●	
PCB160_158	●	●
PCB162	●	
PCB165_131	●	
PCB166	●	

Table 10 cont. PCB compounds composing the "Total PCBs" summation for the 2017 Gulf of Mexico survey.

● signifies that the compound was included in the respective dataset.

Compound	Analyzed in 2017	Analyzed in Historical Data
PCB169	●	
PCB170_190	●	●
PCB171	●	
PCB173	●	
PCB174	●	
PCB175	●	
PCB176	●	
PCB177	●	
PCB179	●	
PCB180_193	●	●
PCB181	●	
PCB183	●	
PCB184	●	
PCB185	●	
PCB186_178	●	
PCB187_182	●	●
PCB188	●	
PCB189	●	
PCB191	●	
PCB192_172	●	
PCB194	●	
PCB195	●	●
PCB197	●	
PCB199	●	
PCB200	●	
PCB201	●	
PCB202	●	
PCB203_196	●	
PCB204	●	
PCB205	●	
PCB206	●	●
PCB207	●	
PCB208	●	●
PCB209	●	●

Results - Total PCBs

14.2 Magnitude and Distribution of PCBs in 2017

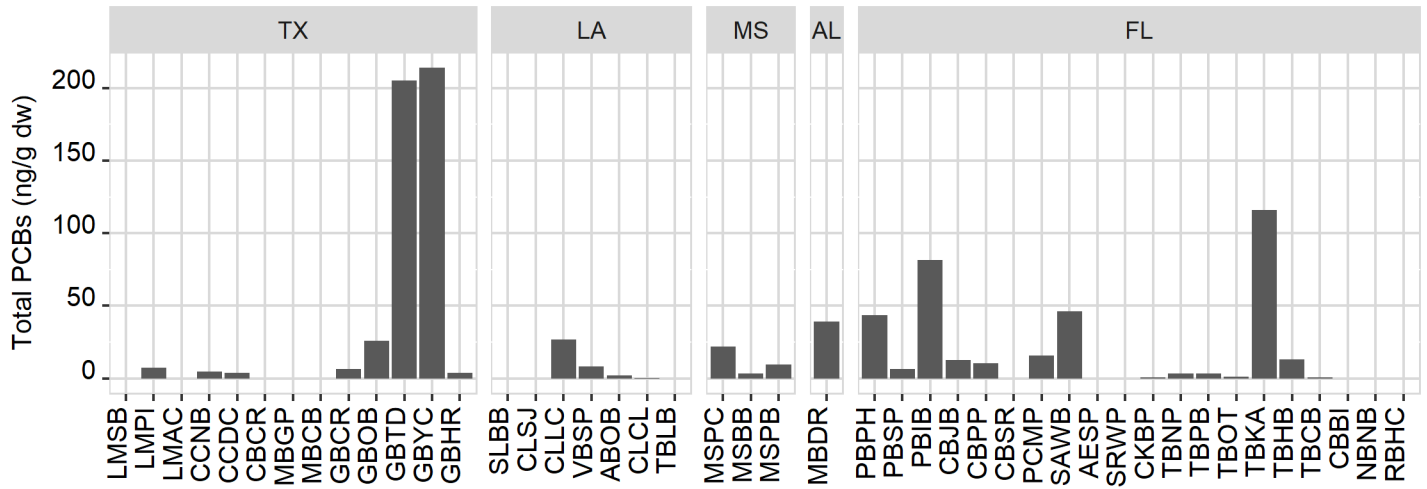


Figure 56. Bar graph showing magnitude of Total PCB concentration sums detected in oyster tissue in the Gulf of Mexico in 2017. Sites are listed geographically from west to east, following the coastline.

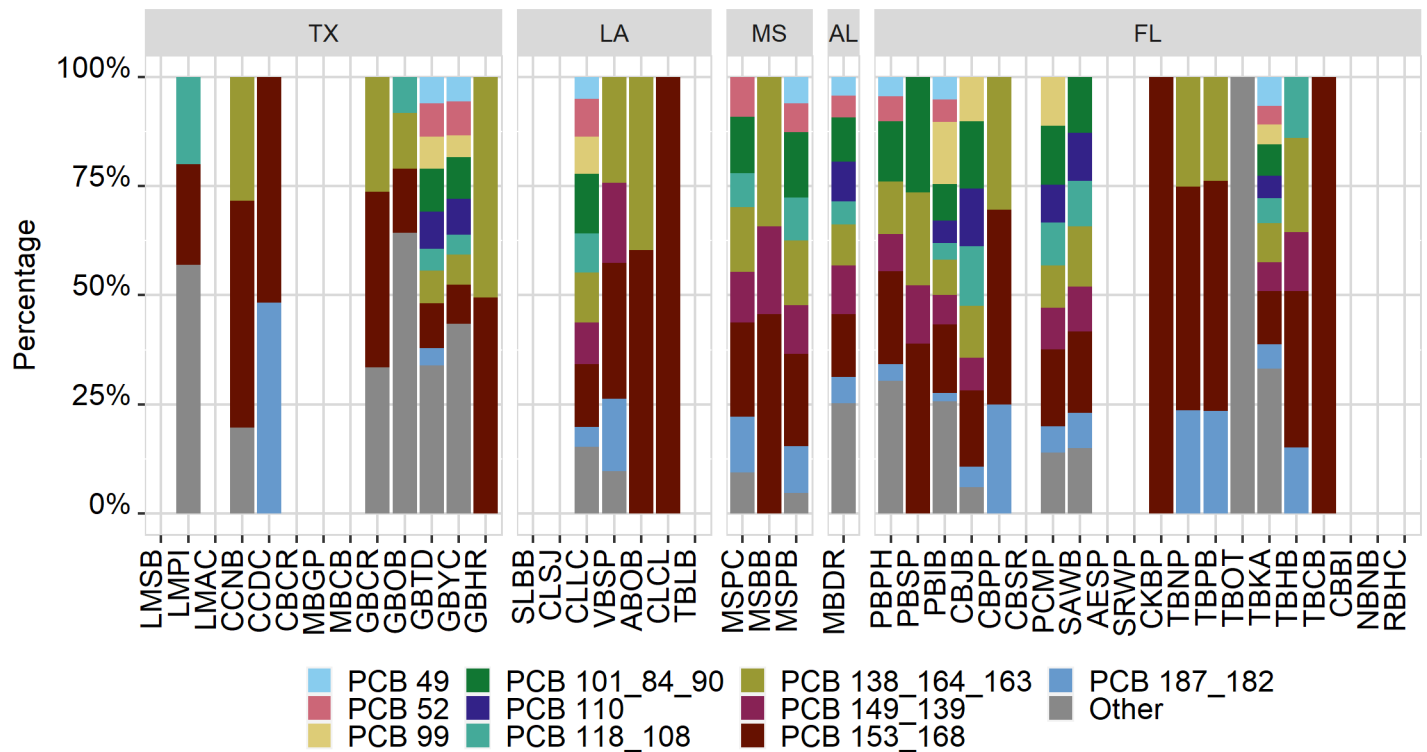


Figure 57. Percent composition of individual PCB contaminants in Total PCB concentration sums per site in the Gulf of Mexico in 2017. Sites are listed geographically from west to east, following the coastline. The 10 most prevalent PCB compounds were chosen to be included in this stacked boxplot, the remaining compounds were grouped together as "Other".

Results - Total PCBs

14.3 Historical Context of PCBs Magnitude and Distribution

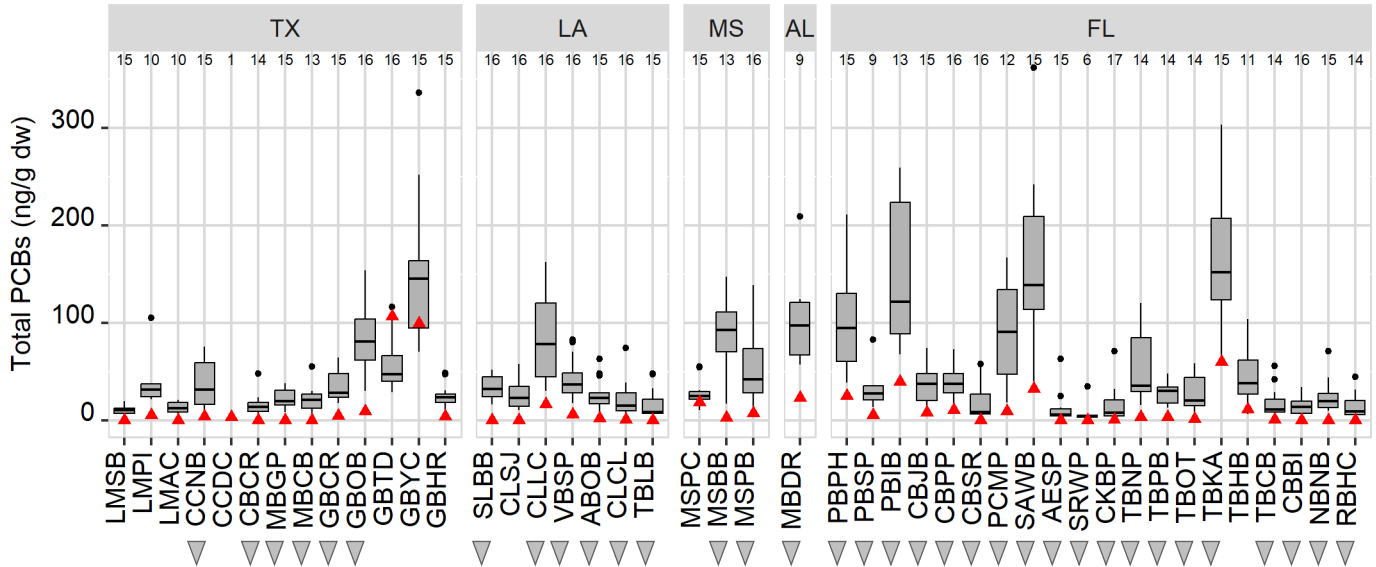


Figure 58. Total PCB concentration sums in 2017 (red triangles) compared to boxplots of the historic Total PCB concentration sums for each site (1988 - 2012) and results of the trend analysis showing significant increasing or decreasing contamination trends over time (gray triangles). Sites are listed geographically from west to east, following the coastline. The number of years each site has been sampled since 1988 is noted at the top of the plot for each site.

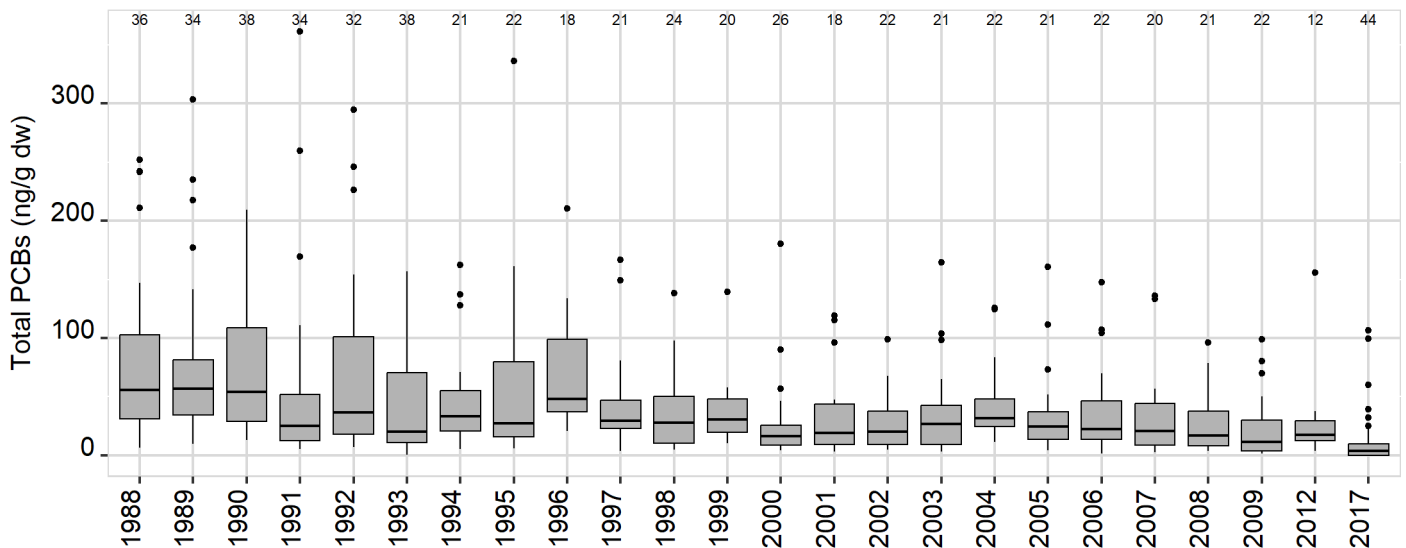


Figure 59. Boxplots representing the historic Total PCB concentration sums of the sites analyzed in this study. The number of sites that were sampled in each year is noted at the top of the plot for each year.

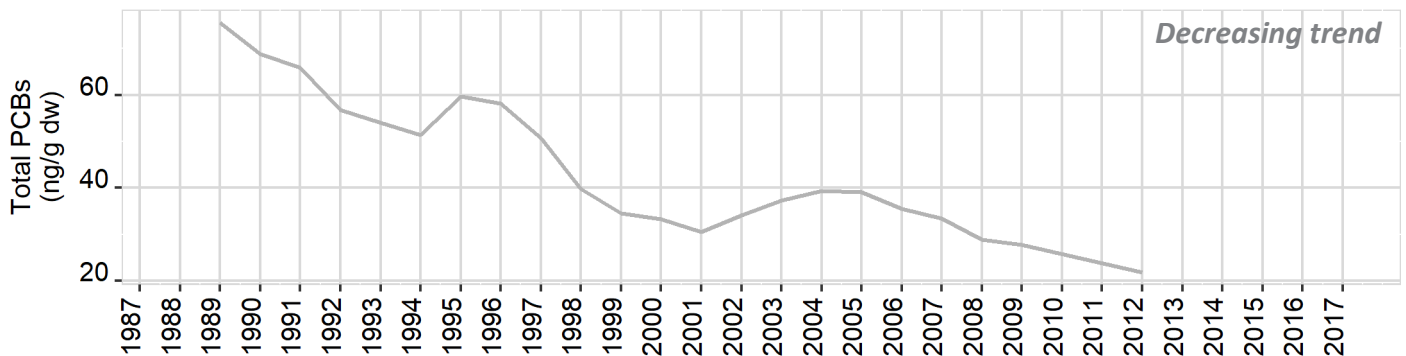


Figure 60. Three-point moving average of the yearly mean Total PCB concentration sums in the sites analyzed in this study.

Results - Total PCBs

14.4 PCBs Summary

2017 Results (sum of 156 compounds):

- Total PCBs were detected at 68.2% of 44 sites analyzed
- Total PCBs concentration sums stats (Figure 56; Figure 57):
 - Range: 0.44 – 214.04 ng/g
 - Total PCB were not detected at 14 sites
 - Maximum concentration sum was detected at site GBYC (Galveston Bay, Yacht Club)
 - Median: 3.60 ng/g
 - Mean \pm SD: 21.29 \pm 47.40 ng/g

Historical Context (sum of 18 compounds):

- 2.3% sites were above their historic median concentrations in 2017
- 34 sites showed decreasing temporal trends of Total PCB concentration sums at $\alpha = 0.05$ (Figure 58; Figure 59; Table A6)
- 0 sites showed increasing temporal trends of Total PCB concentration sums at $\alpha = 0.05$ (Figure 58; Figure 59; Table A6)
- There was a significant decreasing regional trend over time for sites in 2017 ($p < 0.001$, $\rho = -0.87$) (Figure 60)
- The percentage of sites in the Gulf of Mexico in 2017 grouped into each cluster based on historic data (Figure 61):
 - 93.2% of sites in low cluster (0.0 – 41.1 ng/g)
 - 6.8% of sites in medium cluster (41.7 – 188.9 ng/g)
 - 0.0% of sites in high cluster (195.3 – 767.5 ng/g)

General Observations:

- Total PCBs concentration sums were mostly low or not detected throughout the Gulf of Mexico (Figure 61).
- Total PCBs were clustered into the medium cluster at three sites within the Gulf of Mexico (GBTD, GBYC, and TBKA), possibly suggesting localized point sources of contamination (Figure 61).
- Total PCB concentration sums at 4 sites exceeded the US EPA screening values for subsistence fishers and no sites exceeded US EPA recommended screening values for recreational fishers (Table A2; Table A3).

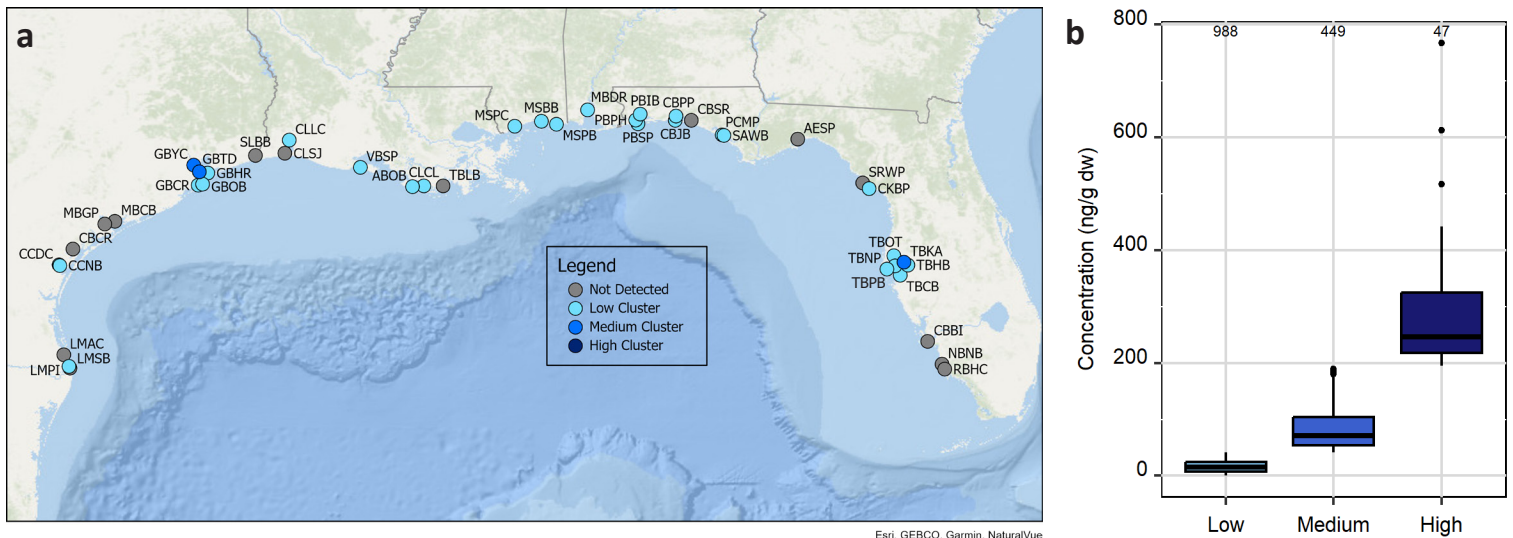


Figure 61. (a) Total PCB concentration sums in 2017 compared to the (b) historic national MWP *Crassostrea virginica* Total PCB concentration sums (ng/g dw). 127 national sites sampled between 1988 - 2017 for a total of 1484 samples.

15.0 SUMMARY

Oysters are good indicators of water quality; hence, they have been used worldwide as sentinel species for chemical pollution in aquatic systems. In this study, oyster tissue samples (*Crassostrea virginica*) were assessed for a suite of legacy organic contaminants. The oyster samples were collected at historic MWP monitoring sites located within the Gulf of Mexico. Sample collection was conducted by TDI Brooks International following standard protocols (Apeti et al., 2012). Oyster tissue was analyzed from 44 monitoring sites for legacy organic contaminants. Separate result summaries for each legacy organic contaminant group can be found in the Summary subsection of each legacy organic contaminant group section within this document. This summary attempts to integrate all contamination results into one analysis to assess overall contamination of sites in the Gulf of Mexico, both with respect to the other sites analyzed within this study and to the larger historical Mussel Watch dataset. Overall site contamination analysis was conducted in two ways. For both analyses, a multivariate cluster analysis was conducted for each contaminant group at each site and represented in a heatmap and then a second multivariate cluster analysis was conducted using the total cluster value of each site to group sites into five clusters with statistically different degrees of overall contamination (not detected, low, medium, high, very high). This two-pronged analysis was conducted (1) as respective of sites within this study and (2) as respective to all historic MWP contamination analyzed in oysters.

The first observation of note is that all sites assessed for legacy organic contaminants in the Gulf of Mexico had at least one contaminant group present, emphasizing the ubiquity of these contaminants even with environmental legislation and restrictions, often imposed many years prior. This showcases how long lasting these contaminants can be in the environment and why they still merit continuous monitoring (Table 11; Figure 62; Figure 64). Also notable is that there were no detections of dieldrins, endosulfans, or mirex at any sites in the Gulf of Mexico (Table 11).

For analyses conducted respective only to sites analyzed in this 2017 study, 25.0% of sites were clustered into the low contamination category, 34.1% in the medium category, 20.5% in the high category, and 20.5% in the very high category (Figure 63). In general, sites categorized as having high or very high contamination tended to be closer to more densely populated areas such as Tampa Bay, FL, Galveston Bay, TX, and along the Alabama, Mississippi, and Florida panhandle coastline (Figure 63). Oppositely, sites categorized as having low or medium contamination tended to be in less populated areas.

Table 11. Frequency of detection (% of total) of legacy contaminant groups in oyster tissue at sites within the Gulf of Mexico.

Contaminant Group	Sites with Detects	Sites Analyzed	Detection Frequency
Total Butyltins	17	41	41%
Total Chlordanes	13	44	30%
Total Chlorobenzenes	1	44	2%
Chlorpyrifos	3	44	7%
Total DDTs	36	44	82%
Total Dieldrins	0	44	0%
Total Endosulfans	0	44	0%
Total HCHs	2	44	5%
Mirex	0	44	0%
Total PAHs	44	44	100%
Total PCBs	30	44	68%

Summary

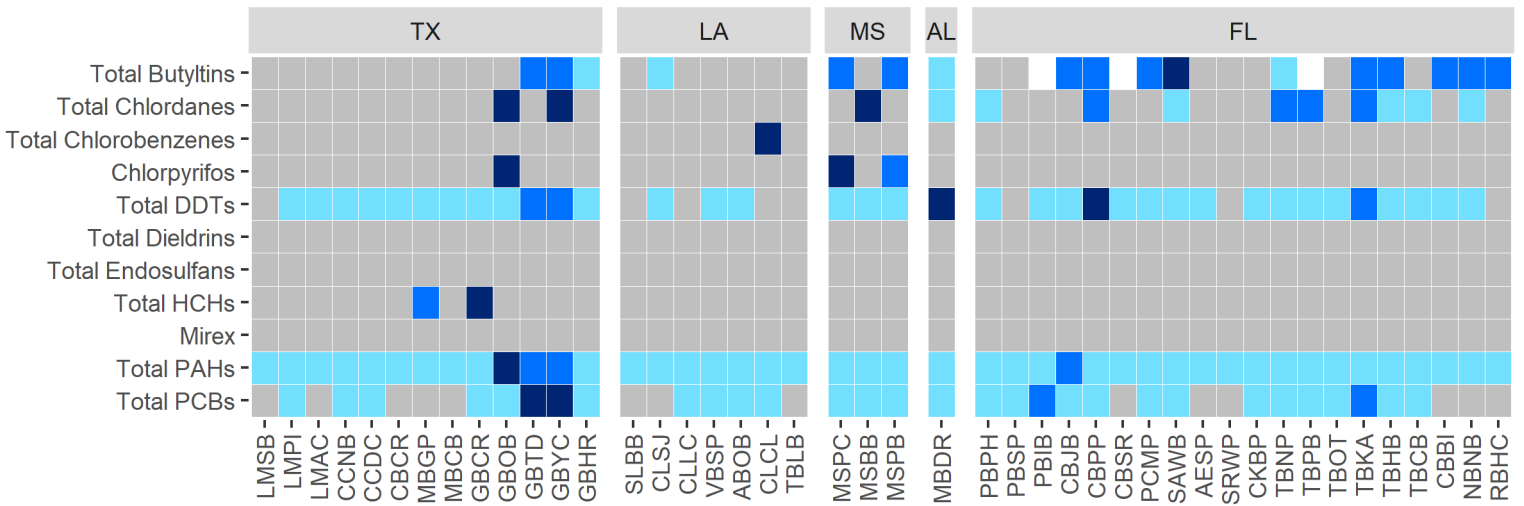


Figure 62. Distribution map showing the distribution and magnitude of 2017 concentrations detected in the Gulf of Mexico, respective to one another in this study. Colors represent contamination magnitude as follows: low (light blue), medium (medium blue), high (dark blue), and not detected (grey). White represents missing or non-quantifiable data.

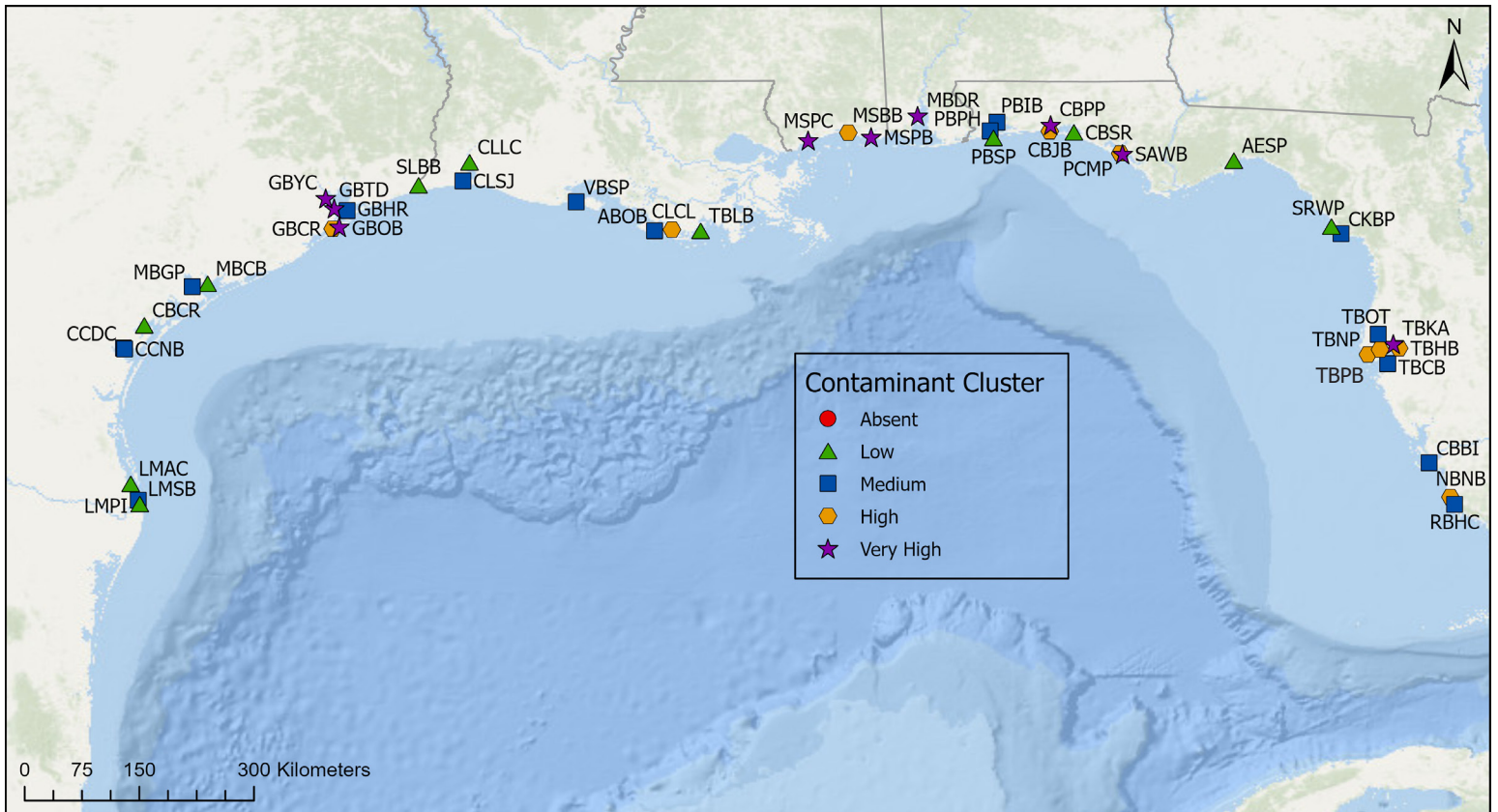


Figure 63. Map of sites sampled in the Gulf of Mexico in 2017 highlighting locations with Low, Medium, High, and Very High degrees of contamination, respective to one another in this study. Esri, GEBCO, Garmin, NaturalVue

Summary

While it is useful to know which sites in the Gulf of Mexico are most contaminated relative to other sites in the region, it is equally important to put contamination of a region into perspective with other available data nationwide and historically. When compared to all legacy organic contaminant data collected by the MWP for oysters since 1986, 31.8% of sites were clustered into the low contamination category, 25.0% in the medium category, 25.0% in the high category, and 18.2% in the very high category (Figure 65). In general, sites categorized as having high or very high contamination tended to be closer to more densely populated areas such as Tampa Bay, FL, Galveston Bay, TX, and along the Alabama, Mississippi, and Florida panhandle coastline (Figure 65). This is similar to results seen when only comparing sites analyzed in 2017 to one another (Figure 63).

Most contaminant groups have decreased in concentration over time; however, several contaminant groups have persisted at background levels in the environment, suggesting that achieving further significant reductions may be challenging (Table 12). Conversely, the only increasing trend was seen at site TBLB (Terrebonne Bay Lake Barre) for Total Chlorobenzenes (Table 12). The numerous decreasing trends of many of the legacy organic contaminants demonstrate the importance of long-standing monitoring programs and provide a point of reference for current sampling. These trends allow us to observe the effects of regulations on the concentrations of these contaminants in the environment and to put into perspective any recently detected concentrations. The absence of detections of Mirex, Total Dieldrins, and Total Endosulfans in the Gulf of Mexico in 2017 suggest that these contaminants may no longer be widely present in the region at detectable levels, but further sampling may be needed to confirm (Figure 62; Figure 64). Oppositely, even when compared to national historic oyster data, three legacy organic contaminant groups were present at medium to high levels at several sites in the Gulf of Mexico (Figure 61; Figure 64). These three specifically include chlorpyrifos at 3 sites (GBOB, MSPC, and MSPB), Total DDTs at 3 sites (GBTD, MBDR, and CBPP), and Total PCBs at 3 sites (GBTD, GBYC, and TBKA). Further analysis and sampling may be beneficial to determine the current point sources of contamination of these legacy organic contaminants.

The low concentrations of most legacy organic contaminants detected in the Gulf of Mexico when compared to nationwide NS&T data support the need for consistent monitoring programs that can contextualize any given result. A successful monitoring program must both achieve an understanding of background contamination levels and capture the variability and range of possible contamination. Existing contaminant data has provided an understanding of general background contamination including range, trends, and variability. Further monitoring should aim to continue analyzing the temporal trends of these contaminants on a regional scale (i.e., through periodic sampling and biennial sampling at select sites) but focus on areas of interest that could give further insight on range, variation, and potential localized point sources of contamination. For example, further sampling around Tampa Bay, FL, Galveston Bay, TX, and along the Alabama, Mississippi, and Florida panhandle coastline could help to clarify the anomalously very high overall contamination seen there relative to other areas in the Gulf of Mexico. Through an understanding of both the temporal and spatial variations, a monitoring program can serve its purpose of assessing potential contaminant exposure. This study provides needed data and information for the MWP and provides contamination data required by coastal resource managers as they develop long-term policies to protect the services provided by the coastal environment within this region.

Summary

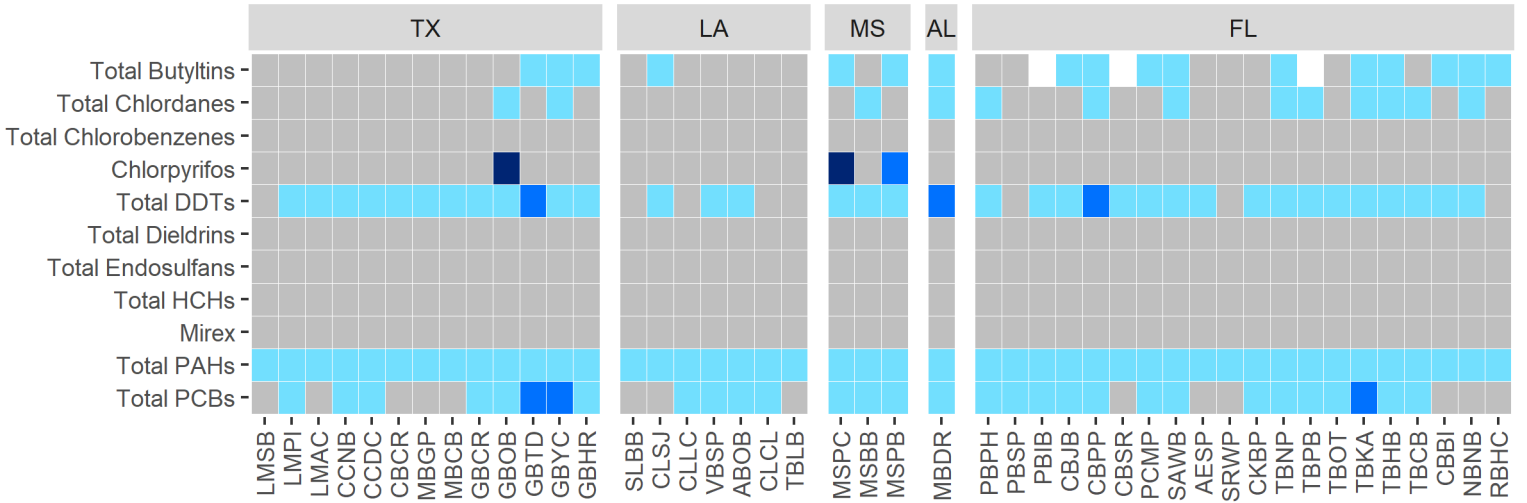
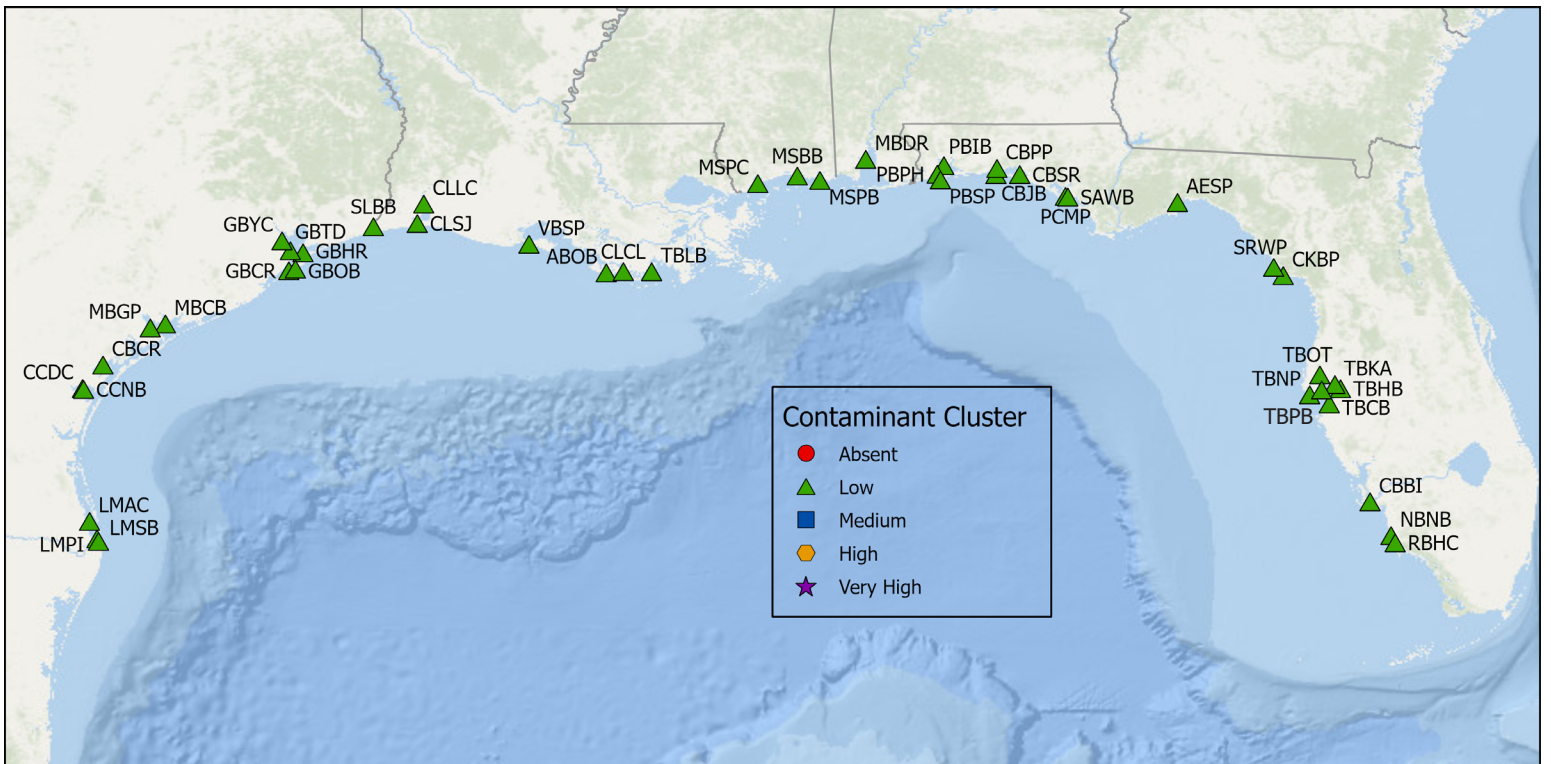


Figure 64. Distribution map showing the distribution and magnitude of 2017 concentrations detected in the Gulf of Mexico, relative to all historic MWP contamination analyzed in *Crassostrea virginica*. Colors represent contamination magnitude as follows: low (light blue), medium (medium blue), high (dark blue), and not detected (grey). White represents missing or non-quantifiable data.



Esri, GEBCO, Garmin, NaturalVue

Figure 65. Map of sites sampled in the Gulf of Mexico in 2017 highlighting locations with Low, Medium, High, and Very High degrees of contamination, relative to all historic MWP contamination analyzed in *Crassostrea virginica*.

Summary

Table 12. Summary of site-based trend analysis for legacy organic contaminants; D = decreasing trend, I = increasing trend.

Site	Total Butyltins	Total Chlordanes	Total Chlorobenzenes	Chlorpyrifos	Total DDTs	Total Dieldrins	Total Endosulfans	Total HCHs	Mirex	Total PAHs	Total PCBs
ABOB		D			D	D		D			D
AESP	D	D			D	D					D
CBBI	D	D			D	D	D				D
CBCR	D	D			D	D	D	D			D
CBJB	D	D			D	D			D		D
CBPP		D	D		D			D		D	D
CBSR		D			D	D					D
CCNB		D			D	D	D	D			D
CKBP		D		D	D	D		D	D		D
CLCL		D			D	D		D			D
CLLC	D	D	D		D	D			D		D
CLSJ		D			D	D		D	D	D	
GBCR	D	D			D	D		D		D	D
GBHR	D				D	D		D			
GBOB	D				D			D			D
GBTD	D							D			
GBYC	D			D		D		D	D		
LMAC					D					D	
LMPI	D				D	D					
LMSB	D	D		D	D	D		D			
MBCB		D			D	D	D			D	D
MBDR	D	D		D					D	D	D
MBGP	D	D			D	D	D	D			D
MSBB	D	D			D			D	D	D	D
MSPB	D	D			D	D			D	D	D
MSPC	D				D			D			
NBNB	D	D			D				D		D
PBIB	D	D			D	D		D	D		D
PBPH	D	D			D			D		D	D
PBSP					D			D			D
PCMP	D	D			D	D			D	D	D
RBHC	D	D			D						D
SAWB	D	D			D	D		D		D	D
SLBB	D	D			D	D		D			D
SRWP					D		D		D	D	D
TBCB	D	D		D	D	D			D		D
TBHB	D			D				D		D	
TBKA	D	D		D	D	D		D	D	D	D
TBLB		D	I		D	D	D	D	D	D	D
TBNP	D	D		D	D	D		D	D	D	D
TBOT	D	D		D	D	D	D	D			D
TBPB	D	D		D	D	D		D	D		D
VBSP	D	D			D	D		D	D		D

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Appendices

APPENDICES

Table A1. Percent dry values for tissue samples at each site collected in the Gulf of Mexico in 2017.

Site	% Dry of Tissue Samples*	Site	% Dry of Tissue Samples*
ABOB	9.68	MBDR	9.40
AESP	4.85	MBGP	4.10
CBBI	12.15	MSBB	6.84
CBCR	9.57	MSPB	6.84
CBJB	10.14	MSPC	4.96
CBPP	7.25	NBNB	9.82
CBSR	8.89	PBIB	10.09
CCDC	11.20	PBPH	7.21
CCNB	10.14	PBSP	8.82
CKBP	14.52	PCMP	10.22
CLCL	9.35	RBHC	3.88
CLLC	5.71	SAWB	8.80
CLSJ	7.14	SLBB	3.68
GBCR	7.58	SRWP	12.70
GBHR	7.48	TBCB	12.39
GBOB	7.14	TBHB	9.24
GBTD	1.68	TBKA	9.80
GBYC	7.14	TBLB	9.09
LMAC	10.71	TBNP	11.95
LMPI	10.43	TBOT	5.45
LMSB	8.18	TBPB	12.00
MBCB	5.52	VBSP	6.50

* $conc. (ng/g ww) = conc. (ng/g dw) \times (\% dry / 100)$

Table A2. US FDA Action and Tolerance levels and US EPA Screening Values (SVs) for chemical contaminants in fish and shellfish (ng/g wet weight) compared to the maximum detected concentration in the Gulf of Mexico in 2017 dataset (FDA, 2011; EPA, 2000).

Contaminant Group	Compounds	Maximum Detected Value	US FDA Action Level	US FDA Tolerance Level	US EPA Recreational Fishers Values - Noncarcinogenic	US EPA Recreational Fishers Values - Carcinogenic	US EPA Subsistence Fishers Values - Noncarcinogenic	US EPA Subsistence Fishers Values - Carcinogenic
Total Butyltins	Tributyltin	5.13			1200		147	
Total Dieldrins	Aldrin	0.00	300					0.307
	Dieldrin	0.00				2.5		
Total Dieldrins 2	Endrin	0.00			1200		147	
Total Endosulfans	Endosulfan I	0.00			24000		2949	
	Endosulfan II	0.00						
Total Chlorodanes 2	Heptachlor	0.00	300			4.39		0.54
	Heptachlor Epoxide	0.00						
Total Chlorobenzenes	Hexachlorobenzene	0.00				250		3.07
Total HCHs	Lindane / γ-HCH	0.00				30.7		3.78
	Mirex	0.00	100		800		98	
Total PAHs*	15 compounds**	20.07				5.47		0.673
Total Chlorodanes	Alpha-Chlordane							
	Gamma-Chlordane							
	Cis-Nonachlor	4.51	300			114		14
	Trans-Nonachlor							
	Oxychlordane							
Total DDTs	2,4'-DDT							
	4,4'-DDT							
	2,4'-DDE	9.12	5000			117		14.4
	4,4'-DDE							
	2,4'-DDD							
Total PCBs	4,4'-DDD							
	18 congeners***	6.89		2000		20		2.45

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

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

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

Appendices

Table A3. Total concentrations of PAHs and PCBs in the Gulf of Mexico in 2017 datasets compared to the US EPA Screening Values (SVs) for chemical contaminants in fish and shellfish (ng/g wet weight) (FDA, 2011; US EPA, 2000).

Site	State	Total PAHs Concentration	PAHs Flag	Total PCBs Concentration	PCBs Flag	Site	State	Total PAHs Concentration	PAHs Flag	Total PCBs Concentration	PCBs Flag
ABOB	LA	0.02		0.18		MBDR	AL	0.35		2.07	
AESP	FL	0.04		0.00		MBGP	TX	0.31		0.00	
CBBI	FL	0.34		0.00		MSBB	MS	0.58		0.19	
CBCR	TX	0.02		0.00		MSPB	MS	0.57		0.51	
CBJB	FL	0.87	*	0.81		MSPC	MS	0.52		0.92	
CBPP	FL	2.58	*	0.77		NBNB	FL	0.77	*	0.00	
CBSR	FL	0.02		0.00		PBIB	FL	0.09		4.05	*
CCDC	TX	0.36		0.40		PBPH	FL	0.21		1.82	
CCNB	TX	1.39	*	0.37		PBSP	FL	0.11		0.49	
CKBP	FL	0.02		0.07		PCMP	FL	0.63		0.91	
CLCL	LA	0.02		0.04		RBHC	FL	0.24		0.00	
CLLC	LA	0.29		0.93		SAWB	FL	1.03	*	2.72	*
CLSJ	LA	0.03		0.00		SLBB	LA	0.25		0.00	
GBCR	TX	0.11		0.33		SRWP	FL	0.00		0.00	
GBHR	TX	0.03		0.27		TBCB	FL	0.04		0.10	
GBOB	TX	20.07	**	0.66		TBHB	FL	0.90	*	1.04	
GBTD	TX	0.30		1.79		TBKA	FL	2.48	*	5.71	*
GBYC	TX	1.14	*	6.89	*	TBLB	LA	0.02		0.00	
LMAC	TX	0.00		0.00		TBNP	FL	0.37		0.43	
LMPI	TX	0.09		0.45		TBOT	FL	0.61		0.05	
LMSB	TX	0.00		0.00		TBPB	FL	0.79	*	0.42	
MBCB	TX	0.01		0.00		VBSP	LA	0.20		0.39	

* exceeds the US EPA Subsistence Fishers Screening Value

** exceeds the US EPA Recreational Fishers Screening Value

Appendices

Table A4. Breakdown of cluster analysis for oyster tissue in the Gulf of Mexico respective to all historic MWP contamination analyzed in *Crassostrea virginica*. The first section of the table is the cluster value assigned for each chemical class. The second section of the table is the calculations conducted to normalize the chemical class cluster sums by number of chemical classes assessed at each site. The final column is the overall chemical contamination cluster rank assigned to each site. 0 = not detected, 1 = low, 2 = medium, 3 = high, 4 = very high. Total ChlBenz = Total Chlorobenzenes. Total Endo. = Total Endosulfans.

Site	Total Butyltins	Total Chlordanes	Total ChlBenz	Chlor-pyrifos	Total DDTs	Total Dieldrins	Total Endo.	Total HCHs	Mirex	Total PAHs	Total PCBs	Cluster Sum	#Classes Analyzed	Normalized Cluster	Overall Cluster
ABOB	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	2
AESP	0	0	0	0	1	0	0	0	0	1	0	2	11	6.1	1
CBBI	1	0	0	0	1	0	0	0	0	1	0	3	11	9.1	2
CBCR	0	0	0	0	1	0	0	0	0	1	0	2	11	6.1	1
CBJB	1	0	0	0	1	0	0	0	0	1	1	4	11	12.1	3
CBPP	1	1	0	0	2	0	0	0	0	1	1	6	11	18.2	4
CBSR		0	0	0	1	0	0	0	0	1	0	2	10	6.7	1
CCDC	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	2
CCNB	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	2
CKBP	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	2
CLCL	0	0	0	0	0	0	0	0	0	1	1	2	11	6.1	1
CLLC	0	0	0	0	0	0	0	0	0	1	1	2	11	6.1	1
CLSJ	1	0	0	0	1	0	0	0	0	1	0	3	11	9.1	2
GBCR	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	2
GBHR	1	0	0	0	1	0	0	0	0	1	1	4	11	12.1	3
GBOB	0	1	0	3	1	0	0	0	0	1	1	7	11	21.2	4
GBTD	1	0	0	0	2	0	0	0	0	1	2	6	11	18.2	4
GBYC	1	1	0	0	1	0	0	0	0	1	2	6	11	18.2	4
LMAC	0	0	0	0	1	0	0	0	0	1	0	2	11	6.1	1
LMPI	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	2
LMSB	0	0	0	0	0	0	0	0	0	1	0	1	11	3	1
MBCB	0	0	0	0	1	0	0	0	0	1	0	2	11	6.1	1
MBDR	1	1	0	0	2	0	0	0	0	1	1	6	11	18.2	4
MBGP	0	0	0	0	1	0	0	0	0	1	0	2	11	6.1	1
MSBB	0	1	0	0	1	0	0	0	0	1	1	4	11	12.1	3
MSPB	1	0	0	2	1	0	0	0	0	1	1	6	11	18.2	4
MSPC	1	0	0	3	1	0	0	0	0	1	1	7	11	21.2	4
NBNB	1	1	0	0	1	0	0	0	0	1	0	4	11	12.1	3
PBIB		0	0	0	1	0	0	0	0	1	1	3	10	10	2
PBPH	0	1	0	0	1	0	0	0	0	1	1	4	11	12.1	3
PBSP	0	0	0	0	0	0	0	0	0	1	1	2	11	6.1	1
PCMP	1	0	0	0	1	0	0	0	0	1	1	4	11	12.1	3
RBHC	1	0	0	0	0	0	0	0	0	1	0	2	11	6.1	1
SAWB	1	1	0	0	1	0	0	0	0	1	1	5	11	15.2	3
SLBB	0	0	0	0	0	0	0	0	0	1	0	1	11	3	1
SRWP	0	0	0	0	0	0	0	0	0	1	0	1	11	3	1
TBCB	0	1	0	0	1	0	0	0	0	1	1	4	11	12.1	3
TBHB	1	1	0	0	1	0	0	0	0	1	1	5	11	15.2	3
TBKA	1	1	0	0	1	0	0	0	0	1	2	6	11	18.2	4
TBLB	0	0	0	0	0	0	0	0	0	1	0	1	11	3	1
TBNP	1	1	0	0	1	0	0	0	0	1	1	5	11	15.2	3
TBOT	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	2
TBPB		1	0	0	1	0	0	0	0	1	1	4	10	13.3	3
VBSP	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	2

Appendices

Table A5. Breakdown of cluster analysis for oyster tissue in the Gulf of Mexico respective to one another in this 2017 study. The first section of the table is the cluster value assigned for each chemical class. The second section of the table is the calculations conducted to normalize the chemical class cluster sums by number of chemical classes assessed at each site. The final column is the overall chemical contamination cluster rank assigned to each site. 0 = not detected, 1 = low, 2 = medium, 3 = high, 4 = very high. Total ChlBenz = Total Chlorobenzenes. Total Endo. = Total Endosulfans.

Site	Total Butyltins	Total Chlordanes	Total ChlBenz	Chlorpyrifos	Total DDTs	Total Dieldrins	Total Endo.	Total HCHs	Mirex	Total PAHs	Total PCBs	Cluster Sum	#Classes Analyzed	Normalized Cluster	Overall Cluster
ABOB	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	2
AESP	0	0	0	0	1	0	0	0	0	1	0	2	11	6.1	1
CBBI	2	0	0	0	1	0	0	0	0	1	0	4	11	12.1	2
CBCR	0	0	0	0	1	0	0	0	0	1	0	2	11	6.1	1
CBJB	2	0	0	0	1	0	0	0	0	2	1	6	11	18.2	3
CBPP	2	2	0	0	3	0	0	0	0	1	1	9	11	27.3	4
CBSR		0	0	0	1	0	0	0	0	1	0	2	10	6.7	1
CCDC	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	2
CCNB	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	2
CKBP	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	2
CLCL	0	0	3	0	0	0	0	0	0	1	1	5	11	15.2	3
CLLC	0	0	0	0	0	0	0	0	0	1	1	2	11	6.1	1
CLSJ	1	0	0	0	1	0	0	0	0	1	0	3	11	9.1	2
GBCR	0	0	0	0	1	0	0	3	0	1	1	6	11	18.2	3
GBHR	1	0	0	0	1	0	0	0	0	1	1	4	11	12.1	2
GBOB	0	3	0	3	1	0	0	0	0	3	1	11	11	33.3	4
GBTD	2	0	0	0	2	0	0	0	0	2	3	9	11	27.3	4
GBYC	2	3	0	0	2	0	0	0	0	2	3	12	11	36.4	4
LMAC	0	0	0	0	1	0	0	0	0	1	0	2	11	6.1	1
LMPI	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	2
LMSB	0	0	0	0	0	0	0	0	0	1	0	1	11	3.0	1
MBCB	0	0	0	0	1	0	0	0	0	1	0	2	11	6.1	1
MBDR	1	1	0	0	3	0	0	0	0	1	1	7	11	21.2	4
MBGP	0	0	0	0	1	0	0	2	0	1	0	4	11	12.1	2
MSBB	0	3	0	0	1	0	0	0	0	1	1	6	11	18.2	3
MSPB	2	0	0	2	1	0	0	0	0	1	1	7	11	21.2	4
MSPC	2	0	0	3	1	0	0	0	0	1	1	8	11	24.2	4
NBNB	2	1	0	0	1	0	0	0	0	1	0	5	11	15.2	3
PBIB		0	0	0	1	0	0	0	0	1	2	4	10	13.3	2
PBPH	0	1	0	0	1	0	0	0	0	1	1	4	11	12.1	2
PBSP	0	0	0	0	0	0	0	0	0	1	1	2	11	6.1	1
PCMP	2	0	0	0	1	0	0	0	0	1	1	5	11	15.2	3
RBHC	2	0	0	0	0	0	0	0	0	1	0	3	11	9.1	2
SAWB	3	1	0	0	1	0	0	0	0	1	1	7	11	21.2	4
SLBB	0	0	0	0	0	0	0	0	0	1	0	1	11	3.0	1
SRWP	0	0	0	0	0	0	0	0	0	1	0	1	11	3.0	1
TBCB	0	1	0	0	1	0	0	0	0	1	1	4	11	12.1	2
TBHB	2	1	0	0	1	0	0	0	0	1	1	6	11	18.2	3
TBKA	2	2	0	0	2	0	0	0	0	1	2	9	11	27.3	4
TBLB	0	0	0	0	0	0	0	0	0	1	0	1	11	3.0	1
TBNP	1	2	0	0	1	0	0	0	0	1	1	6	11	18.2	3
TBOT	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	2
TBPB		2	0	0	1	0	0	0	0	1	1	5	10	16.7	3
VBSP	0	0	0	0	1	0	0	0	0	1	1	3	11	9.1	2

Appendices

Table A6. Results of Spearman's Rank tests on site specific temporal trends for sites sampled in the Gulf of Mexico in 2017. All trends were decreasing except for Total Chlorobenzenes at site TBLB (*).

Chemical Group	Site	p value	rho value	Chemical Group	Site	p value	rho value
Total Butyltins	AESP	p = 0.020	rho = -0.61	Total Chlordanes	MBCB	p < 0.001	rho = -0.86
Total Butyltins	CBBI	p = 0.010	rho = -0.64	Total Chlordanes	MBDR	p = 0.036	rho = -0.70
Total Butyltins	CBCR	p = 0.025	rho = -0.62	Total Chlordanes	MBGP	p < 0.001	rho = -0.82
Total Butyltins	CBJB	p < 0.001	rho = -0.87	Total Chlordanes	MSBB	p = 0.008	rho = -0.65
Total Butyltins	CLLC	p = 0.030	rho = -0.58	Total Chlordanes	MSPB	p = 0.005	rho = -0.63
Total Butyltins	GBCR	p < 0.001	rho = -0.81	Total Chlordanes	NBNB	p = 0.010	rho = -0.60
Total Butyltins	GBHR	p = 0.024	rho = -0.62	Total Chlordanes	PBIB	p < 0.001	rho = -0.80
Total Butyltins	GBOB	p < 0.001	rho = -0.83	Total Chlordanes	PBPH	p = 0.004	rho = -0.69
Total Butyltins	GBTD	p = 0.002	rho = -0.76	Total Chlordanes	CLSJ	p = 0.003	rho = -0.67
Total Butyltins	GBYC	p = 0.006	rho = -0.72	Total Chlordanes	GBCR	p = 0.002	rho = -0.70
Total Butyltins	LMPI	p = 0.002	rho = -0.84	Total Chlordanes	LMSB	p < 0.001	rho = -0.77
Total Butyltins	LMSB	p = 0.002	rho = -0.76	Total Chlordanes	MBCB	p < 0.001	rho = -0.86
Total Butyltins	MBDR	p = 0.010	rho = -0.80	Total Chlordanes	MBDR	p = 0.036	rho = -0.70
Total Butyltins	MBGP	p < 0.001	rho = -0.82	Total Chlordanes	MBGP	p < 0.001	rho = -0.82
Total Butyltins	MSBB	p < 0.001	rho = -0.96	Total Chlordanes	MSBB	p = 0.008	rho = -0.65
Total Butyltins	MSPB	p < 0.001	rho = -0.81	Total Chlordanes	MSPB	p = 0.005	rho = -0.63
Total Butyltins	MSPC	p = 0.049	rho = -0.55	Total Chlordanes	NBNB	p = 0.010	rho = -0.60
Total Butyltins	NBNB	p < 0.001	rho = -0.97	Total Chlordanes	PBIB	p < 0.001	rho = -0.80
Total Butyltins	PBIB	p < 0.001	rho = -0.88	Total Chlordanes	PBPH	p = 0.004	rho = -0.69
Total Butyltins	PBPH	p < 0.001	rho = -0.84	Total Chlorobenzenes	CBPP	p = 0.043	rho = -0.48
Total Butyltins	PCMP	p = 0.002	rho = -0.87	Total Chlorobenzenes	CLLC	p = 0.027	rho = -0.55
Total Butyltins	RBHC	p = 0.028	rho = -0.63	Total Chlorobenzenes	TBLB	p < 0.037	rho = 0.49*
Total Butyltins	SAWB	p = 0.011	rho = -0.68	Chlorpyrifos	CKBP	p = 0.034	rho = -0.67
Total Butyltins	SLBB	p = 0.003	rho = -0.73	Chlorpyrifos	GBYC	p = 0.049	rho = -0.71
Total Butyltins	TBCB	p = 0.031	rho = -0.60	Chlorpyrifos	LMSB	p = 0.009	rho = -0.77
Total Butyltins	TBHB	p = 0.001	rho = -0.87	Chlorpyrifos	MBDR	p = 0.048	rho = -0.71
Total Butyltins	TBKA	p = 0.001	rho = -0.77	Chlorpyrifos	TBCB	p = 0.025	rho = -0.73
Total Butyltins	TBNP	p = 0.009	rho = -0.67	Chlorpyrifos	TBHB	p = 0.001	rho = -0.89
Total Butyltins	TBOT	p = 0.023	rho = -0.62	Chlorpyrifos	TBKA	p = 0.011	rho = -0.76
Total Butyltins	TBPB	p = 0.019	rho = -0.64	Chlorpyrifos	TBNP	p < 0.001	rho = -0.92
Total Butyltins	VBSP	p < 0.001	rho = -0.88	Chlorpyrifos	TBOT	p = 0.029	rho = -0.68
Total Chlordanes	ABOB	p < 0.001	rho = -0.94	Chlorpyrifos	TBPB	p = 0.029	rho = -0.68
Total Chlordanes	AESP	p = 0.046	rho = -0.52	Total DDTs	ABOB	p < 0.001	rho = -0.98
Total Chlordanes	CBBI	p < 0.001	rho = -0.85	Total DDTs	AESP	p < 0.007	rho = -0.66
Total Chlordanes	CBCR	p = 0.001	rho = -0.74	Total DDTs	CBBI	p < 0.001	rho = -0.82
Total Chlordanes	CBJB	p < 0.001	rho = -0.78	Total DDTs	CBCR	p < 0.001	rho = -0.77
Total Chlordanes	CBPP	p = 0.002	rho = -0.68	Total DDTs	CBJB	p < 0.002	rho = -0.73
Total Chlordanes	CBSR	p = 0.008	rho = -0.60	Total DDTs	CBPP	p < 0.002	rho = -0.69
Total Chlordanes	CCNB	p < 0.001	rho = -0.80	Total DDTs	CBSR	p < 0.003	rho = -0.66
Total Chlordanes	CKBP	p < 0.001	rho = -0.92	Total DDTs	CCNB	p < 0.001	rho = -0.87
Total Chlordanes	CLCL	p < 0.001	rho = -0.90	Total DDTs	CKBP	p < 0.001	rho = -0.89
Total Chlordanes	CLLC	p < 0.001	rho = -0.77	Total DDTs	CLCL	p < 0.001	rho = -0.86
Total Chlordanes	CLSJ	p = 0.003	rho = -0.67	Total DDTs	CLLC	p < 0.001	rho = -0.76
Total Chlordanes	GBCR	p = 0.002	rho = -0.70	Total DDTs	CLSJ	p < 0.001	rho = -0.80
Total Chlordanes	LMSB	p < 0.001	rho = -0.77	Total DDTs	GBCR	p < 0.001	rho = -0.82

Appendices

Table A6 cont. Results of Spearman's Rank tests on site specific temporal trends for sites sampled in the Gulf of Mexico in 2017. All trends were decreasing except for Total Chlorobenzenes at site TBLB (*).

Chemical Group	Site	p value	rho value	Chemical Group	Site	p value	rho value
Total DDTs	GBHR	p = 0.035	rho = -0.51	Total Dieldrins	PBIB	p < 0.001	rho = -0.85
Total DDTs	GBOB	p = 0.003	rho = -0.69	Total Dieldrins	PCMP	p = 0.004	rho = -0.76
Total DDTs	LMAC	p = 0.006	rho = -0.76	Total Dieldrins	SAWB	p < 0.001	rho = -0.86
Total DDTs	LMPI	p = 0.021	rho = -0.68	Total Dieldrins	SLBB	p < 0.001	rho = -0.81
Total DDTs	LMSB	p < 0.001	rho = -0.82	Total Dieldrins	TBCB	p < 0.001	rho = -0.81
Total DDTs	MBCB	p = 0.011	rho = -0.68	Total Dieldrins	TBKA	p = 0.005	rho = -0.68
Total DDTs	MBGP	p = 0.001	rho = -0.72	Total Dieldrins	TBLB	p < 0.001	rho = -0.77
Total DDTs	MSBB	p = 0.002	rho = -0.74	Total Dieldrins	TBNP	p = 0.030	rho = -0.56
Total DDTs	MSPB	p < 0.001	rho = -0.80	Total Dieldrins	TBOT	p = 0.001	rho = -0.76
Total DDTs	MSPC	p = 0.033	rho = -0.52	Total Dieldrins	TBPB	p = 0.018	rho = -0.55
Total DDTs	NBNB	p = 0.002	rho = -0.69	Total Dieldrins	VBSP	p < 0.001	rho = -0.84
Total DDTs	PBIB	p < 0.001	rho = -0.84	Total Endosulfans	CBBI	p = 0.003	rho = -0.86
Total DDTs	PBPH	p < 0.001	rho = -0.80	Total Endosulfans	CBCR	p = 0.034	rho = -0.85
Total DDTs	PBSP	p = 0.010	rho = -0.80	Total Endosulfans	CCNB	p = 0.012	rho = -0.87
Total DDTs	PCMP	p < 0.001	rho = -0.83	Total Endosulfans	MBCB	p = 0.015	rho = -0.85
Total DDTs	RBHC	p < 0.001	rho = -0.79	Total Endosulfans	MBGP	p = 0.003	rho = -0.93
Total DDTs	SAWB	p < 0.001	rho = -0.87	Total Endosulfans	SRWP	p = 0.041	rho = -0.89
Total DDTs	SLBB	p < 0.001	rho = -0.82	Total Endosulfans	TBLB	p = 0.050	rho = -0.71
Total DDTs	SRWP	p = 0.042	rho = -0.83	Total Endosulfans	TBOT	p = 0.007	rho = -0.85
Total DDTs	TBCB	p < 0.001	rho = -0.89	Total HCHs	ABOB	p < 0.001	rho = -0.71
Total DDTs	TBKA	p < 0.001	rho = -0.77	Total HCHs	CBCR	p < 0.043	rho = -0.51
Total DDTs	TBLB	p < 0.001	rho = -0.77	Total HCHs	CBPP	p < 0.001	rho = -0.72
Total DDTs	TBNP	p = 0.003	rho = -0.71	Total HCHs	CCNB	p < 0.004	rho = -0.66
Total DDTs	TBOT	p = 0.007	rho = -0.66	Total HCHs	CKBP	p < 0.007	rho = -0.60
Total DDTs	TBPB	p < 0.001	rho = -0.78	Total HCHs	CLCL	p < 0.001	rho = -0.71
Total DDTs	VBSP	p < 0.001	rho = -0.77	Total HCHs	CLSJ	p < 0.001	rho = -0.79
Total Dieldrins	ABOB	p < 0.001	rho = -0.89	Total HCHs	GBCR	p < 0.001	rho = -0.80
Total Dieldrins	AESP	p = 0.037	rho = -0.54	Total HCHs	GBHR	p < 0.001	rho = -0.89
Total Dieldrins	CBBI	p < 0.001	rho = -0.79	Total HCHs	GBOB	p < 0.010	rho = -0.62
Total Dieldrins	CBCR	p = 0.026	rho = -0.55	Total HCHs	GBTD	p < 0.001	rho = -0.79
Total Dieldrins	CBJB	p < 0.001	rho = -0.88	Total HCHs	GBYC	p < 0.001	rho = -0.87
Total Dieldrins	CBSR	p = 0.025	rho = -0.53	Total HCHs	LMSB	p = 0.019	rho = -0.55
Total Dieldrins	CCNB	p < 0.001	rho = -0.83	Total HCHs	MBGP	p = 0.005	rho = -0.65
Total Dieldrins	CKBP	p < 0.001	rho = -0.83	Total HCHs	MSBB	p = 0.006	rho = 0.67
Total Dieldrins	CLCL	p < 0.001	rho = -0.80	Total HCHs	MSPC	p < 0.001	rho = -0.75
Total Dieldrins	CLLC	p = 0.009	rho = -0.63	Total HCHs	PBIB	p < 0.001	rho = -0.78
Total Dieldrins	CLSJ	p = 0.002	rho = -0.68	Total HCHs	PBPH	p < 0.001	rho = -0.81
Total Dieldrins	GBCR	p = 0.001	rho = -0.71	Total HCHs	PBSP	p = 0.004	rho = -0.85
Total Dieldrins	GBHR	p = 0.025	rho = -0.54	Total HCHs	SAWB	p = 0.002	rho = -0.71
Total Dieldrins	GBYC	p = 0.004	rho = -0.66	Total HCHs	SLBB	p = 0.020	rho = -0.54
Total Dieldrins	LMPI	p = 0.021	rho = -0.68	Total HCHs	TBHB	p < 0.001	rho = -0.88
Total Dieldrins	LMSB	p < 0.001	rho = -0.78	Total HCHs	TBKA	p < 0.001	rho = -0.91
Total Dieldrins	MBCB	p < 0.001	rho = -0.80	Total HCHs	TBLB	p = 0.010	rho = -0.59
Total Dieldrins	MBGP	p < 0.001	rho = -0.80	Total HCHs	TBNP	p = 0.021	rho = -0.59
Total Dieldrins	MSPB	p < 0.001	rho = -0.75	Total HCHs	TBOT	p = 0.005	rho = -0.69

Appendices

Table A6 cont. Results of Spearman's Rank tests on site specific temporal trends for sites sampled in the Gulf of Mexico in 2017. All trends were decreasing except for Total Chlorobenzenes at site TBLB (*).

Chemical Group	Site	p value	rho value
Total HCHs	TBPB	p = 0.003	rho = -0.66
Total HCHs	VBSP	p = 0.002	rho = -0.69
Mirex	CBJB	p < 0.001	rho = -0.74
Mirex	CKBP	p < 0.007	rho = -0.62
Mirex	CLLC	p < 0.001	rho = -0.55
Mirex	CLSJ	p < 0.001	rho = -0.48
Mirex	GBYC	p < 0.002	rho = -0.51
Mirex	MBDR	p < 0.002	rho = -0.81
Mirex	MSBB	p < 0.003	rho = -0.62
Mirex	MSPB	p < 0.001	rho = -0.77
Mirex	NBNB	p < 0.001	rho = -0.48
Mirex	PBIB	p = 0.002	rho = -0.61
Mirex	PCMP	p = 0.001	rho = -0.72
Mirex	SRWP	p = 0.025	rho = -0.85
Mirex	TBCB	p = 0.004	rho = -0.92
Mirex	TBKA	p = 0.021	rho = -0.85
Mirex	TBLB	p < 0.001	rho = -0.54
Mirex	TBNP	p < 0.001	rho = -0.85
Mirex	TBPB	p < 0.001	rho = -0.85
Mirex	VBSP	p < 0.001	rho = -0.73
Total PAHs	CBPP	p < 0.001	rho = -0.80
Total PAHs	CLSJ	p = 0.030	rho = -0.58
Total PAHs	GBCR	p = 0.022	rho = -0.63
Total PAHs	LMAC	p = 0.032	rho = -0.65
Total PAHs	MBCB	p = 0.017	rho = -0.67
Total PAHs	MBDR	p = 0.030	rho = -0.72
Total PAHs	MSBB	p = 0.003	rho = -0.80
Total PAHs	MSPB	p = 0.029	rho = -0.58
Total PAHs	PBPH	p = 0.022	rho = -0.63
Total PAHs	PCMP	p = 0.043	rho = -0.65
Total PAHs	SAWB	p < 0.001	rho = -0.86
Total PAHs	SRWP	p = 0.037	rho = -0.90
Total PAHs	TBHB	p = 0.039	rho = -0.63
Total PAHs	TBKA	p = 0.014	rho = -0.64
Total PAHs	TBLB	p = 0.043	rho = -0.55
Total PAHs	TBNP	p = 0.006	rho = -0.69
Total PCBs	ABOB	p = 0.001	rho = -0.75
Total PCBs	AESP	p < 0.001	rho = -0.85
Total PCBs	CBBI	p = 0.002	rho = -0.72
Total PCBs	CBCR	p = 0.019	rho = -0.62
Total PCBs	CBJB	p = 0.006	rho = -0.68
Total PCBs	CBPP	p = 0.009	rho = -0.63
Total PCBs	CBSR	p = 0.039	rho = -0.52
Total PCBs	CCNB	p < 0.001	rho = -0.89
Total PCBs	CKBP	p = 0.005	rho = -0.64

Chemical Group	Site	p value	rho value
Total PCBs	CLCL	p = 0.006	rho = -0.65
Total PCBs	CLLC	p = 0.001	rho = -0.73
Total PCBs	GBCR	p < 0.001	rho = -0.84
Total PCBs	GBOB	p = 0.031	rho = -0.54
Total PCBs	MBCB	p = 0.018	rho = -0.64
Total PCBs	MBDR	p = 0.050	rho = -0.67
Total PCBs	MBGP	p = 0.003	rho = -0.70
Total PCBs	MSBB	p < 0.001	rho = -0.84
Total PCBs	MSPB	p < 0.001	rho = -0.83
Total PCBs	NBNB	p = 0.024	rho = -0.58
Total PCBs	PBIB	p = 0.002	rho = -0.77
Total PCBs	PBPH	p < 0.001	rho = -0.81
Total PCBs	PBSP	p = 0.007	rho = -0.82
Total PCBs	PCMP	p = 0.017	rho = -0.67
Total PCBs	RBHC	p = 0.018	rho = -0.62
Total PCBs	SAWB	p < 0.001	rho = -0.85
Total PCBs	SLBB	p < 0.029	rho = -0.54
Total PCBs	SRWP	p = 0.005	rho = -0.94
Total PCBs	TBCB	p = 0.002	rho = -0.75
Total PCBs	TBKA	p = 0.006	rho = -0.67
Total PCBs	TBLB	p < 0.001	rho = -0.79
Total PCBs	TBNP	p < 0.001	rho = -0.89
Total PCBs	TBOT	p = 0.018	rho = -0.62
Total PCBs	TBPB	p = 0.029	rho = -0.58
Total PCBs	VBSP	p = 0.004	rho = -0.68



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