Maryland Department of the Environment

Young of the Year Fish Monitoring in Maryland Freshwaters and Estuaries: A Means of Observing Change in Hg Availability

Data Report
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Executive Summary

To track the effectiveness of the combined Maryland Healthy Air Act and federal rule Mercury and Toxic Standards (MATS), a standardized young-of-the-year (YOY) predatory fish sampling and analysis project was initiated to assess year-to-year and long-term trends in methylmercury (MeHg) bioaccumulation in nine freshwater sites using largemouth bass (*Micropterous salmoides*) and four estuarine sites using white perch (*Morone americana*). This project primarily utilizes existing Maryland Department of Natural Resources (MDNR) monitoring programs. The sites were chosen for both spatial coverage and site diversity.

This data report summarizes the results of 2017 collections and compares the 2017 data with that obtained from 2008-2016. This data provides an opportunity to look for early trends in response to the implementation of Hg emission reduction plans.

Not all reservoir sites in 2017 provided the target number of fish (25). For largemouth bass only16 were collected from Prettyboy and 5 from Lake Lariat. At this young age, fish size or mass does not appear to correlate with fish Hg concentration making normalization to a standard fish size impossible. This may stem from growth dynamics of young fish. Mercury is not accumulated at the same rate in rapidly growing fish as adult fish. At any one site, the Hg concentrations in the fish collected in 2017 varied by approximately 20%. The current sample size of 25 fish will need to be maintained for future statistical analysis to be effective. Reservoir operational information will also be needed along with lake water residence time, area precipitation and lake temperature for refining future models and determining what fish should be used in determining long term trends.

Fish data has now been collected for 10 years, but wet deposition of Hg only recently appears to be changing. Mercury concentrations in rain is now showing a significant downward trend but loading, which is likely more important to accumulation in the food web, is lagging behind as it is also dependent on precipitation amount. Fluctuations in volume increase the inter-annual variability in the data.

At freshwater sites, Hg concentrations in fish have largely not changed with decreases sustained at Cash Lake. In some cases fish size has increased as has concentration such as in Lake Lariat and Loch Raven. The Hg burden (average mass of mercury in each fish) has not changed. Piney Reservoir is an interesting case, were Hg concentrations appear to have undergone some sort of rapid change or reset. The average size of the perch collected from the bay sites have increased at 3 of the 4 sites but the Hg concentration has remained the same or has decreased. As a result the fish Hg burden is unchanged. Detailed modeling with more localized wet and dry Hg deposition estimates along with reservoir operations is needed for an accurate evaluation of the drivers behind the year to year variability in fish concentration we have observed. This is not part of the current study scope. In a limited study, when available, a small set of fish were digested and analyzed for mercury and selenium. Differences in Hg concentration in fish among reservoirs appears negatively related to the presence of selenium, with selenium potentially retarding Hg accumulation.

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1. Introduction

A standardized young-of-the-year (YOY) predatory fish sampling and analysis project was initiated to assess year-to-year and long-term trends in methylmercury (MeHg) bioaccumulation in selected Maryland fresh and salt water fish species. This project primarily utilizes existing Maryland Department of Natural Resources (MDNR) monitoring programs and will implement the standard methods for mercury (Hg) analysis used by Maryland research laboratories. This technical document provides a rationale, approach, and results from 2008 through 2017.

2. Rationale

Federal and state regulations to control mercury releases to the environment have recently been promulgated. These new regulations are in response to elevated levels of MeHg in fish and other animals, and the resulting increased risk to people and wildlife (Scheuhammer et al. 2007; Mergler et al. 2007). At the Federal level, the Clean Air Interstate Rule (CAIR) and Clean Air Mercury Rule (CAMR) of 2005 target reduction in Hg emissions from coal-fired power plants; however, this was never implemented and instead the Mercury and Air Toxics Standards (MATS) was passed late in 2011 and the final rule for existing plants implemented as of February 16, 2012 and effective as of April 16, 2012. Complete implementation will result in a 95% reduction in emissions but utilities had up to 4 years to comply, thus until April 16, 2016. At the State level, the Maryland Healthy Air Act required reductions in Hg emissions on a faster time schedule with 90% reduction by 2013. As a result, emissions reductions in the US will occur over a protracted period. Emissions reductions resulting from reductions in sulfur emissions likely have already had some impact.

While the recent regulatory efforts are an important step in working to decrease the amount of Hg released to the environment, evaluation of the effectiveness of these efforts will be the ultimate measure of success. Supporting documents for CAMR (U.S. EPA CAMR NODA, Feb., 2005) demonstrate that current models provide widely divergent assessments of both (1) the effectiveness of U.S. emissions controls on the U.S. deposition rates, and (2) the effectiveness of decreases in Hg deposition on MeHg concentrations in fish. The 1997 *Report on Mercury to Congress* listed long-term monitoring of Hg deposition and effects as a very high priority.

The EPA Office of Air and the Society for Environmental Toxicology and Chemistry (SETAC) convened a group of research scientists in fall 2003 to develop a coherent monitoring and assessment framework for Hg. In addition to long-term deposition monitoring, the EPA strategy called for ecosystem-level monitoring at a few key sites across the U.S. in order to assess the timing and magnitude of ecosystem responses to changes in the rates of Hg deposition (Mason et al. 2005; Harris et al. 2007). While the existing Mercury Deposition Network (MDN) is providing data to assess changes in wet deposition, it will not provide the information needed to assess the impact of these changes on biological receptors. The complexity of Hg cycling in ecosystems means that linearity of response, either in space or time cannot be assumed (Orihel et al. 2006; Paterson et al 2006; Munthe et al. 2007).

Because reductions in Hg emissions started to take place throughout the region, 2008 was a critical year for establishing a pre-compliance ecosystem baseline. Appropriate monitoring tools and indicators for ecosystem-level Hg monitoring were laid out briefly in Mason et al. (2005) and in more detail in the Harris et al. (2007). The SETAC workgroup identified yearling fish, collected from fixed locations, as the single best indicator of the aquatic ecosystems response to short-term changes (years vs. decades) in Hg loads (Harris et al. 2007). A workshop convened by the Chesapeake Bay Program's Scientific and Technical Advisory Committee (http://www.chesapeake.org/stac/MercuryWorkshop.html) also recommended increased mercury monitoring, including ecosystem-level monitoring similar to the existing SERC program and complementing the existing fisheries monitoring studies to allow assessment of annual change in MeHg levels in fish. Young-of-the-year (YOY) fish, collected in the fall, reflect approximately 1/2 year of MeHg exposure, while older fish integrate multiple years of exposure, making it more difficult to assess the impact of other variables on fluctuations in MeHg fish tissue levels. Measuring MeHg in YOY piscivorous fish species captures the accumulation of MeHg through long food webs over a relatively short time frame, providing a more rapid indicator of change than could be observed using older fish that are routinely surveyed to assess risk to human consumers.

3. Study Design

This study established a statewide YOY fish monitoring project designed to detect changes in MeHg bioaccumulation in response to anticipated long-term emission reductions over a time scale of about a decade. At the onset of the study in 2008, a rough estimate of the range of potential MeHg reductions in Maryland fish by 2015 was predicted to be between 5% and 45% (see Section 4). Clearly the delays in implementing reductions have changed this outlook. The proposed YOY monitoring study is still believed to be best suited to detect this level of change. The relative homogeneity of YOY fish populations collected in fixed locations will help to minimize sample variability, while analytical methods that include high levels of quality assurance (QA) will ensure low analytical variability.

Fish Species: In Chesapeake Bay, YOY white perch (*Morone americana*) serve as the indicator species. White perch are a good indicator of MeHg accumulation because they are piscivorous and resident in the system. Also, for more than 20 years, Maryland DNR has been running a long-term collection program for YOY white perch and striped bass (*Morone saxatilis*) at fixed locations throughout the Chesapeake Bay. This on-going sampling program and long-term database provides critical information on sampling densities and interannual population dynamics that allowed for site selection for the MeHg YOY study and could aid in data interpretation.

In Maryland reservoirs, YOY largemouth bass (*Micropterous salmoides*) serve as the indicator species. This species was selected as it is ubiquitous in Maryland's fresh waters and information on MeHg bioaccumulation in adults is already available through prior sampling efforts by UMD/SERC and MDNR/MDE.

Site Choice: Four locations in Chesapeake Bay were selected to collect YOY white perch and nine Maryland reservoirs were selected from which to collect YOY largemouth bass (Table 1, Figure 1). Site selection was focused on reservoirs with pending TMDLs and/or with substantial Hg TMDL reduction requirements, as well as areas where models suggest that Hg deposition is currently high, and thus likely to show the most declines through time. Deep Creek Lake was added in 2009 and Loch Raven in 2010. Watershed characteristics are summarized in Table 2.

Site	Latitude	Longitude	Map Number
Sharptown-nanticoke	38.53876	75.72741	1
Plum-Point Head of Bay	39.48696	76.11385	2
Mill Town Patuxent River	38.63302	76.69111	3
Eagle Harbor Patuxent River	38.57051	76.68219	4
Tuckahoe Lake	38.96854	75.94462	5
Piney Reservoir	39.70842	79.0018	6
Savage River Reservoir	39.54327	79.13751	7
Liberty Reservoir	39.44576	76.88376	8
Prettyboy Reservoir	39.65239	76.74183	9
Cash Lake	39.03199	76.79729	10
Lake Lariat	38.37774	76.42265	11
Deep Creek	39.55807	79.35482	12
Loch Raven	39.46250	76.57814	13

Table 1. Site names and locations, shading indicates white perch sites.

Site	Lake Area	Watershed Area	Ratio
	Km^2	Km^2	
Sharptown-nanticoke	NA	NA	NA
Plum-Point Head of Bay	NA	NA	NA
Mill Town Patuxent River	NA	NA	NA
Eagle Harbor Patuxent River	NA	NA	NA
Tuckahoe Lake	0.35	223	640
Piney Reservoir	1.21	28	23
Savage River Reservoir	1.46	270	185
Liberty Reservoir	12.57	424	34
Prettyboy Reservoir	6.07	206	34
Cash Lake	0.21	6	29
Lake Lariat	0.39	7	18
Deep Creek	18	163	9
Loch Raven	9.71	789	81

Table 2. Watershed characteristics, shading indicates white perch sites. NA denoted not available.

Sampling

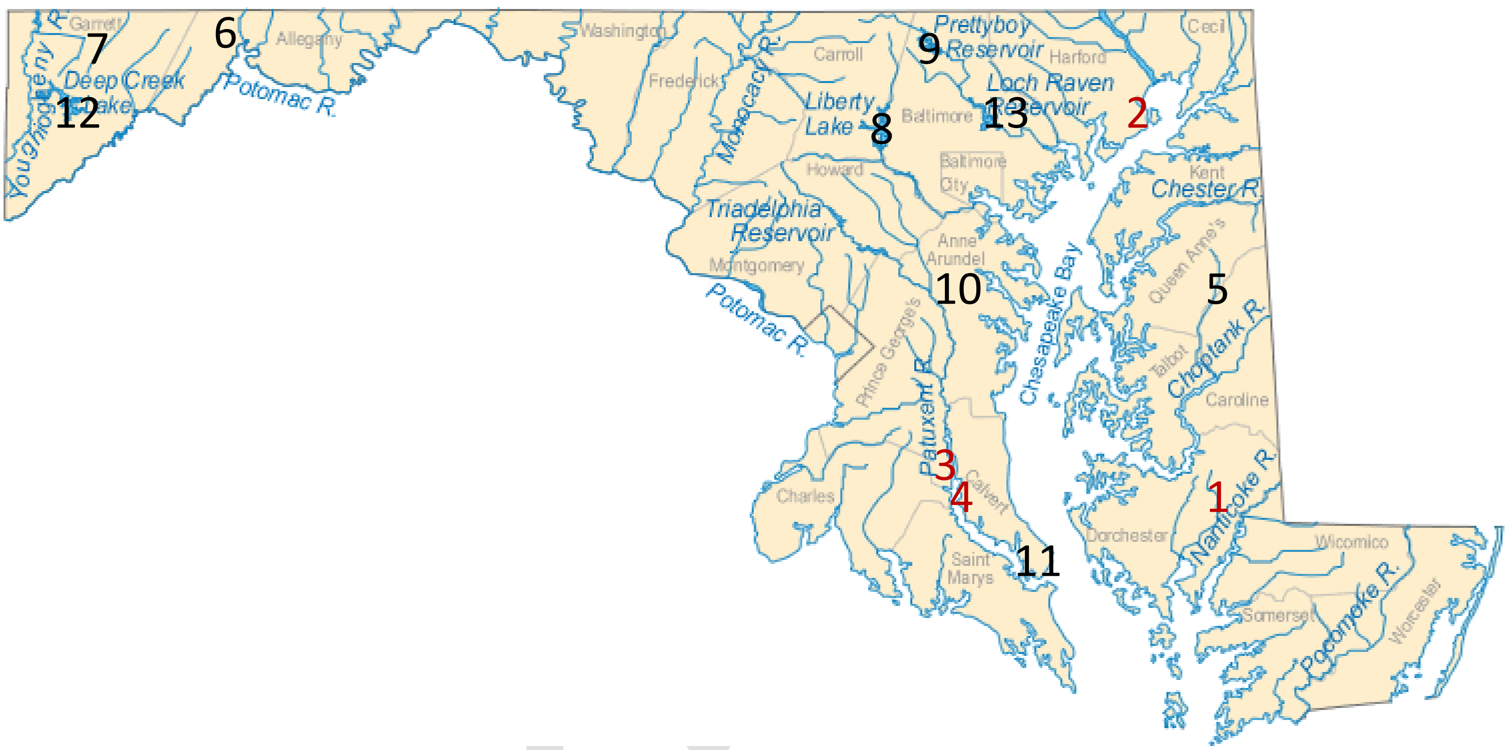


Figure 1. Locations of sampling sites described in Table 1.

4.0 Estimated Reductions in Fish Tissue MeHg Concentrations in Response to Emission Regulations at the onset of the Study

A rough estimate of the range of potential reductions in fish MeHg levels in Maryland's freshwater lakes/reservoirs is 5-30% and 20-45% in tidal waters. The current total deposition of Hg to Maryland watersheds was estimated using a CALPUFF-based model (Sherwell et al. 2006). The model also provides an estimate of the Hg deposition derived from Electric Generating Units (EGUs). The CALPUFF model suggests that about 50% of the overall Hg emissions in Maryland, and about 75% in the Chesapeake Bay watershed come from EGUs. For the purpose of estimating future emission levels, it was assumed that the Hg emissions reductions from EGUs would occur at the rate described by federal and state regulations. It was further assumed that deposition derived from global background would stay constant over the time period, at roughly 6 $ug/m^2/year$.

Based on the METAALICUS study (Harris et al. 2007), it can be hypothesized that MeHg levels in Maryland fish would respond rapidly to changes in the direct Hg deposition to surface waters. However, the Hg concentrations present in surface runoff would respond very slowly to such changes. Thus, it was assumed that by 2015, Hg concentrations in runoff would not change at all. What remains uncertain is whether the Hg in runoff is as bioavailable as the Hg delivered via direct deposition; most of the Hg delivered in surface runoff is in particulate form, which is generally less available to Hg methylating bacteria than the aqueous Hg phase.

The amount and rate of the estimated reductions are very uncertain including uncertainties in the Hg budget for the Chesapeake Bay, modeled Hg deposition rates for Maryland, Hg yields from Maryland watersheds, and the availability of Hg from various sources for methylation. Thus, while the minimum estimates for reduction in MeHg concentrations in fish are reasonably well constrained, the maximum estimates are no more than an informed guess.

Chesapeake Bay. Roughly 50% of the Hg load to Chesapeake Bay is delivered through direct deposition to the surface of the Bay (Mason et al. 1999). Using the CALPUFF model outputs and applying reductions required by existing regulations, the Hg load to the Chesapeake watershed was expected to decline by about 30% by 2010 and about 40% by 2015. The Garrison model assumes that about 50% of Hg emissions in the Bay region are from EGUs; these values may be somewhat different for the Bay itself. If Hg delivered to the Bay in surface runoff is equally available for methylation and bioaccumulation as Hg deposited directly to Chesapeake Bay surface waters, then MeHg in YOY fish should have declined by about 15% by 2010 and 20% by 2015. However, if Hg delivered to the Bay in surface runoff is less available for methylation and bioaccumulation, reductions in MeHg fish concentrations would be greater. If the availability of Hg in runoff is 20% of that in direct deposition, MeHg levels in fish could decline by as much as 45% by 2015.

Summary of estimated reductions in Chesapeake Bay MeHg concentrations in fish by 2015:

Minimum reduction by 2015: 20%
Maximum reduction by 2015: 45%

Lakes/Reservoirs. Here we use Prettyboy Reservoir as a model freshwater system. Prettyboy has a surface area of 6.1 square kilometers (3% of the watershed area) and a drainage area of 206.5 square kilometers. According to the Garrison model, Prettyboy watershed has one of the highest rates of Hg deposition in Maryland (total Hg deposition of 33 ug/m^2 per year). Assuming that 50% of regional Hg emissions are from EGUs (the average for the entire Chesapeake Bay watershed), and that by 2015 most of the regional emissions sources that contribute to deposition to the Prettyboy Reservoir watershed will be reduced by 90%, total Hg load to the watershed is expected to decline by a little less than 40%. Given the high Hg load to this watershed, and its proximity to EGU sources, it might be reasonable to assume that more than 50% of regional emissions come from EGUs.

As a mercury budget has not been developed for this reservoir (or any other MD reservoir), the key parameters used to estimate future MeHg bioaccumulation in fish tissue have been extracted from the literature (Harris et al. 2007, Munthe et al. 2007). Assuming that all Hg loads to the reservoir contribute equally to MeHg bioaccumulation in fish and that surface runoff constitutes about 15% of the Hg load to the lake, at a minimum, the Prettyboy Reservoir would experience a ~5% decline in MeHg in YOY fish by 2015.

However, if more than 50% of regional emissions sources that contribute to Prettyboy watershed deposition are EGUs, or if Hg delivered by surface runoff is not as available as Hg in direct deposition, the reduction in fish tissue MeHg levels could be higher. If EGUs contribute 75% of regional emissions, and watershed Hg is only 20% as bioavailable as direct deposition, actual reductions in fish tissue MeHg could be as much as 30% by 2015.

Summary of estimated reductions in Prettyboy MeHg concentrations in fish by 2015:

Minimum reduction by 2015: 5%
Maximum reduction by 2015: 30%

2016 update on 2008 Initial Model

Since emission reduction goals in Maryland EGUs were not achieved until early 2012 and bordering states implementation into 2016, the decreases predicted with the 2008 model must be extended forward in time. The model assumes external fluxes remain constant or decrease, which has not likely been the case, but data to estimate the changes is lacking.

5.0 Analytical Methods

Quality assurance of fish MeHg data, especially analysis of certified reference materials (CRMs) and intercalibration among labs, are crucial for collecting high quality data. Since almost all of the Hg in muscle tissue in piscivorous fish is MeHg (Bloom 1992; Storelli et al. 2002, 2003; Baeyens et al. 2003; Agah et al. 2007), and since total Hg analysis is substantially less expensive and somewhat more accurate than MeHg analysis (Horvatt et al. 1993), fish collected for this project have been analyzed for total Hg.

CBL analyzes all of the Hg samples. Details of the procedures for analysis of total Hg are described in a number of publications (Gilmour et al. 1998; 2000; Heyes et al. 2004, 2006; Orihel et al. 2007, 2008; Harris et al. 2007). The methods are derived from EPA Method 1631 for total Hg and include the following procedures:

- Fish and sediment samples are digested prior to analysis via open pan or microwave digestion with a 7:4 HNO_3:H_2SO_4 acid mix.
- Analyses are accomplished by Tekran 2600. Total Hg is reduced to elemental Hg with $SnCl_2$, stripped into the gas phase onto gold-coated bead columns and heated into the Tekran to drive the Hg off in single pulse.

The labs' analytical performance has been repeatedly and rigorously tested through formal and informal inter-laboratory calibrations (SERC: Florida DEP; METAALICUS project isotope intercalibrations; CBL: Florida DEP, METAALICUS, CALFED). Detection limits for most matrices are <0.05 ng/L or ng/g. Routine QA/QC includes 10% blanks and duplicates/replicates, and analysis of CRMs in every run.

For mercury and selenium analysis comparison, a separate digest was required as sulfuric acid is not recommended for use on ICP-MS instruments. Whole fish where freeze dried and a subsample of fish microwave digested (Milestone Ethos) using nitric acid. The digest was diluted and analyzed for Hg as above (Tekran 2600) and Se by ICP-MS (Agilent 7500).

6.0 Results

6.1 Mercury in wet deposition

Mercury deposition is being measured at three locations in Maryland as part of the National Atmospheric Deposition Network (NADP). The sites are Beltsville (MD99), Piney Reservoir (MD08) and SERC (MD00). These sites by no means cover the range in deposition likely to exist in Maryland but are representative of Urban, Chesapeake Bay shore line and western Maryland. The data collected from these sites is available at (http://nadp.sws.uiuc.edu/). For this study we are interested in the trends in regional deposition as this is a key driver in fish Hg concentrations. MD99 and MD08 have been in service since 2004 and MD00 since 2007. The average annual Hg concentration in rain has fluctuated by nearly a factor of 2 among years and sites. The concentration of Hg at the Beltsville site decreased from between 8 and 10 ng L^{-1} to a concentration closer to 7 ng L^{-1} after 2006, becoming similar to the other two sites (Figure 2). In recent years 2013 to 2016, T-Hg concentration has decreased in wet deposition at Beltsville, but as the precipitation at this site has exceeded the other two Maryland sites, the result is greater deposition. From this data we can see that while Hg loading is slowly decreasing with time, the load to any particular ecosystem remains highly variable.

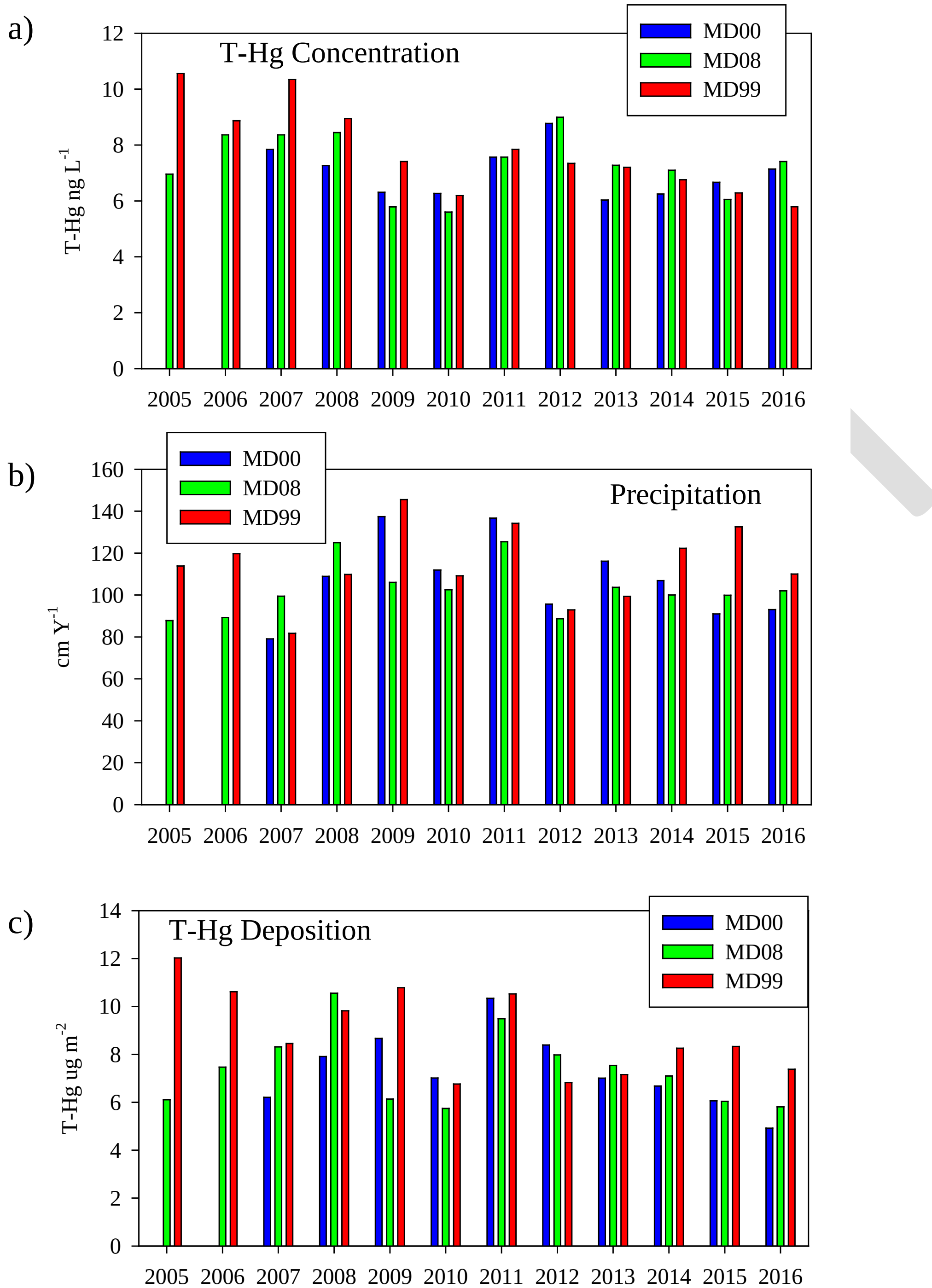


Figure 2. Average Hg concentration in rain, annual precipitation and mercury load in wet deposition at the three NADP sites in Maryland. Data was extracted from http://nadp.sws.uiuc.edu/ and calculated using NADP protocol.

The variability of precipitation both in time and space has a large impact on Hg loading and responsible for higher loadings at Beltsville in recent years (Figure 2). Wet Hg deposition has trended downward over the last 5 years at both SERC and Piney to ~ 6 ug m^{-2} yr^{-1} but similar Hg loads have been recorded at these sites in the past. We cannot say that Hg load has in fact decreased over the past 6 years across the state, only it appears to be trending downward. Further clouding the issue is wet deposition only accounts for a portion of atmospheric Hg inputs. Dry deposition is believed to be of a similar magnitude as wet deposition (Graydon et al. 2008). Early measures from NADP suggest dry deposition ranging from 9 to 15 ug m^{-2} y r^{-2} which is on par with wet deposition ranging from 6 to 12 ug m^{-2} yr^{-1}but we cannot address this portion of the Hg load here. Given Hg load has not decreased everywhere; no change in fish concentrations is expected state wide. Below we will discuss what we have observed in the YOY fish study which provides wider spatial coverage.

6.2 Estuary (White Perch) Sites

The average, median and standard deviation of concentrations for fish collected in 2017 are presented in Table 3 along with the 9 year mean. The white perch data for the entire study are summarized in Figure 3. In 2017, the distribution of Hg concentrations among the four sites remained the same as in past years. Perch from the Plum Point station are consistently lower than the other three sites. Concentrations of Hg in fish from three sites are near the lowest levels observed during the study period, except for Plum Point. Trends at each of the sites are examined in more detail in the sections below, but T-Hg concentrations tend to be lower now than earlier in the study but fish size has also increased.

Site	Arithmetic Mean	Standard Deviation	Sample Size	9 Year Mean
	ng/g wet wt.			ng/g wet
Sharptown-nanticoke	6.85	0.83	25	9.96
Plum-Point Head of Bay	5.47	1.04	25	4.00
Mill Town Patuxent River	5.97	1.22	25	7.85
Eagle Harbor Patuxent River	9.94	1.36	25	12.88
Tuckahoe Lake	21.63	7.08	25	18.69
Piney Reservoir	46.65	13.57	25	46.07
Savage River Reservoir	80.57	21.17	25	143.48
Liberty Reservoir	26.30	11.78	25	26.18
Prettyboy	10.18	2.19	15	11.94
Cash Lake	85.06	16.78	25	85.30
Lake Lariat	63.28	8.71	5	71.70
Deep Creek	35.09	8.90	25	28.86
Loch Raven	16.16	9.29	25	19.30

Table 3. Concentrations of Hg in YOY fish collected from the 13 sites in 2017.

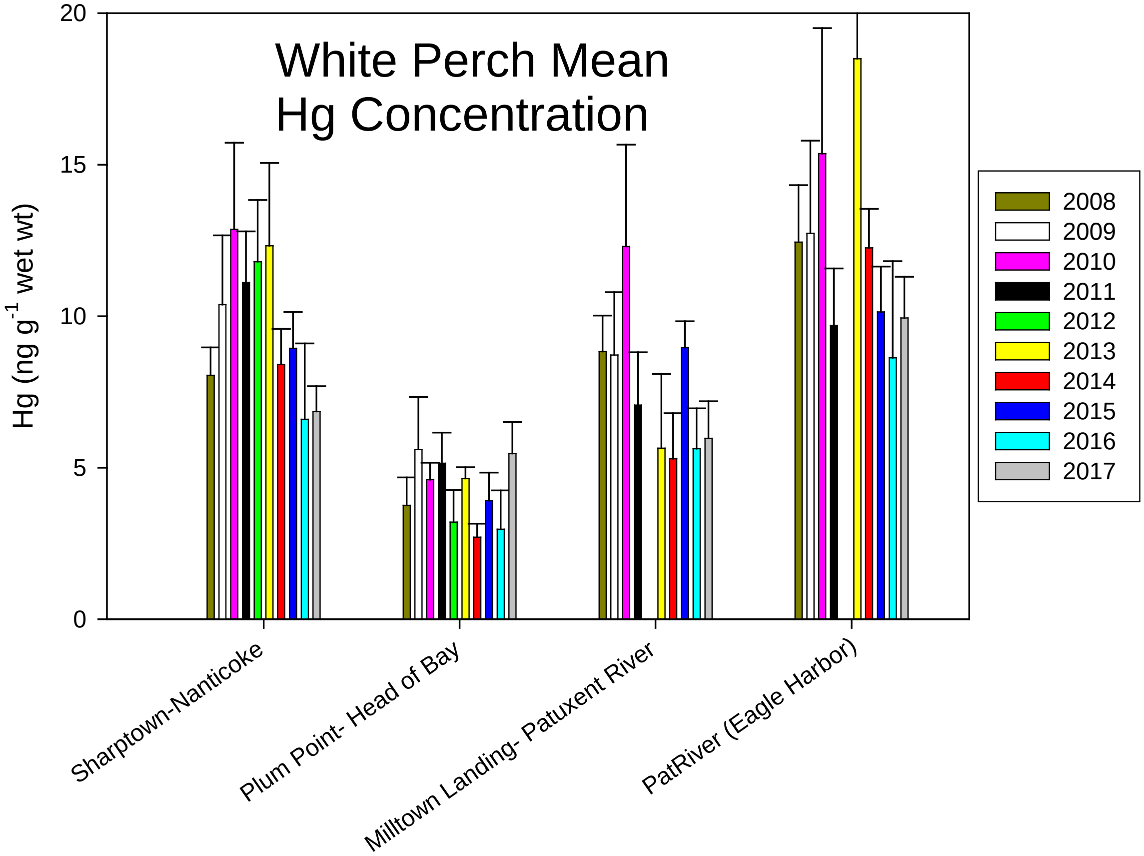


Figure 3. Mean concentrations of Total-Hg in White perch collected in 2008 to 2017. Error bars are the standard deviation around the mean.

6.2.1 Sharptown-nanticoke

White perch were successfully collected from the Sharptown site on the Patuxent River in ten successive years 2008 to 2017. No changes in site characteristics or factors that might influence fish health or Hg concentrations have been reported. A strong relationship between fish weight and length within and across years was maintained in 2017. Fish size in 2017 where the largest collected (2.50 g) relative to previous years (running mean being 1.68 g) (Figure 4). Fish Hg concentrations had trended upward from an average of 8.0 ng g^{-1} in 2008 to 12.3 ng g^{-1} in 2013 but the concentration dropped to 8.4 ng g^{-1} in 2014 and Hg concentrations have trended downward to 6.85 ng g^{-1} (Figure 4). There is no relationship between either fish size or fish weight and fish Hg concentration within a year or among years (Figure 5) and therefore no way to standardize the Hg concentration data based on size with these young fish. The fish Hg burden (the amount of Hg in each individual fish) increased with fish size through 2012 (Figure 6). After 2012, fish Hg burden also decreases at this site while fish size does not drastically change.

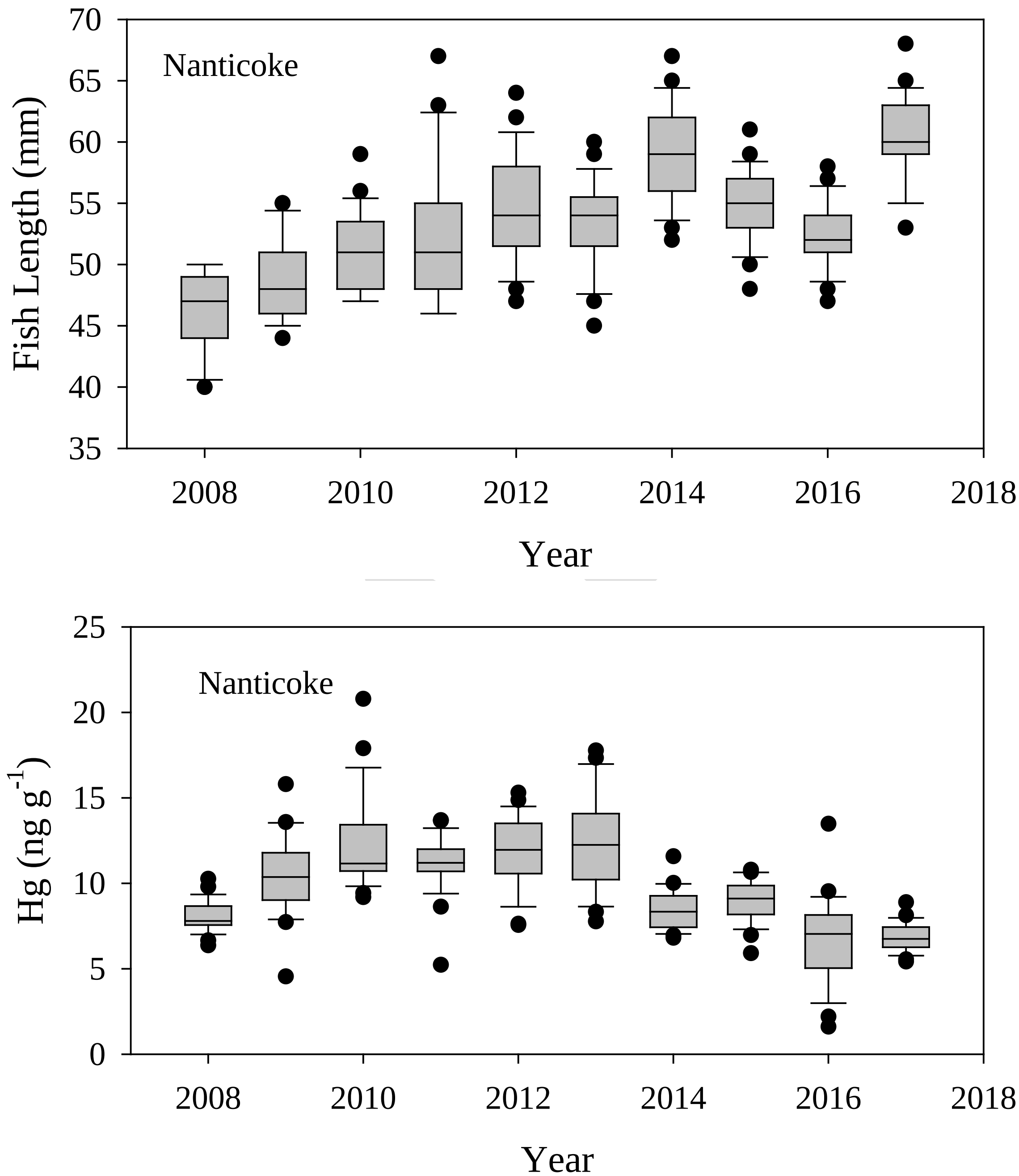


Figure 4. Box plots of Nanticoke fish size (upper) and fish Hg concentration (lower) 2008 through 2017. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers.

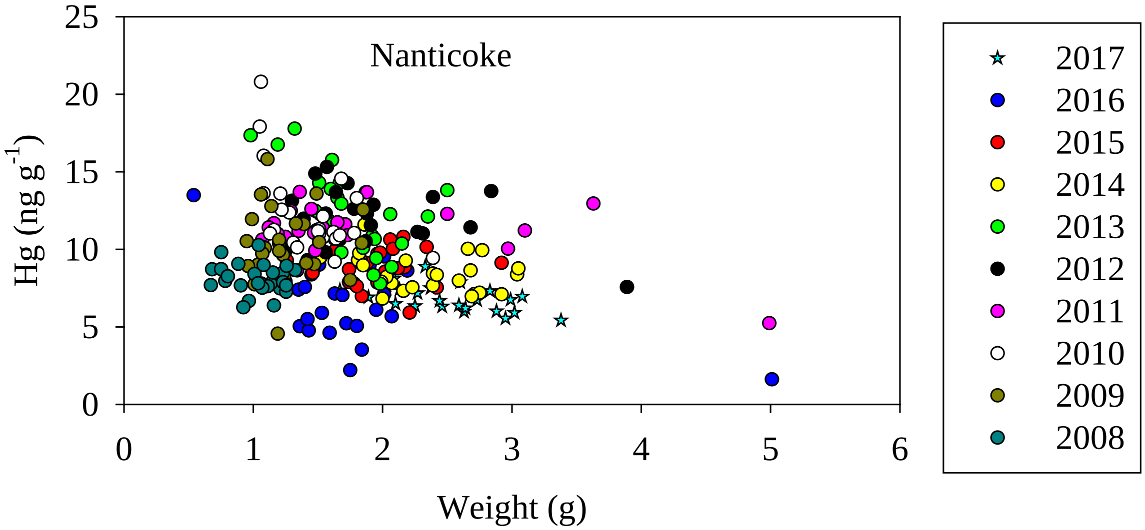


Figure 5. Fish Hg-weight relationship for individual fish collected at Sharptown-nanticoke.

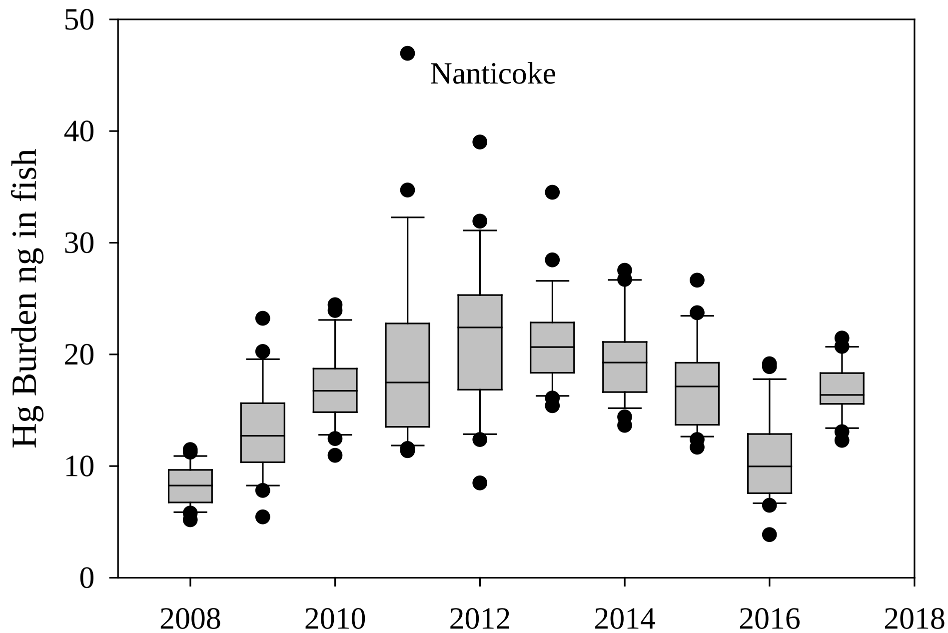
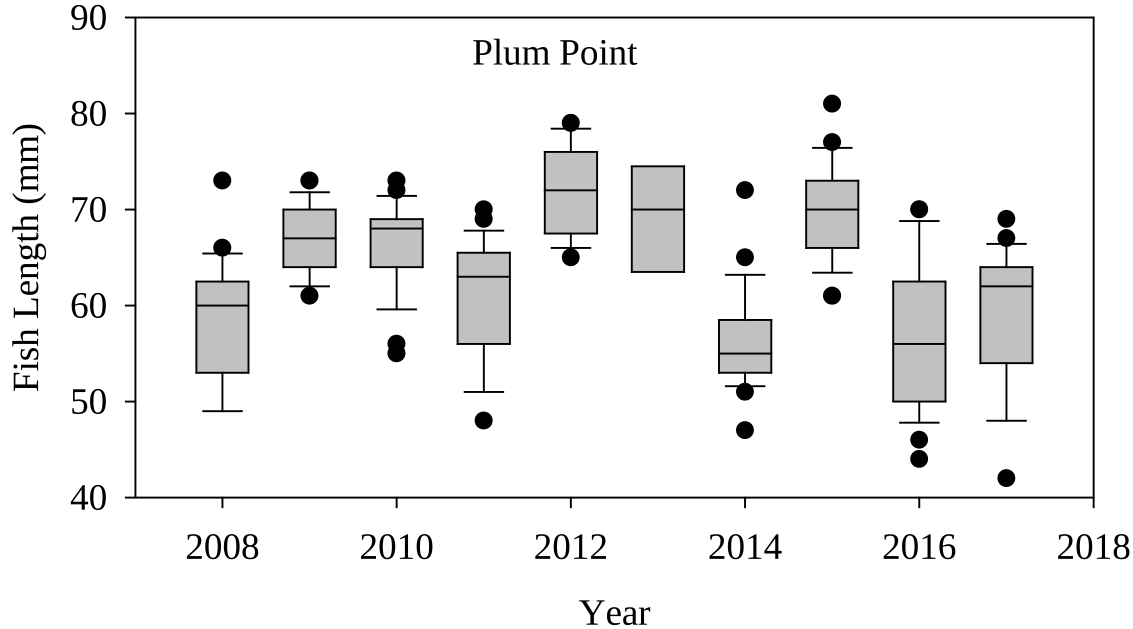


Figure 6. Box plots of Nanticoke fish Hg burden 2008 through 2017. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers.

6.2.2 Plum Point Head of Bay

For fish collected from the Plum Point site in 2017, the same strong relationship between fish length and weight is maintained. The average fish size collected across the study period was highly variable, and in 2017, the fish were smaller than most years averaging 58.8 mm and 2.28 grams compared to the study average of 63.6 mm and 3.06 g (Figure 7). Fish Hg concentrations were greater in 2017 than fish in any other year except for 2009 (Figures 7 and 8). No relationship between fish size and Hg concentration in any one year, nor among years, is evident. Mercury concentrations are consistently lower at the Plum Point site than any other site but no temporal trend is apparent (Figure 7 lower). This site is unique in the fact that fish are not increasing in size over the study period. This is also reflected in the fish Hg burden (Figure 9).



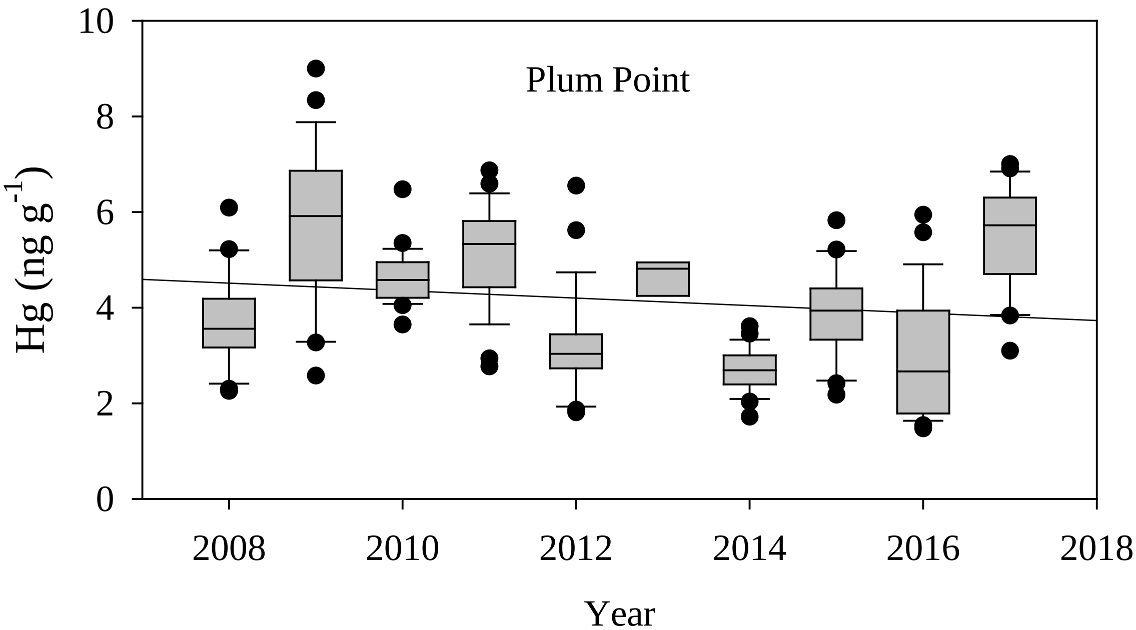


Figure 7. Box plots of Plum Point fish size (upper) and Fish Hg concentration (lower) 2008 through 2017. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers. The regression line is not significant.

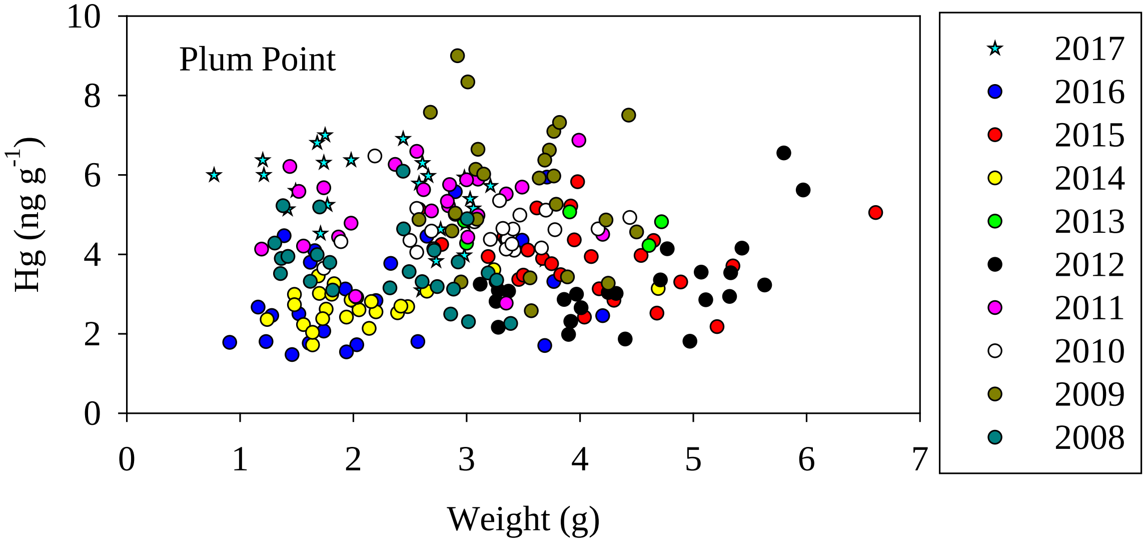


Figure 8. Fish Hg-weight relationship for individual fish collected at Plum Point.

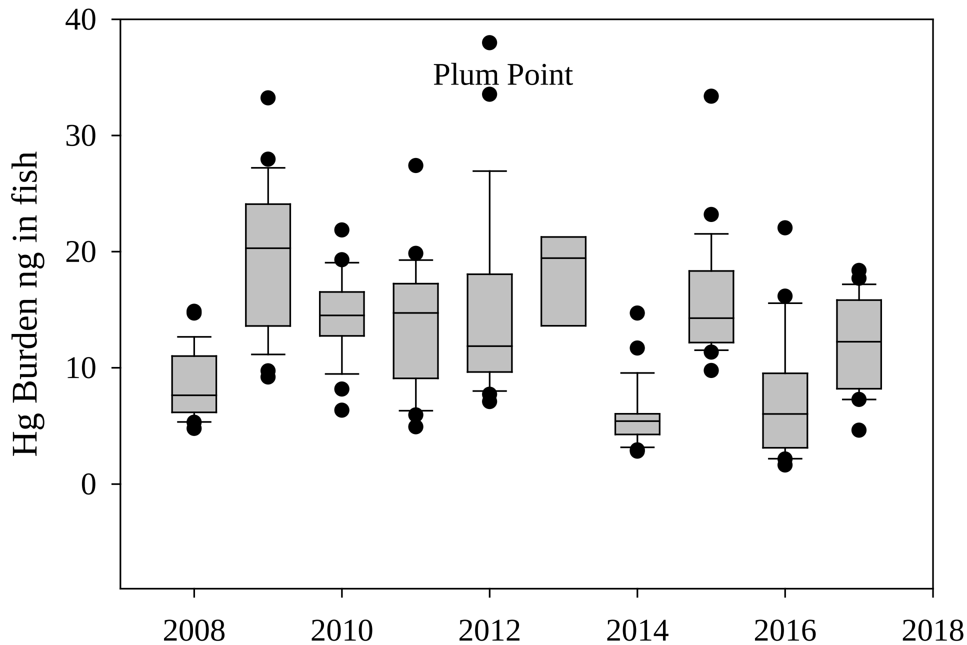


Figure 9. Box plots of Plum Point fish Hg burden 2008 through 2017. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers

6.2.3 Milltown Patuxent River

The perch collected in 2017 were larger than most years, and an increasing trend of fish size is apparent over the study period. The length to weight relationship remains consistent with past years. (Figure 10). Mercury concentrations in the fish continue trending downward (r^2 0.18 and significant $p< 0.01$) despite a large variation in fish size between years. There appears to be no relationship between Hg concentration and fish size within a year but across years, Hg concentration appears to be decreasing as fish size increases (Figure 11) and as Hg burden does not appear to change, increase in fish size is likely the major cause (Figure 12).

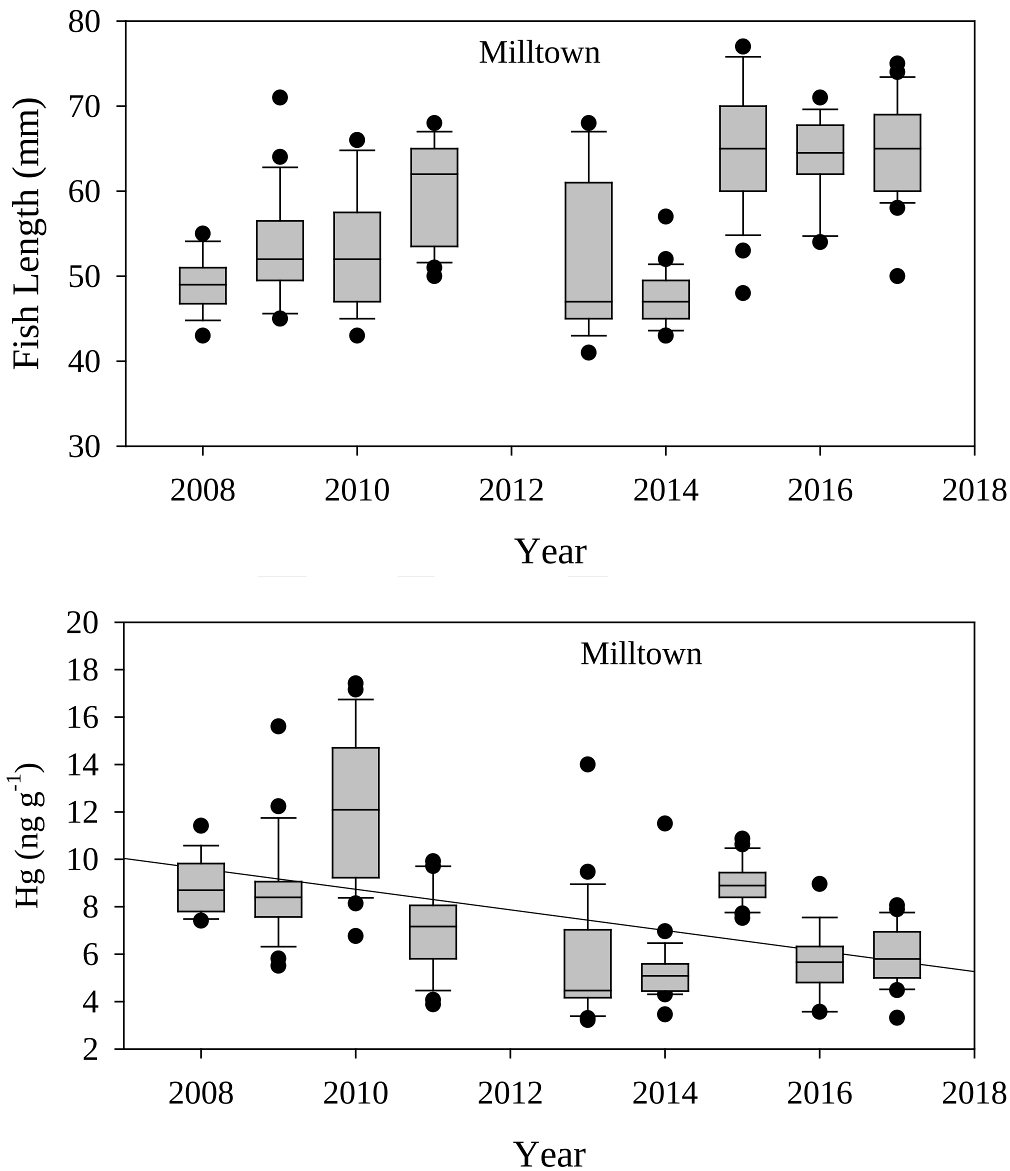


Figure 10. Box plots of Milltown fish size (upper) and Hg concentration (lower) 2008 through 2017. No samples were collected in 2012. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers. The regression line has an r^2 of 0.2 (p value 0.000).

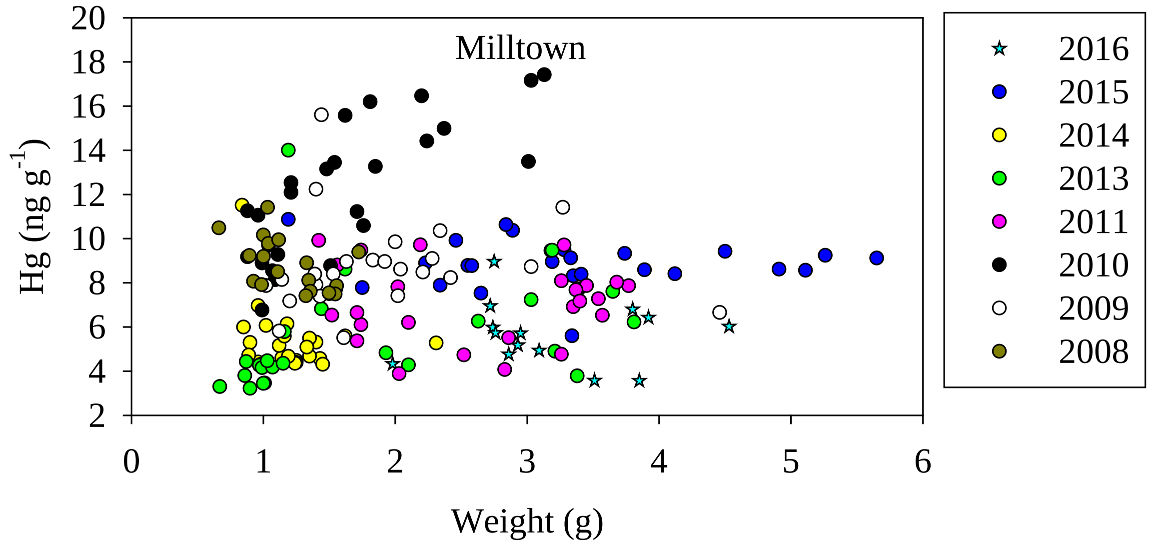


Figure 11. Fish Hg-weight relationship for individual fish collected at Milltown Patuxent River.

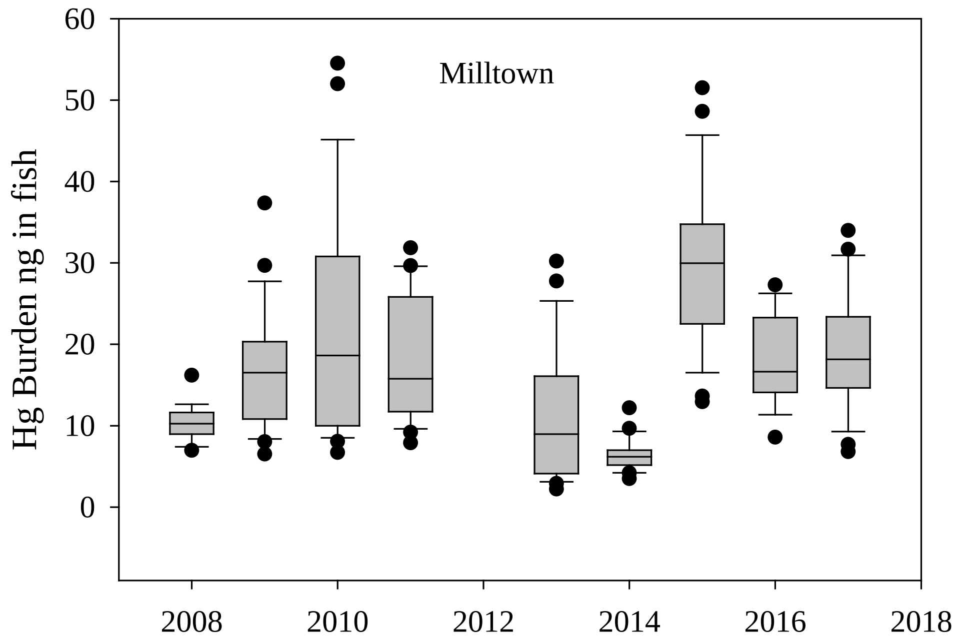


Figure 12. Box plots of Milltown fish Hg burden 2008 through 2017. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers.

6.2.4 Eagle Harbor Patuxent River

Perch collected from Eagle Harbor in 2017 were larger than previous years but had the same strong length weight relationship (Figure 13). Fish were not collected in

2012 because seining was unsuccessful and attributed to low reproductive success. From 2013 onward the perch collected from Eagle Harbor have increased in size. Following a peak in 2013, Hg concentrations have trended downward. This trend was treated with caution as the sample size for 2016 is only 9. However, with 25 fish in 2017, the trend continued and overall the decrease in concentration is significant (r^2 0.07 p <0.01). There is no relationship between individual perch size and Hg concentration within a year or even between years (Figure 14). The average size of the fish is increasing over time as Hg concentration decreases, but unlike at other Patuxent sites, an increase in Hg burden also occurs (Figure 15).

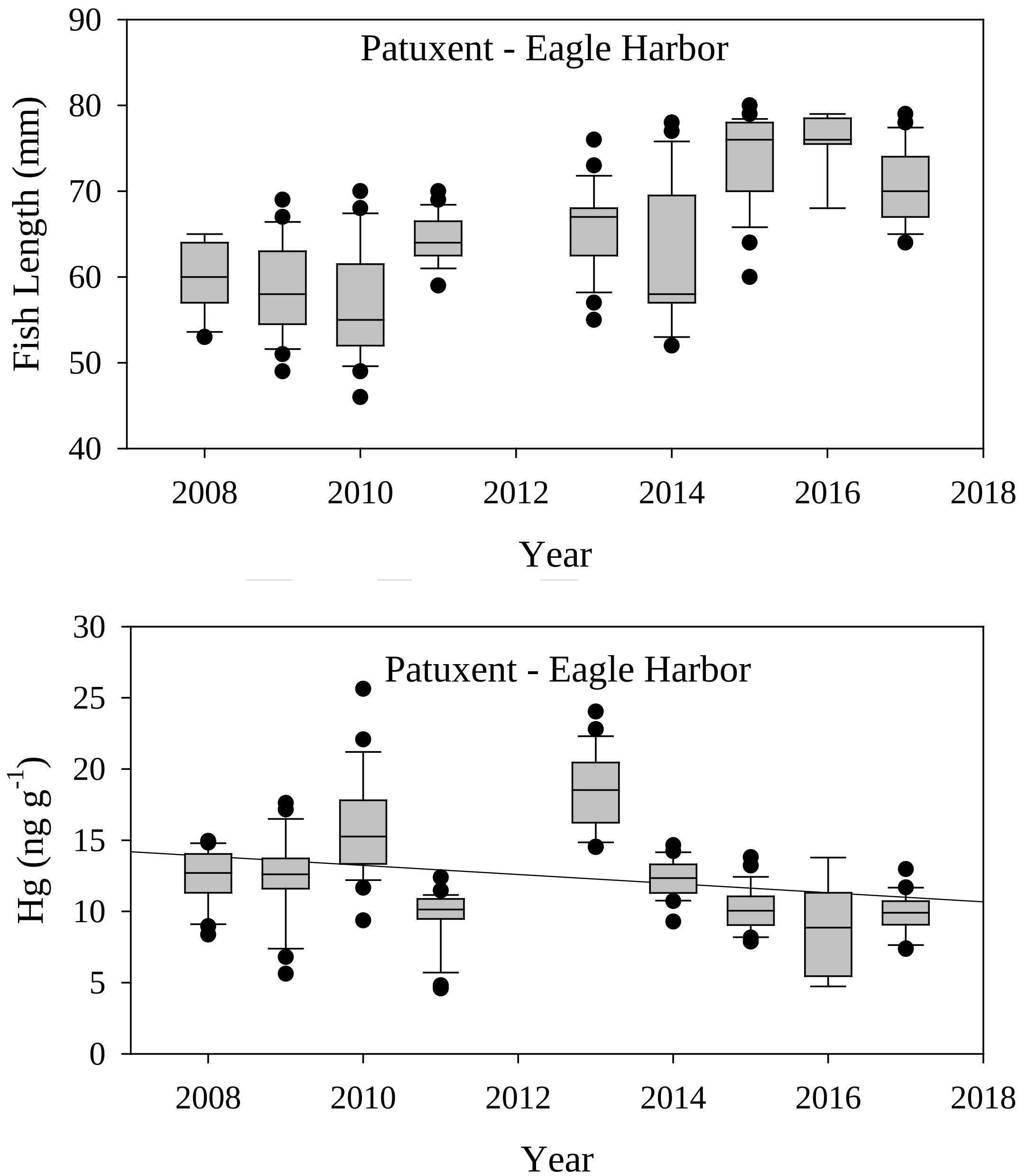


Figure 13. Patuxent River Eagle Harbor Box plots of fish size (upper) and fish Hg concentration (lower) 2008 through 2017. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers.

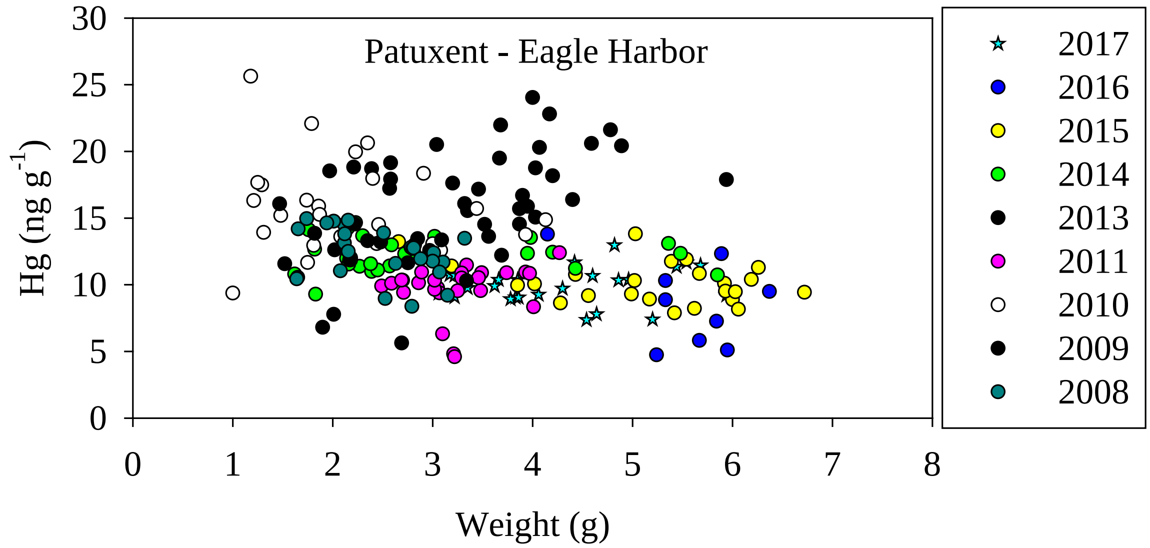


Figure 14. Fish Hg-weight relationship for individual fish collected at Eagle Harbor Patuxent River.

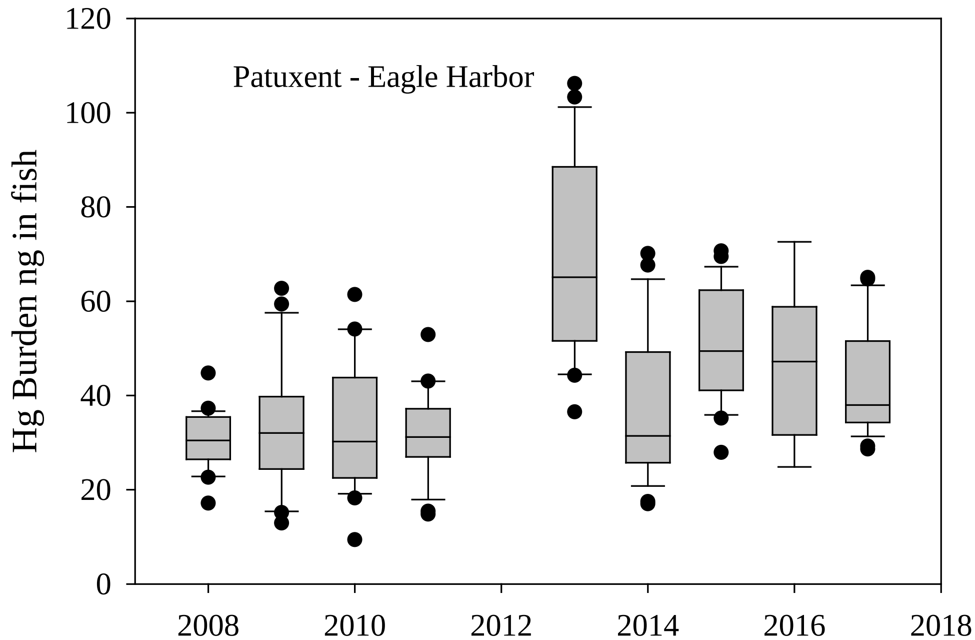


Figure 15. Box plots of Patuxent Eagle Harbor fish Hg burden 2008 through 2017. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers.

6.3 Freshwater (Largemouth Bass) Sites

Concentrations of Hg in Largemouth Bass of nine reservoirs have shown the same relative distribution. Bass from the Savage Reservoir continue to have the highest Hg concentrations followed by fish from Cash Lake and Lake Lariat (Figure 16). These trends suggest factors inherent to each watershed influence the concentrations. One indication of this these differences can be seen in the mercury:selenium ratio discussed at length in section 7.0. Trends in fish Hg concentration among fish in the lakes are variable; some show increasing fish Hg concentrations such as in Tuckahoe Lake, whereas some show decreasing trends such as Savage Reservoir and some show no change such as Prettyboy Reservoir. The detailed data from each site are discussed in the following sections. Overall, no temporal trend is apparent to date. Inter annual fish size fluctuations add to the complexity.

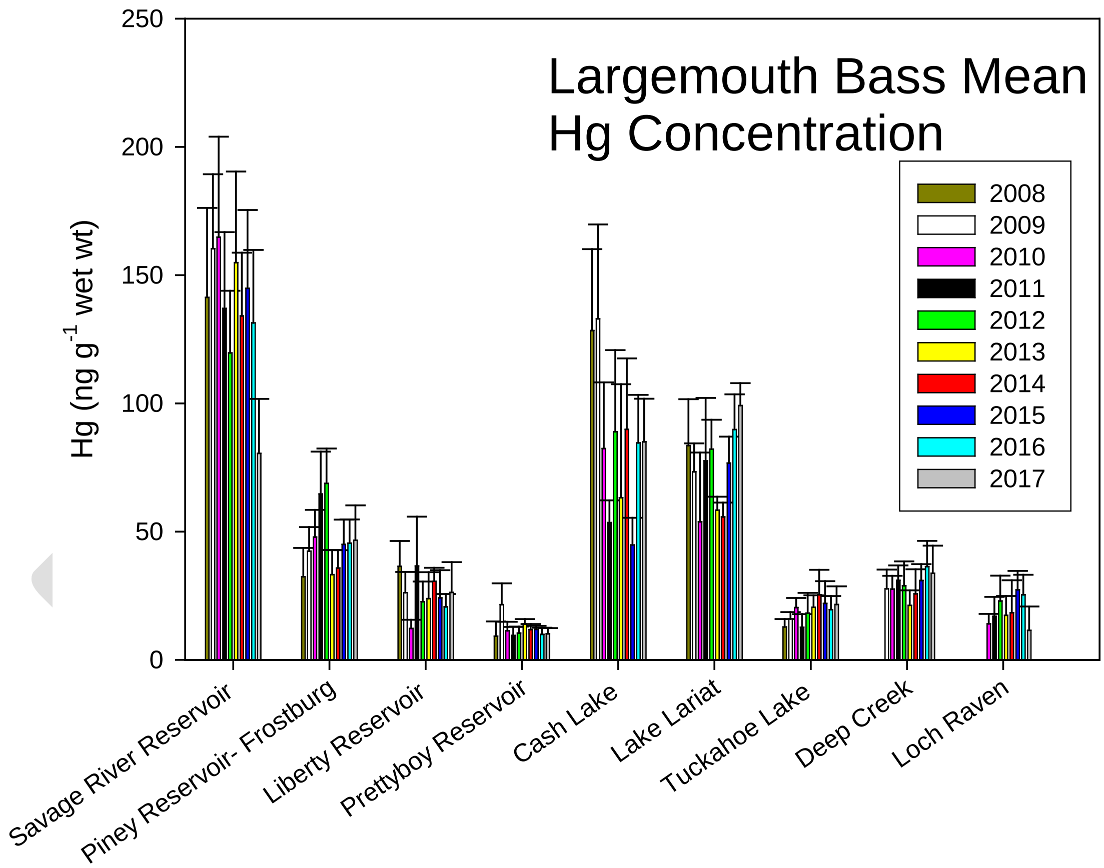


Figure 16. Mean and standard deviation of mercury concentrations in bass collected between 2008 and 2017 from Maryland freshwater lakes and reservoirs.

6.3.1 Tuckahoe Lake

The largemouth bass collected from Tuckahoe Lake in 2017 had the same strong relationship between size and weight as observed in previous years. The fish were larger in size than average (2017 mean 6.6g) relative to other years (mean 5.6g) (Figure 17) but within the range that has been observed. The concentrations of Hg in fish from 2017 are toward the high end of what has been observed in the study, and a trend of increasing Hg concentration is now apparent ($R^2 = 0.14$ $p < 0.01$) (Figure 13). While there is no general trend between Hg concentration and fish size, the highest concentrations do occur in the smallest fish (Figure 18). The wide range in fish sizes collected in a given year results in a wide range in Hg burdens, but no change in burden over time is apparent (Figure 19).

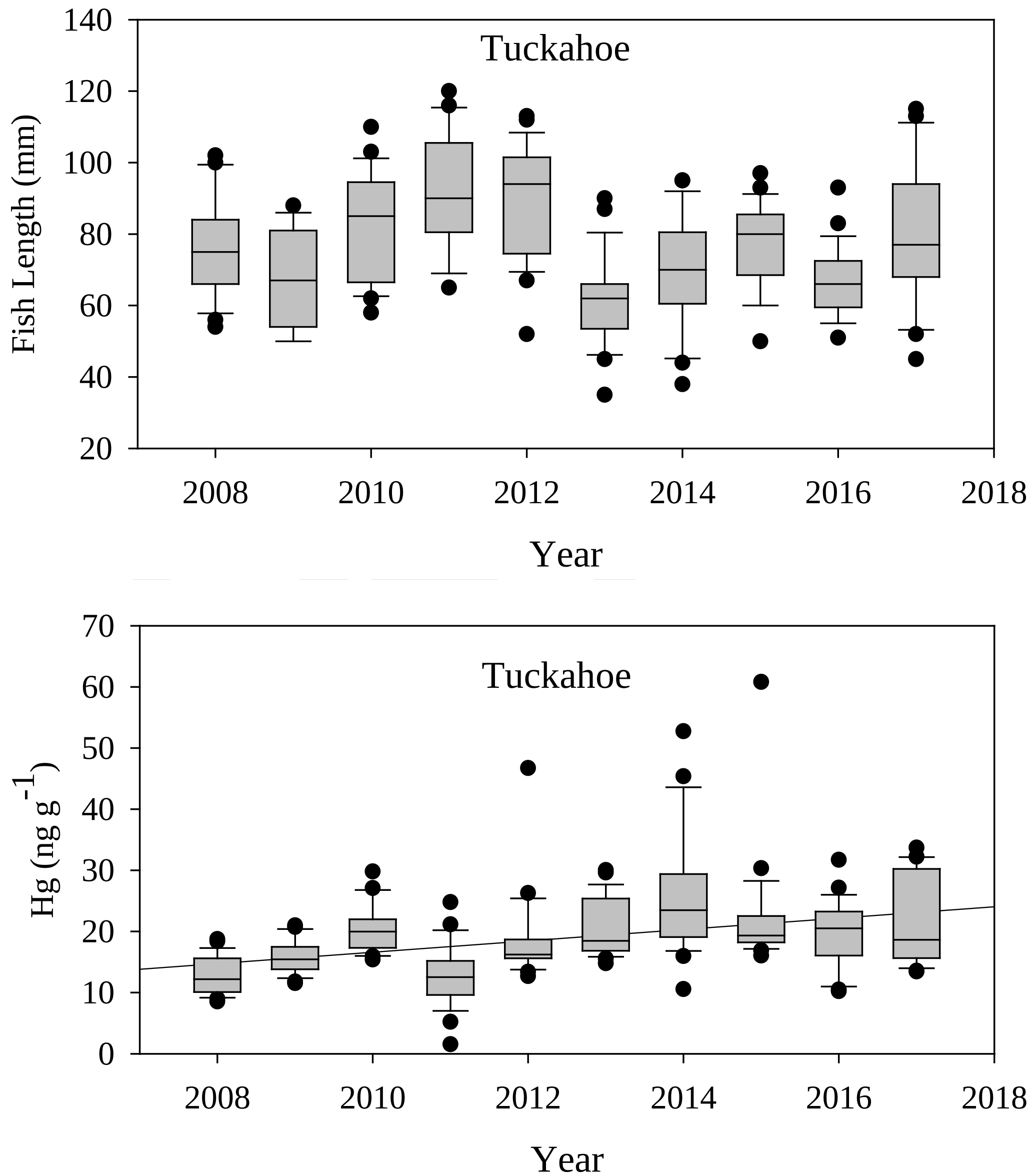


Figure 17. Tuckahoe Lake: Box plots of fish size (upper) and fish Hg concentration (lower) 2008 through 2017. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers.

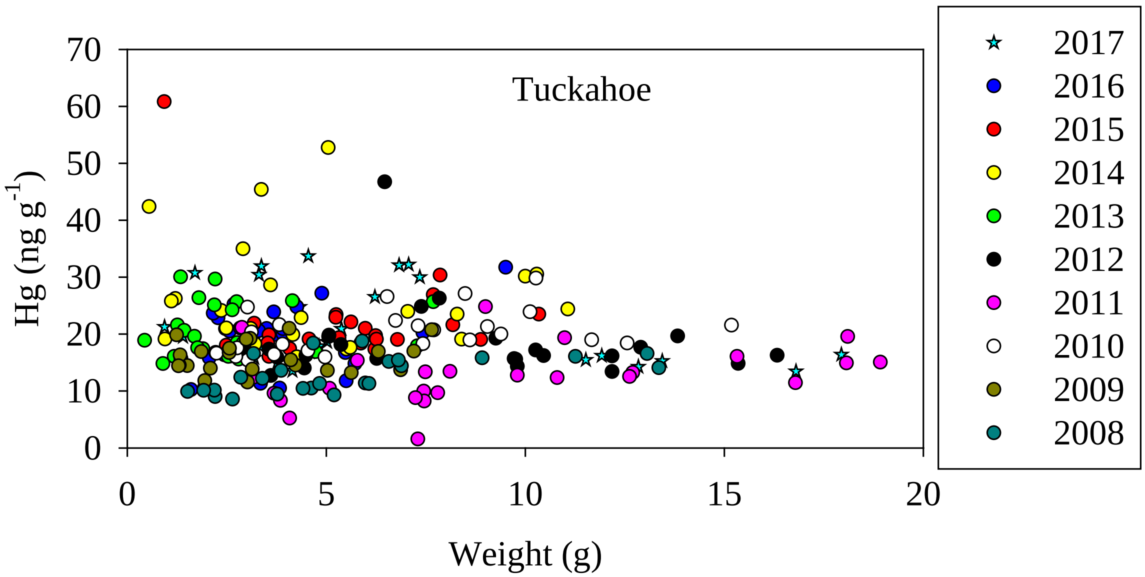


Figure 18.Fish Hg-weight relationship for individual fish collected at Tuckahoe Lake.

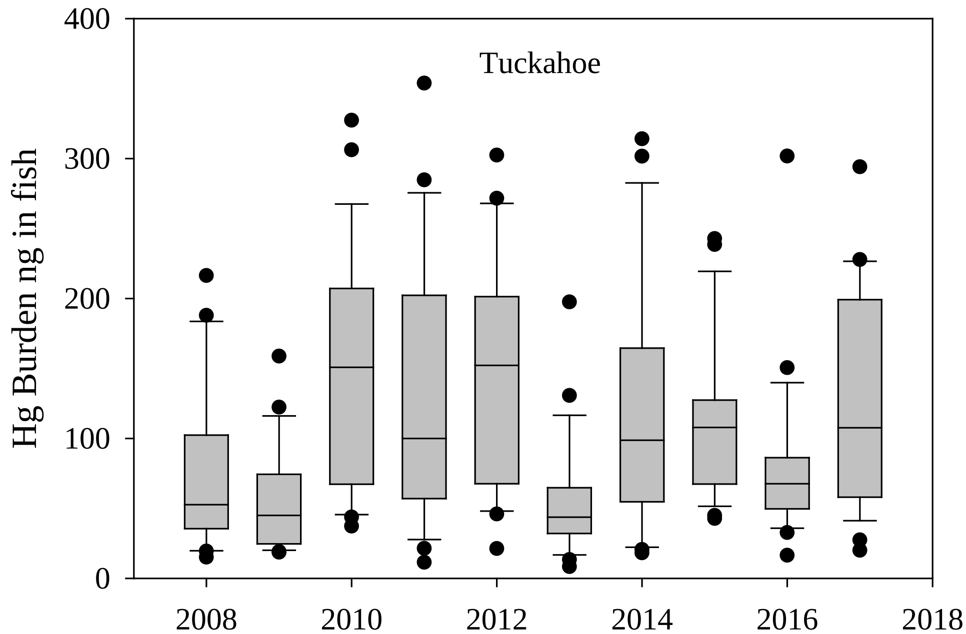


Figure 19. Box plots of Tuckahoe Reservoir fish Hg burden 2008 through 2017. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers.

6.3.2 Liberty Reservoir

The size of the fish, collected in Liberty Reservoir in 2017, were larger (7.1g) compared to previous years (running mean = 4.7g) (Figure 20). The strong length weight relationship (not shown) seen in past years was also apparent in 2017. Concentrations of Hg in fish collected in 2016 were similar to other years and there remains no temporal trend in the data (Figure 20). No relationship between fish size and fish Hg concentration is apparent within or among the study years (Figure 21). Body burdens of Hg are variable but tend to follow fish size (Figure 22).

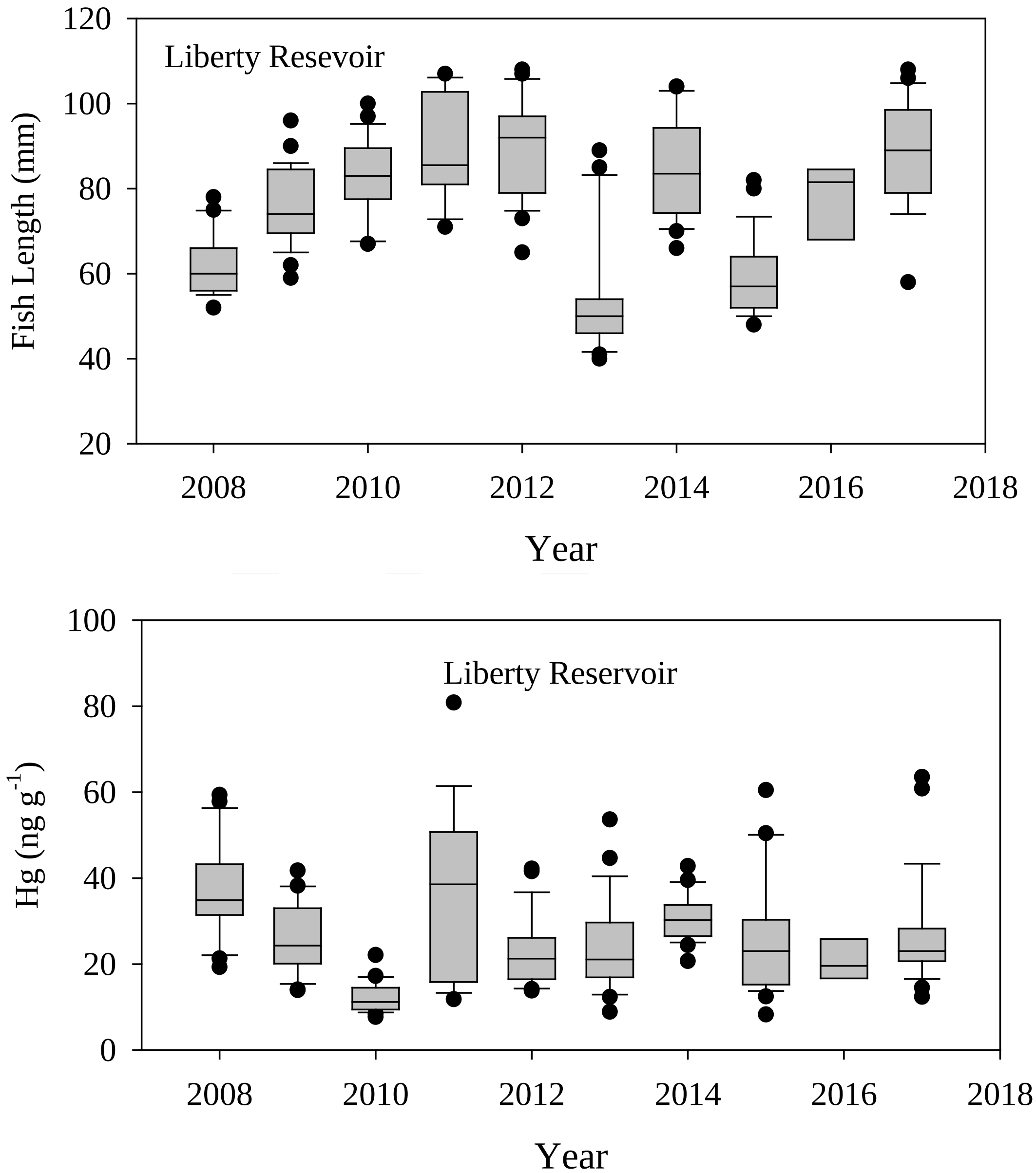


Figure 20. Liberty Reservoir: Box plots of fish size (upper) and Hg concentration (lower) 2008 through 2017. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers.

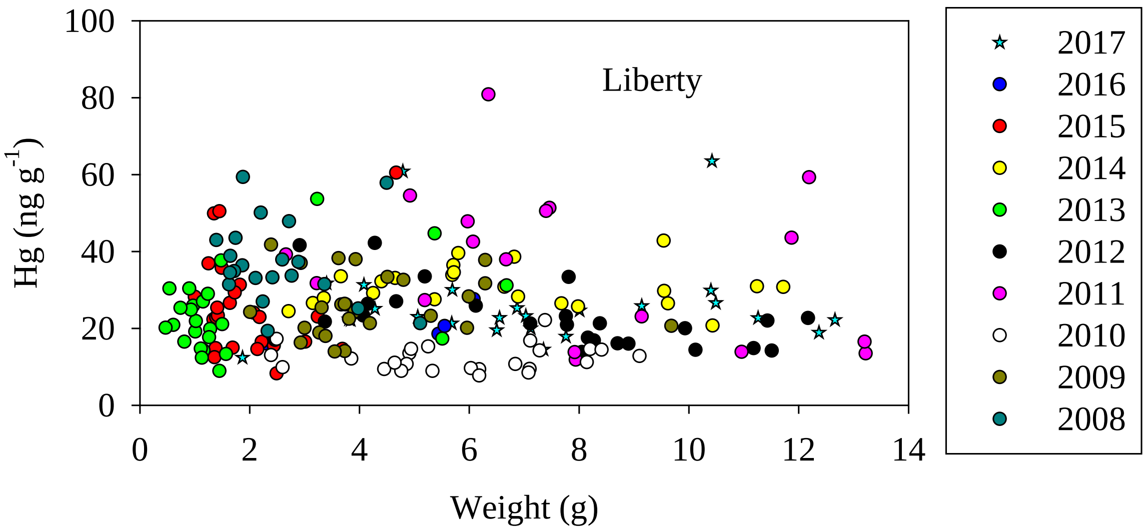


Figure 21. Fish Hg-weight relationship for individual fish collected at Liberty Reservoir.

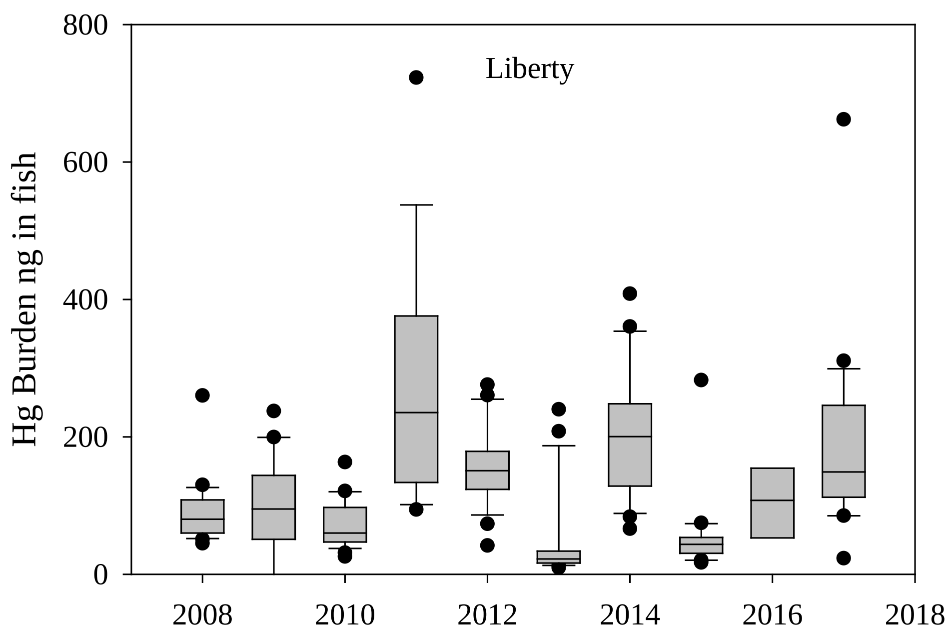


Figure 22. Box plots of Liberty Reservoir fish Hg burden 2008 through 2017. The boundary of the box is the 25^{th} percentile, the whiskers indicate the 10^{th} and 90^{th} percentile and circles indicate outliers.

6.2.3 Prettyboy Reservoir

Largemouth Bass collected in 2017 were larger (5.9 g) when compared to the average of other years (4.7 g) (Figure 23). The range in size over the study is substantial, being a factor of 2. A strong weight to length relationship exists in 2017 as in previous years (not shown). Mercury concentrations remained relatively stable over the entire study, with 2009 being anomalous, even though size of the fish size has varied. The scatter plot of mass and individual concentrations from all years reveals no trend within or among years (Figure 23). Fish Hg burdens also have changed little over time (Figure 24)

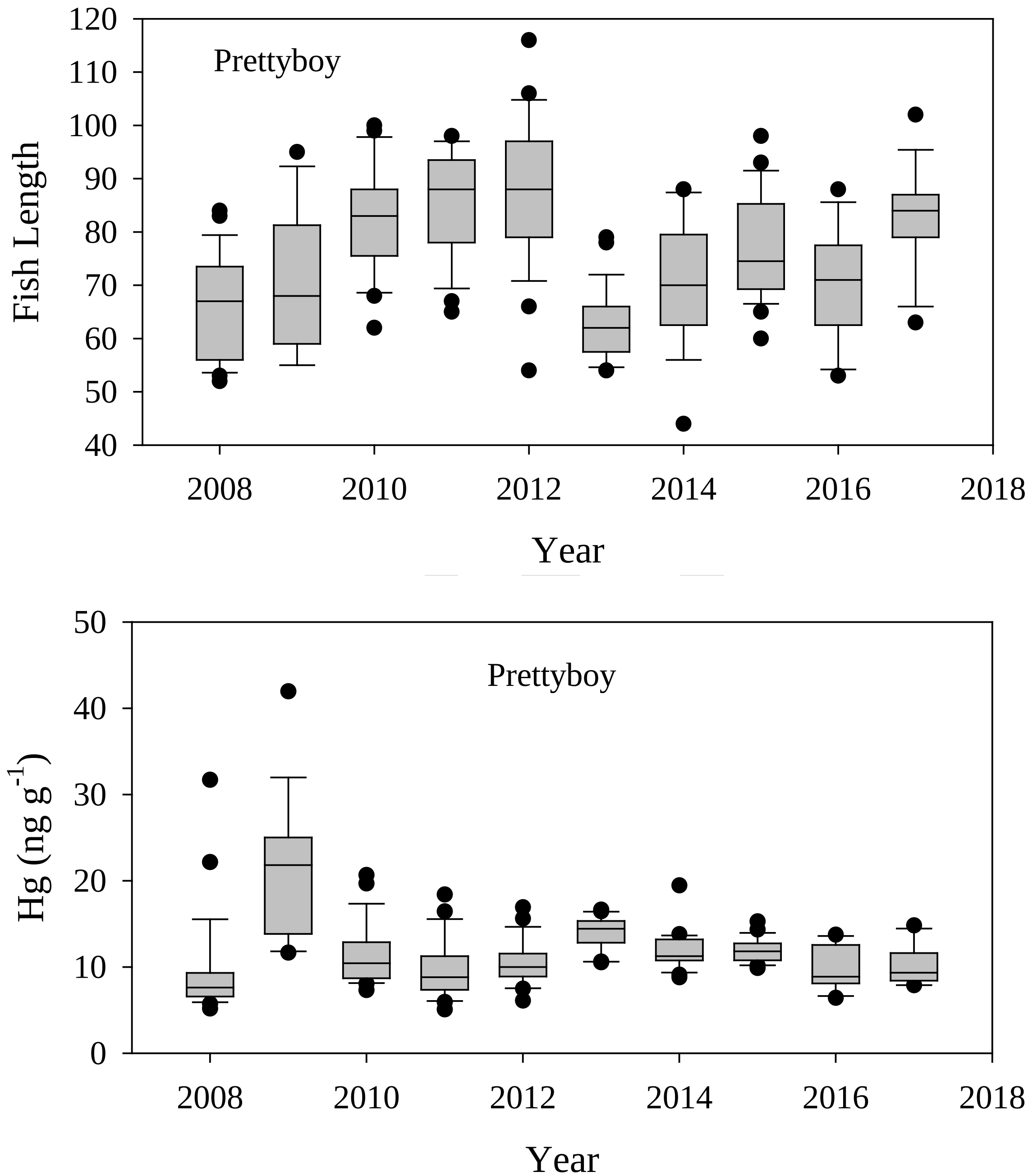


Figure 23. Prettyboy Reservoir: Box plots of fish size (upper) and Fish Hg concentration (lower) 2008 through 2017. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers.

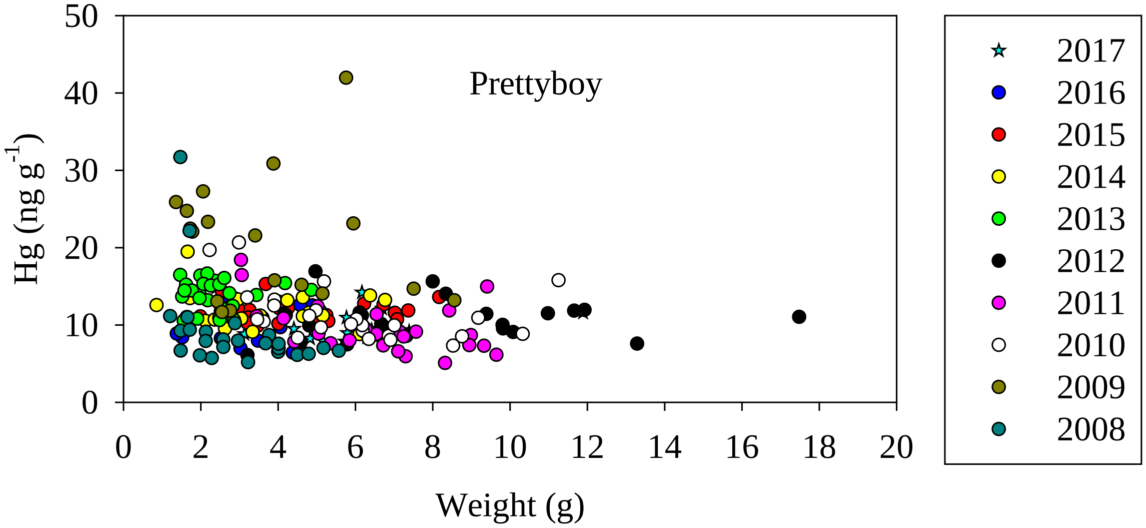


Figure 24. Fish Hg-weight relationship for individual fish collected at Prettyboy Reservoir.

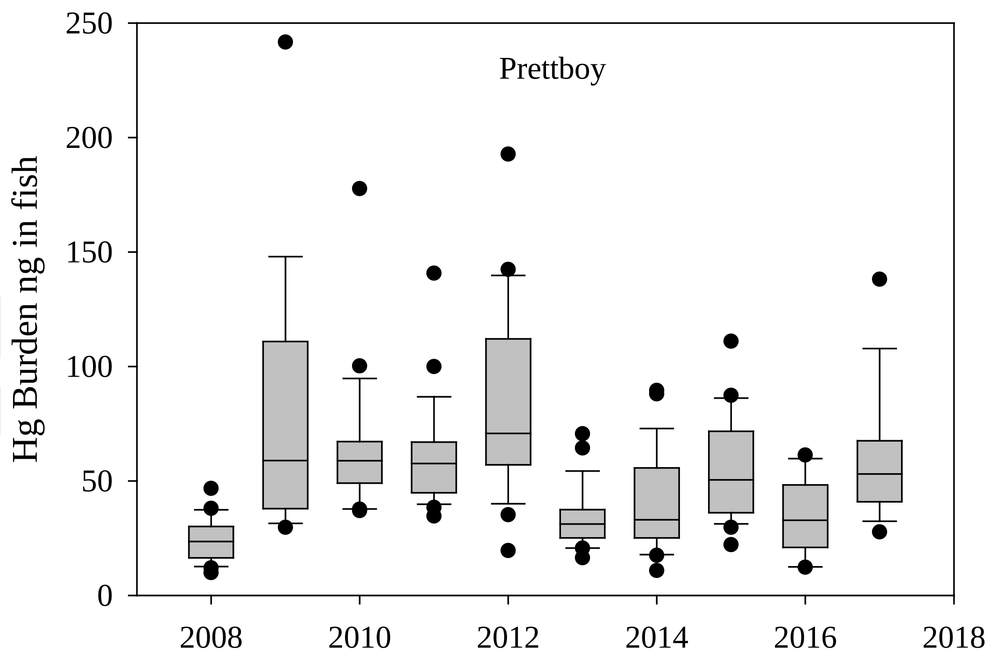
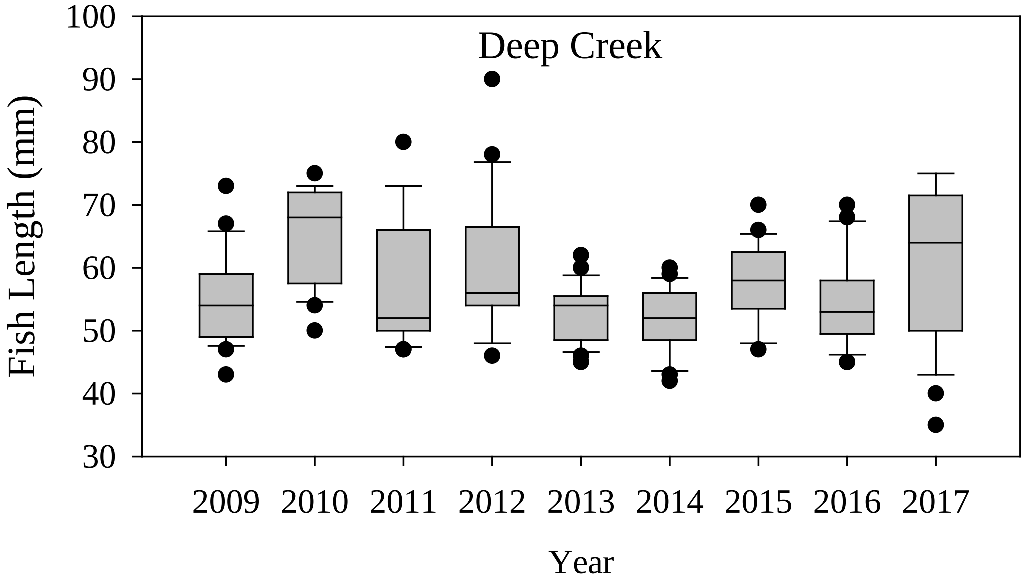


Figure 25. Box plots of Prettyboy Reservoir fish Hg burden 2008 through 2017. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers

6.2.4 Deep Creek

Sampling in Deep Creek started in 2009. The fish length weight relationship (not shown) remains strong in 2017 as it did in previous years. Fish Hg concentrations were similar to previous years and there is no trend with time (Figure 26). The variability in fish Hg concentrations among individuals collected from a single year remains large. There is no consistent trend between fish size and fish Hg concentration (Figure 27) and no temporal trend in fish Hg burden (Figure 28).



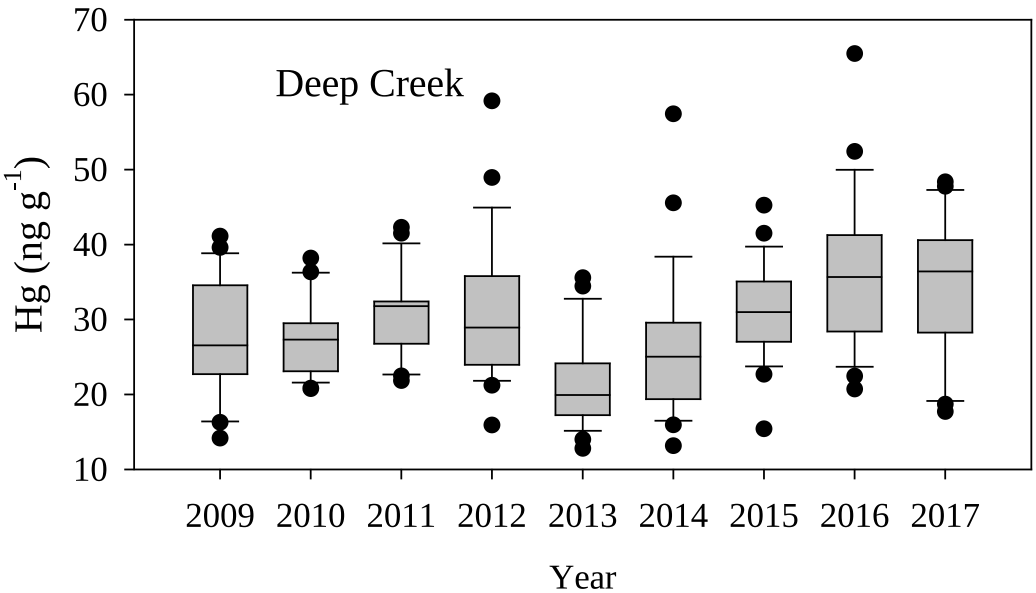


Figure 26. Deep Creek Reservoir: Box plots of fish size (upper) and fish Hg concentration (lower) 2009 through 2016. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers.

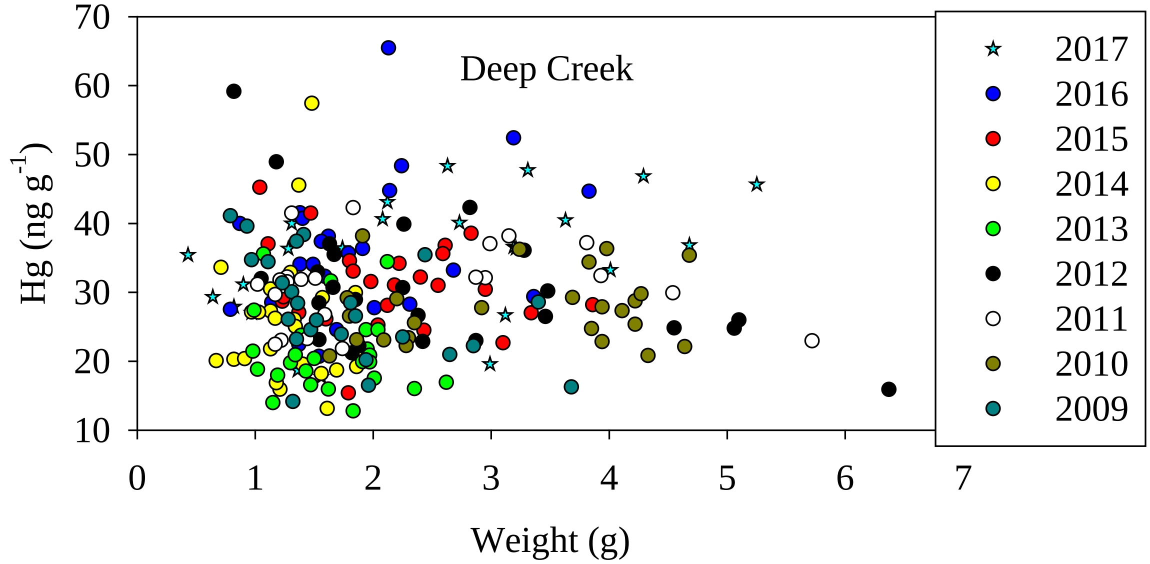


Figure 27. Fish Hg-weight relationship for individual fish collected at Deep Creek Reservoir.

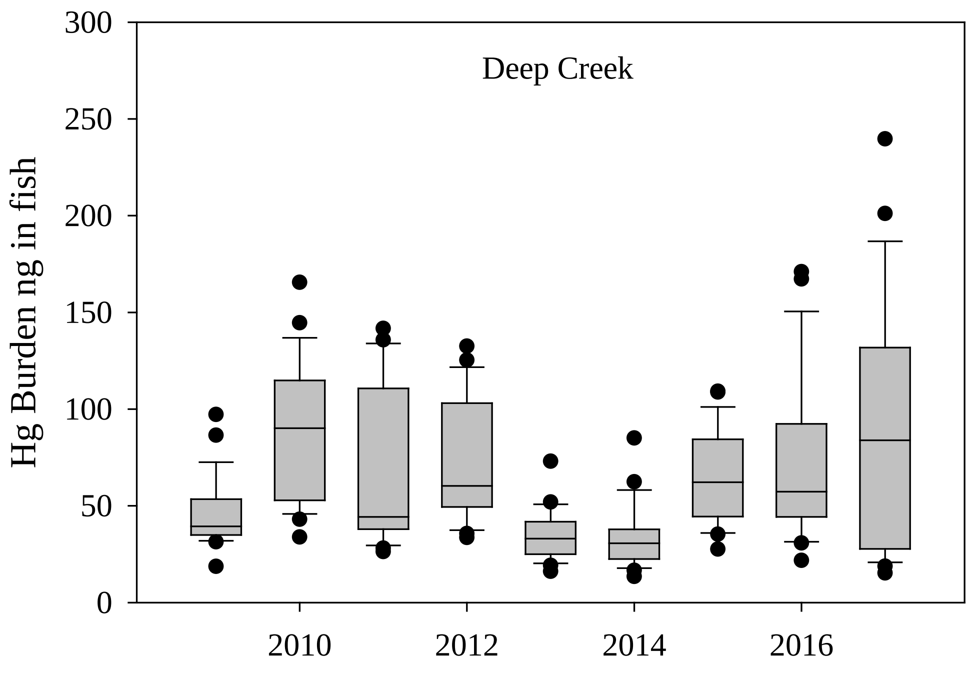
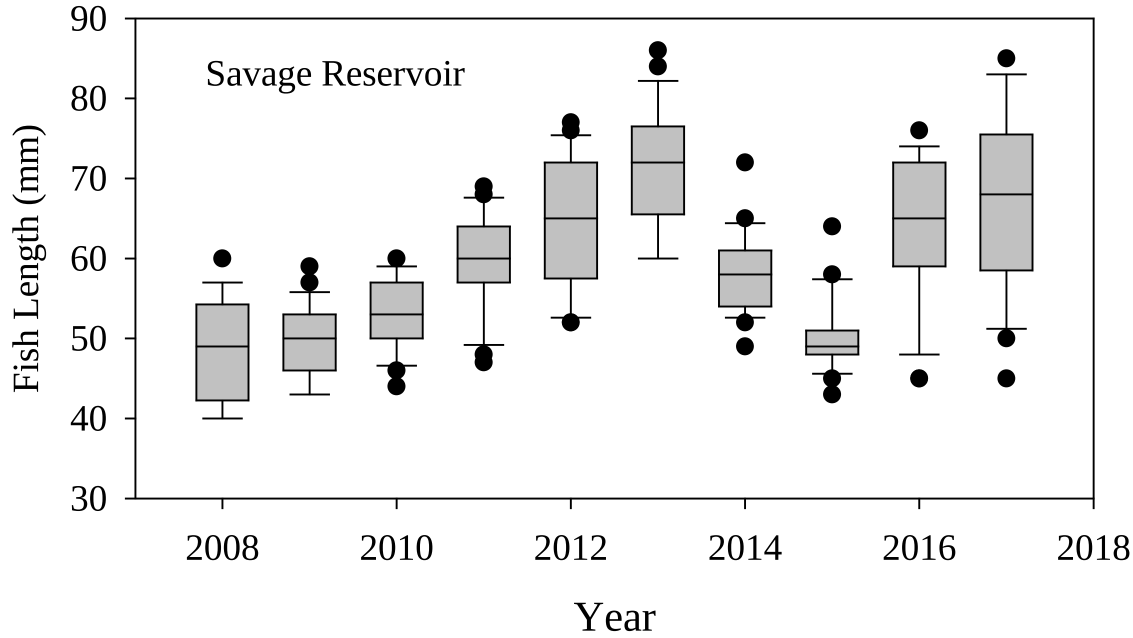


Figure 28. Box plots of Deep Creek Reservoir fish Hg burden 2008 through 2017. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers.

6.2.5 Savage River Reservoir

The fish collected from the Savage River Reservoir in 2017 were larger in mass (3.7g) compared to previous years (2.3g) and length (Figure 28). The size of the YOY bass collected had increased each year from 2009 to 2013. However, in 2014 and 2015 a step wise decrease in the size of fish sampled occurred. In 2016, the fish size increased to 3.1 g and to 3.7g in 2017. The length-mass relationship is strong and remains consistent with what has been observed in the previous five years. The 2017 fish varied widely in size, a condition common in previous years. The average fish Hg concentration varies from year to year but oscillates around a value of approximately 150 ng g^{-1} (Figure 29). In 2017, the average concentration was only 80.5 ng g^{-1} a marked change from previous years. The within year variation in individual YOY fish Hg concentrations in Savage Reservoir remains high compared to the other sites sampled in this study (Table 3). Even with a drop in Hg concentrations fish size and size variability was similar to previous years (Figure 30). There are no temporal trends in the concentrations of Hg in Savage Reservoir fish (Figure 29) or fish Hg burden (Figure 31). Fish Hg burden peaked in 2013 coincident with maximum fish size and the variability in Hg burden appears to be increasing coincident with increasing variability in fish size. The peak is contrary to a biodilution explanation (discussed in section 7) and may reflect changes in fish diet with increased size.



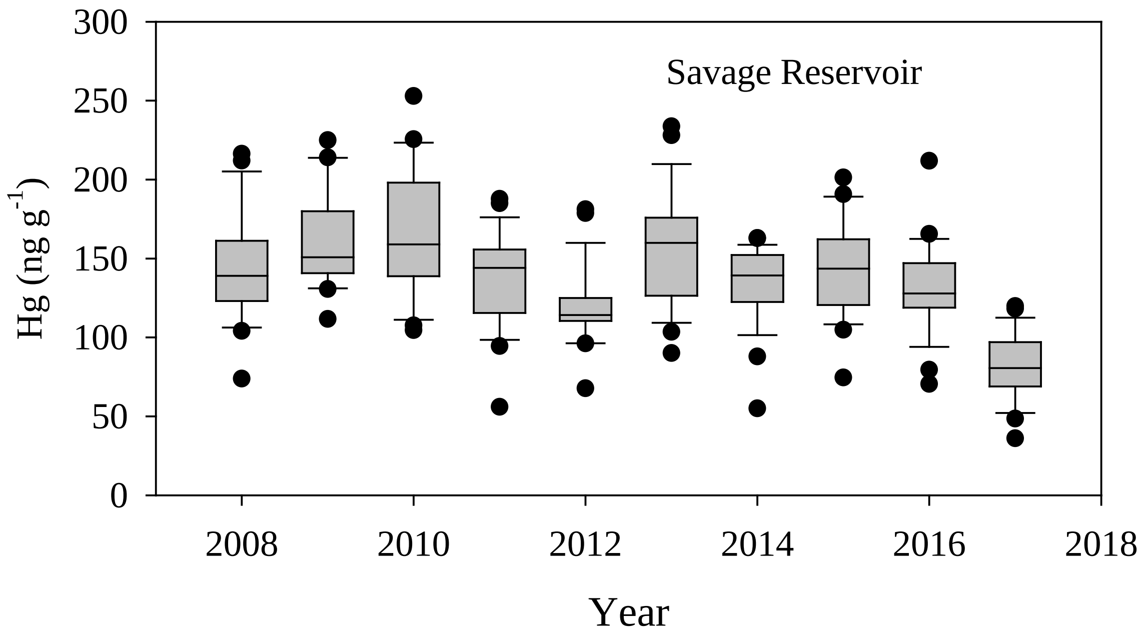


Figure 29. Savage Reservoir: Box plot of fish size (upper) and fish Hg concentration (lower) 2008 through 2016. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers.

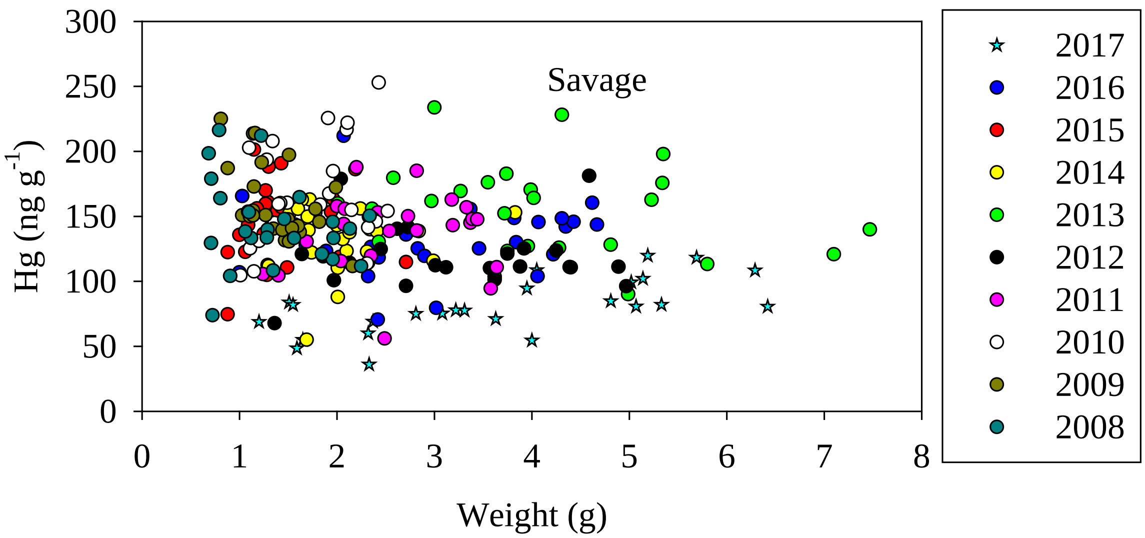


Figure 30. Fish Hg-weight relationship for individual fish collected at Savage Reservoir.

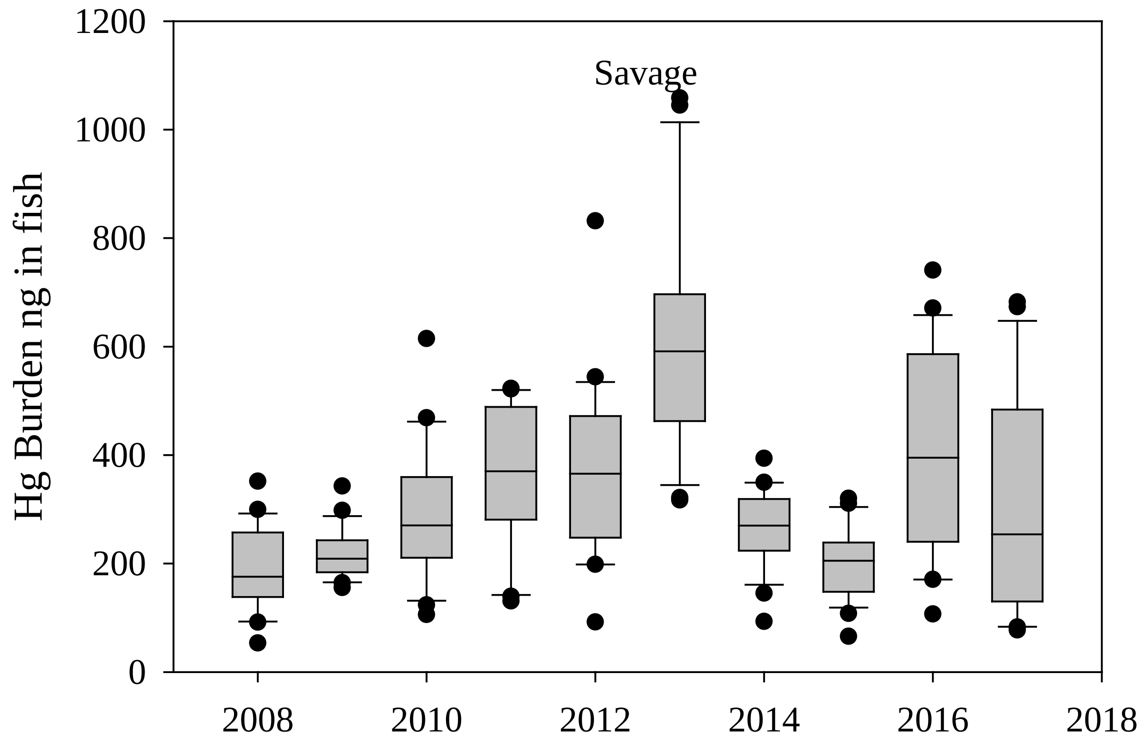


Figure 31. Box plots of Savage River Reservoir fish Hg burden 2008 through 2017. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers.

6.2.6 Piney Reservoir (Frostburg)

The fish collected from the Piney Reservoir in 2017 had a strong weight length relationship and were similar in size to fish collected in previous years (Figures 32, 33). Concentrations of Hg in fish create and interesting temporal pattern, as Hg concentrations increase from 2008 to 2012 but in 2013 concentrations appear to have “reset” to 2008 levels before increasing in subsequent years (Figure 32). Concentrations in fish collected in the last three years are similar (Figure 32) but have lower Hg burdens (Figure 34). Otherwise, fish Hg burdens have the same pattern as fish Hg concentration. Size does not explain the cycles as there is no relationship between fish size and fish Hg concentration (Figure 33). The cause maybe a result of a system “resets” perhaps the result of a water level variation as part of operations and warrants investigation.

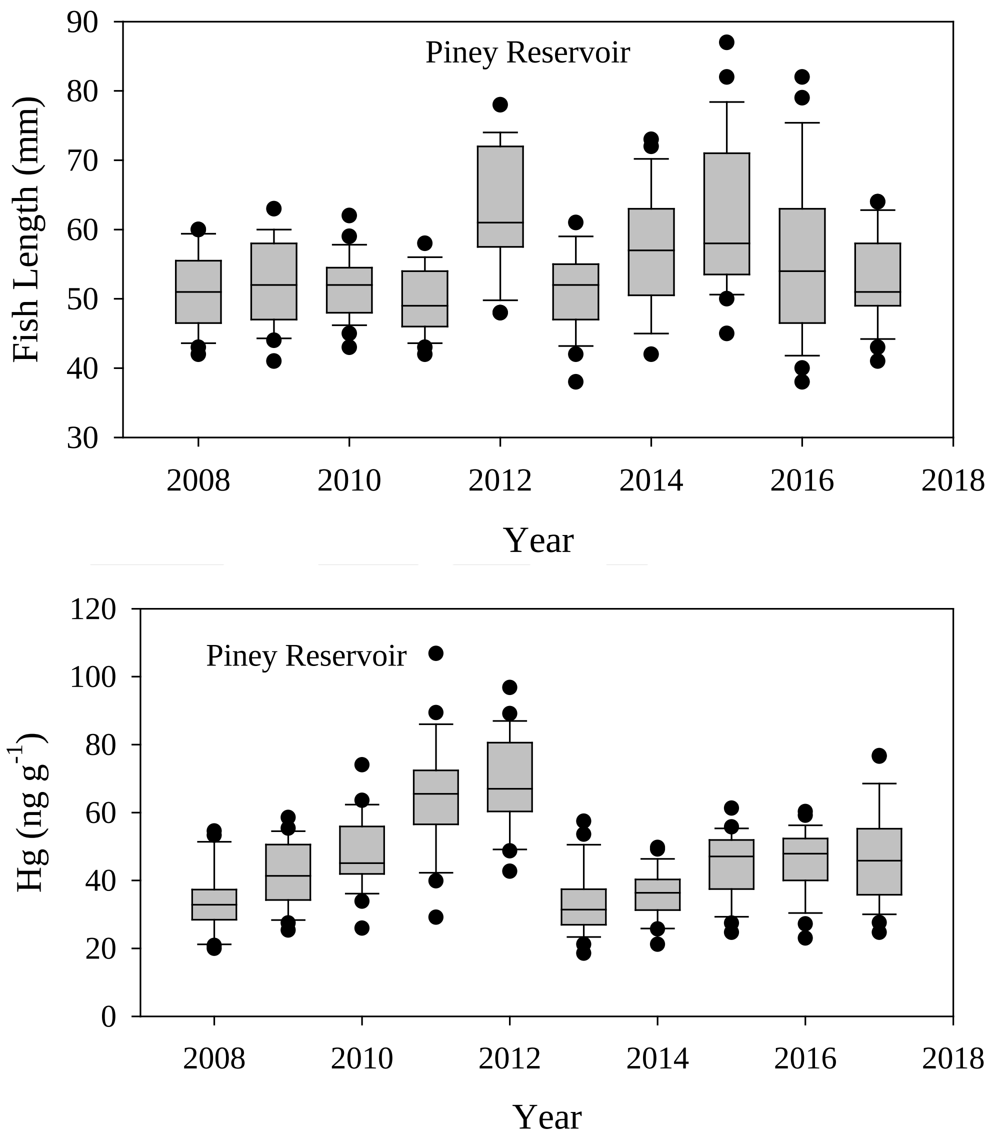


Figure 32. Piney Reservoir: Box plots of fish size (upper) and fish Hg concentration (lower) 2008 through 2017. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers.

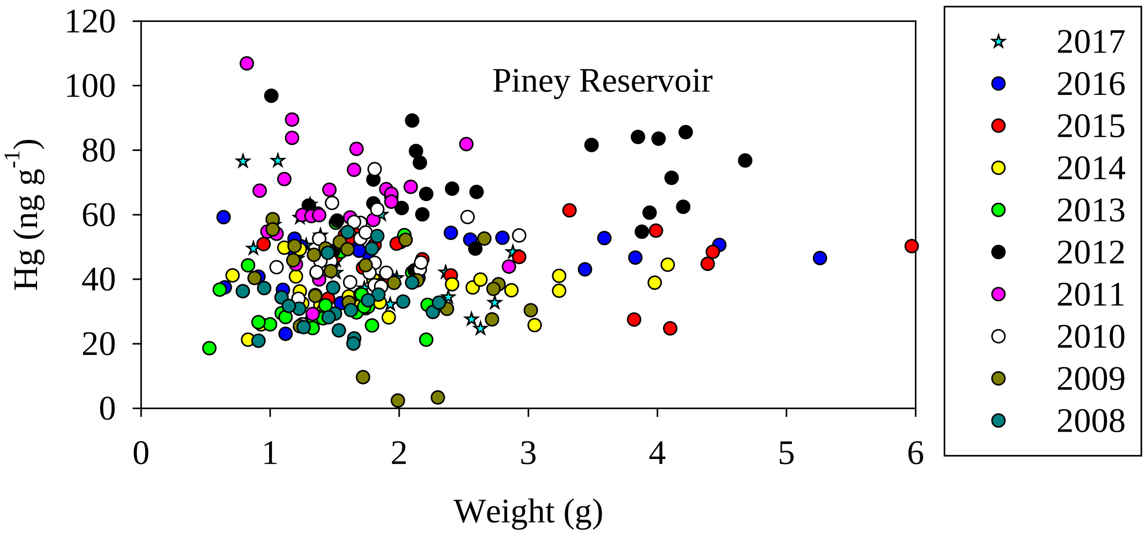


Figure 33. Fish Hg-weight relationship for individual fish collected at Piney Reservoir.

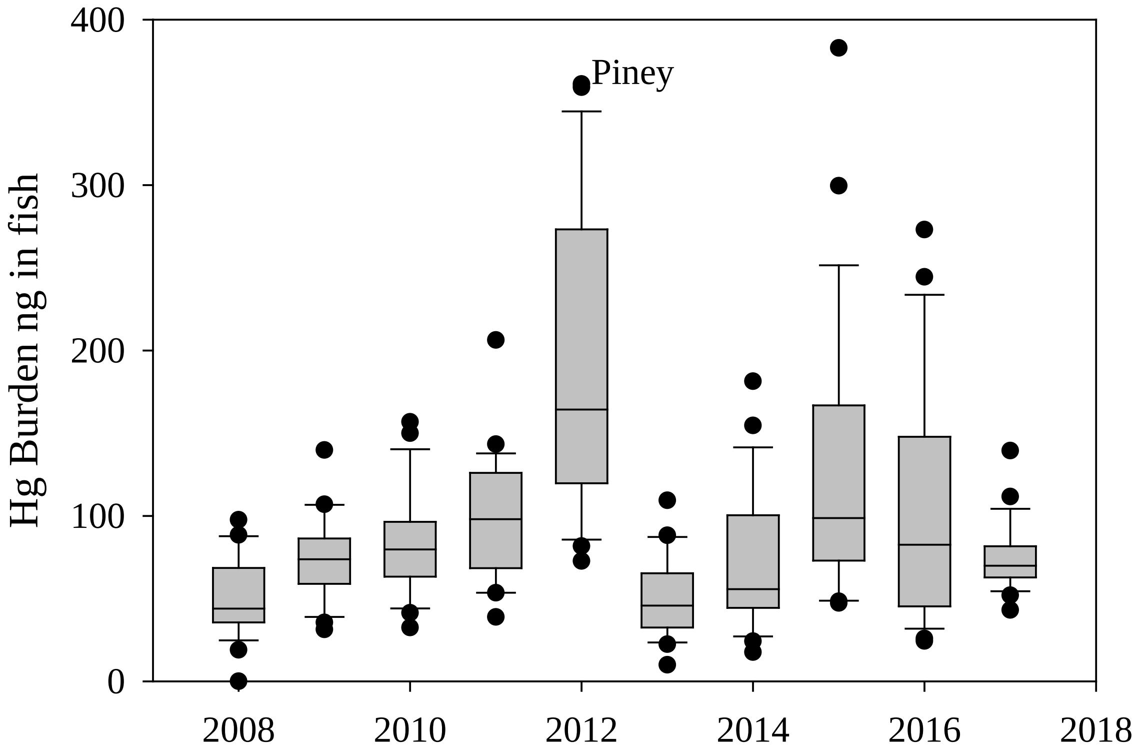


Figure 34. Box plots of Piney Reservoir fish Hg burden 2008 through 2017. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers.

6.2.7 Cash Lake

The bass collected from Cash Lake in 2017 have the same strong length to weight relationship displayed in previous years (not shown). The size of bass collected from Cash Lake in 2017, were typical of other study years (Figure 35). The Hg concentrations in the fish collected in 2017 were typical of the past few years. The downward trend in Hg concentration is driven by the decrease observed in 2010 (R^2 = 0.16 p=0.00). Even though the size of fish varies greatly, there is no relationship between fish size and Hg concentration within a year or between years (Figure 36). The large range in fish size results in a wide range of fish Hg burdens (Figure 37).

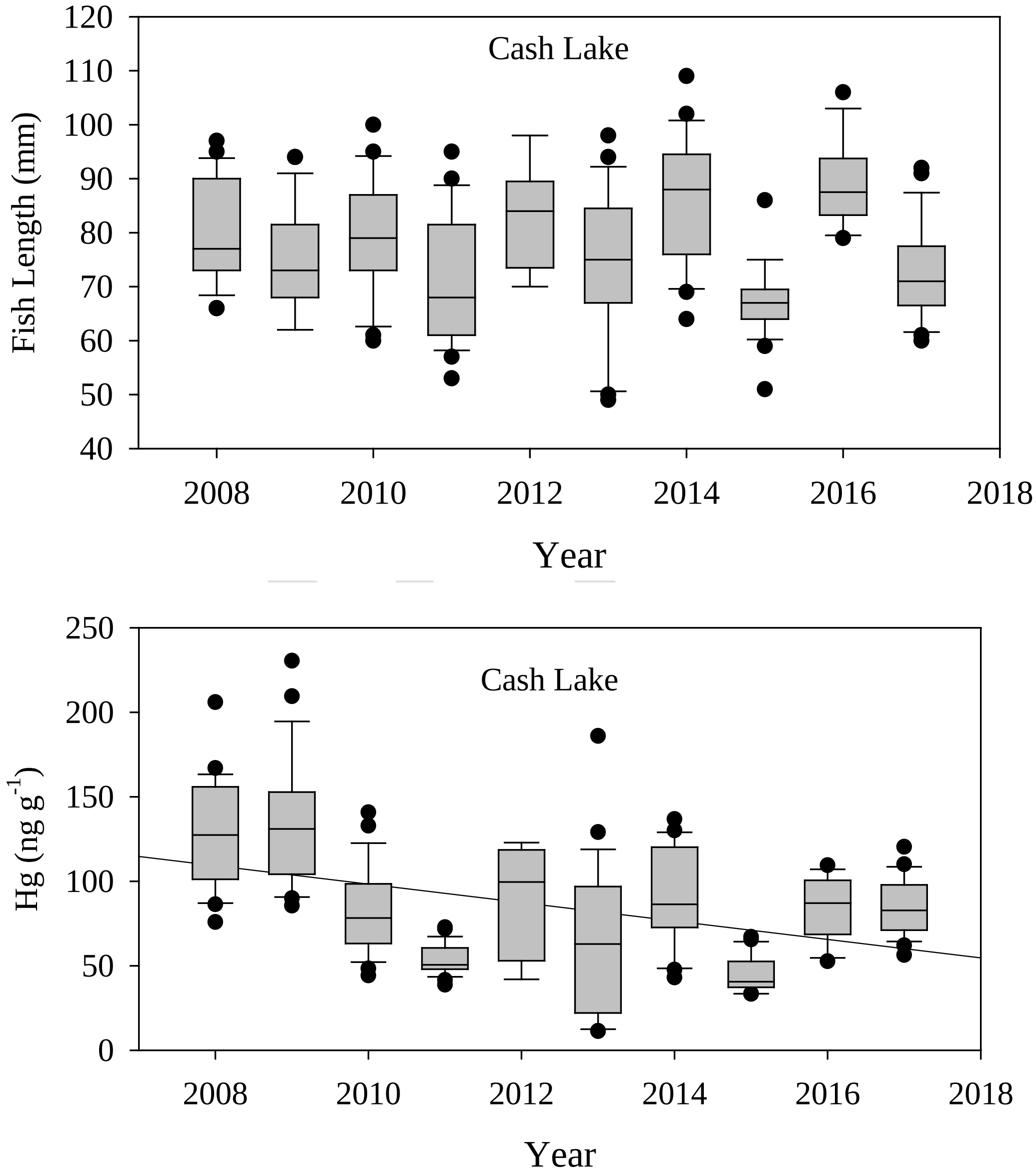


Figure 35. Cash Lake: Box plots of fish size (upper) and fish Hg concentration (lower) 2008 through 2016. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers.

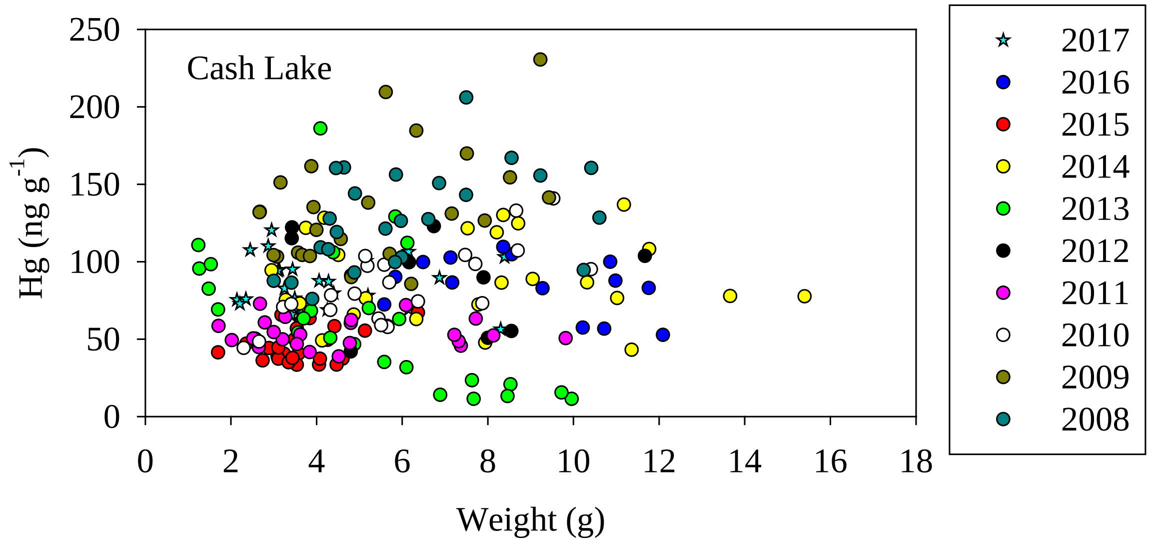


Figure 36. Fish Hg-weight relationship for individual fish collected at Cash Lake.

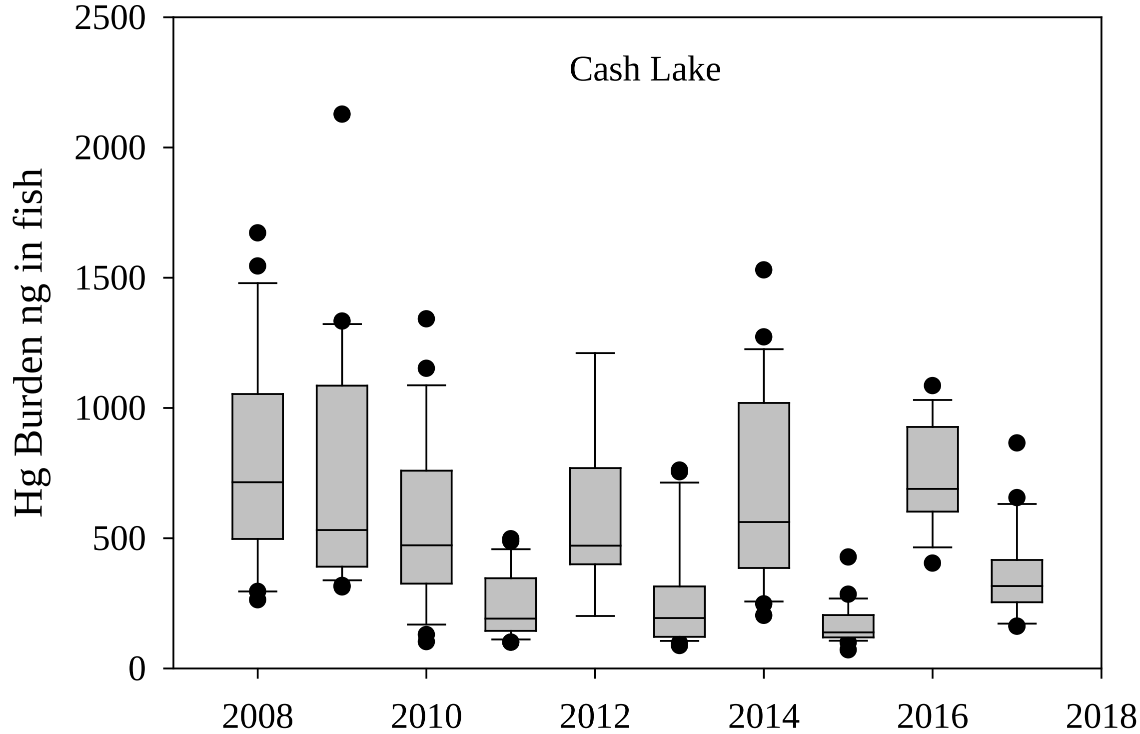


Figure 37. Box plots of Cash Lake fish Hg burden 2008 through 2017. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers.

6.2.8 Lake Lariat

The bass collected from Lake Lariat in 2017 have a strong weight to length relations, which we have also seen in previous years. The bass collected from Lake Lariat in 2017 were about average compared to all study years (Figure 38). The size of the fish collected increased each year from less than 80 mm in 2008 to more than 110 mm in 2012 but from 2014 onward the fish have been similar in size. The average Hg concentration in bass collected in 2017 was about average for the study period being 63 ng g^{-1} compared to 71 ng g^{-1} for the entire period (Table 3). Hg concentrations tend to be oscillating around 70 ng g^{-1} rather than trending in any direction. There is no trend between fish size and fish Hg concentration apparent within or among the study years (Figure 39) and fish mercury burdens appear driven by fish size, peaking in 2011 and 2012 (Figure 40).

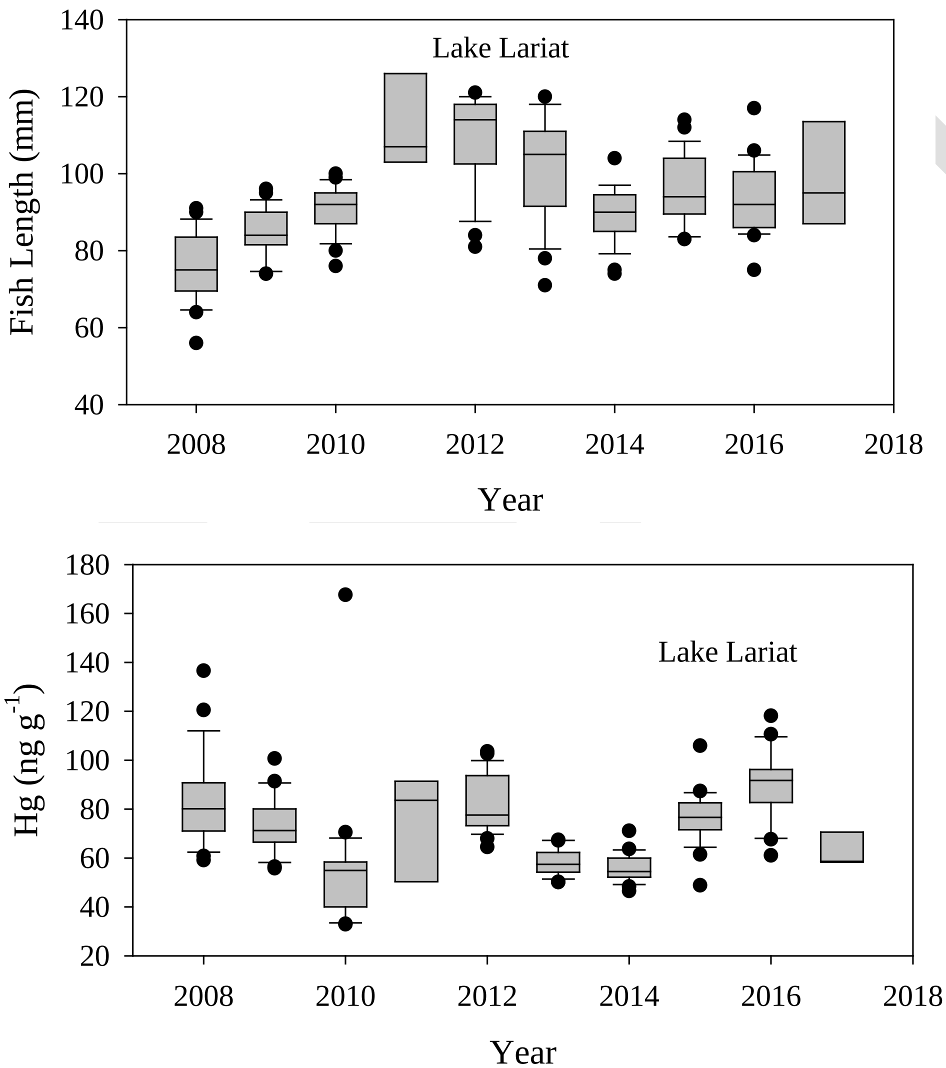


Figure 38. Lake Lariat: Box plots of fish size (upper) and fish mercury concentration (lower) 2008 through 2016. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers. The sample size for 2011 was 7, whereas it was 25 for the other years.

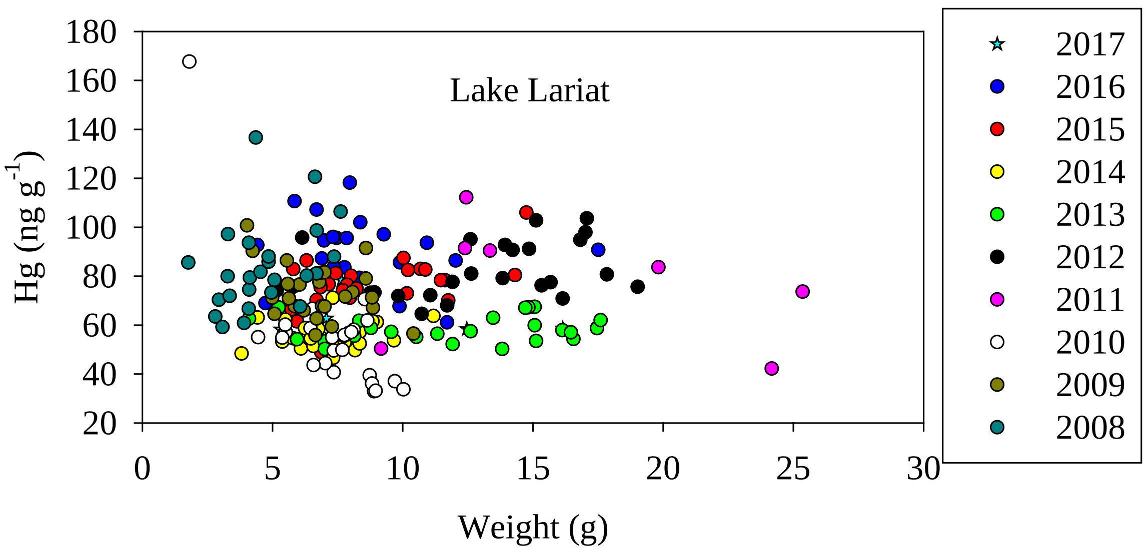


Figure 39. Fish Hg-weight relationship for individual fish collected at Lake Lariat.

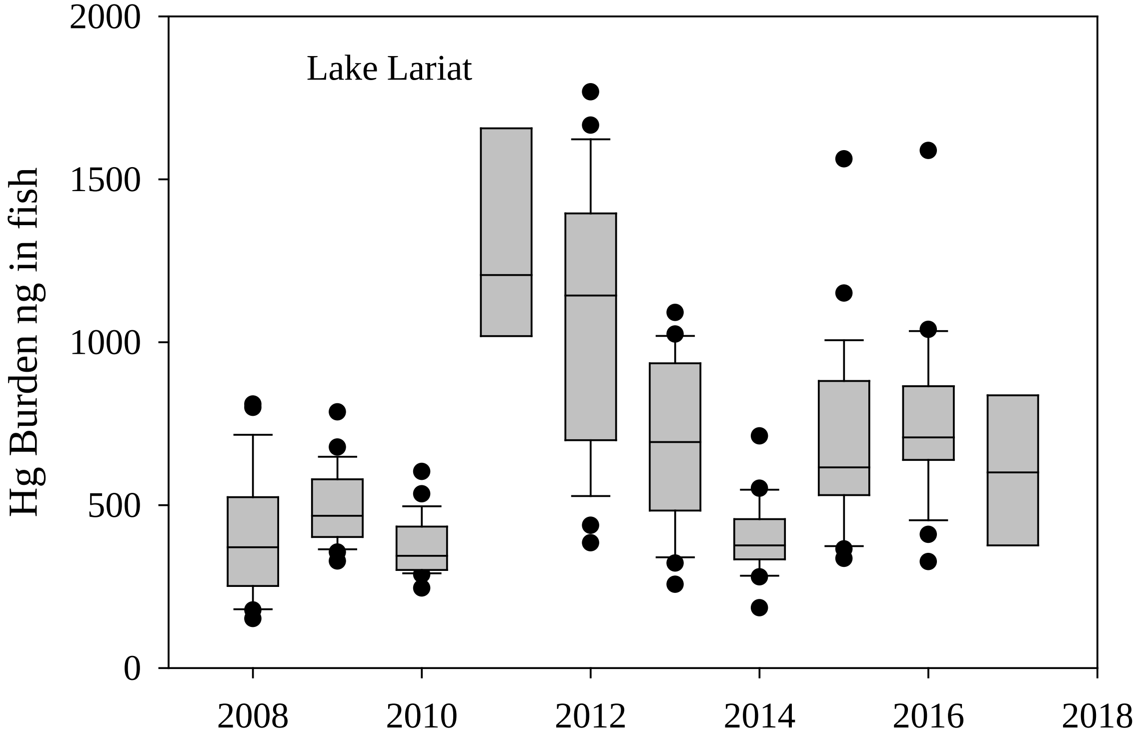


Figure 40. Box plots of Lake Lariat fish Hg burden 2008 through 2017. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers.

6.2.9 Loch Raven

The bass collected from Loch Raven in 2017 had a strong size to weight relationship. The size of the 2017 fish were the largest (6.5g) relative to other collection years (mean all years = 3.9g) (Figure 41). The average concentration of Hg in fish was the lowest measured to date, and a weak (r^2=0.1 p=0.01) but significant trend of increasing Hg concentrations over time is still apparent (Figure 41). Fish Hg concentrations appear unrelated to fish mass despite the high variability in both (Figure 42). The fish Hg burden in 2017 is greatest averaging 110 ng compared to an average of 79 ng for previous years (Figure 43).

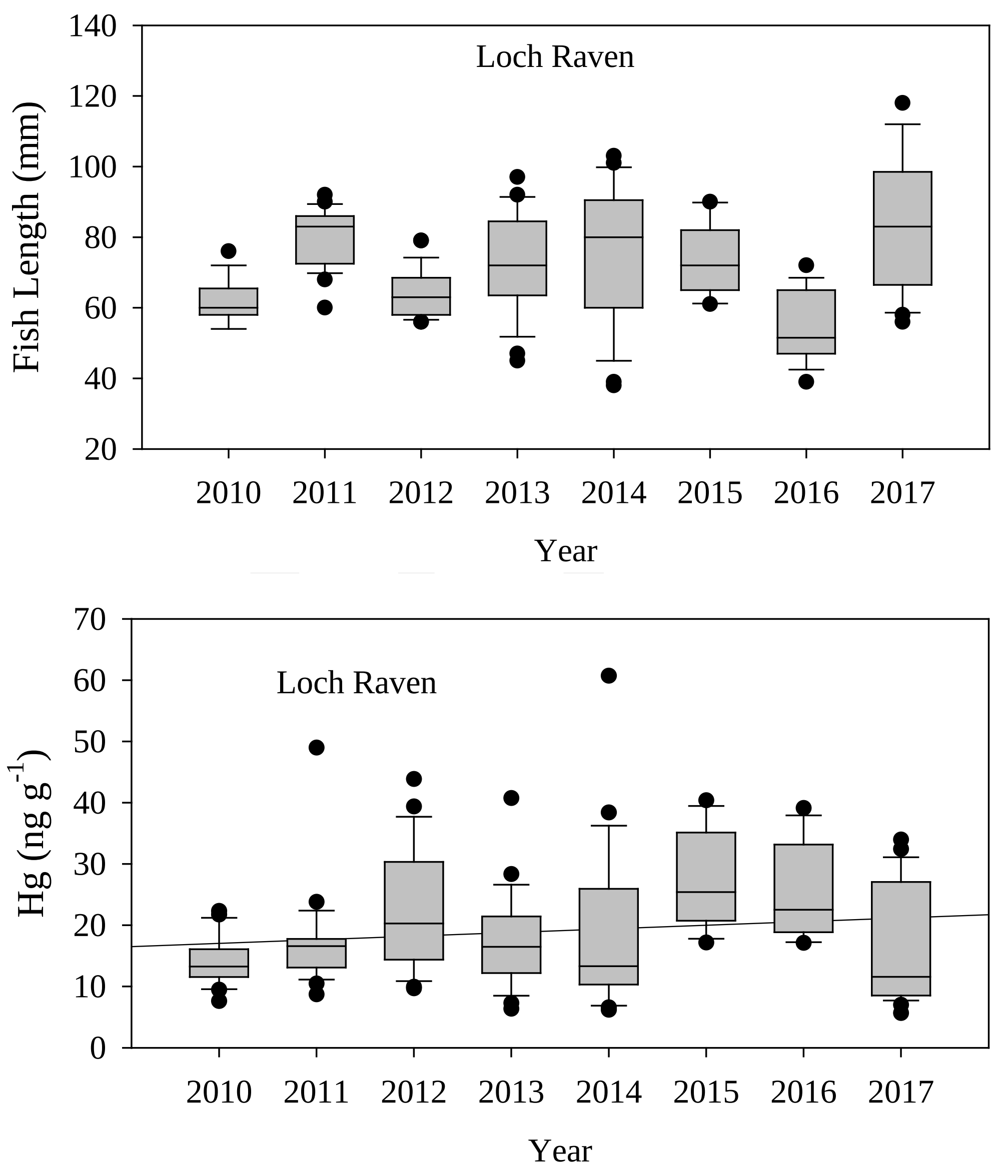


Figure 41. Loch Raven: Box plots of fish size (upper) and fish Hg concentration (lower) 2010 through 2016. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers.

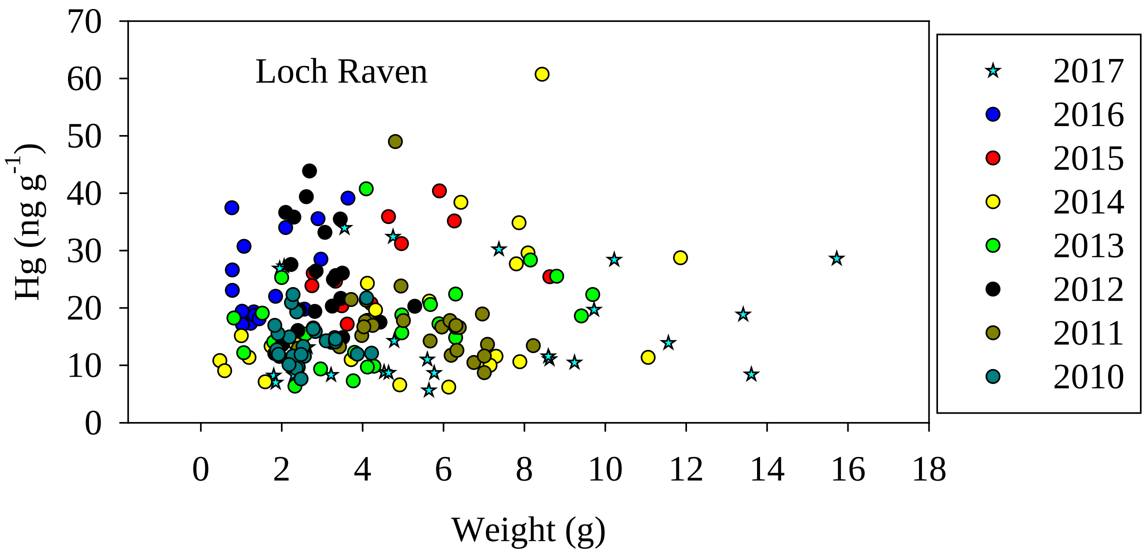


Figure 42. Fish Hg-weight relationship for individual fish collected at Loch Raven.

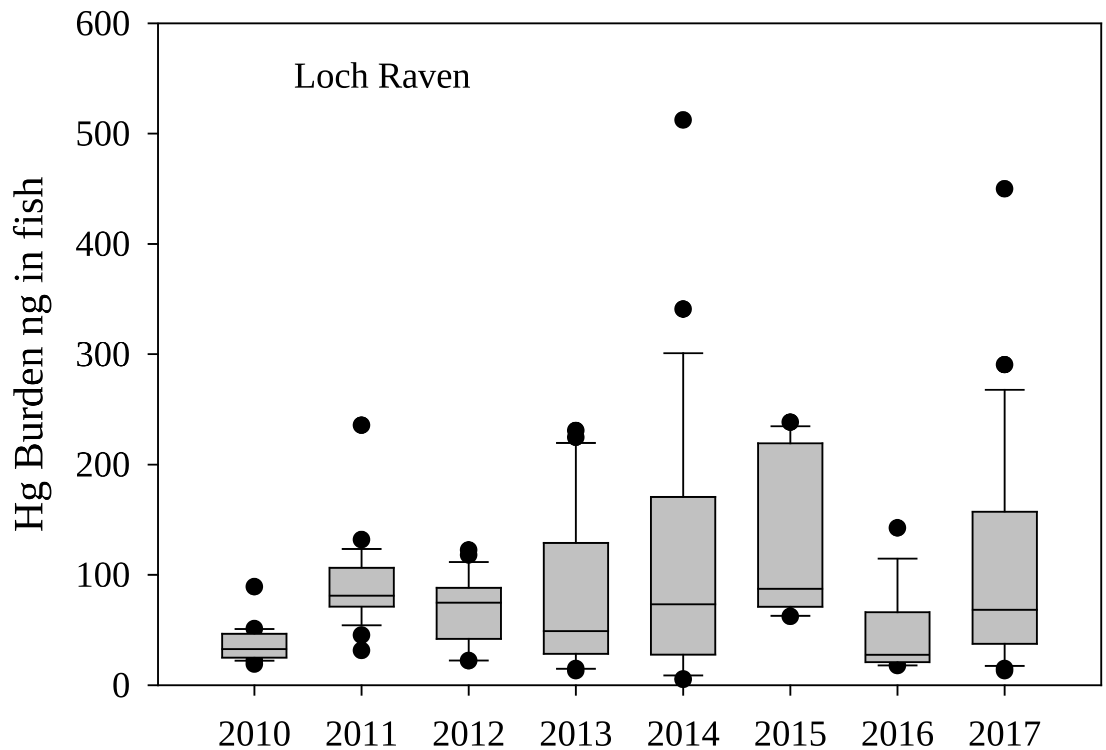


Figure 43. Box plots of Loch Raven Reservoir fish Hg burden 2008 through 2017. The boundary of the box is the 25th percentile, the whiskers indicate the 10th and 90th percentile and circles indicate outliers.

7.0 Temporal and Spatial Trends in Fish Mercury

7.1 Biodilution

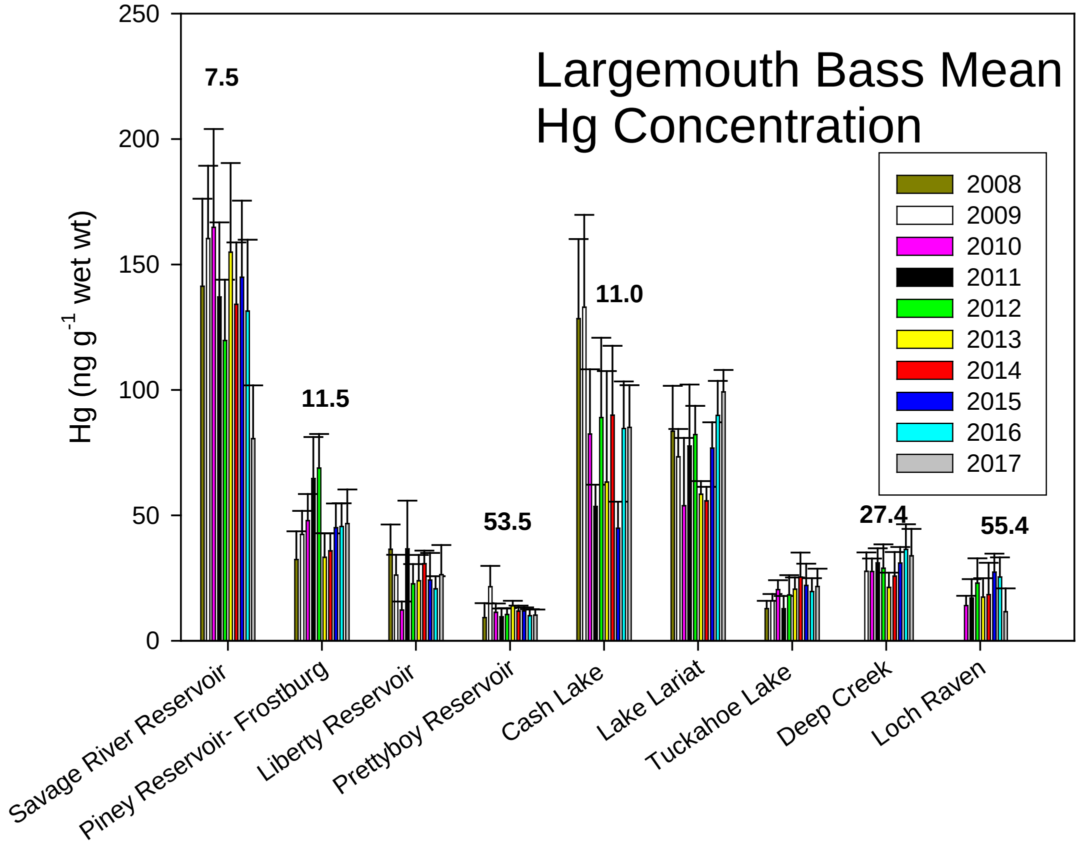
One common aspect of the results obtained in this study to date is the increase in the size of the YOY fish collected with time, despite being collected at a similar date each year. Year to year variability in size was expected but not the clear trends as seen at some sites such as Lake Lariat and Savage Reservoir. In some cases this has coincided with a decrease in the concentration of mercury in fish but no change in the fish Hg burden, such as occurred in the Savage River Reservoir. In others cases such as the Prettyboy reservoir, the picture isn't as clear. Biodilution can be caused by changes in food quality resulting in faster growth per calorie taken in. This would result in a decrease in the toxin concentration independent of any change in toxin availability (Karimi et al 2007).

7.2 Mercury selenium

In 2017, five extra YOY bass were collected when available, which resulted in six sites being examined. These fish were freeze dried and a subsample microwave digested in nitric acid in order to analyze mercury (AF) and selenium (ICP-MS) on the same extract. Because the standard digestion employs sulfuric acid, these samples cannot be analyzed by ICP-MS, the instrument used to measure selenium. From these concentrations molar ratios were calculated and these are shown on Figure 44. The concentration of Se only varies by a factor of 3 (1 to 3.5 ug/g) which some have argued as being to low to effect Hg concentrations in adult fish (Okelsrud et al 2017). However, these are young fish with both elements only recently being taken up.

The correlational impact of Se on fish Hg concentration has been well documented (Yang et al 2008). In a study of lakes with differing Hg and Se loading, Belzile et al (2006) reported a connection with lower concentrations of MeHg in response to elevated Se in fish but less of a connection lower down the food web. We have observed a direct response of Hg in frogs in controlled experiments (Rowe and Heyes 2017 and others have for shrimp (Bjerregaard and Christensen 2012). The mechanism of selenium intervention at both the cellular and ecosystem level is in question. It isn't clear if elevated Se acts to depress mercury at the level of the individual at higher levels in the food web, by depressing Hg throughout the food web, or even through a geochemical level by depressing Hg methylation. Since Hg can be effectively lowered in frogs and shrimp, it suggests an intervention in the food web is possible, although it is not known how far down the food web Se and Hg actually interact. Selenium and mercury could be ingested independently, at higher trophic levels, and generate organismal responses at any step in the food web. Regardless higher tissue selenium coincides with substantially lower mercury concentrations.

There is no geographic explanation for the Se:Hg interaction. Although Loch Raven and Prettyboy reservoirs are close together, Cash Lake is also in the same area with low Se:Hg ratios as Savage Reservoir from Western MD.



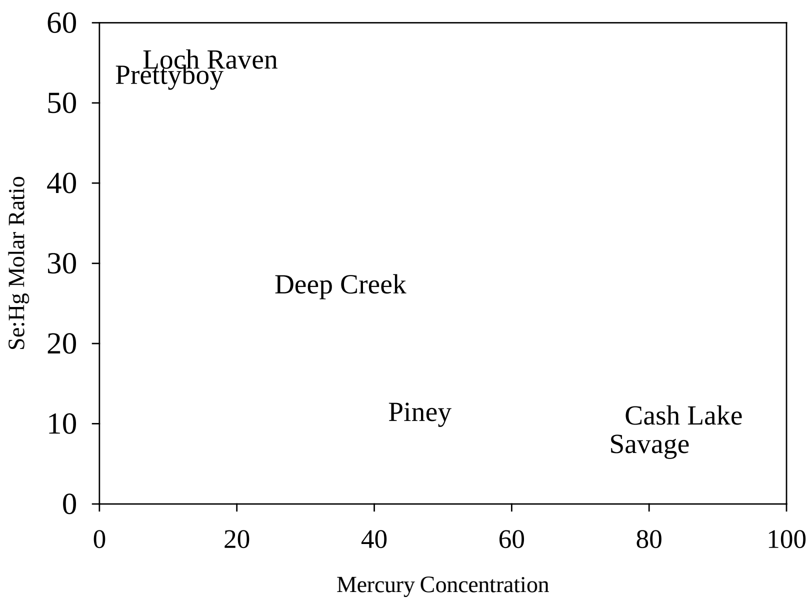


Figure 44. Average mercury concentrations and 2017 Se:Hg Molar ratios placed above mercury concentrations (upper) and Se:Hg Molar ratio against average Hg concentration in 2017 (lower).

8.0 Mercury Deposition and Fish Hg Concentrations: Summary of Findings

In studies using adult fish, Hg concentrations are often normalized to a standard fish size in order for comparisons to be made between systems. Since these fish are all young of the year one might expect a less than ideal mass to mercury relationship within a year for both white perch and largemouth bass, but the Hg concentration versus size relationships are exceedingly weak. Even in Lake Lariat, where the fish range in size from 2-20 grams, there is no relationship between fish size and fish Hg concentration. The Hg concentration in fish at any one site varies by between 20 and 30% of the mean concentration in any given year (standard deviation/mean). In laboratory studies the Hg concentration in fish is clearly reflect the Hg content in their diet however as fish age an decrease in the rate of accumulation occurs, likely driven by metabolic changes as the diet remained the same (Steffanson et al 2013). In the wild, fish diet varies and so does the Hg load per unit energy gained. For example decreases in Hg concentration and increase in food quality would have the same effect, that being to decrease fish Hg concentrations (Karimi et al 2007). Mercury burdens of the YOY fish do not show trends in general, either up or down, at any of the sites thus provide no further insight. Given the increase in YOY fish size growth dilution cannot be ruled out but without measures of diet Hg concentration and "quality", we cannot address whether food quality or food with lower Hg concentration has created the change.

Within a site, year to year variability in mercury concentrations appear to be reflected across any years fish population as a whole. Site specific activities such as lowering or raising water levels (Kelly et al 2007, KSLA 2009) may alter Hg bioavailability. Differences in Hg concentrations among sites appear consistent, and site specific aspects may contribute to Hg transfer or bioavailability. One interesting aspect is the role of selenium where by elevated selenium has been shown to correlate with lower fish Hg concentrations. This relationship has been observed before but seldom is it so clear (Petersen et al 2007). Furthermore it is not clear why this occurs, whether it is a direct interaction between Hg and Se in the gut of organisms or occurs at the cellular level within the organism. The interaction has been shown experimentally in amphibians (Rowe and Heyes 2017) and by Bjerregaard and Christensen (2012) in shrimp, thus Hg is perhaps lowered throughout the food web. There are few food web studies, and certainly none across a distinct gradient. Both Se and Hg are bioaccumulated (Økelsrud et al. 2017) but mercury has stronger factors. The food web studies do not take into account the chemical forms of both mercury and selenium, and how this might impact uptake and retention (Belzile et al 2006).

While atmospheric deposition of Hg is trending downward, changes in Hg loading are less clear across the region during the period of study. A reduction in Hg emission was reported in (Castro and Sherwell 2015) as well as changes in speciation. However, the magnitude of the reduction in Hg load that we anticipated at the start of the project has not yet materialized. Changes in fish Hg concentrations are only likely to occur post 2013, as more than one year has passed since emission reduction technologies were largely put into practice. Since to date, we have not seen a large reduction in Hg loading across the state, we can only speculate as to the reasons why such as: either local (State)

Hg emissions are less important when compared to the regional signal or regional and global sources than predicted; and/or regional and global emissions have risen to offset the benefit of local emission reductions. Given the high year to year variability in Hg loading, a continuation of monitoring is required if the reductions are to be accurately recorded and understood. We do not have the comparable data for dry deposition, which may account for a load of similar magnitude. Whether this load is equally variable is not yet known.

A tertiary geographic analysis of the current wet deposition and fish concentrations across the site yields no trends. Wet deposition in 1 year is not directly reflected in the fish of the same year or even the following year. In Cash Lake, fish Hg concentrations dropped in coincidence with the implementation of emission reduction technology in Maryland in 2010, but there is no trend in regional deposition to support this as the cause. Without more accurate site specific modeling to account for dry and wet deposition site specific cause and effects will remain undocumented. However, this particular project was designed to assess the impact of a sustained decrease in load, not year to year changes as many other pieces of information such as reservoir operational information, lake water residence time, lake temperature, degree of lake stratification would be needed to develop site specific models. Local effects are clearly important given the variations in fish Hg concentration independent of a change in load. The downward trend at some reservoir sites and the Chesapeake Bay sites suggests some water bodies may be responding. Given the Bays large surface area to watershed ratio, it may respond fastest to changes in regional deposition. It is imperative monitoring of changes in Hg deposition and its impacts on fish be continued in order to maintain data integrity through this important period of change.

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