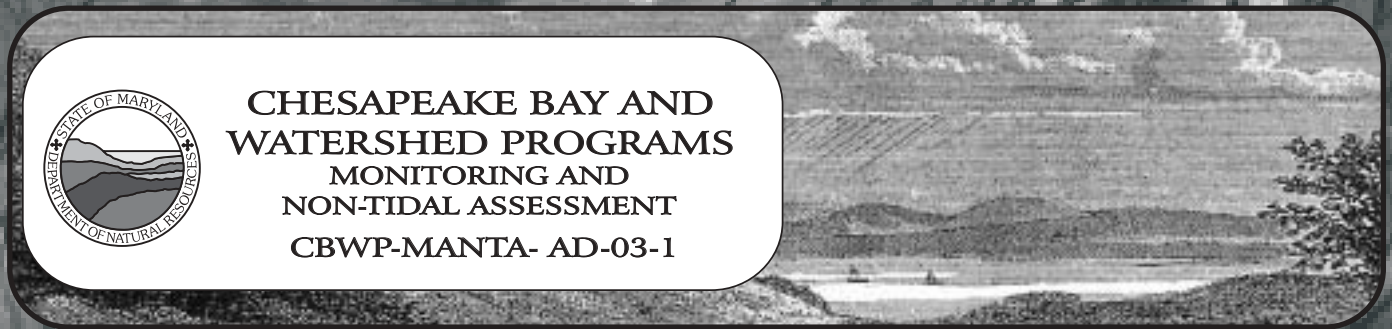
MERCURY AND METHYLMERCURY CONCENTRATIONS IN WATER AND LARGEMOUTH BASS IN MARYLAND RESERVOIRS



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Mercury and Methylmercury Concentrations in Water and Largemouth Bass in Maryland Reservoirs.

Final Report

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LIST OF ABBREVIATIONS AND ACRONYMS

CBL	University of Maryland, Chesapeake Biological Laboratory
DO	Dissolved Oxygen
DOC	Dissolved Organic Carbon
Hg	Mercury
HgT	Total Mercury
MeHg	Methylmercury
MDNR	Maryland Department of Natural Resources
PON	Particulate Nitrogen
SRM	Standard Reference Material
TSS	Total Suspended Solids
USEPA	United States Environmental Protection Agency
USFDA	United States Food and Drug Administration
POC	Particulate Carbon
WHO	World Health Organization

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EXECUTIVE SUMMARY

Concerns for human and ecosystem health have provided the basis for increased attention on studying mercury (Hg) in the environment. Public health warnings and guidelines for consumption of fish with elevated levels of methylmercury (MeHg) have been issued by many organizations. However, until recently (12/2001) there were no Hg based fish consumption advisories posted in Maryland even though atmospheric deposition of Hg in Maryland is higher than in most other regions of the USA. Data from this study contributed to the advisory issuance. The sources of Hg to the aquatic systems are both natural and anthropogenic. However, the most important source of MeHg is not external but is in situ production within aquatic systems by sulfate-reducing bacteria. The amount of MeHg varies among ecosystems and is not directly related to the amount of atmospheric Hg deposition, as the ability of the ecosystem to convert that Hg to MeHg, and for that MeHg to bioaccumulate, depends on many physicochemical variables. Given the above, the project was formulated to determine the concentrations of total Hg and MeHg in muscle tissue of largemouth bass, a representative top predator fish in Maryland reservoirs, and how these concentrations vary with fish size. In addition the study was designed to probe between-lake variability in fish concentration and investigate potential influential factors controlling MeHg in fish. The results indicate that, while fish concentration increased with size, there was substantial variability in the rate of increase between reservoirs. Many chemical factors, such as water Hg and MeHg concentration, pH, dissolved organic carbon, nutrient and major anion concentrations were investigated, as well as physical factors such as reservoir location, shape and morphology and watershed characteristics. Biological factors studied included size and growth rate of fish and food preferences. The results of this initial limited study indicate that there is not one specific variable that controls reservoir fish MeHg concentration and that many, potentially interlinked factors, are important in understanding the level of MeHg in fish. The most important factors were water MeHg and sulfate concentration and lake morphology. More studies are needed to further investigate the conclusions of this study, and to investigate other potentially important factors.

CHAPTER 1: INTRODUCTION

1.1 Background

In recent years, concerns for human and ecosystem health have provided a basis for an increased attention to studying mercury (Hg) in the environment. Hg occurs naturally in a variety of inorganic and organic compounds and not only in solid or dissolved states, but also in liquid and gas phases (Meili, 1994). The sources of Hg to the atmosphere are both natural and anthropogenic. Although some Hg is emitted to the atmosphere from natural sources, such as from volcanic eruptions, forest fires, biogenic emissions, degassing from water surfaces and wind entrainment of dust particles (Rasmussen, 1994), the anthropogenic emissions of Hg, mostly from coal combustion, municipal and medial waste incineration and smelting (Lindquist et al., 1991), exceed the inputs from the natural sources at least three-fold (Mason et al., 1994) with higher levels in developed areas and around point sources (Mason et al., 1997a). Present concentrations of Hg are elevated as a result of enhanced anthropogenic activities (Meili, 1994). Population growth and urbanization have contributed to significantly elevated levels of Hg in the atmosphere and it has been estimated that Hg derived from anthropogenic activities in the atmosphere is up to 80% of the total Hg in the atmosphere (Mason et al., 1994).

The enhanced atmospheric deposition of Hg is often the dominant source of Hg to aquatic systems (Hakanson et al., 1988; Rolfhus and Fitzgerald, 1995). Hg has been found to possess high toxicity to aquatic organisms (Mason et al., 1996) and in its organic form, methylmercury (MeHg), has a large capacity for biomagnification along food webs either through uptake from water or diet (Rodgers and Beamish, 1983). Even though most of the Hg in freshwater environments consists of inorganic Hg, almost all of the Hg found in fish is MeHg (Choi et al., 1990). Sources of MeHg to the aquatic system have been identified to be from precipitation, in-lake methylation and runoff from wetlands (Watras et al., 1995b; Rudd, 1995).

It has been estimated that sediment is an important sink for both Hg and MeHg in the aquatic environment (Mason et al., 1999) and after atmospheric deposition and runoff from surrounding catchments, Hg can be converted to MeHg from in situ production by natural bacteria in anoxic sediments and soils (Gilmour et al., 1992). The amount of MeHg in aquatic regions varies among ecosystems, as does atmospheric Hg deposition. Therefore, MeHg bioaccumulation in fish not only depends on how much Hg enters the ecosystem, but also on the ability of the ecosystem to convert that Hg to MeHg (Heyes and Gilmour, 1999). For example, methylation of Hg has been found to be enhanced in wetlands but can be produced in other anoxic regions as well. Increased runoff from highly urbanized areas and as the result of impervious surfaces in and around the watershed may contribute to higher than normal concentrations of Hg and MeHg into aquatic systems.

There are at least two possible pathways for the accumulation of MeHg in fish (Downs et al., 1998). MeHg can be directly absorbed through the gills and/or skin of animals, or

indirectly via the food chain. Whereas MeHg has high affinity for particles and organic matter, the extent to which sediment is a source of MeHg to the fish largely depends on the size of particles and organic matter content of the sediment (Benoit et al., 1998; Mason, 2001). Absorption of ionic species through gills is a potential route of entry (Xun et al., 1987), although ingestion of food does seem to be the primary route, with 85% to 90% of MeHg in fish and benthic invertebrates coming from food sources (Lawrence and Mason, 2001; Rodgers, 1994; Mason, 2001).

Once MeHg has been taken up by organisms low in the food chain (such as phytoplankton and zooplankton), it is efficiently accumulated and transferred to organisms higher in the food chain (Mason, 2001). Accumulation of MeHg by fish is of concern since consumption of MeHg-contaminated fish is the major route for transfer of mercury from the aquatic environment to fish-eating birds and mammals, including humans (Rodgers, 1994). Exposure to high levels of MeHg has been found to cause neurological damage, as well as fatalities, among adults. Prenatal life and small children are even more susceptible to brain damage due to their enhanced sensitivity to the neurotoxin (Weiss et al., 1999).

The burden of MeHg in fish is suspected to depend on many physicochemical variables of the watershed environment and water-column. Water chemistry is likely to be an important factor controlling bioaccumulation rate and the concentration of MeHg in fish at any one time. Hg and MeHg concentrations, water temperature, dissolved oxygen (DO), dissolved organic carbon (DOC), pH, total suspended solids (TSS), particulate organic carbon (POC), particulate organic nitrogen (PON), and sulfate concentrations are among chemical variables that might influence both methylation of Hg and the uptake and accumulation of MeHg by fish from sediment and water-column. DOC and pH are probably the most important chemical variables for MeHg accumulation by fish (Mason, 2000), although many other chemical parameters that influence Hg speciation in the water column and ultimately affect the bioavailability of MeHg to biota, could also be contributing factors. Physical parameters of watershed-surroundings can influence the amount of MeHg in the aquatic environment but are unlikely to have direct impact on bioaccumulation of MeHg in aquatic organisms.

Public health warnings and guidelines for consumption of fish with elevated levels of MeHg have been issued by the World Health Organization (WHO), in parts of Scandinavia and Canada, by the U.S. Food and Drug Administration (USFDA), the US Environmental Protection Agency (USEPA) and numerous other agencies and governments. Until recently, USFDA regulations stated that fish containing less than 1ppm MeHg is safe for human consumption (USFDA, 2002). In March 2001, USEPA and USFDA posted a consumer advisory about the risk of mercury in fish, advising pregnant women, and women of childbearing age who may become pregnant, and young children not to eat large fish that accumulate the highest levels of MeHg (USEPA and USFDA, 2002). In the U.S. there are more fish consumption advisories posted for Hg than of any other contaminant. However, until recently there were no Hg based fish consumption advisories posted in Maryland even though atmospheric deposition of Hg in Maryland is higher than in most other regions of the

U.S. (Mason et al., 1997a). In December 2001, the state of Maryland released an advisory based on the preliminary results from this study and others that showed that Hg concentrations in large piscivorous fish from the state's freshwater lakes exceeded a common advisory level of 0.3 mg MeHg/kg fish, and in some cases exceeded the USFDA action level of 1 mg MeHg/kg fish tissue (Gilmour, 1999; Sveinsdottir, 2002; this study).

Mercury (Hg) contamination in freshwater fish stocks has been recognized as a problem in Europe and North America for over three decades. Hg concentration in fish is of heightened concern, because consumption of fish is the largest source of mercury in human diet (WHO, 1990) and that of fish-eating wildlife. Atmospheric transport and deposition appear to be the major source of inorganic mercury to remote lakes. Sources of methylmercury (MeHg) to the aquatic system have been identified to be from precipitation, in-lake methylation and runoff from wetlands (Rudd, 1995) but, in most instances, in-situ methylation is the dominant source (Benoit et al., 2002). MeHg differs from the inorganic form in that it is more toxic, more mobile, more readily bioaccumulated by aquatic organisms and it accounts for 95 – 99% of the total mercury found in the muscle tissue of higher trophic level freshwater fish (Grieb et al., 1990; Bloom 1992).

1.2 Objectives

In light of the discussion above, the project described here was formulated under the following objectives:

1) Determine the concentrations of total Hg and MeHg in muscle tissue of top predator fish in Maryland reservoirs, and how these concentrations vary with fish size;

2) Determine the relationships that control between-lake variability in fish concentrations for fish of the same size;

3) Determine the concentration of Hg and MeHg in representative food organisms and in the water in conjunction with measurement of water quality parameters and reservoir characteristics, so that the principal factors influencing bioaccumulation can be elucidated.

To achieve these goals and provide the required information for regulatory decision, it was desirable to collect the same fish species from all reservoirs. The target species was largemouth bass of a size above the legal limit so that the fish analyzed is only those that might be consumed by humans. Further, as it was not possible to obtain the same fish species from all reservoirs, two fish species were collected from each lake so that there was overlap at a two-species level. In this report, the focus is on the largemouth bass as this species was found in all except one reservoir. However, data for the other fish species collected, crappie and bluegill, from each water body are given in the appendix. To simplify the between-lake comparisons, and to allow comparison with fish from other locations and states, and to ensure

that a range of fish were caught, three size classes were chosen for each species, and five fish from each size class were sought to the extent possible in each reservoir.

To achieve objectives 2 and 3, water quality parameters and total Hg and MeHg in water were determined on all reservoirs, and in the inlet and outflow waters. Also, the concentrations of Hg and MeHg in the main food species of the fish were measured on a subset of the reservoirs to see if there were similar bioaccumulation factors across systems between food and top predators. Stomach content analysis was used to ascertain, in a qualitative way, what the primary food for the bass was for each water body. Differences in food concentrations could be related to differences in water chemistry and trophic status, which will allow an investigation of the extent of food type or environmental conditions in controlling fish concentration.

Overall, these studies were designed to provide the necessary information for evaluation of the extent of the Hg problem in MD. Clearly, while the information collected was not sufficient for a comprehensive and exhaustive analysis, the data can be used by managers to set regulations, if necessary. In addition, the study has provided further information to allow the continual refinement of our understanding of the factors regulating Hg and MeHg fate and transport in the environment, and of the propensity for fish concentrations to either increase, decrease or remain the same in the future. This study thus forms a starting point and building block for future endeavors to understand the factors controlling MeHg concentration in Maryland fish.

CHAPTER 2: MATERIAL AND METHODS

2.1 Study Sites

The 20 reservoirs used in this study are all located in the state of Maryland (Fig. 2.1; Table 2.1 & 2.2). Largemouth bass are found in all waters of Maryland, from freshwater to brackish water. They are fish-eating predators as adults, although their diets also include invertebrates and amphibians. As they are also important recreational sport fish, they were the target species for this study. The reservoirs sampled range widely in depth (2.62 m – 54.5 m), volume ($3.2*10^4$– $3.8*10^8$ m^3) and in surface area (50 – 8960 acres) and all have inflow from rivers and outflow controlled by a dam (Table 2.1). Such a large variation in reservoir size makes intercomparisons somewhat difficult without some normalization. They vary substantially in shape and this affects the dynamics of Hg methylation and bioaccumulation, as methylation is mostly confined to sediments, and bioaccumulation depends on diet, which is influenced by reservoir shape. Also shape determines the degree of stratification and oxygen depletion during summer.

If the water bodies were perfectly cubic in shape, then the ratio of volume (V)/surface area (A) would be equivalent to depth (D). We therefore developed the parameter D/(V/A) or D*A/V (actual depth/hypothetical depth) as a measure of the “shape” of the reservoir, using

the data in Table 2.1. A “cubic-shaped” water body would have a value of 1. Most had similar values for this parameter, which generally ranged from 2-3, representing the more cone shaped morphology of lakes in general. Johnson’s Pond, Big Pool and Potomac #4 had values close to 1. Three lakes, Clopper, Piney Run and St. Mary’s Lake had values greater than 10 indicating that these are steep systems, although St. Mary’s is not deep like the other two lakes (50-60 m). Piney (Frostburg) had a value of 4.3. Thus, for these unusual systems, morphology may be a contributing factor to differences in MeHg in fish in these reservoirs compared to the others, as the extent of methylation is related to stratification and sediment area/water volume. It is likely that the deep, steeply sided lakes will stratify more quickly and that the stratification would persist longer than the more shallow lakes and this may increase the methylation potential of the system. However, shallow lakes have a higher surface area/volume ratio and this affects the dynamics of Hg methylation as methylation is mostly confined to sediments. Furthermore, bioaccumulation depends on diet, which is influenced by reservoir shape.

Another parameter that was considered important was sediment area/water volume as the higher this value, the more important in-situ methylation will be and the more likely benthic organisms will be part of the fish diet. Based on our qualitative survey, bass appear to prefer crayfish to other food. While it was not possible to determine sediment area/volume directly from the information available, one can conclude, overall, considering the various geometries that exist for aquatic systems, that this parameter is maximized in shallow systems. However, shallow systems do not stratify in summer and oxygen depletion is less likely and this would mitigate the effects of large surface area. Overall, therefore, relatively shallow lakes that stratify are the most likely candidates for high in-situ methylation. These include Broadford Lake, Piney (Frostburg), Tridelphia, St. Mary’s, Lake Lariat, and Centennial Lake. Johnson’s Pond and Tuckahoe are very shallow and are also likely candidates for higher methylation. While Big Pool and Potomac #4 are shallow, these are impoundments on a large river (Potomac) and are highly dynamic, with a rocky bottom and without substantial sediment.

Watershed characteristics of the reservoirs can be important, especially factors such as the extent of wetlands surrounding the reservoir, or in the watershed, as wetlands have been shown to be important regions for mercury methylation. However, given the location of these reservoirs, and their large size compared, for example, to the seepage lakes in Wisconsin (Watras et al., 1995b), the effect of wetlands in terms of overall methylation is of less importance. For the reservoirs where the information was available, wetlands were a small percentage of the watershed area, typically less than 1% (MDNR data). In addition, most of the watersheds are forested or rural, with little commercial or urban influence nearby (<20%), given that many of the reservoirs are used for water supply. In general, the reservoirs in western Maryland are largely forested, while the two reservoirs on the eastern shore (Tuckahoe and Johnson’s Pond), and some of the reservoirs within the middle region of Maryland (such as Liberty, Prettyboy and Loch Raven) have more agricultural activity in the watershed (>40%).

Other potential factors influencing MeHg levels in fish are the watershed area/lake area as this indirectly controls Hg inputs, and has been shown to be related to water column concentration (Mason and Sullivan, 1997). As Hg is strongly bound to particulate, only a small fraction of the Hg deposited (typically, <20%) to a watershed is exported (Lawson et al., 2001; Lawson and Mason, 2001; Hurley et al., 1995; 1996). However, for systems with a large watershed/lake area, the contribution of runoff relative to direct deposition is important and thus these systems may have higher MeHg in fish as a result of higher input. All these factors are considered in this study to be important in terms of determining fish concentration.

In total, 249 largemouth bass (*Micropterus salmoides*) were collected from 20 reservoirs in Maryland during late spring and summer of 2000, 2001 and 2002. The reservoirs sampled are listed in Table 2.2. Information about number of fish analyzed, their mean length and weight, are detailed in Appendix I. Fish were collected in three size classes and the minimum size was the minimum size defined for recreational fishing. This approach was chosen to ensure that the fish collection obtained fish over a range in sizes by predetermining that 5 fish should be collected in each size class. It was not possible in all cases to obtain fish in each size class, as shown in Appendix I. In terms of results and discussion, the size classes were not considered, as this approach was only devised as a suitable sampling strategy.

The 20 reservoirs are located in the four main regions of Maryland: Eastern Maryland, Central Maryland, Southern Maryland and Western Maryland. The reservoirs were chosen for study by MD DNR personnel. Most of the reservoirs serve as a water source for local communities in addition to being used for recreational activities and flood control. The reservoirs range in age from 133 to 12 years old (Table 2.1). With the exception of three reservoirs (Piney Run, built in 1990, Centennial Lake, built in 1986 and Big Pool, which is a natural lake) it can be assumed that the physicochemical characteristics of the reservoirs resemble that of a natural lake, as the so called “reservoir effect” (i.e. a dramatic increase of MeHg in all compartments in concert with the flooding of the reservoir) typically lasts for 20-25 years (Rosenberg et al., 1987). However six reservoirs (Rocky Gap, Duckett, Loch Raven and Piney (Frostburg), Potomac #4 and Tridelphia) were modified in the last 20 years (1988, 1986 , 1986, 1990, 1994 and 1999 respectively), which might have influenced not only the physical characteristics of the reservoirs, but also perhaps the chemistry of the impoundments. The impact of reservoir age on fish concentration is well known, and in this study, such an impact was considered, as is discussed below.

2.2 Fish Collection and Analysis

Fish were collected by electroshock techniques by MDNR personnel in early spring/summer 2000, 2001 and 2002. Largemouth bass were collected at each reservoir, ranging in size from 276 to 563 mm in length, and weighing from 224.1 to 2688.0 grams. While there was some concern given the fact that fish were collected over multiple years and both early and late in the season, it was felt that the impact on fish concentration would not be significant for the larger fish. Indeed, in a study in western MD, Mason et al. (2000) found no significant change in MeHg concentration in fish collected in three different seasons. In the

field, all fish were handled by gloved personnel, and after rinsing and measurements, each fish was bagged in a plastic Ziploc bag. In an attempt to keep the fish as cold as possible, each fish was wrapped in aluminum foil and then bagged in a second Ziploc bag. Fish were kept on ice and shipped overnight to The Chesapeake Biological Laboratory where the weight of each fish was measured. Once in the laboratory, fish were filleted and muscle tissue from both sides of each individual fish (representing the portion of fish normally consumed by humans) was removed and homogenized in blender in a non-contaminating environment and stored and frozen in a Ziploc bag until further analysis. All sampling equipment, such as stainless steel knifes and food processors were acid cleaned prior to use and in between deployments.

In an effort to assess food effects, in the first two years of the project,106 largemouth bass were sampled for gut content. Only half contained food remnants in the gut. Of these, 70% was identified to be crayfish, 17% sunfish and less than 1% channel catfish.

To assess the importance of differences in diet, crayfish and small forage fish were collected from these reservoirs, where possible (Appendix III). The small fish were collected during the electro-shocking for the larger fish while the crayfish were collected by hand from the shallow, rocky portions of the reservoirs, where possible. This was only done in the latter part of the second year of study and only 8 lakes were sampled. The average length of crayfish collected was 38.3 ± 51.9 mm (ranging from 7.04 to 138.15 mm) and average MeHg concentration was 22.1 ± 13.5 ng/g wet weight (ranging from 7.24 – 44.58 ng/g wet weight). Average length of forage fish (blue gill, black crappie, golden shiner, yellow perch, white sucker and pumpkin seed) was found to be 59.5 ± 72.0 mm (ranging from 3.23 – 216.0).

For MeHg analysis of the fish approximately 1 g of sub-sampled axial muscle was placed in a Teflon ® vial. Samples were digested in an alkaline digest (Bloom, 1989) prior to derivitization with sodium tetraethylborate to convert nonvolatile MeHg to gaseous MeHg (Bloom, 1989). The volatile adduct was then purged from solution and collected onto a graphitic carbon trap. The MeHg was then thermally desorbed from the trap and analyzed by isothermal gas chromatography separation with CVAFS.

Fish were also analyzed for total Hg. Approximately 0.2–0.4 g of fish was sub-sampled from the axial muscle and placed in a Teflon ® vial. Each fish sample was digested in 70% sulfuric acid/ 30% nitric acid solution overnight, to ensure complete digestion of organic matter (Bloom and Crecelius, 1983) in a VWR Scientific forced air oven at 60^{o}C. HgT samples were oxidized using bromine monochloride (BrCl) and let sit for at least ½ hour. The excess oxidant was neutralized with 10% hydroxylamine hydrochloride, then reduced with $SnCl_2$ solution, and lastly purged with argon to remove elemental Hg to a gold trap. The amount of Hg on the gold trap was then determined by cold vapor atomic fluorescence detection (CVAFS) (Bloom, 1989) in accordance with protocols outlined in EPA Method 1631 (USEPA, 1995; Bloom and Fitzgerald, 1988).

In this report the MeHg data are only discussed, for two reasons. Firstly, as MeHg is the form of Hg of concern from a health perspective, this information is more accurate and valuable than HgT, even if essentially all the Hg in these fish is MeHg. Secondly, we have found that, despite the general contention by some that HgT analyses are more accurate, in our laboratory the analysis for MeHg in fish is consistently more accurate and precise that the HgT determination. This is because the MeHg method measures a particular form of Hg and is therefore less prone to random contamination. Further, for fish analyzed by alkaline digestion there is little difference in the amount of preparation and analysis time. For the first year samples, the average % MeHg in fish was 103 ± 53%. With the outlier samples removed (>140% or <60% MeHg) the fraction was 91 ± 28%. We reanalyzed all fish in which the ratio of MeHg was greater than 140% or less than 60% and found in the majority of cases that it was the HgT measurement and not the MeHg measurement that was inaccurate. For this reason, we used the MeHg data in this report. The HgT measurements were made purely as internal QA check.

The following QA/QC was performed. Laboratory blanks and duplicates were part of all analytical runs, as were matrix spikes and analysis of standard reference materials (Table 2.3). Blanks were typically a small fraction of the sample concentration. An analysis of the concentration of MeHg in one fish sample was done eleven times during a 17 month period. Results from this analysis show that MeHg concentrations in the fish sample remained stable over the course of the period and concentration did not decrease with time (ave=20.5 ± 4.7 ng/g wet weight, slope not significantly different from 0). Analysis of SRM's typically yielded a value within the certified variance.

2.3 Water Sampling and Analysis

Water sampling was conducted during mid-summer 2001 and late spring/early summer 2002 and 2003 in an attempt to sample thermally stratified water when the degree of eutrophy might be the strongest. The sampling was done in accordance with EPA Method 1669 (USEPA, 1998). Surface samples were collected by hand, and the deeper samples were obtained with a GOFLO ® apparatus that had been acid cleaned and rinsed with Q-water prior to sampling. At each sampling location, the GOFLO ® bottle was submerged in the water and rinsed for 15 minutes. Samples within each reservoir were either obtained from surface waters near banks for the shallow reservoirs, or at the location of the deepest depth of the reservoir where samples were taken from three depths. For most reservoirs, water samples were collected by boat away from the shore. Where possible, and for the deeper reservoirs only, one sample was obtained from bottom-water, one from mid-water and one from surface-water. It is well known that the concentration of Hg and specifically MeHg can change seasonally and that the collection of one set of samples may not be adequate to accurately assess the variability on a seasonal basis. However, the budget for the project was limited and this did not allow more than one collection.

Clean double-bagged 2L Teflon bottles, partially filled with dilute trace metal grade HCl, were used for water collection. Prior to sampling, each bottle was emptied of the HCl downstream from the sampling location. Next, the bottle was rinsed three times with reservoir water and, after being filled with sample-water, the bottle was recapped, double-bagged and stored in a cooler for transport back to the laboratory.

Approximately 1 L of sample from each bottle was filtered for Hg and MeHg onto quartz fiber filters (pre-cleaned by combustion in a furnace overnight at 600° C) using acid-cleaned glassware and other filtering equipment. All equipment used for filtering was acid washed between samples and rinsed with Q-water. Filtered and unfiltered water was stored in acid-cleaned Teflon bottles and both water samples were spiked with Optima HCl acid and stored in a refrigerator until analyses were performed.

For MeHg analysis, water samples were distilled with additions of a 50% sulfuric acid solution and a 20% potassium chloride solution (Horvat et al., 1993). The MeHg in the distillate was derivitized with sodium tetraethylborate to convert it to volatile methyl-ethyl-mercury (Bloom, 1989). The volatile adduct was then purged from solution and collected onto a graphitic carbon trap. The MeHg was then thermally desorbed from the trap and analyzed by isothermal gas chromatography separation with CVAFS.

Total Hg was measured in water samples after bromine monochloride (BrCl) oxidation of samples (0.5 mL of 2N BrCl added; Bloom and Crecelius, 1983) and pre-reduction with hydroxalamine. The samples were then reduced with tin chloride solution, purged to remove elemental Hg to a gold trap, and the amount of Hg in samples then determined by two-stage gold amalgamation CVAFS (Bloom and Fitzgerald, 1988).

Standard calibration curves with an r^2 of at least 0.99 were run daily and a standard addition spike was added to one in every 20 samples to check for matrix interferences. Laboratory duplicates and external certified standard reference materials (SRM) (digestates of IAEA SRM 142) of known HgT and MeHg concentrations were analyzed daily to ensure the accuracy of results. QA/QC data are given in Table 2.3. Duplicate analysis of 10% of the MeHg samples yielded no significant difference and 95% of all SRM replicates analyzed for HgT and MeHg fell within the certified ranges. Detection limits for HgT and MeHg were based on three standard deviations of sample blank measurements. Aqueous detection limits for MeHg were 0.01 ng/L for water and 0.11 ng/g for fish. Field and travel blanks were typically less than the detection limits.

Not all reservoirs' water were sampled as part of this study. For Conowingo Dam, water samples were not collected as samples were collected by our group over a period of a year as part of a study of Hg biogeochemistry in rivers flowing into the Chesapeake Bay (Lawson et al., 2001). The collection and analytical techniques were comparable with this study. No water samples were taken from Rocky Gap and information for total Hg, collected

by Castro et al. (2002) using similar techniques, is used in the comparison of data discussed below. Big Pool and Potomac #4 are on the Potomac River in relatively close proximity to each other and only one set of water samples was collected for these sites. Fish from Centennial Lake were only collected in the latter part of the project and because of season, no water samples were collected from this reservoir.

2.4 Ancillary Sampling and Analysis

The pH of the water was measured at the laboratory using standard techniques. The pH meter was calibrated using buffered solutions on each occasion. Particulate organic carbon (POC), nitrogen (PON) and total suspended solids (TSS) were sub-sampled from an unfiltered water sample and collected onto a glass fiber filter (GF/F 0.7 μm). DOC, dissolved nutrients, chloride and sulfate were sub-sampled from filtered water and stored frozen until subsequent analysis. All ancillary parameters were measured by CBL Analytical Services. DOC was measured using a Shimadzu TOC-5000 Total Organic Carbon Analyzer after high temperature combustion (680^{o}C) in the presence of a platinum catalyst (Sugimura and Suzuki, 1988). Samples for POC and PON were combusted in pure oxygen under static conditions using the Exeter Analytical, Inc (EAI) CE-440 Elemental Analyzer. TSS were determined by drying the pre-weighed filters and re-weighing, according to APHA (1975) Method No. 280 D and USEPA (1979) Method No. 160.2. Sulfate, chloride and nitrate were analyzed with a Rainin Co. Inc./Dionex Hybrid Ion Chromotography System according to method USEPA (1987) section 11.0 and test method USEPA No. 300.0 as described in Pfaff et al., 1991. Determination of ammonium was done with a Technicon TrAAcs-800 Nutrient Analyzer according to Technicon Industrial Method No. 804-86T. Alkalinity and cations were not measured as part of this study as previous work had suggested that correlations of Hg and MeHg were primarily with those of sulfate, as a result of the influence of sulfate on bacterial activity. The role of nutrients in controlling reservoir productivity was considered important and therefore these parameters were measured.

2.5 Statistical Analysis

Pearson correlation analysis was used to determine p-values between length and weight of fish and fish concentration. Concentration in largemouth bass was correlated with various chemical parameters of reservoirs using SAS multiple regression analysis and Pearson correlation analysis. Fish mercury concentration was normalized to the length of fish, using a range of 362 to 377 mm, to remove variability based on the fish size for the regression analysis. For reservoirs with three or more fish in this size range, the average concentration was used. For reservoirs without three fish in this size, the linear regressions between fish size and concentration for all fish were used to estimate the average fish concentration for a fish of median size within this range. For three reservoirs (Big Pool, Potomac #4 and Centennial Lake), there was insufficient information to determine a normalized concentration and these were emitted from the analysis.

Table 2.1. Physical characteristics of the reservoirs. a = depth of water (m) impounded at the normal operating pool elevation, b= area (acres) of the lake surface at the normal operating pool elevation, c- volume of water (m3) stored below the normal operating pool elevation. Reference: Maryland Power Plant Research Program (PPRP) Inventory of Maryland Dams, CD-ROM, Version 1.2.

Reservoir	**Use**	**Year Completed/ modified**	**Normal Depth (m)[a]**	**Surface Area (acres)[b]**	**Normal Capacity (m^3)[c]**
Deep Creek Lake	Hydro/Recreational	1925	15.2	4500	1.1E+08
Broadford Lake	Recreational	1937	4.4	20	1.5E+05
Savage	W Supply/Rec./Other	1952	46.1	360	2.5E+07
Piney (Frostburg)	W Supply	1934/1990	16.6	110	1.7E+06
Rocky Gap	W Supply/ Recreational	1969/1988	25.0	209	6.6E+06
Big Pool	Recreational	n/a	1.5	47	2.9E+05
Potomac #4	Hydro/ Recreational	1869/1994	2.8	675	9.0E+06
Clopper	Rec/Flood Control	1975	53.0	350	2.0E+06
Tridelphia	W supply/Hydro/Rec.	1943/1999	15.8	800	2.3E+07
Piney Run	W supply	1990	54.5	298	7.4E+06
Duckett	W supp/Recreational	1953/1986	22.6	773	2.1E+07
Liberty	W supply/Recreational	1953	40.5	3106	1.6E+08
St. Mary's Lake	Rec/Flood Control/Other	1975	6.4	250	3.9E+05
Prettyboy	W supply/ Recreational	1936/1936	30.0	1500	7.4E+07
Lake Lariat	Recreational	1965	9.1	97	1.9E+06
Centennial Lake	Rec./Flood Control	1985	7.3	50	6.3E+05
Loch Raven	W supply/ Recreational	1923/86	23.2	2400	9.0E+07
Conowingo	W supply/Hydro./Rec.	1928/1983	30.0	8960	3.8E+08
Tuckahoe	Recreational	1975	2.7	86	3.2E+04
Johnson’s Pond	Recreational	1936	2.620	104.0	1.1E+06

Table 2.2. Names and location of the reservoirs sampled in the study.

Reservoir	County	Longitude	Latitude
Deep Creek Lake	Garrett County	79º39'	39º30'
Broadford Lake	Garrett County	79º22'	39º24'
Savage	Garrett County	79º08'	39º30'
Piney (Frostburg)	Garrett County	79º01'	39º35'
Rocky Gap	Allegany County	78º39'	39º42'
Big Pool	Washington	78º00'	39º36'
Potomac #4	Washington	77º83'	39º50'
Clopper	Montgomery County	77º15	39º08'
Tridelphia	Howard/Montgomery	77º00'	39º11'
Piney Run	Carrol County	76º98'	39º39'
Duckett	Prince Georges County	76º 88'	39º 12'
Liberty	Baltimore County	76º56'	39º26'
St. Mary's Lake	St. Mary's County	76º 56'	38º 25'
Prettyboy	Baltimore County	76º44'	39º38'
Lake Lariat	Calvert County	76º42'	38º36'
Centennial Lake	Howard County	76º30'	39º42'
Loch Raven	Baltimore County	76º23'	39º26'
Conowingo	Harford/Cecil	76º10'	39º39'
Tuckahoe	Caroline County	75º56'	38º58'
Johnson’s Pond	Wicomico County	75º36'	38º22'

Table 2.3: Quality control parameters. The detection limit, percentage relative standard deviation for laboratory and field duplicates, typical spike recoveries and field blanks are given for mercury (Hg) and methylmercury (MeHg).

Metal	DL Water*	DL Fish	% RSD	% Recovery of Matrix Spike	Field Blank*
Hg	0.1	0.05	<20	80-120	<1
MeHg	0.01	0.015	<20	80-120	<DL

*Detection limit (DL) and blank values are given in ng/L and ng/g.

CHAPTER 3: RESULTS AND DISCUSSION

3.1 Methylmercury in Largemouth Bass

Information about length, weight and MeHg concentration in each fish from the 20 reservoirs is contained in Appendix I. Length of the fish ranged from 276–563 mm (mean=376 ± 54, n=249) (Table 3.1). Mean length (mm) was highest in Lake Lariat 438.1 ± 80.4 mm (n=7), and lowest in Big Pool, 304.0 ± 16.4 mm (n=5) (Table 3.2). Weight of the bass ranged from 224 to 2688 g (mean=777 ± 391, n=249) with the mean weight highest in Lake Lariat, 1246 ± 669 g (n=7) (Table 3.2) and lowest in Big Pool, 322 ± 72 g (n=5) (Table 3.2). Correlation between total length (mm) and weight (g) of largemouth bass revealed a significant positive correlation when fish from all reservoirs were combined ($p<0.01$, r^2 = 0.92, n = 249; slope = 6.99) (Figure 3.2a). Such a strong relationship between length and weight in all the reservoirs could be taken as indicative that the fish live under similar conditions and none of the fish populations are under significant stress. Alternatively, all populations could be stressed, although this is unlikely. On a regional basis there are small differences in the length-weight relationship. For western MD, the slope (s) of the relationship (Fig. 3.2b; s=5.80) is lower than for the reservoirs in central MD (Fig. 3.2c; s=6.58) and coastal MD (Fig. 3d; s=7.86). So, overall, fish of the same length weigh more for the coastal reservoirs than for the others, and those in western MD reservoirs weigh the least. Such differences may be expected given that the fish were caught in late spring/early summer and the reservoirs in the coastal zone likely warm up more quickly, and are more productive, earlier in the season because of differences in climatic factors (such as elevation, amount of snow, and the mediating coastal effects on temperature). Comparable strong relationships between the weight and length of largemouth bass has been documented in other states, such as in Massachusetts's lakes (Rose et al., 1999) and for lake trout in Tadenac Lake, Canada (MacCrimmon et al., 1983).

These relationships suggest that all of the fish are not necessarily growing at the same rate, although as suggested above this is possibly to be a temporary difference that reflects the time of sampling. Growth rates for largemouth bass, which were not assessed during this study, appear to be relatively similar across the study reservoirs (Klotz and Johnson, 2000) and are close to the statewide average (Elser, 1962) (Figure 3.3). However, the differences may influence the bioaccumulation rate, as discussed below. Closer examination of the growth curves show that the growth rates for fish from western MD (Deep Creek and Piney) are somewhat slower than that of the reservoirs from the central region of the state. For example, for a 500 mm fish, extrapolation of the growth data indicates that ages range from somewhat more than 8 years to about 11 years. This is a fairly important difference for the larger fish. For the smaller fish, the differences are less (for 200 mm fish, ages range from 2-3 years). No growth rate data is available for the reservoirs of the coastal plain. If the fish are growing more slowly, then it should be expected that the concentration of MeHg in the tissue would be higher as bioaccumulation is more a factor of age than size. Indeed, the concept of growth dilution is important, as the excretion of MeHg by fish is very slow (Hudson et al., 1994). Thus, the slower growth of the fish in western MD may lead to a higher concentration for a fish of a particular size. As discussed below, there is little trend between concentration

and reservoir location and the highest concentration fish are from costal reservoirs which is contrary to expectation based on growth rate.

MeHg concentrations in the fish fillet samples ranged from 9.0 to 2077 ng/g wet weight (mean = 325 ± 283 ng/g, n = 249) (Table 3.1). These values are similar to the documented range of MeHg in largemouth bass in Massachusetts (Rose et al., 1999), Maine (Stafford and Haines, 1997) and in Connecticut (Ward and Neumann, 1999). A significant statistical difference was observed between the reservoirs containing the average highest and lowest MeHg in the largemouth bass ($p<0.001$). Mean MeHg concentrations in the fish were highest in Lake Lariat, 1044 ± 580 ng/g wet weight (n = 7) and lowest in fish from Centennial Lake, 98.5 ± 59.1 ng/g wet weight (n = 7) (Table 3.2). Potential reasons for these differences are discussed below, but include differences in fish size. Results from a study by Becker and Bigham (1995) indicated that an analysis of MeHg concentration in fillets and whole bodies of fish did not show a significant difference and therefore, fillets can provide acceptable estimates of MeHg concentration in whole bodies of the fish they studied (*Dorosoma cepedianum, Morone american, Cyprinus carpio, Ictalurus punctatus, Lepomis macrochirus, Micropterus dolomieui* and *Stizostedion vitreum*). However, in the context of this study, it was important to determine the filet concentration as the data was designed, in addition to understanding factors controlling fish concentration, to be useful to managers interested in setting consumption advisories, if fish concentration deemed these necessary.

There was a correlation, on both on a linear and a $\log_{10}/\log_{10}$ basis, between fish Hg concentration and length and weight for all largemouth bass (Fig. 3.4; n = 249; for length, linear relationship $r^2 = 0.26$; for weight, linear relationship, $r^2 = 0.28$; for log plot, $r^2 = 0.53$; all $p<0.01$) (Fig. 3.4). It is possible that the relationship with weight shows an exponential relationship ($r^2 = 0.35$; $p<0.01$). A positive correlation with size and concentration of MeHg in fish has been documented in many other instances such as for largemouth bass (Rose et al., 1999; Hanten et al., 1998), largemouth bass and chain pickerel (Horwitz et al., 1995) and for other predator fish species (Stafford and Haines, 1997; Ward and Neumann, 1999) and is most likely related to the accumulation of MeHg in the fish tissue as the fish grows (Hueter et al., 1995; Stafford and Haines, 1997; Somers and Jackson, 1993). In Figures 3.5 and 3.6, the relationship between length and weight and total MeHg concentration in largemouth bass in each reservoir is illustrated. Fifteen reservoirs showed significant correlation ($p< 0.05$) between the concentration and the size variables. However, the rate of accumulation of MeHg into the fish (shown as regression slopes) (ranging from 0.68 to 6.04 for length, and from 0.09 to 0.76 for weight) show that accumulation rates vary substantially between reservoirs, even those in close proximity.

The reservoirs in Western Maryland showed considerable variability among their regression slopes and had a large range of values. Deep Creek Lake (DCL) and Broadford Lake (BFL), which are geographically close, had similar slopes on a weight basis (0.39 & 0.21, respectively; Fig. 3.6a) and on a length basis, they were similarly within a factor of two (Fig. 3.5a). Overall, the differences between fish concentration, and length or weight, showed

similar variability across reservoirs and either is therefore a reasonable means of comparison. Therefore, the relationships will be discussed mostly in terms of length although the information for the relationships with weight is contained in the figures. Savage (SAV) and Piney (Frostburg) (PINF), which are also in the vicinity of DCL and BFL (Fig. 3.1), were found to have relatively steeper regression slopes, of 3.59 and 2.55, respectively. However, Big Pool (BP) and Rocky Gap (RGP) which are located east of the other four reservoirs, did not have a significant relationship between length and concentration, and the fish showed little change in concentrations with size. Concentrations were also low compared to other reservoirs. Similar results have been documented by Simonin et al (1994) and Rose et al (1999), where regression slopes between MeHg concentration in tissue of largemouth bass, and brown bullhead, and weight in a variety of lakes were found to be unequal.

The variability of slopes implies individual trends in each of the reservoirs and possibly effects of chemical and/or physical characteristics of the reservoir on the MeHg accumulation by the fish. Additionally, biological factors such as growth rate, age, size, physiology and diet might influence the accumulation and final concentration of MeHg in largemouth bass. As seen in Figure 3.5, the regression lines do not intercept at the y-axis at 0. This effect is most likely due to the fact that largemouth bass change their food preferences with age and feed at higher trophic levels when older. Thus, the MeHg concentration they are being exposed to increases with age. Also, as they age, the rate of growth decreases and thus the effect of growth dilution on concentration is smaller. Therefore, it is expected that the relationship between length and concentration is non-linear and increases with age. However, as the largemouth bass in this study are those that have reached harvestable length (circa 305 mm = 4 years, Klotz and Johnson, 2000), it is reasonable to approximate the slope of the relationships for these older fish with a linear equation. This approach is validated by comparison of the estimated MeHg concentrations in small largemouth bass from PINF, DCL and RGP (Castro et al., 2002; only total Hg was measured and is has been assumed that 90% of HgT in the fish muscle is MeHg) with the data collected in this study (Fig. 3.7). The notion that MeHg concentrations are relatively low in small fish is confirmed, as is the suggestion of a non-linear curve. This indicates that these fish are foraging lower in the food chain.

While the slope of the relationships in Fig. 3.7 for fish over a larger size range indicate a strong rate of increase for the fish in PINF with age (s=2.11), it is also apparent that the bass in RGP (s=0.27), and to a lesser degree, DCL (s=1.0), do not increase markedly in concentration as their length increases. Note that the slopes of all the data are slightly lower than those for the data from this study only, and this is expected based on the discussion above. Changes and differences in diet could account for these differences between lakes, although other factors are also likely important. While stomach analysis was done in an observational fashion in this study, the results, and other anecdotal evidence, suggest that largemouth bass have a preference for crayfish. These omnivorous invertebrates are “lower” in the food chain than small fish, as demonstrated in a stream study in western Maryland (Mason et al., 2000). The crayfish were found to have a lower bioaccumulation factor and a lower %MeHg in tissue than either predatory insects or small fish, such as dace and sucker. So, a bass feeding predominantly on crayfish should have a lower burden than one feeding exclusively on small fish, all else being equal. Thus, given the caveat that a one time analysis

of food in fish stomachs gives no real indication of feeding preferences, it is possible that differences in food availability between lakes may be one factor accounting for the differences in both the concentration and the rate of accumulation with size.

In a study in Sweden (Lindqvist et al., 1990) different slopes in the Hg burden-fish size relationship were also observed. Furthermore, this study was able to show that differences in slope between fish species, and within the overall growth of a single species could be related to feeding strategies. Roach had little change in Hg content with size, or even showed a decrease in concentration in older fish, and this was explained by the fact that the fish changed diet from zooplankton when young to a diet combination of herbivory and feeding on benthic macroinvertebrates when older. Pike accumulated Hg at a fairly constant rate as their diet was fairly invariant with age, being fish almost exclusively. Finally, perch show an intermediate trend as they switch diet from zooplankton when young to macroinvertebrates and fish when older. Changes in the rates of accumulation within a species were shown to be related to these changes in diet.

As mentioned above, in this study, for convenience, the slopes of the correlations lines are assumed to be linear but it is possible that shift in diet of the fish with age leads to a more complicated relationship. The current study was not designed to examine the relationship between diet and concentration in detail and the sample size is insufficient to determine this. However, there is a suggestion in the data of an increase in slope with size for some of the reservoirs, especially those in western MD where data for smaller fish is also available, which would be expected for fish switching diet from crayfish and invertebrates to small fish.

The rate of MeHg accumulation in largemouth bass in Central Maryland was found to differ from the rates in Western Maryland, although a range in rates of increase in concentration with length were found. Loch Raven (LRV) and Prettyboy (PBY) had similar regression slopes (2.33 & 2.53, respectively) as shown in Figure 3.4d. However, Liberty (LIB), Tridelphia (TRI), and Piney Run (PRN), also in central Maryland, had similar slopes but they were lower compared to the other two (1.73, 1.02 & 0.81, respectively) (Figure 3.4f & g). Duckett (DUC) had a higher rate of accumulation of 3.59, while the fish from Centennial Lake (CEN) were all small and no relationship was evident. In Southern Maryland, accumulation rates of MeHg in largemouth bass were found to be the highest of the 20 reservoirs tested (Lake Lariat (LLR), 6.04 and 3.70 in St. Mary's Lake (SML)) (Figure 3.4 h). Johnson's Pond (JHP) and Tuckahoe (TUC), both on Maryland's Eastern Shore, had similar regression slopes (2.24 & 2.03, respectively) while the regression slope for largemouth bass in Conowingo (CON) was merely 0.68, the lowest slope that was significant (Figure 3.4i).

The differences in the accumulation rates of MeHg in largemouth bass across the 20 reservoirs might be caused by many variables, such as the chemistry of the water, condition of the environmental surroundings and physical characteristics of the reservoirs. There is no clear sense from the relationships shown above that this is entirely dependent on the reservoir

location. The five highest slopes were from throughout the state (in decreasing order, LLR, SML, DUC, SAV and BFL. At the opposite end, the five lowest significant slopes were from, in increasing order, CON, PRN, DCL, LIB and TUC. Five reservoirs had no significant relationships but this is likely the function of either too little data, or too little spread in fish size. Combining the data of this study and that of Castro et al. (2002), RGP had the lowest slope for concentration versus length at 0.27.

As mentioned before, food sources of the largemouth bass may differ between the reservoirs. Since aquatic organisms low in the food chain generally contain lower concentrations of MeHg than a higher-trophic-level organisms, fish feeding on low-trophic-level animals consequently bioaccumulate less MeHg into their tissues during their lifetime than fish feeding on high-trophic-level organisms.

In an effort to assess food effects, in the first two years of the project,106 largemouth bass were sampled for gut content. Only half contained food remnants in the gut. Of these, 70% was identified to be crayfish, 17% sunfish and less than 1% channel catfish. Note must be taken that the fish was captured at the same time in both years (late spring, early summer) and gut content might not be representative of food sources largemouth bass consume during the whole year. In addition, sunfish and other soft tissue organism are more easily digested than crayfish, and that might explain the prevalence of crayfish remnants in the stomachs over that of other fish. Furthermore, largemouth bass are known to be opportunistic feeders and have been found to eat both terrestrial and aquatic organisms. However, gut content analysis can give an idea of the MeHg concentration in the diet of largemouth bass in the reservoirs that were studied. To assess the importance of differences in diet, crayfish and small forage fish were collected from these reservoirs, where possible (Appendix III). The average length of crayfish collected was 38.3 ± 51.9 mm (ranging from 7.04 to 138.15 mm) and average MeHg concentration was 22.1 ± 13.5 ng/g wet weight (ranging from 7.24 – 44.58 ng/g wet weight). Average length of forage fish (blue gill, black crappie, golden shiner, yellow perch, white sucker and pumpkin seed) was found to be 59.5 ± 72.0 mm (ranging from 3.23 – 216.0) and average MeHg concentration was 42.7 ± 23.9 ng/g wet weight (ranging from 9.41-108.17 ng/g wet weight), higher than for crayfish, as expected. The mean HgT concentration in the forage fish was 73.6 ng/g wet weight (ranging from 9.10-164.86 ng/g (Appendix III).

The average MeHg concentrations of forage sunfish in each reservoir (where available; n=8) were plotted against average MeHg concentration of largemouth bass that had been adjusted for length (362-377 mm) in the corresponding reservoir (Figure 3.8). A weak positive correlation was observed and the regression coefficient was found to be small (r^2=0.24). As seen in the figure, there is an outlier (PINF) that influences the overall correlation. If largemouth bass from PINF were excluded, the regression was significant (r^2 = 0.74). Other researchers have documented similar results between MeHg concentrations in piscivorous fish, such as largemouth bass, and foraging fish (Mathers and Johansen, 1985; Cope et al., 1990).

As discussed above, largemouth bass from PINF reservoir had high MeHg concentrations, possibly as a result of flooding of terrestrial land in 1990. It has been shown in studies in lakes in Canada that under comparable conditions, methylation rates were stimulated by the decomposition of flooded terrestrial material (Bodaly et al., 1997; Hecky et al., 1991). As enhanced MeHg production in the water column and sediments is often translated into amplified fish MeHg concentrations (Xun et al., 1987), older fish, which are long-term integrators of exposure, have concentrations that reflect to some degree past rather than current conditions in the reservoirs.

The poor correlation between the forage fish and largemouth bass for this reservoir might be an indicator of decreasing levels of aquatic and sedimentary MeHg in the reservoir since the flooding occurred. The older (the largemouth bass captured in Piney was determined to be 4 - 10 years old; Klotz, 2000) and larger largemouth bass have accumulated MeHg over a long time-period including immediately after inundation, when water and sediment might have contained high MeHg concentrations. On the other hand, the omnivorous, smaller and shorter-lived forage fish are likely more representative of the recent levels of MeHg in the water and sediments. Consequently, it can be assumed that the forage fish better represents the current MeHg levels in the reservoir while the concentration in the largemouth bass correspond to an average, which includes the high MeHg concentration in the reservoir in the last decade. However, there is no linear relationship between small bluegill concentration and dissolved MeHg in water, and this was also true for the crayfish. Given differences in DOC, pH and other chemical factors between reservoirs, and the known importance of speciation in determining bioavailability, such a simple relationship is unlikely to exist. Theoretical models predict returns of MeHg concentrations to background levels after impoundment of reservoir to be 20 years for lake trout and 30 years for northern pike (Anderson et al., 1995). Data collected sixteen years after impoundment of Smallwood Reservoir in Canada showed that the Hg levels in flesh of non-piscivores was similar to background Hg levels while piscivores were still elevated above un-impounded lakes (Anderson et al., 1995). Other reasons for the lack of correlation can be either that the chosen forage fish were not representative of the diet, and/or other factors.

In addition to the above, a more thorough examination of the fish growth rate data (Fig. 3.2) shows that bass in PINF grow at a slightly slower rate than fish in most of the other reservoirs. If fish are growing slower because of a higher physiological cost, then they will likely have a higher MeHg burden, given the same food, as growth dilution is their major concentration “loss” mechanism, as depuration is very slow (Hudson et al., 1994). However, the differences are likely insufficient to account for inconsistency for this reservoir with regard to the fish-food relationship. Overall, the comparison of the growth rate data in concert with the slope of the regression lines (Figs. 3.2, 3.5 & 3.6) support the notion that the concentrations of MeHg are higher relatively (higher slope to the regression line) in slower growing fish. However, there is one reservoir that does not fit this trend. Clopper (CLO) has slow growing fish with a low MeHg concentration. As mentioned earlier, both CLO and PRN are deep lakes with relatively low surface area and perhaps the dynamics of these lakes lead to slow growing fish, which have relatively low MeHg. Growth data is not available for PRN but

its bass do have relatively low MeHg concentration. Further investigation is required to assess this.

Overall, the average MeHg concentration of the fish for the 20 reservoirs (Table 3.2) shows substantial variability, as discussed above. The large variability obtained for each of the reservoirs is caused by the variability in the size of the fish. No significant statistical difference was obtained in MeHg concentrations in largemouth bass across the reservoirs except for the fish in Lake Lariat, Savage and Piney (Frostburg). Exceptionally high concentrations were measured in fish from these reservoirs compared to the other 17 reservoirs. To show the data in a more comparable fashion, the estimated concentration for fish of around 370 mm (362-377 mm) is shown in Fig. 3.9. These data reinforce the conclusions above that the fish concentrations are highest in two of the coastal reservoirs, LLR and SML, and is also elevated above the others for two reservoirs in western MD, SAV and PINF. This suggests that growth rate influence over fish concentration is not the only factor as, as stated above, the length-weight relationships and the growth rate curves are most different for the coastal region compared to the colder, more mountainous western MD region. The fish in PINF are characterized by a diverse age and size structure, with older, larger individuals in the population (Klotz and Johnson, 2000).

3.2 Methylmercury and Total Mercury in Reservoir Water

Much evidence points to the importance of direct atmospheric Hg deposition into aquatic environment in Maryland (Mason et al., 1997a. Mason et al., 2000b) and the regional wet depositional flux for the Chesapeake Bay is higher than found for many regional locations in the mid-west (Hoyer et al., 1995). However, indirect input (runoff from the watershed) is also important. Hultberg et al. (1994) reported, for example, that direct atmospheric deposition, in addition to watershed inputs, accounted for most of the Hg found in lakes in southwestern Sweden. Thus, the relative size of the watershed can impact the fate of mercury within the reservoir. A number of relationships between fish mercury concentrations and watershed area have been noted (Suns and Hitching, 1990). The lake size to watershed ratio will influence the ratio of the amount of Hg directly deposited into the system through dry or wet deposition versus indirect runoff, and differences in Hg loading to the systems are related to differences in watershed area to lake area ratios (Suns and Hitching, 1990; Rudd, 1995; Rose et al., 1999). Under anoxic conditions, such as frequently found in wetlands, enhanced methylation of Hg can occur. The export of MeHg from riparian wetlands has been identified as a major source of MeHg for drainage lakes (St. Louis et al., 1994) and therefore, the relative size of the wetlands surrounding the reservoirs compared to the size of the watershed is an important factor that may modify MeHg levels. Other factors, such as productivity (influenced by the availability of nutrients (French et al., 1999)) and DOC, can stimulate MeHg production in the aquatic system and subsequently alter mercury partitioning between sediment, water and biota.

Concentrations of mercury (HgT) and methylmercury (MeHg) in the dissolved fraction of the water samples are shown in Table 3.3 (raw data is contained in Appendix II).

Average HgT concentrations ranged from 0.4 ± 0.1 ng/L (Rocky Gap; Castro et al., 2002) to 19.5 ± 6.0 ng/L (Duckett). Average HgT concentration was 3.6 ± 5.7 ng/L, which is similar to values found for Chesapeake Bay tributaries (Lawson et al., 2001) and in lakes in Wisconsin (Watras et al., 1995), although somewhat lower concentrations were found in reservoirs in Western Maryland (Castro et al., 2002).

The range of MeHg concentrations observed in the water samples ranged from 16.5 ± 1.5 pg/L (Lake Lariat) to 292 ± 381 pg/L (Tridelphia), with an average concentration of 131 ± 85 pg/L. Similar concentrations have been found in surface waters in the Lake Gardsjön watershed on the west coast of Sweden (Lee and Hultberg, 1990), in freshwater systems in Wisconsin (Watras et al., 1995a), in the Chesapeake Bay (Mason et al., 1999) as well as in tributaries to the Chesapeake Bay (Lawson et al., 2001). MeHg concentrations were exceptionally high in Piney (Frostburg) and Deep Creek Lake in Western Maryland and in Tridelphia and Piney Run in Central Maryland.

3.3 Reservoir Chemistry

Ancillary data from the 20 reservoirs are shown in Table 3.4 (raw data is contained in Appendix II). pH was similar across the 20 reservoirs, with no reservoirs having low pH's such as found in some lakes in the Midwestern USA. Overall, the reservoirs can be considered to be circum-neutral. The average pH values ranged from 6.7 ± 0.8 (Broadford Lake, n=2) to 8.7 ± 1.0 (Loch Raven, n=3), with the overall average for the 20 reservoirs of 7.5 ± 2.0 (n=38). Statistically, no significant difference in pH was found among the four Maryland regions.

Dissolved organic carbon (DOC) concentrations were also relatively consistent across the reservoirs. Average DOC concentrations were 3.66 ± 1.96 mg/L (n=37), ranging from 1.95 ± 0.6 mg/L in Prettyboy (n= 3) to 6.51 ± 0.2 mg/L in St. Mary's Lake (n=2) (Table 3.4). No significant difference in DOC concentrations was observed across the four Maryland regions. However, values in Western Maryland were generally low while DOC concentrations in Southern Maryland reservoirs were higher (Table 3.4). These DOC concentrations are fairly low compared to values obtained in lakes in Wisconsin (average about 7 mg/L, Watras et al., 1995b) but similar to concentrations found in other Maryland streams (Mason, 2000).

Particulate nitrogen (PON) was low in all reservoirs, ranging from 0.03 ± 0.01 mg/L in Rocky Gap (n=3) to 0.37 ± 0.41 mg/L in Potomac #4 (n=2). Average PON concentration for all of the reservoirs was 0.12 ± 0.07 mg/L (n=38). Particulate carbon (POC) was found to be relatively low across all of the reservoirs (1.13 ± 0.78 mg/L, n=25), and in four of the reservoirs concentrations were at or below detection limits (Table 3.4 and Appendix II).

Nitrite and nitrate concentrations were quite consistent in the reservoirs (Table 3.4). Nitrite was highest in Duckett (1.27 ± 0.16 mg/L, n=3) and below detection limits in Broadford Lake, Savage, Rocky Gap and Loch Raven. Nitrate concentrations across the reservoirs were consistent as well, ranging from below detection limits (Duckett and Clopper) to 6.51 mg/L (St. Mary's Lake) with an average of 1.52 ± 1.70 mg/L (n=31) (Table 3.4). Ammonia concentrations ranged from 0.01 mg/L in Piney (Frostburg) to 0.36 mg/L in Big Pool, with an average of 0.05 ± 0.05 mg/L (n= 38).

Sulfate concentrations in the reservoirs ranged from 3.43 ± 0.44 mg/L in Prettyboy (n=3) to 36.8 ± 1.56 mg/L (Potomac #4, n=2) with an average concentrations of 9.49 ± 2.80 mg/L (n=38). Comparable values have been documented in various aquatic systems such as in New York, northern Wisconsin, and Massachusetts (Simonin et al., 1994; Watras et al., 1995a and Rose et al., 1999) and seem to be within normal ranges of freshwater systems. Thus, it is not likely that any of the reservoirs are sulfate-limited, and thus, limited compared to the other systems in terms of mercury methylation.

Phosphate levels were found to be relatively low in the tested reservoirs. Average concentrations were 0.05 ± 0.18 mg/L (n=38) with lowest levels detected in Piney Run Reservoir (0.0012 mg/L) and highest in Liberty (0.34 ± 0.51 mg/L, n=3) (Table 3.4). These phosphate levels are well within the documented range found in non-polluted natural waters (1 *u*g/L to 200 mg/L) (Wetzel, 1983). Total suspended concentrations (TSS) in the reservoirs were relatively low. The average TSS concentration was 7.8 ± 10.0 mg/L (n=37) and was lowest in Piney (Frostburg) (0.9 ± 0.3 mg/L, n=2) and highest in Loch Raven (27.9 ± 19.9 mg/L, n=3). These values are in agreement with TSS concentrations observed in samples from other Maryland streams (Mason, 2001).

Chloride concentrations (Cl^-), contrary to all of the other ancillary measurements, ranged widely across the reservoirs. Average concentration across all of the reservoirs was 33.8 ± 28.6 mg/L (n=38). Values in Liberty were found to be lowest (1.74 ± 0.12 mg/L, n=3) while average values for Clopper were 135.8 ± 6.93 mg/L (n=3). These values are relatively high compared to lakes in northern Wisconsin (0.11-4.0 mg/L, Watras et al., 1995a). However, similar values have been found in lakes in Massachusetts (1-35 mg/L, Rose et al., 1999).

Several factors could be responsible for the high amount of chloride in the reservoirs. Proximity to the ocean might have a bearing on the high chloride concentrations because of atmospheric inputs. However, data from three reservoirs in Western Maryland which are far removed from the influence of seasalt input (Piney (Frostburg), Rocky Gap and Deep Creek Lake) still show a 1:1 relationship between chloride and sodium concentrations (Castro et al., 2002), suggesting that the chloride found in these reservoirs comes from salt that is used for melting of snow in urban communities. No correlation was observed between the chloride concentrations and nitrate/nitrite or phosphate concentrations in the water, suggesting that the

chloride is not derived from detergents and water softening agents used by local residents or industries, as sometimes is evident.

Two decades of water quality data have indeed shown a general increase in the chloride concentration in the reservoirs surrounding Baltimore (Bill Stacks, pers. comm.) and the data reported here for Liberty are low relative to those data, which show that the concentration is more similar to that of Prettyboy and Loch Raven. Road salt is the likely culprit for these noted increases. Thus, the low concentrations of chloride in Liberty as opposed to the relative high concentrations in Loch Raven and Clopper, while of interest could just represent a sampling artifact. Alternatively, the cause for the discrepancies between the reservoirs might be based on the differences in population densities around the reservoirs as well as their volume. Population density around Loch Raven, for example, (1.83 people/acre) is significantly higher than for Liberty (0.7 people/acre; MDNR, 2001) and this could account for the dramatic differences in chloride concentrations between the two reservoirs because of salt usage on roads. Furthermore, Liberty holds twice as much volume as does Loch Raven and thus dilution might be an additional plausible cause for the differences in the chloride concentration between the two reservoirs. Likewise, in Western Maryland, a large range of chloride values was obtained from the three reservoirs. Rocky Gap had a low average value of only 3.3 ± 0.93 mg/L while Piney (Frostburg) had an average value of 33.1 ± 11.1 mg/L. Reservoir proximity to major roads (or population density) does not seem to be at fault here, since equal values have been documented for both reservoirs (0.28 people/acre) (MDNR, 2001). However, there is a storage mound of road salt in the vicinity of Piney (Frostburg) and that could account for the high concentration in this reservoir (Castro, pers. comm.). While the watershed characteristics are similar for both Piney and Rocky Gap, Piney is much shallower (normal depth = 9.7 m vs. 25 m) and contains only one third of the volume of Rocky Gap (Table 2.1).

No significant geographic gradient was observed for any of the chemicals in the reservoirs and concentrations of the chemicals were in an agreement with water quality measurement done by the Maryland Department of Natural Resources in conjunction with Versar (MDNR/Versar).

3.4 Statistical Analysis

Chemical and physical parameters of the reservoirs were run as Pearson correlates in combination with the MeHg concentration in largemouth bass at each reservoir. Many studies in the literature have attempted to find correlations between fish concentration and physical and chemical characteristics and a number of parameters have been found to be of primary importance in different locations. For example, pH is a variable that has been associated with fish concentration in some locations. However, pH does not vary significantly between the reservoirs in this study and thus no significant correlation was expected, or found (Table 3.5). The effect of pH on accumulation of MeHg in fish has been extensively studied (Wren et al., 1991; Weiner et al., 1990), although the driving mechanisms are still debated by many

researchers (Richman et al., 1988). The effect of acidity on MeHg concentration in fish is complex as acid lakes tend to have longer food chains, and different foodwebs, and acidity will also enhance the fraction of MeHg bound to chloride complexes (Mason et al., 1996). Most studies in acidic lakes report an inverse correlation between pH and mercury in fish (Watras and Bloom, 1992; Spry and Weiner, 1991;Weiner et al., 1990; Grieb et al., 1990; Suns and Hitchin, 1990; Hakanson et al., 1988). The lack of any significant correlation of pH with MeHg accumulation in largemouth bass in this study is also perhaps a result of the pH of the reservoirs falling into a range in which the relationship to Hg dynamics is unclear (Allen-Gil et al., 1995).

Overall, however, there was no significant simple linear correlation with any of the reservoir chemical characteristics and the normalized MeHg in the largemouth bass (Table 3.5) with the highest value for the regression coefficient being <0.3. This contrasts the results of the initial phase of the study where a correlation, for example, with chloride concentration was apparent. Also, HgT and MeHg concentrations in the water did not significantly correlate with the fish MeHg for all reservoirs while in the initial study MeHg concentrations in largemouth bass in eight Maryland reservoirs showed a positive correlation with MeHg concentration in the dissolved fraction of the water (Sveinsdottir, 2002). Since bioavailability of MeHg to phytoplankton and other animals in the base of the food chain depends mostly on uptake from water, it is expected that MeHg concentration in the dissolved fraction of the water be closely related to MeHg concentration in fish, although the effects of speciation on uptake need to be considered. For example, it has been suggested that DOC binds MeHg and makes it less available for bioaccumulation. In addition, it must be noted that water sampling was only done on one occasion while fish concentration is an integration of years of accumulation so the lack of correlation may be due to some degree to a lack of sufficient water column data. While this may be true for other water quality parameters, the data obtained in this study was comparable to other data collected previously by others (MDNR data) and thus these parameters are likely to be a more true representation of the water quality in the reservoirs.

No correlation was found between MeHg concentrations in the fish and DOC concentrations of the waters (Table 3.5). Other studies have shown that DOC can potentially be an important factor affecting the extent of bioaccumulation of MeHg in fish (Driscoll et al., 1995), with the presence of DOC potentially reducing the uptake of MeHg by fish due to the polar nature and large size of the MeHg-DOC compound (Choi et al., 1998). Although DOC concentrations in the study lakes are relatively low, they are sufficient (Hudson et al., 1994) to bind most of the dissolved MeHg as an organic complex (approx. 97%). MeHg-hydroxide compounds will account for approximately 2% while, the last 1% is taken up by MeHg-chloride compounds. This supposition is based on thermodynamic equilibrium estimations using literature values for the DOC equilibrium constant (Benoit et al., 2002; Dyrssen & Wedborg, 1991 and Stumm & Morgan, 1996).

A significant correlation was not detected between MeHg concentrations in the largemouth bass and the chloride concentrations of the reservoirs. Such a relationship had

previously been observed in eight Maryland reservoirs (Sveinsdottir, 2002) and predicted by Mason et al. (1996), where increased MeHg concentrations in fish could be explained by the superior uptake efficiency of chloride compounds (neutral species) via passive diffusion as opposed to the hydroxide complexes, or organically-complexed MeHg. There have been a number of studies, for example, Watras et al. (1998), that have provided a contradictory conclusion i.e. that passive diffusion is not the most important uptake mechanism for MeHg into plankton.

Pearson correlation analysis (Table 3.5) indicated that MeHg in largemouth bass was not significantly correlated with any of the physical characteristics (depth, surface area, volume and surface area/volume ratio) of the reservoirs (Table 2.1). This contradicts several findings (Rose et al., 1999, Suns and Hitchin, 1990) where significant correlation between MeHg concentration in largemouth bass and the size of the watershed, lake area as well as area in the watershed occupied by wetlands were observed.

In addition to linear regression analysis, multiple regression approaches were used to study the interactions between variables. As noted in Table 3.5, while no correlations were statistically significant, the highest correlations were found with: total MeHg, POC, sulfate, surface area, depth and DOC. Unfortunately, the POC dataset was too small to examine this variable further. The other variables were examined in combination, and the "best fit" combination of variables is shown in Table 3.6. As these relationships are not highly significant, this evaluation was not pursued in its entirety. A relationship between water column MeHg and fish MeHg indicates the lack of importance of such factors as DOC differences in modifying the bioavailability of MeHg in these reservoirs, which is not unexpected given the fairly constant DOC concentration and pH in these systems. The lack of importance of Cl, even given its high variability across the reservoirs, is counter to expectation, and early results, and suggests that even though DOC concentrations are low, they are sufficient to negate any effect of differences in chloride concentration on bioaccumulation at the base of the food chain. Modeling results (e.g. Hudson et al., 1994) support such a conclusion.

The negative relationship with sulfate suggests that sulfate concentrations are inversely effecting MeHg in fish, while all the other interactions are positive. While the negative correlation could indicate that in the higher sulfate reservoirs MeHg concentration is negatively impacting methylation rate, the relationship between sulfate and MeHg in water is positive, while not significant (data not shown). This suggests that the fish concentration-sulfate relationship is reflecting another factor. Given the importance of sulfate in rain in contributing sulfate to the watershed, the relationship may indicate a correlation such as relative watershed contribution of Hg and MeHg. The exact cause for the relationship is not evident from the current analysis of the data.

As discussed above, the relative shape and size of the reservoir impacts the MeHg in fish through a complex series of interactions. In the multiple variable analysis, the surface

area to volume ratio appeared to be the best physical parameter that correlated with fish concentration. This ratio is a reflection to some degree of the relative sediment surface area to volume ratio and likely reflects the various interactions that occur at the sediment-water interface (methylation and bioaccumulation via benthic organisms). The equation with the highest significance, in terms of the F statistic, is:

Fish standard MeHg (ng/g) = 0.501*MeHg(unfil.) (ng/L) –5.603*SO_4 (mg/L) + 24.43*(Surf.Area/Vol.) (m^{-1}) + 258.3; r^2=0.19.

While this relationship is not significant at the 95% confidence level, it is significant at a somewhat lower level of confidence (90%). Overall, this relationship encompasses the impact of methylation and the studied reservoir characteristics in determining MeHg in the largemouth bass. However, only 20% of the variability is explained by this equation and clearly other factors, such as food chain length and structure, must have an important bearing on fish MeHg concentration. In addition, the watershed itself, and the rate of turnover of water within the system, are potentially important variables that could not be sufficiently assessed based on current data about the reservoirs and their watersheds. Future studies should endeavor to focus attention onto these factors and other that may be more important in reservoirs than in natural lakes. For example, it has been shown in Florida that water body "draw down" in summer, with subsequent reflooding, has an impact on the degree of Hg methylation (Krabbenhoft, pers. comm.). Reservoir water levels may fluctuate quite substantially, either from intentional water release or from unintentional factors, such as excessive water removal during a drought.

A detailed study of the factors controlling MeHg in fish should also take into account the feeding habits of the fish, or attempt to minimize the potential effect of this factor on fish concentration. Details on growth rate and age are also important. Other characteristics of water bodies that were not covered in this study but which could impact fish MeHg are bottom substrate and the degree of stratification, and other limnological considerations. If sufficient information were available then a more detailed modeling approach may provide substantial insight. More data is needed to help refine the analysis presented here.

3.5 Summary

The relatively strong relationship between MeHg body burden in largemouth bass and length and/or weight demonstrates that MeHg accumulates in fish with increasing size, as has been previously shown (e.g. Rose et al., 1999; Hanten et al., 1998; Horwitz et al., 1995; Stafford and Haines, 1997; Ward and Neumann, 1999 Hueter et al., 1995; Stafford and Haines, 1997 and Somers and Jackson, 1993). The variability observed between regression slopes of the 20 reservoirs indicate that there are other factors controlling the MeHg accumulation in largemouth bass and its prey than merely their diet. A variety of both physical and chemical characteristics of the reservoirs can be responsible for the variation of MeHg uptake by the largemouth bass and its prey. Such factors include geological influences,

chemical variability (e.g. water quality and mercury biogeochemistry) and physical variability (e.g. lake and watershed size, lake depth, wetland size) (Rose et al., 1999; Wren and MacCrimmon, 1983; Sonesten, 2001; Simonin et al., 1994; Rudd, 1995).

MeHg concentrations measured in the water samples were within documented range for Maryland lakes and rivers (Lawson et al., 2001; Mason et al., 1999). However, the results from the multiple regression analysis are quite different from those reported by others for MeHg in fish (French et al., 1999; Watras et al., 1995b; Bodaly et al, 1993; Lange et al., 1993; and Johnston et al., 1991). No correlation between physical and chemical characteristics of the reservoirs and MeHg bass concentrations were observed, which was contrary to numerous documented instances of correlations; such as with lake area ratios (French et al., 1999; Hanten et al., 1998; Lee and Iverfeld, 1991), DOC and pH (Mason, 2000; Watras et al., 1995a; Watras et al, 1995b, Lange et al., 1993; and others) and chloride (Mason et al., 1996).

DOC and pH are often cited as important parameters in predicting MeHg concentrations in fish (Mason, 2000; Watras et al., 1995a; Watras et al, 1995b, Lange et al., 1993). Mason (2000) determined that the concentration of DOC appeared to have the most influence over MeHg concentration in sunfish from small streams in Maryland. However in the same study, pH was reported as the most important parameter controlling MeHg concentrations in chain and redfin pickerel. It was concluded that the difference in the controlling parameter was based on the fish diet and position in the food chain. As DOC and pH values in the study lakes were relatively low (for DOC) and consistent across the reservoirs, these parameters did not show a good correlation with MeHg body burden in the largemouth bass

Atmospheric deposition of mercury, either directly into the lakes or via their watersheds, has been estimated to be responsible for the largest Hg input in aquatic systems in Maryland (Mason et al., 1997a). By assuming that these input rates are similar among the 20 reservoirs, it can be concluded that other factors than Hg supply play an important role in determining MeHg concentrations in the fish. DOC, pH and low buffering capacity of lakes have typically been correlated with fish Hg concentrations (Rose et al., 1999; Watras et al., 1995a; Watras et al, 1995b, Simonin et al., 1994; Lange et al., 1993; Grieb et al., 1990; Suns and Hitchin, 1990; Hakanson et al., 1988; Wren and MacCrimmon, 1983). Furthermore, water hardness, limnological state of reservoirs, and methylation and demethylation rates have been implicated as potential factors affecting the uptake of Hg by fish (Snodgrass et al., 2000; Rodgers and Beamish, 1983). Temperature, as well as loading rates, watershed ratios and lake size can be considered important variables influencing Hg levels in fish along with previously determined variables. Each of these mechanisms, and possibly more, are probably not mutually exclusive processes but rather, Hg cycling and uptake into fish tissues is governed by an array of interrelated, variables, the relative importance of which can differ from lake to lake.

Table 3.1. Summary of average weight (g), length (mm) and MeHg (ng/g wet weight) concentrations in 249 largemouth bass from 20 Maryland reservoirs.

	Weight (g)	**Length (mm)**	**MeHg (ng/g wet weight)**
Mean	777.4	376	325.0
Median	696.8	375	225.8
Standard Deviation	391.0	54	283.4
Count	249	249	249
Max	2688.0	563	2076.5
Min	224.1	276	9.5

Table 3.2. Summary of average weight (g), length (mm) and MeHg (ng/g wet weight) as well as standard deviation for largemouth bass from 20 Maryland reservoirs.

Reservoir	# of fish analyzed	Average weight (g)	Average length (mm)	Average MeHg conc. (ng/g wet weight)
Deep Creek Lake	13	766.9 ± 267.4	376.1 ± 42.9	320.0 ± 112.0
Broadford Lake	15	665.2 ± 226.9	367.7 ± 40.4	308.1 ± 172.3
Savage	12	609.5 ± 187.9	359.8 ± 35.6	484.2 ± 313.7
Piney (Frostburg)	15	780.4 ± 321.3	373.5 ± 49.1	615.2 ± 202.0
Rocky Gap	13	629.2 ± 198.6	356.6 ± 34.8	107.6 ± 46.0
Big Pool	5	321.9 ± 71.5	304.0 ± 16.4	264.8 ± 116.5
Potomac #4	2	434.2 ± 185.04	330.5 ± 51.6	130.7 ± 52.0
Clopper	15	779.1 ± 280.13	378.3 ± 39.1	197.4 ± 83.3
Tridelphia	14	889.0 ± 366.89	387.4 ± 56.0	193.7 ± 107.0
Piney Run	14	794.6 ± 281.77	374.9 ± 44.1	158.2 ± 63.4
Duckett	15	949.7 ± 483.5	390.8 ± 53.8	327.8 ± 201.1
Liberty	16	830.8 ± 304.4	383.0 ± 49.4	304.8 ± 171.1
St. Mary's Lake	12	883.5 ± 794.5	383.4 ± 93.1	736.0 ± 419.9
Prettyboy	15	759.7 ± 338.6	375.4 ± 56.5	348.4 ± 177.6
Lake Lariat	7	1246.4 ± 668.91	438.1 ± 80.4	1043.7 ± 580.0
Centennial Lake	7	329.43 ± 13.54	421.7 ± 114.8	98.46 ± 59.09
Loch Raven	16	718.5 ± 284.8	375.0 ± 53.3	327.8 ± 201.1
Conowingo	14	736.9 ±350.87	370.7 ± 46.2	118.5 ± 52.0
Tuckahoe	14	891.3 ± 481.3	351.1 ± 65.5	351.1 ± 209.0
Johnsons's Pond	15	879.9 ± 430.8	392.8 ± 56.5	204.1 ± 145.5

Table 3.3. HgT (ng/L) and MeHg (pg/L) in whole and dissolved fraction of water.

Reservoir	HgT (ng/L) Whole	HgT (ng/L) Dissolved	MeHg (pg/L) whole	MeHg (pg/L) Dissolved
Deep Creek Lake	1.95	0.61	382.60	277.37
Broadford Lake	1.32	0.99	159.75	116.97
Savage	1.28	0.61	105.46	72.80
Piney (Frostburg)	2.78	1.41	336.95	262.20
Rocky Gap	0.40	0.40	109.57	47.58
Big Pool	6.79	n/a	105.7	80.4
Potomac #4	1.48	1.17	239.74	209.79
Clopper	n/a	18.05	226.33	94.20
Tridelphia	2.67	1.96	n/a	292.39
Piney Run	1.68	0.97	337.42	254.83
Duckett	n/a	19.53	114.86	49.78
Liberty	1.98	1.89	64.22	75.11
St. Mary's Lake	2.17	0.45	190.42	130.87
Prettyboy	3.95	4.03	46.87	58.04
Lake Lariat	2.42	2.09	123.25	16.45
Centennial Lake	1.67	1.69	97.5	50.7
Loch Raven	5.09	5.63	194.65	160.39
Conowingo	n/a	3.55	n/a	118.58
Tuckahoe	4.08	2.40	125.58	136.55
Johnson’s Pond	3.18	1.71	156.26	113.80

Table 3.4. Average concentrations of pH, dissolved organic carbon (DOC mg/L), particulate nitrogen (PN mg/L), particulate carbon (PN mg/L), nitrite (NO_2^- mg/L) and ammonium (NH_4^+ mg/L) in the reservoirs. N/d = not detectable.

Reservoir	pH	DOC	PN	PC	NO_2	NH_4
		Mg/L				
Deep Creek Lake	6.9	2.94	0.06	n/d	0.04	0.08
Broadford Lake	6.7	3.90	0.17	1.28	0.00	0.03
Savage	7.2	2.38	0.04	0.64	0.00	0.01
Piney (Frostburg)	7.2	4.99	0.13	1.05	0.01	0.01
Rocky Gap	7.1	3.83	0.03	n/d	0.00	0.06
Big Pool	7.0	3.76	0.066	0.467	0.001	0.36
Potomac #4	7.2	2.40	0.37	0.59	0.01	0.04
Clopper	7.5	4.31	0.14	2.78	0.48	0.09
Tridelphia	n/a	n/a	n/a	N/a	n/a	n/a
Piney Run	7.9	3.13	0.07	0.61	0.01	0.04
Duckett	7.6	3.01	0.19	1.15	1.27	0.05
Liberty	7.6	2.5	0.1	N/a	0.0	0.0
St. Mary's Lake	6.8	6.51	0.17	2.10	0.00	0.03
Prettyboy	7.6	1.95	0.10	0.93	0.01	0.02
Lake Lariat	7.8	4.11	0.21	2.14	0.01	0.14
Centennial Lake	8.2	5.06	0	0.96200	0.012	0.02
Loch Raven	8.7	2.14	0.12	1.19	0.00	0.02
Conowingo	8.1	n/a	n/a	N/a	n/a	n/a
Tuckahoe	7.3	5.15	0.09	0.94	0.01	0.09
Johnson’s Pond	7.8	5.95	0.06	0.85	0.05	0.05

Table 3.4 cont. Average concentrations of nitrate (NO^{3-} mg/L), phosphate (PO_4^{3-} mg/L), total suspended solids (TSS mg/L), chloride (Cl^- mg/L) and sulfate (SO_4^{2-} mg/L) in the reservoirs. N/d = not detectable.

Reservoir	NO_3	PO_4	TSS	CL	SO_4
			mg/L		
Deep Creek Lake	0.02	0.022	2.4	6.59	13.67
Broadford Lake	1.13	0.003	3.7	40.75	8.40
Savage	0.58	0.217	6.0	19.80	12.30
Piney (Frostburg)	0.72	0.023	0.9	26.70	7.22
Rocky Gap	0.02	0.016	2.4	3.38	10.59
Big Pool	0.19	0.002	9.6	73.7	24.6
Potomac #4	0.78	0.016	8.0	17.80	36.80
Clopper	n/d	0.003	5.1	135.77	6.17
Tridelphia	n/a	n/a	n/a	N/a	n/a
Piney Run	0.58	0.001	2.4	27.96	6.91
Duckett	n/a	0.003	10.4	61.37	6.03
Liberty	1.7	0.340	2.4	1.73	7.9
St. Mary's Lake	6.51	0.002	11.9	9.00	5.02
Prettyboy	2.26	0.010	15.2	30.50	3.43
Lake Lariat	0.53	0.001	9.6	20.70	12.18
Centennial Lake	0.24	0.003	7.0	22.3	11.31
Loch Raven	2.34	0.020	27.9	70.47	5.03
Conowingo	n/a	n/a	n/a	N/a	n/a
Tuckahoe	2.72	0.030	5.0	16.43	6.87
Johnson’s Pond	2.40	0.010	5.7	22.31	4.24

Table 3.5 Correlation coefficients (r values) for linear regression analysis of fish methylmercury concentration, normalized to a standard length, and physical and chemical variables for the reservoirs.

Variable	**Corr. Coeff.**	**Variable**	**Corr. Coeff.**
HgT – unfilt.	0.10	NO_3	0.10
HgT – filt.	0.058	PO_4	-0.075
MeHg – unfilt.	0.22	TSS	0.12
MeHg – filt.	0.044	Cl	0.067
pH	-0.031	Depth	0.12
DOC	0.16	Surface Area	0.25
POC	0.30	Volume	0.010
SO_4	0.24	Surf. Area/Volume	0.095

Table 3.6 Correlation coefficients (r values) and F statistics for multiple linear regression analysis of fish methylmercury concentration, normalized to a standard length, and physical and chemical variables for the reservoirs. Note that only 19 reservoirs had data for all variables used in this multiple analysis.

Variables	**Corr. Coeff.**	**F value**	**Degrees of Freedom**	**F_{Crit} p=0.05**
MeHg-unfl.	0.19	0.57	16	2.28
MeHg-unfl., SO_4	0.32	0.86	15	2.33
MeHg-unfl., SO_4, Area/Vol	0.44	1.12	14	2.39
MeHg-unfl., SO_4, Area/Vol, Cl	0.45	0.81	13	2.46
MeHg-unfl., SO_4, Area/Vol, Cl, Depth	0.47	0.68	12	2.54

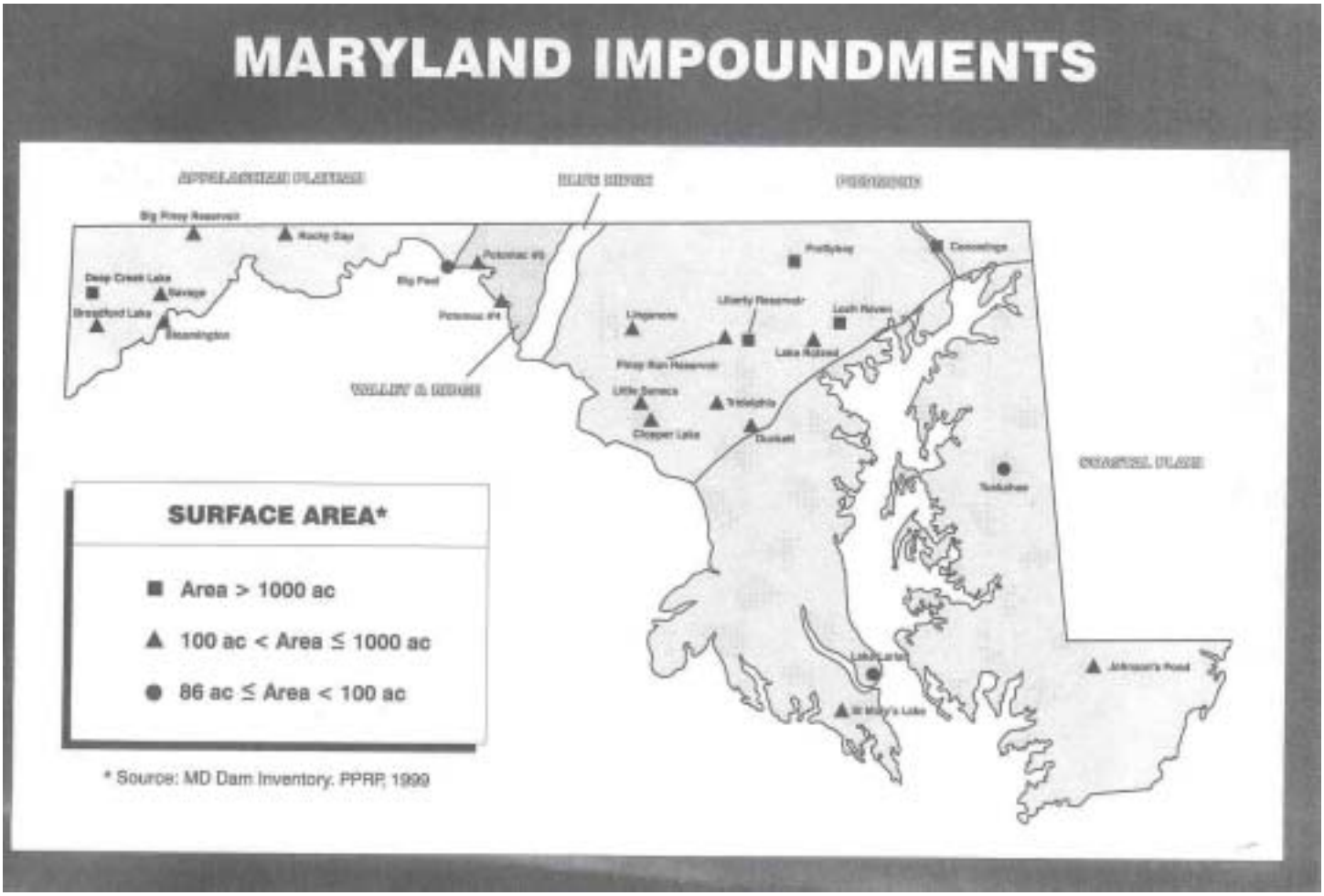


Figure 3.1: Location of the reservoirs sampled in Maryland. Symbol is indicative of reservoir size.

Note: In the following figures, acronyms are used for the reservoirs as follows:
DCL = Deep Creek Lake; BFL =Broadford Lake; SAV = Savage Reservoir; PINF = Piney Dam, Frostburg; RGP = Rocky Gap Park Reservoir (Lake Habeeb); BP = Big Pool, on the Potomac River; PO4 = Potomac #4, on the Potomac River; PBY = Prettyboy Reservoir; LRV = Loch Raven; DUC = Lake Duckett; CEN = Centennial Lake; TRI = Tridelphia Reservoir; LIB = Liberty Reservoir; CLO = Clopper Lake; PRN = Piney Run Reservoir; LLR = Lake Lariat; SML = St Marys Lake; CON = Conowingo Dam; TUC = Lake Tuckahoe; JHP = Johnsons Pond.

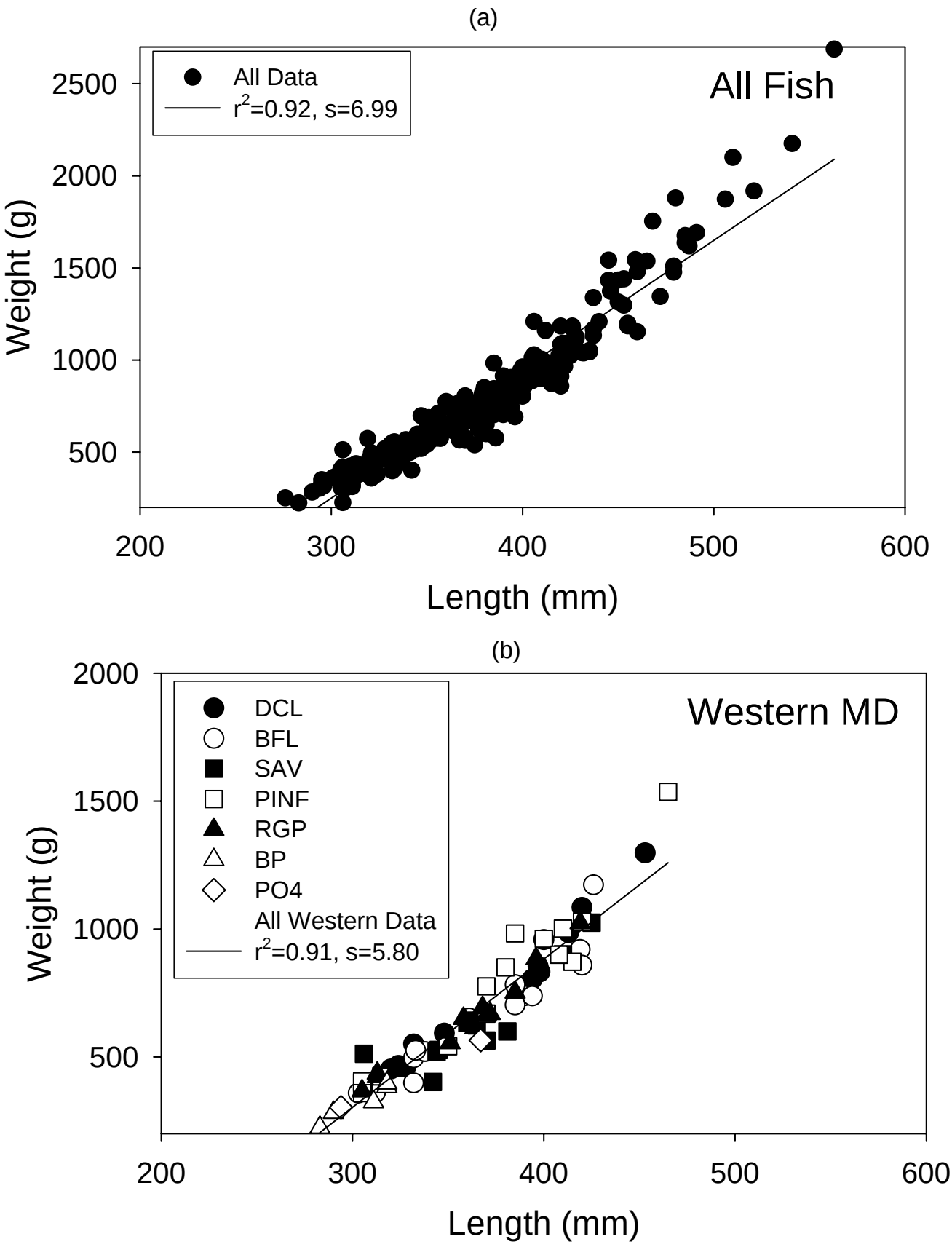
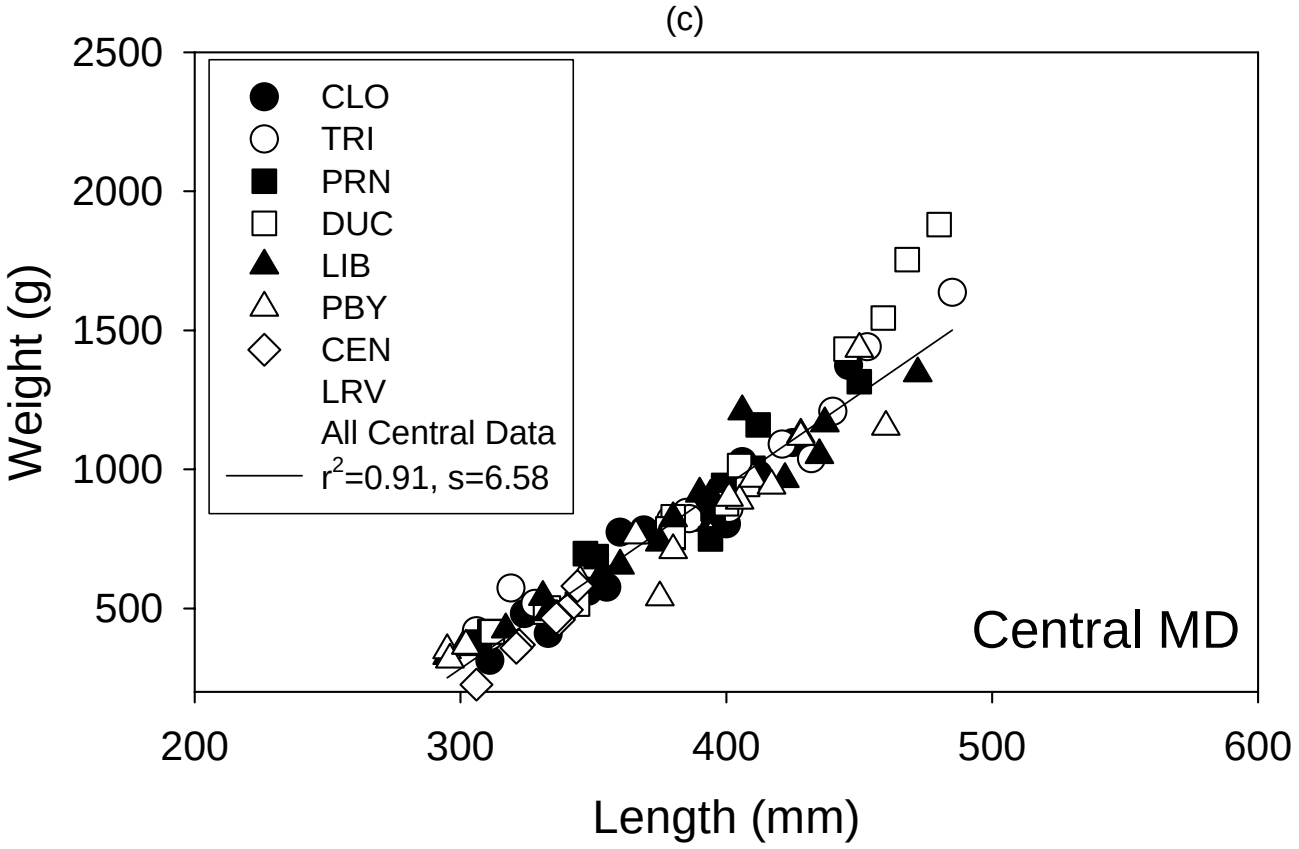


Figure 3.2. Weight (g) vs. length (mm) for largemouth bass from (a) all reservoirs; (b) from Western MD reservoirs,



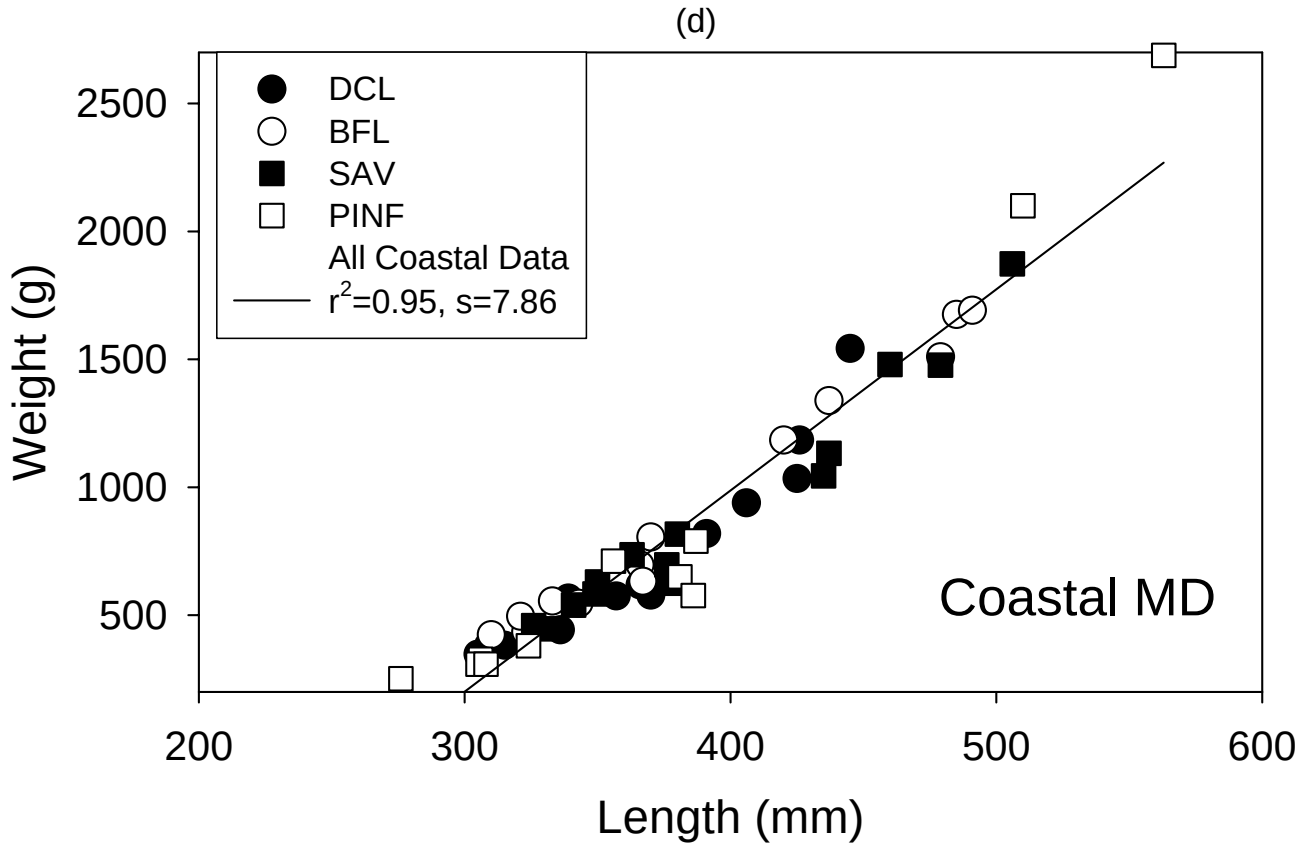


Figure 3.2 cont.: Weight (g) vs. length (mm) for largemouth bass from (c) Central MD reservoirs; (d) from Coastal MD reservoirs,

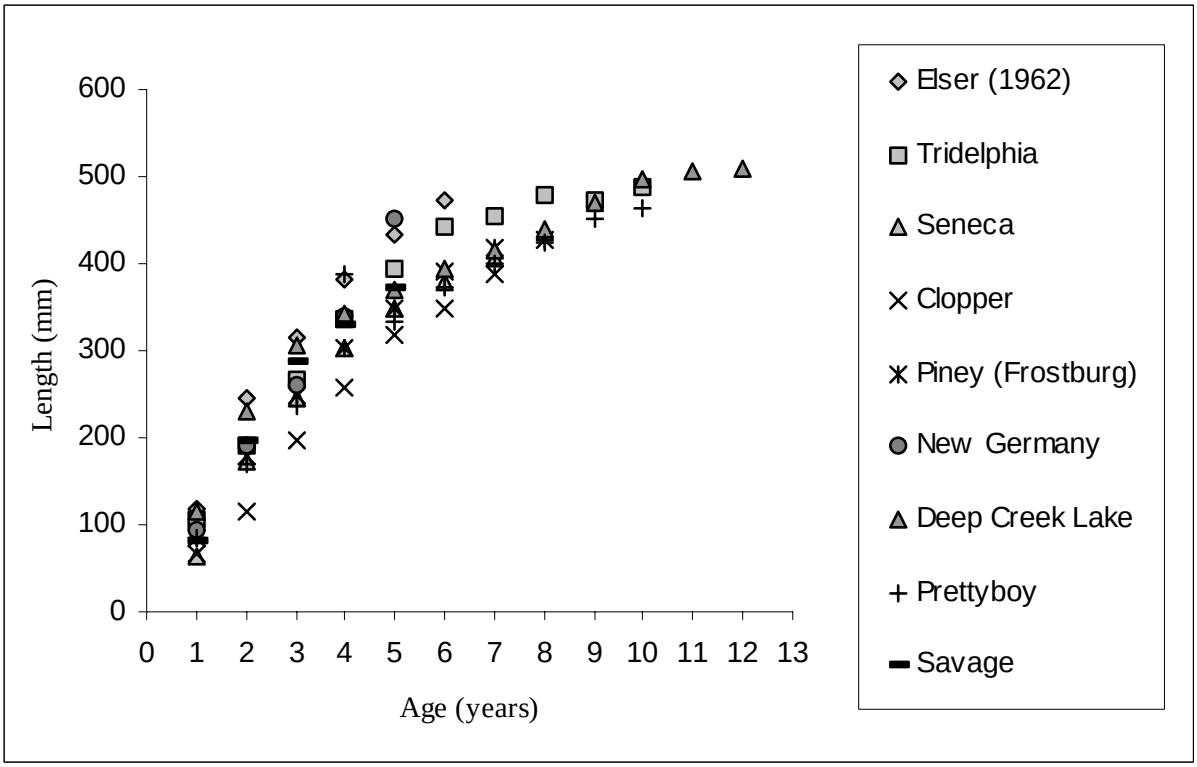


Figure 3.3. Growth rate of largemouth bass in reservoirs and lakes in Maryland. Growth rate reported by Elser (1962) is an average for the state. Reference: MDNR, 2000.

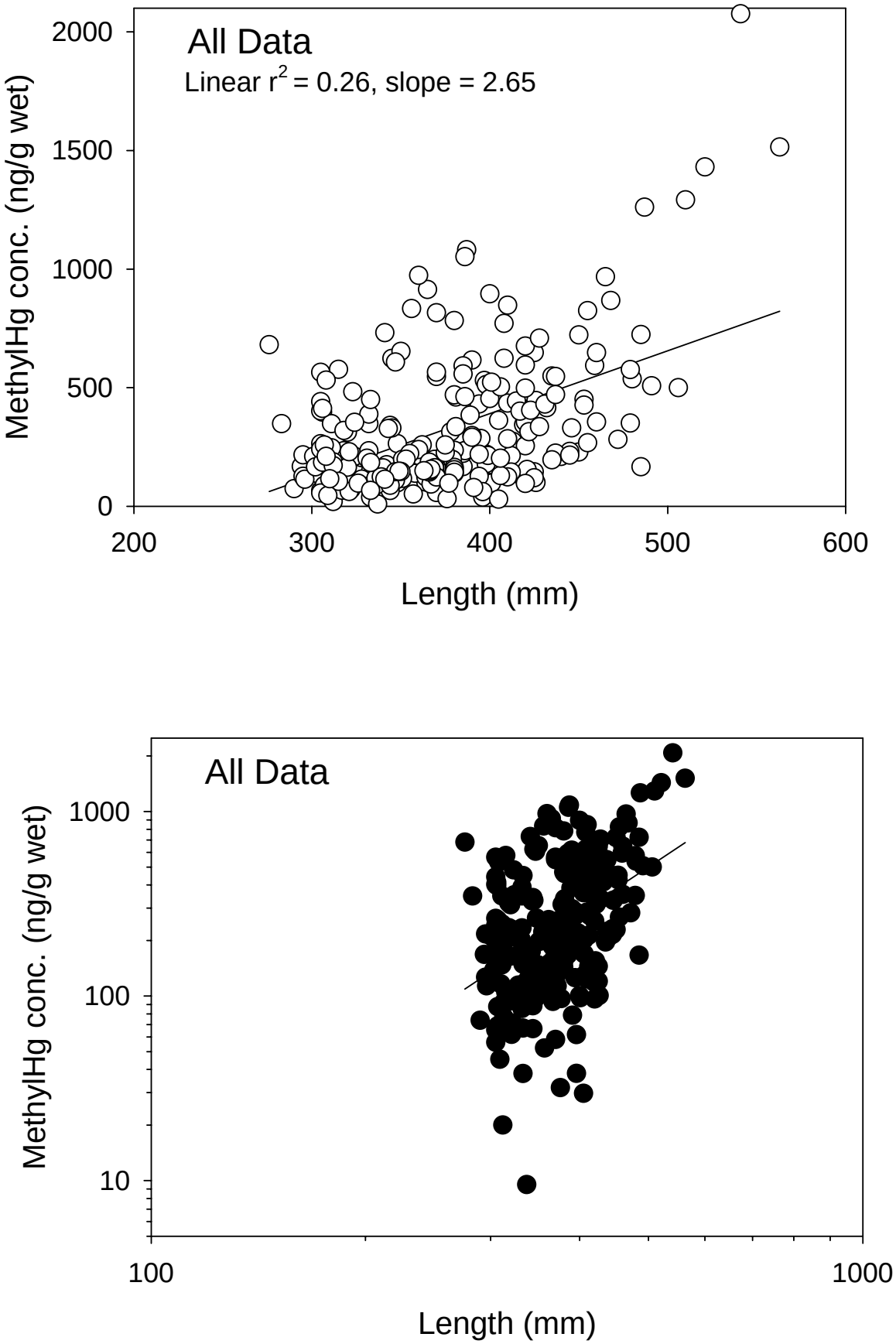


Fig. 3.4: Relationship between methylmercury in bass (ng/g wet weight) and fish length (mm) for all reservoirs sampled. Data shown both as a linear and log-log relationship. Correlations for a linear relationship are given in the top figure ($p<0.01$)

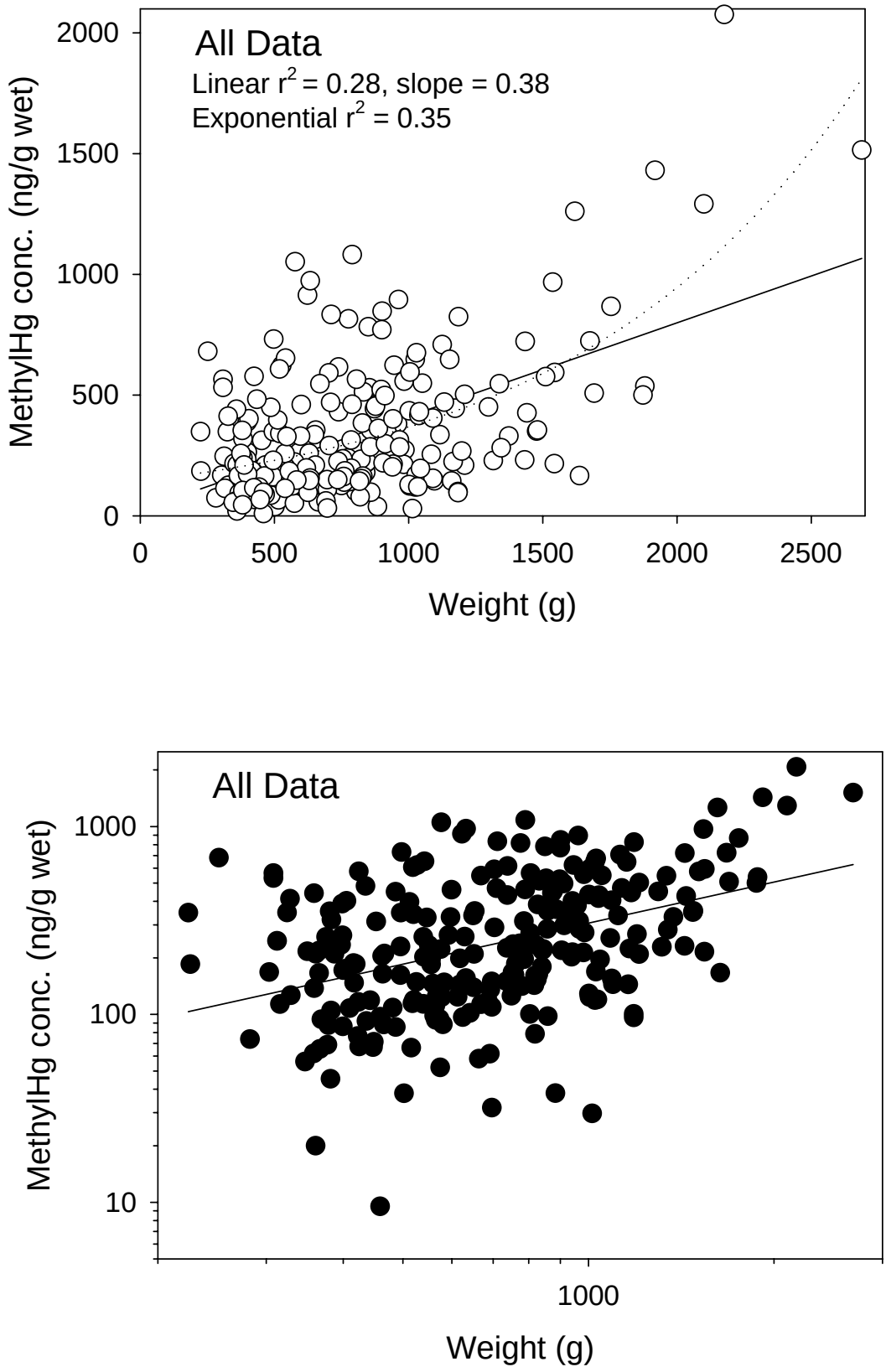


Fig. 3.4 cont: Relationshiop between methylmercury in bass (ng/g wet weight) and fish weight (g) for all reservoirs sampled. Data shown both as a linear and log-log relationship. Correlations for both a linear relationship and an exponentrial relationship are given in the top figure. Both are significant ($p<0.01$)

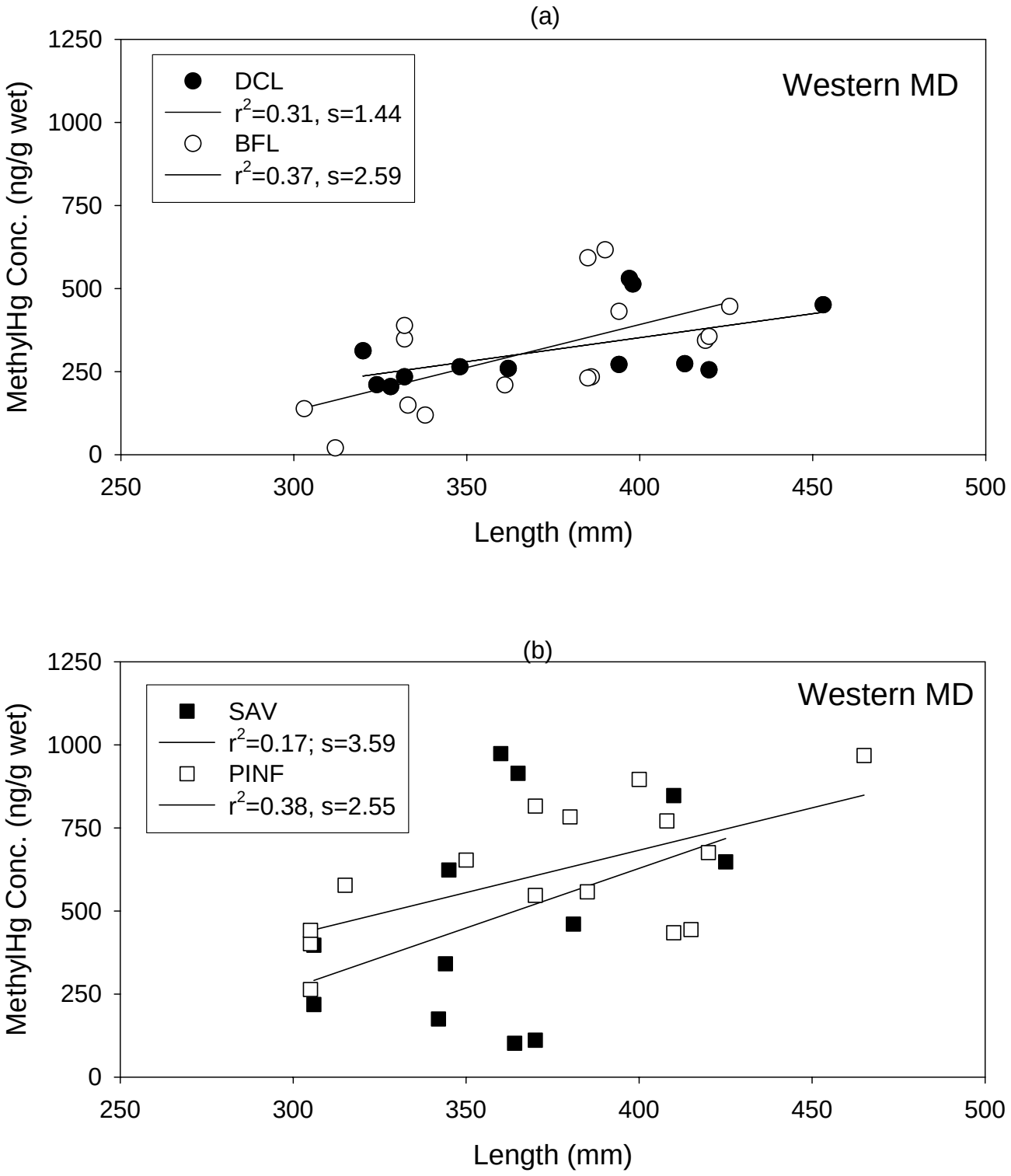


Figure 3.5: Relationship between methylmercury in bass (ng/g wet weight) and fish length (mm) for reservoirs in Maryland. Regression coefficients and slope of regression are given for significant relationships ($p<0.05$). NC = no correlation. Reservoirs are (a) DCL = Deep Creek Lake, BFL = Broadford Lake; (b) SAV = Savage Reservoir, PINF = Big Piney Reservoir outside Frostburg

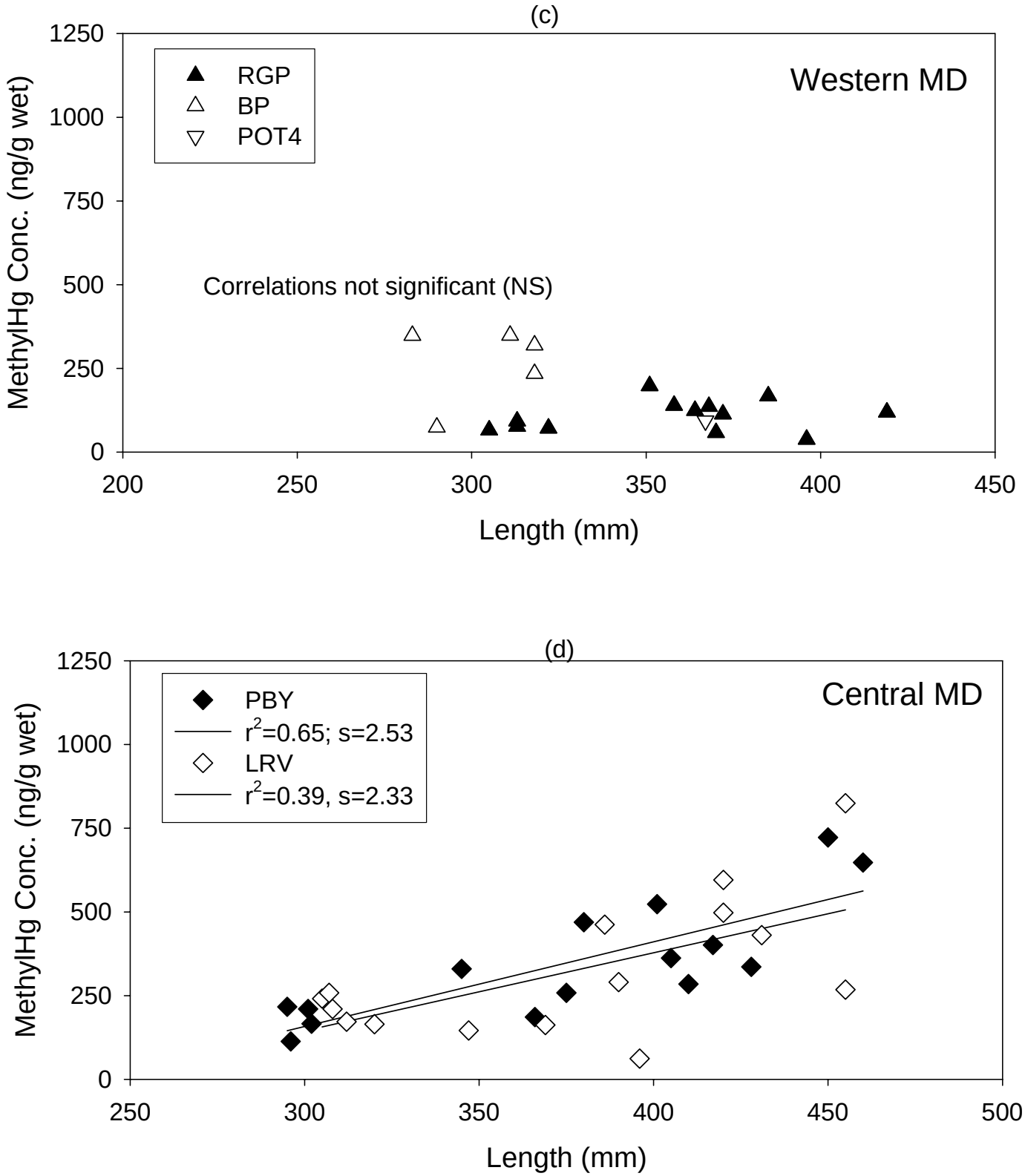
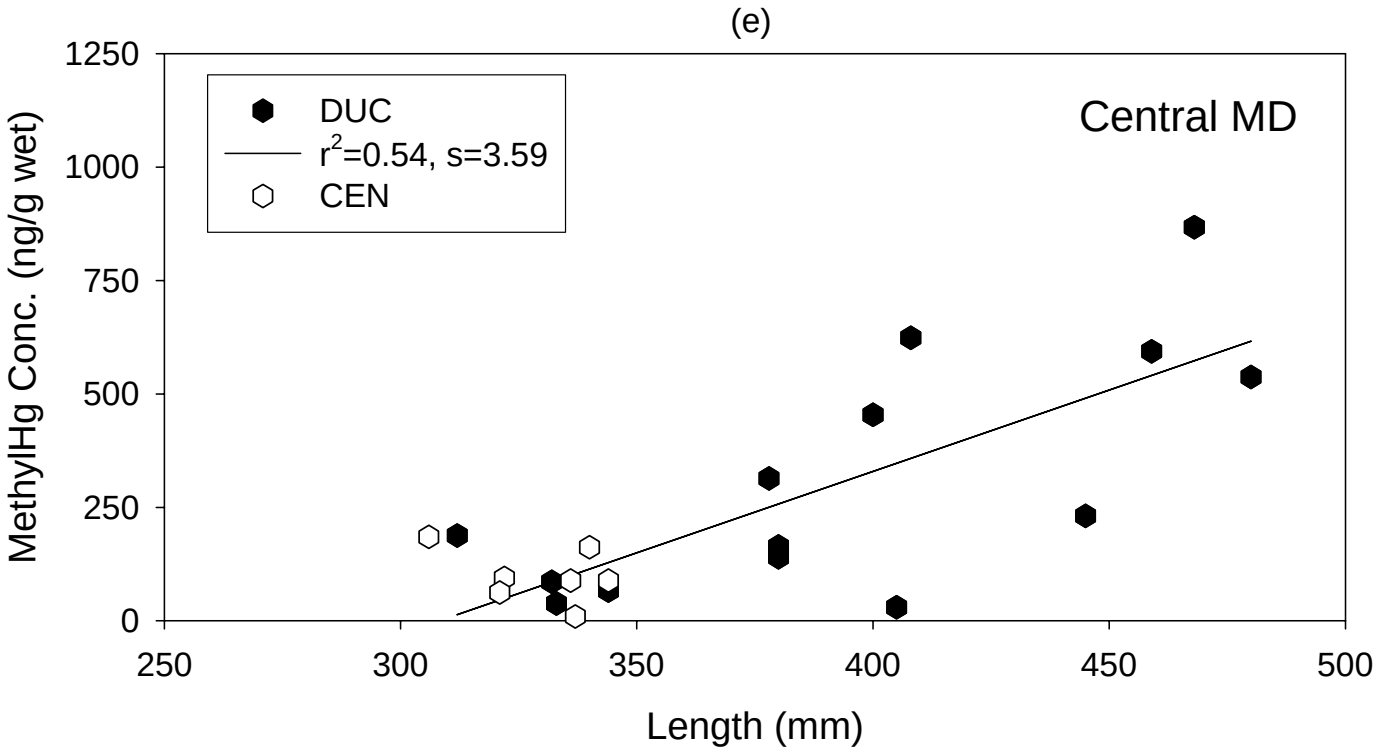


Figure 3.5 cont.: Relationship between methylmercury in bass (ng/g wet weight) and fish length (mm) for reservoirs in Maryland. Regression coefficients and slope of regression are given for significant relationships ($p<0.05$). NC = no correlation. Reservoirs are (c) RGP = Rocky Gap Park, BP = Big Pool and POT4 = Potomac #4, both on the Potomac River; (d) PBY = Prettyboy Reservoir, LRV = Loch Raven



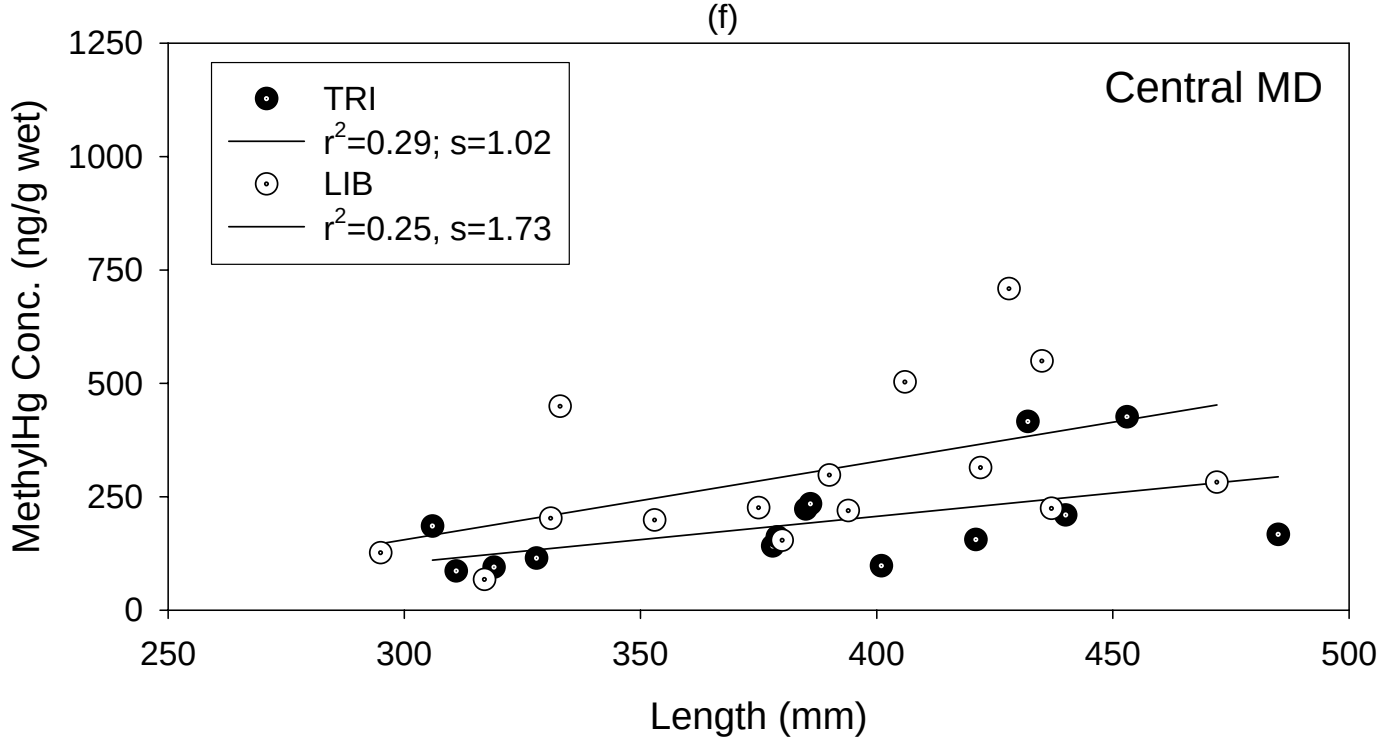
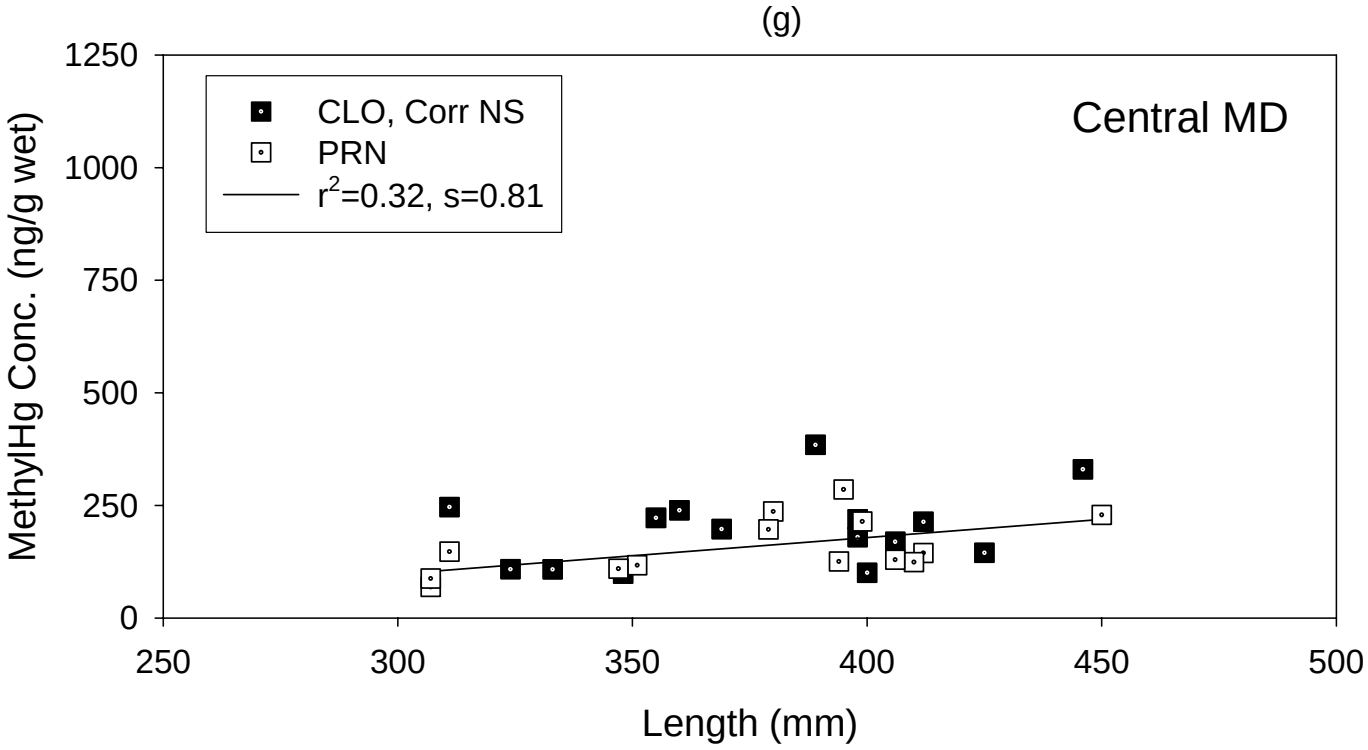


Figure 3.5 cont.: Relationship between methylmercury in bass (ng/g wet weight) and fish length (mm) for reservoirs in Maryland. Regression coefficients and slope of regression are given for significant relationships ($p<0.05$). NC = no correlation. Reservoirs are (e) DUC = Duckett lake, CEN = Centennial Lake; (f) TRI = Tridelphia Reservoir, LIB = Liberty Reservoir



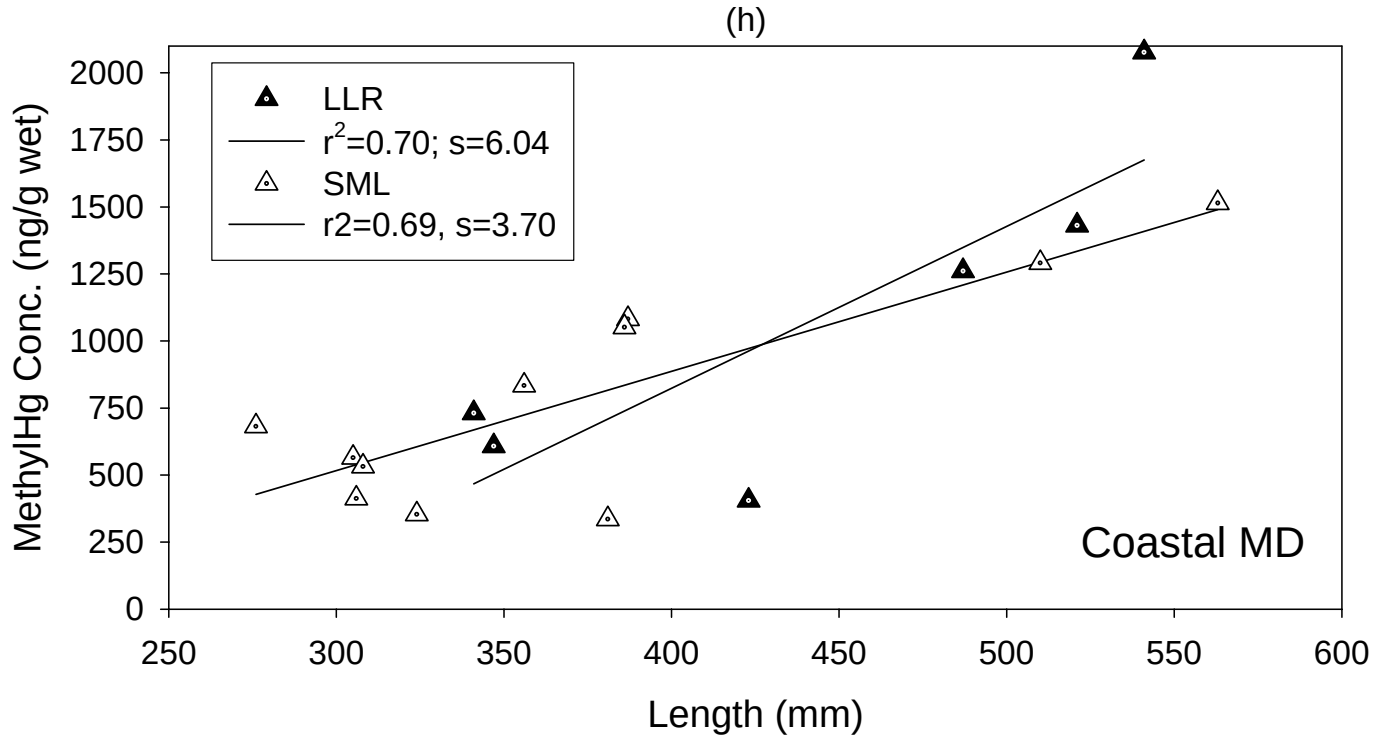


Figure 3.5 cont.: Relationship between methylmercury in bass (ng/g wet weight) and fish length (mm) for reservoirs in Maryland. Regression coefficients and slope of regression are given for significant relationships ($p<0.05$). NC = no correlation. Reservoirs are (g) CLO = Clopper Lake, PRN = Piney Run Reservoir; (h) LLR = Lake Lariat, SML = St Marys Lake

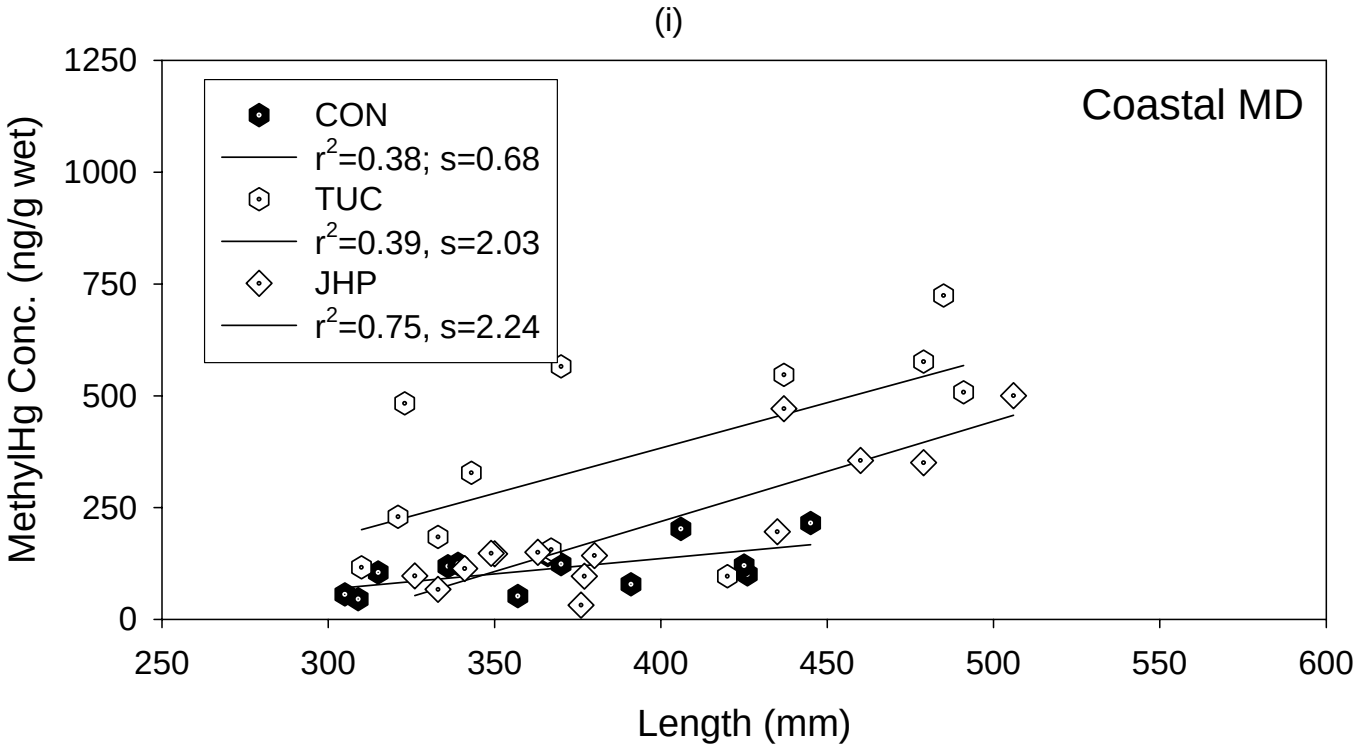


Figure 3.5 cont: Relationship between methylmercury in bass (ng/g wet weight) and fish weight (g) for reservoirs in Maryland. Regression coefficients and slope of regression are given for significant relationships ($p<0.05$). NC = no correlation. Reservoirs are (i) CON = Conowingo Dam, TUC = Tuckahoe Reservoir and JHP = Johnsons Pond.

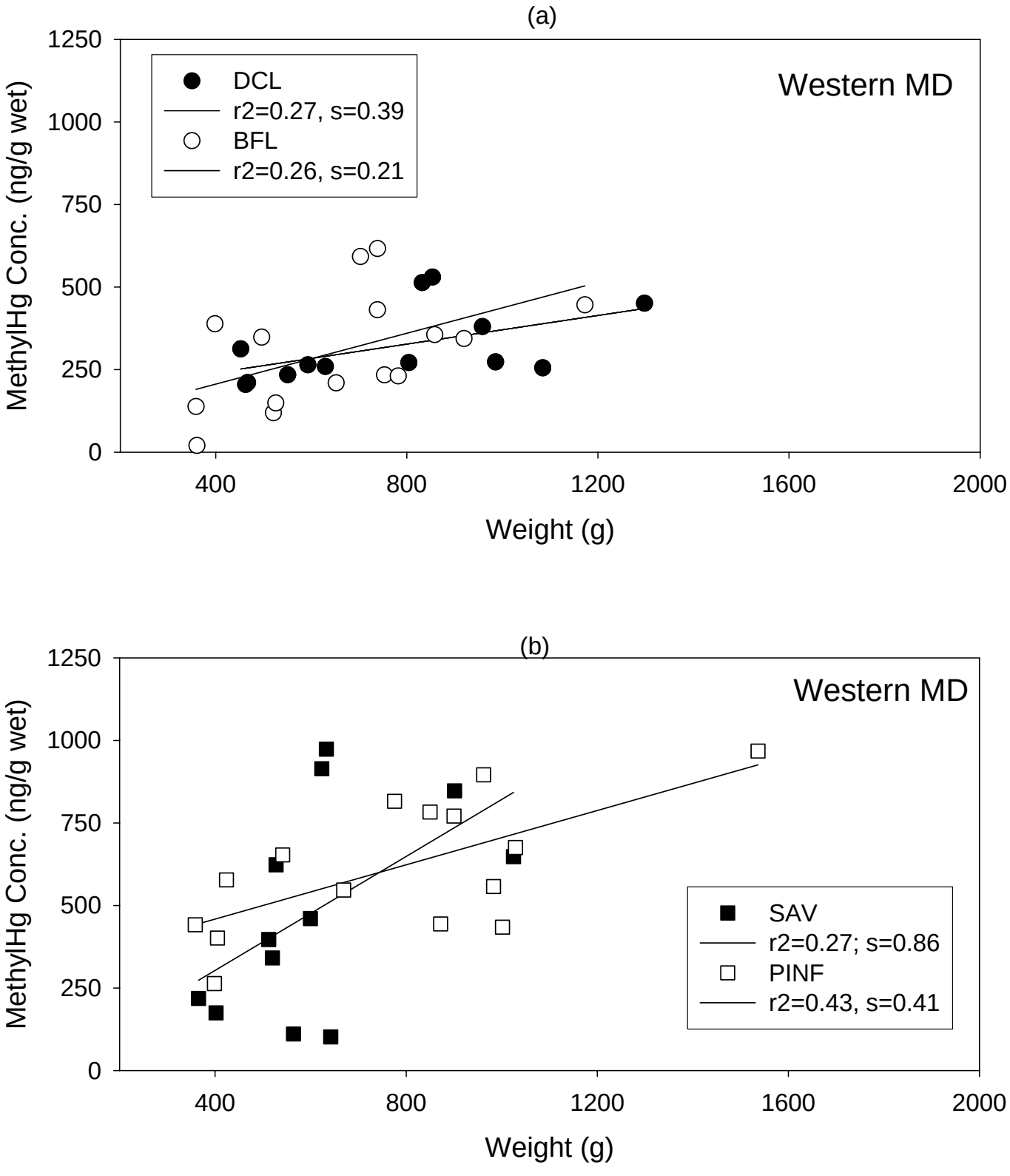


Figure 3.6: Relationship between methylmercury in bass (ng/g wet weight) and fish weight (g) for reservoirs in Maryland. Regression coefficients and slope of regression are given for significant relationships ($p<0.05$). NC = no correlation. Reservoirs are (a) DCL = Deep Creek Lake, BFL = Broadford Lake; (b) SAV = Savage Reservoir, PINF = Big Piney Reservoir outside Frostburg

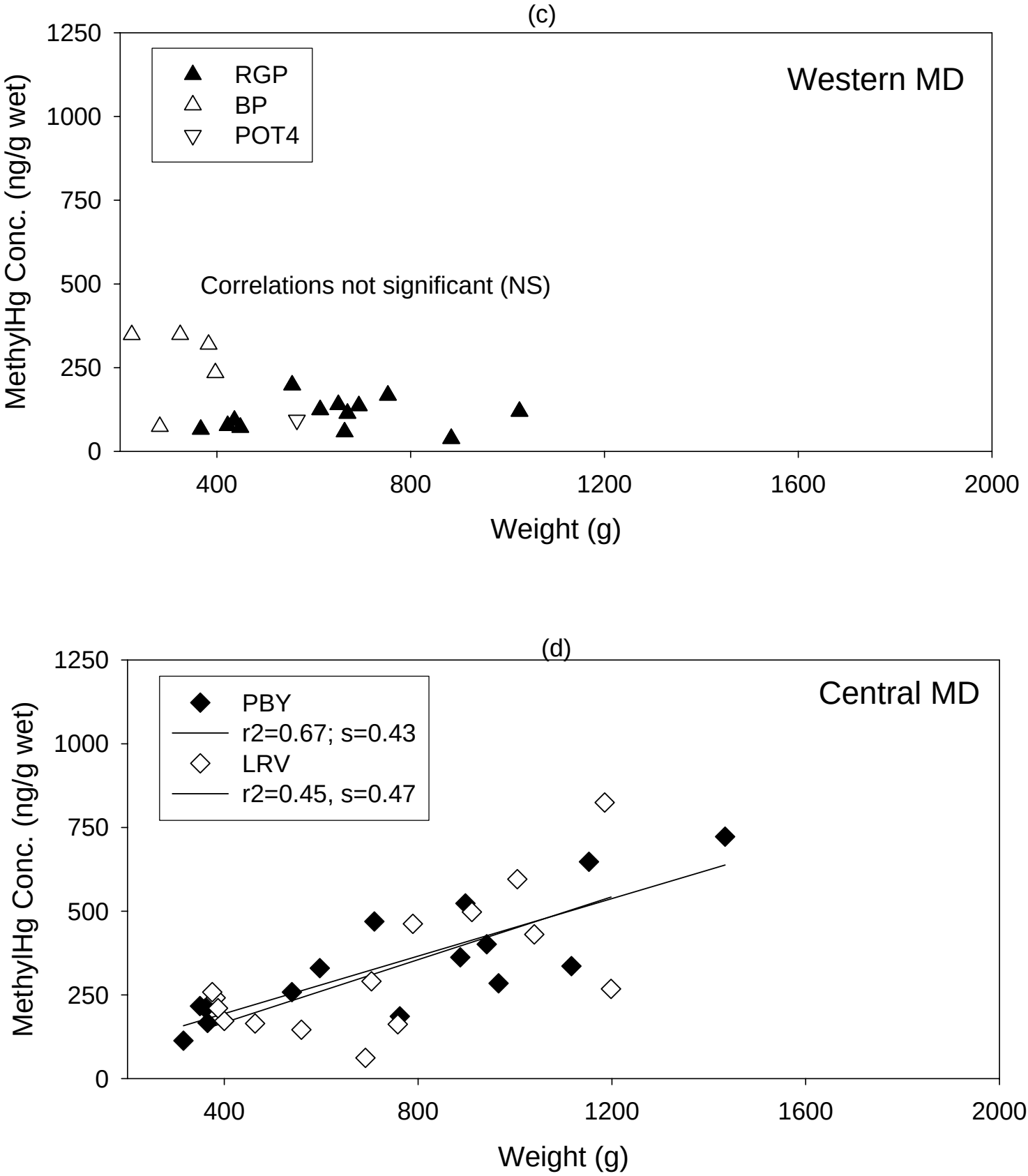


Figure 3.6 cont.: Relationship between methylmercury in bass (ng/g wet weight) and fish weight (g) for reservoirs in Maryland. Regression coefficients and slope of regression are given for significant relationships ($p<0.05$). NC = no correlation. Reservoirs are (c) RGP = Rocky Gap Park, BP = Big Pool and POT4 = Potomac #4, both on the Potomac River; (d) PBY = Prettyboy Reservoir, LRV = Loch Raven

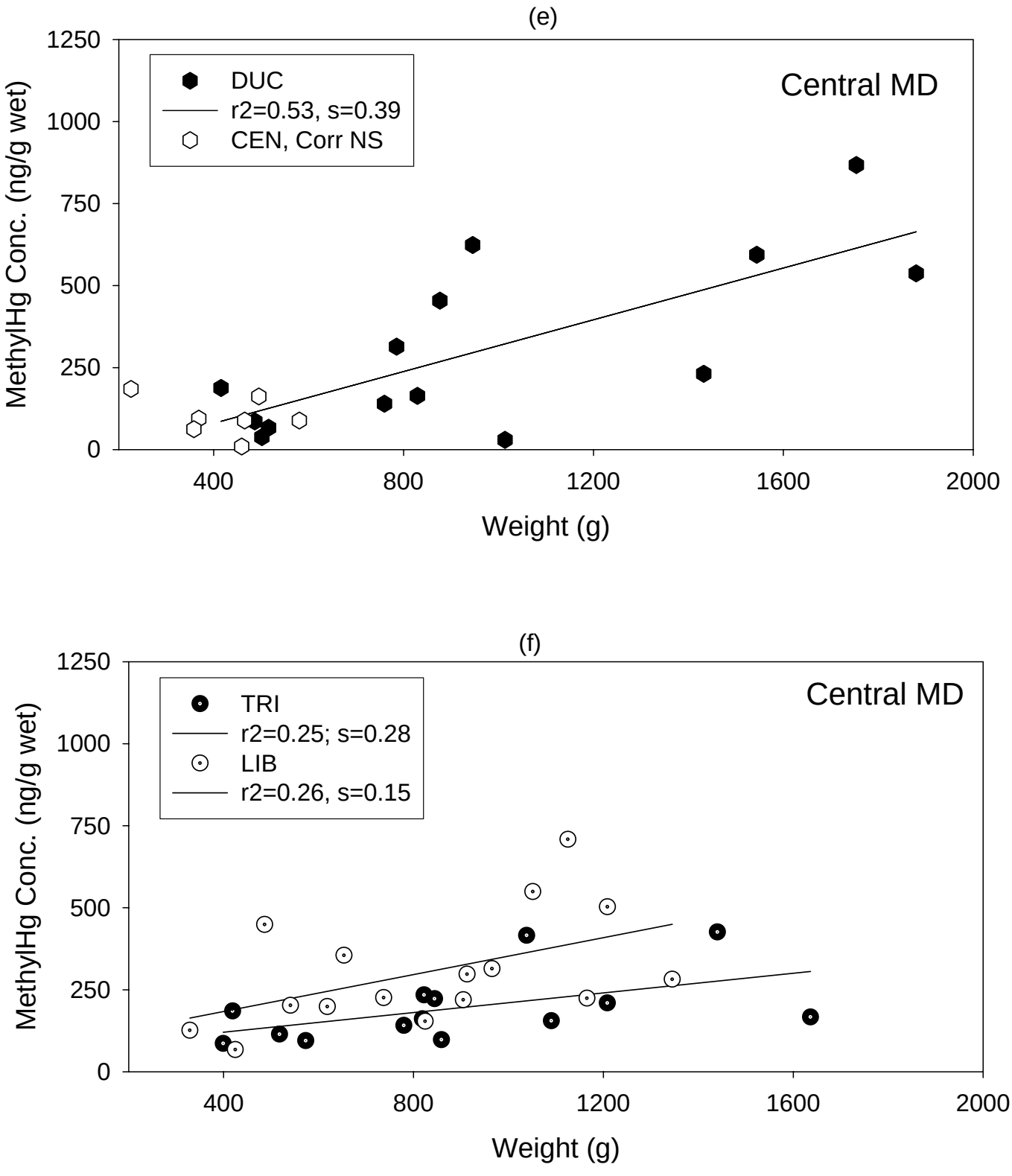
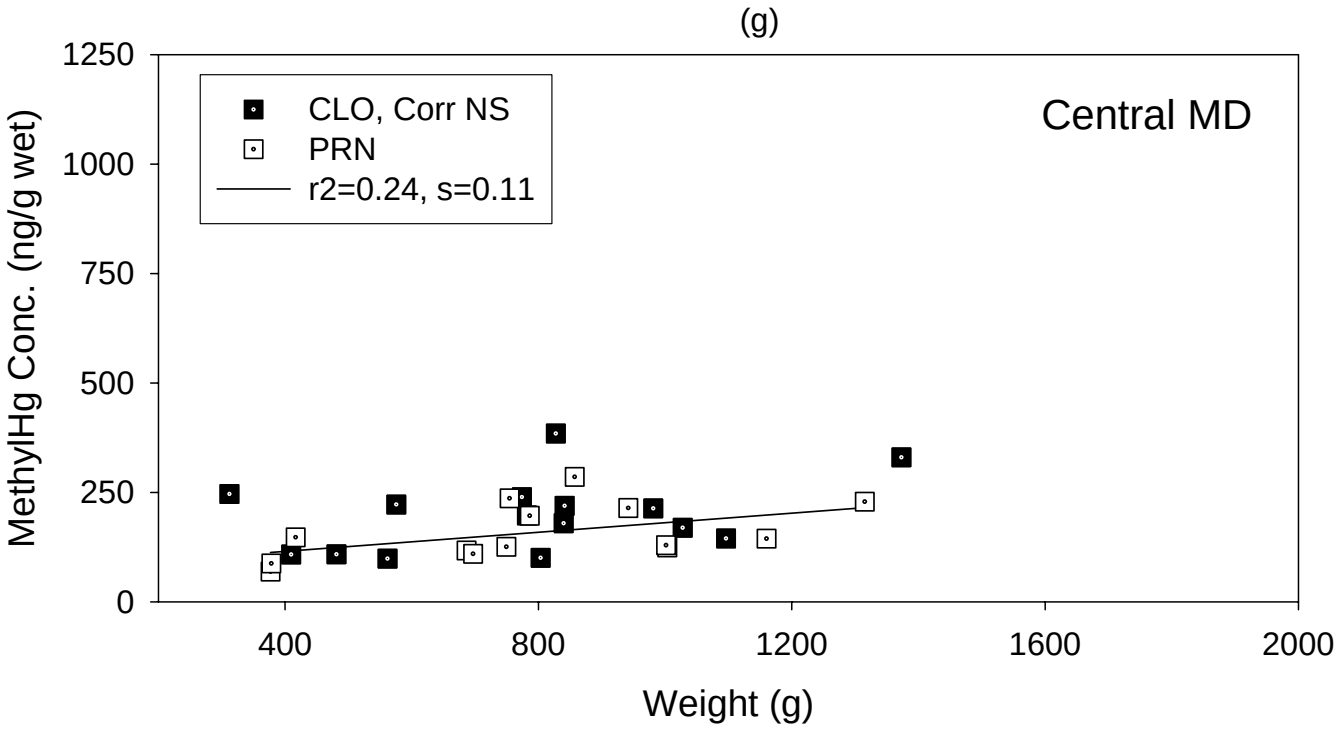


Figure 3.6 cont.: Relationship between methylmercury in bass (ng/g wet weight) and fish weight (g) for reservoirs in Maryland. Regression coefficients and slope of regression are given for significant relationships ($p<0.05$). NC = no correlation. Reservoirs are (e) DUC = Duckett lake, CEN = Centennial Lake; (f) TRI = Tridelphia Reservoir, LIB = Liberty Reservoir



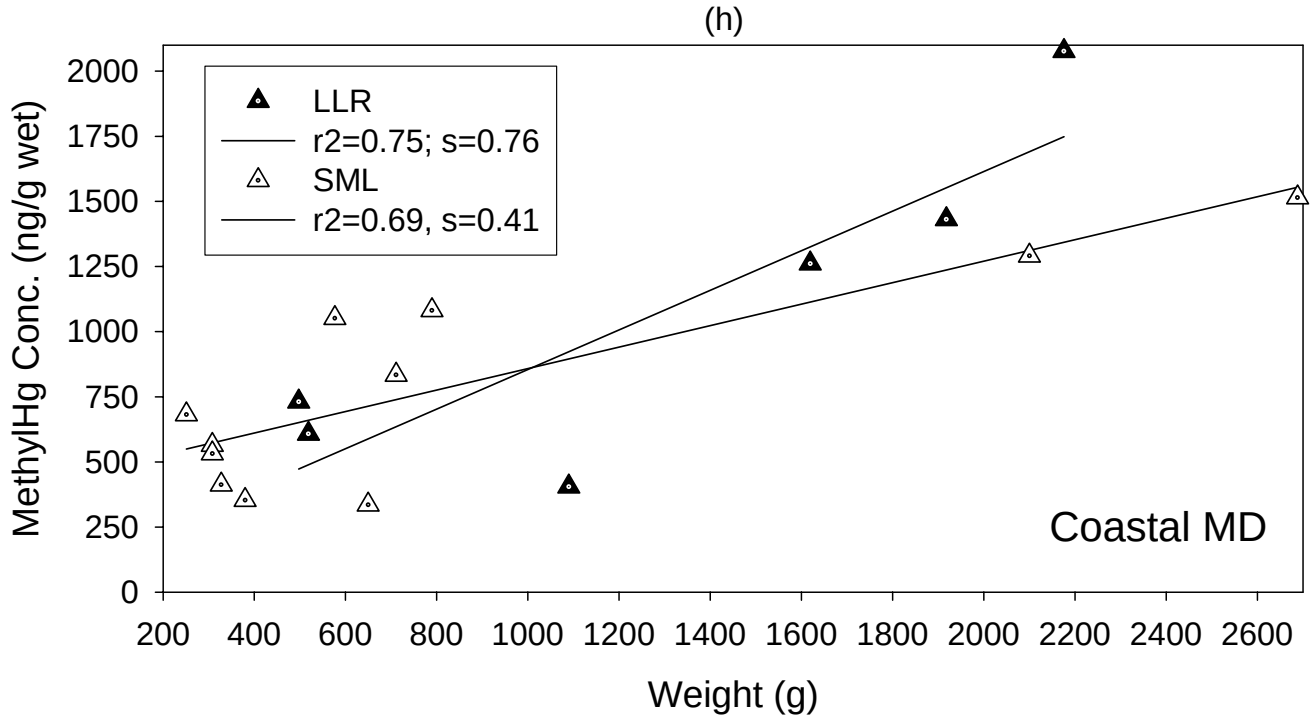


Figure 3.6 cont.: Relationship between methylmercury in bass (ng/g wet weight) and fish weight (g) for reservoirs in Maryland. Regression coefficients and slope of regression are given for significant relationships ($p<0.05$). NC = no correlation. Reservoirs are (g) CLO = Clopper Lake, PRN = Piney Run Reservoir; (h) LLR = Lake Lariat, SML = St Marys Lake

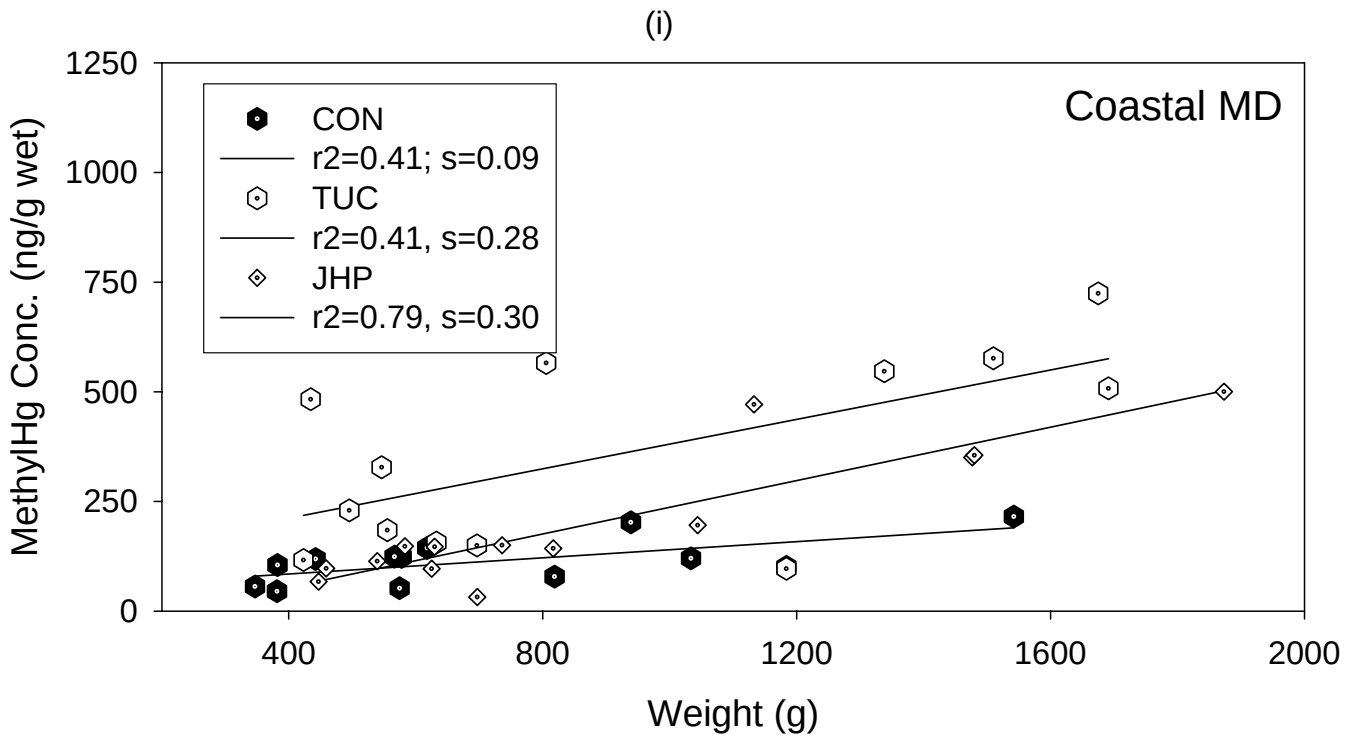


Figure 3.6 cont: Relationship between methylmercury in bass (ng/g wet weight) and fish weight (g) for reservoirs in Maryland. Regression coefficients and slope of regression are given for significant relationships ($p<0.05$). NC = no correlation. Reservoirs are (i) CON = Conowingo Dam, TUC = Tuckahoe Reservoir and JHP = Johnsons Pond.

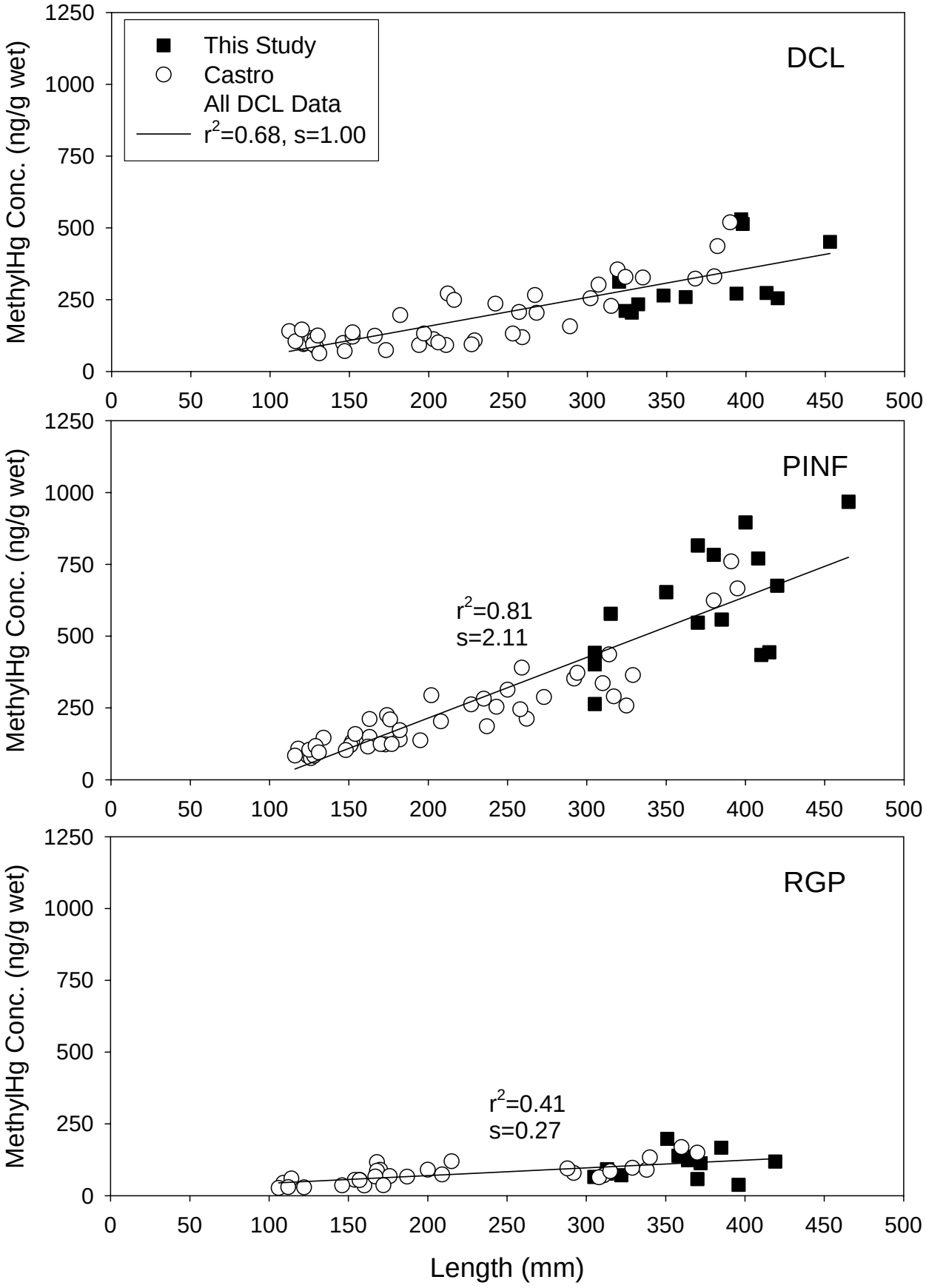


Figure 3.7. Comparison of data, in terms of concentration and length, generated in this study with that of Castro et al. (2002) for three reservoirs

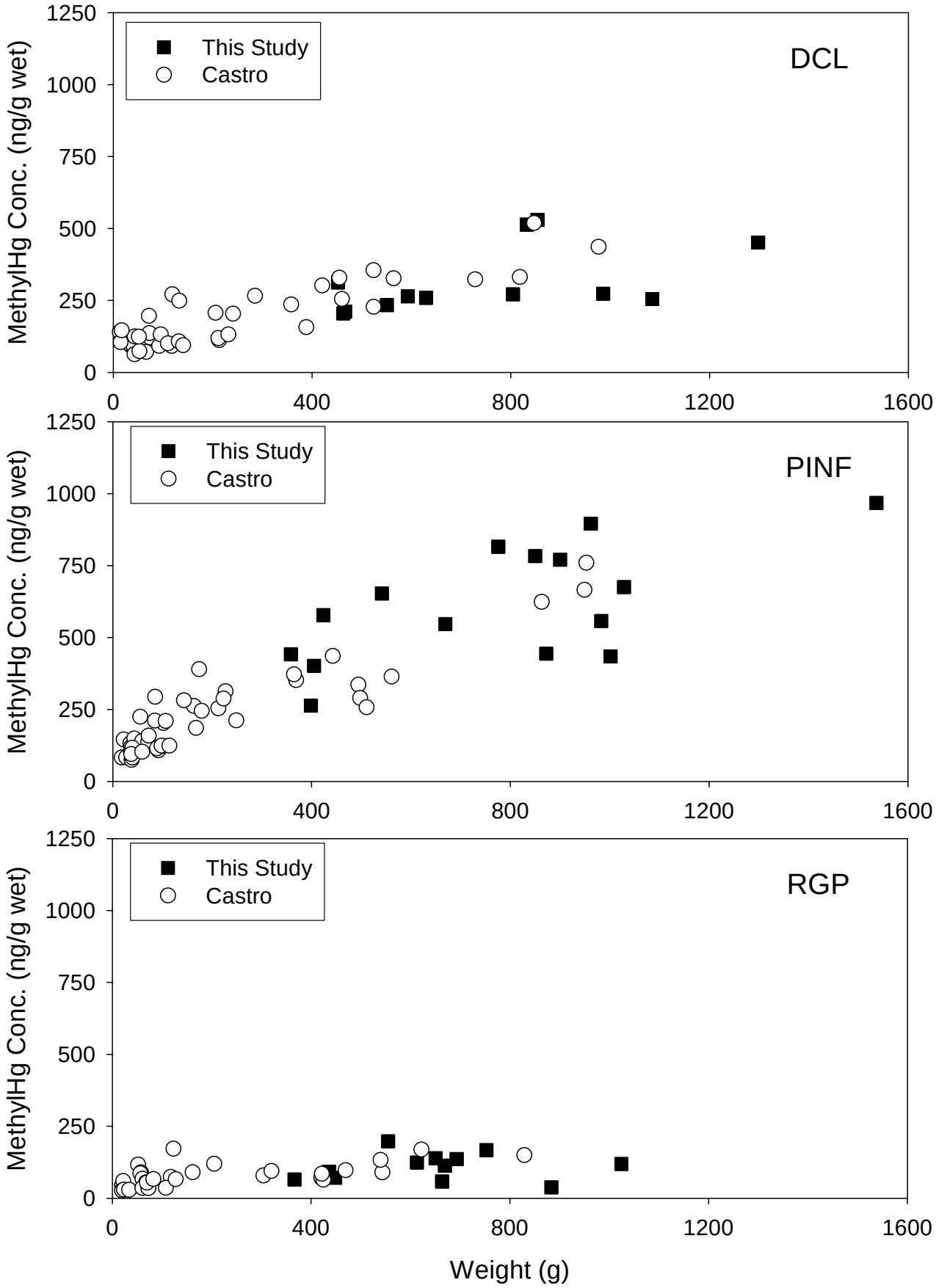


Figure 3.7cont. Comparison of data, in terms of concentration and weight, generated in this study with that of Castro et al. (2002) for three reservoirs

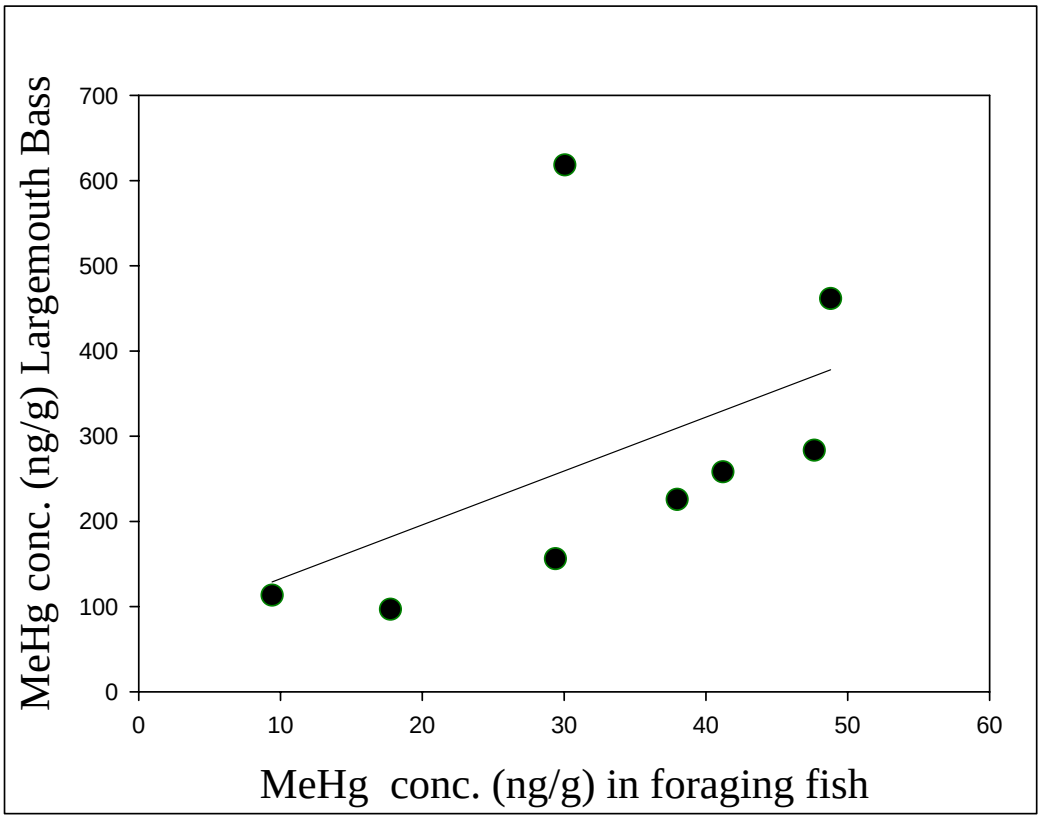


Figure 3.8: Relationship between methylmercury concentrations in bass of standardized length and that in small bluegills from the same reservoir. For the bluegills, whole fish were analyzed and concentrations represent that of a composite sample of at least 10 fish. The correlation is not significant if the one high value, for PINF is included. Without this datapoint, $r^2 = 0.74$ ($p<0.05$).

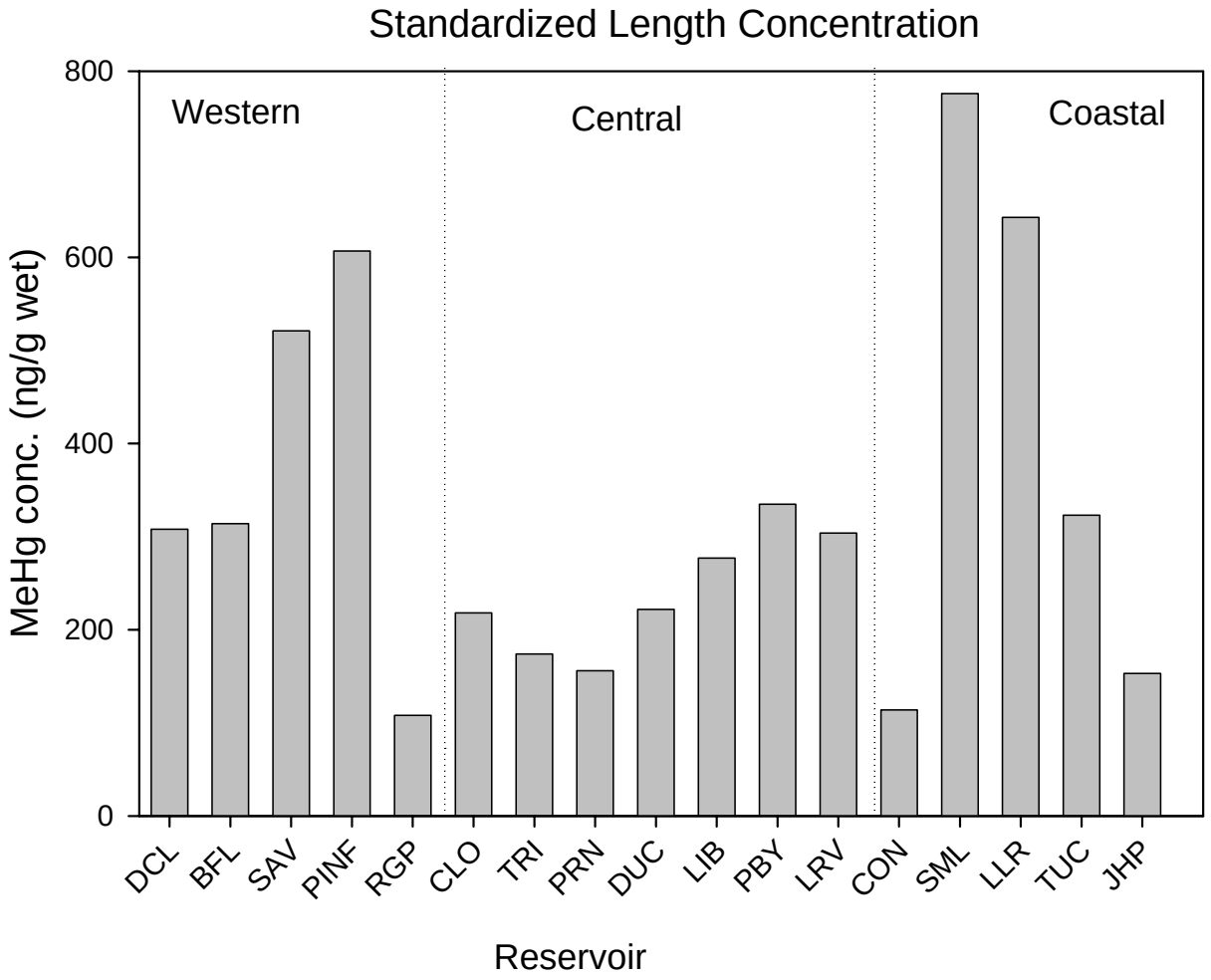


Figure 3.9. Calculated MeHg standard length concentrations (ng/g wet weight) of largemouth bass in reservoirs in Maryland where sufficient fish were analyzed to make the estimation. n=17.

REFERENCES

Allen-Gil, S.M., Gilroy, D.J. and Curtis, L.R. 1995. An ecoregion approach to mercury bioaccumulation by fish in reservoirs. *Arch. Environ. Contam. Toxicol*. 28:61-68.

Anderson, M.R., Scruton, D.A., Williams, U.P., and Payne, J.F. 1995. Mercury in fish in the Smallwood reservoir, Labrador, Canada, twenty- one years after impoundment. *Water Air Soil Poll*. 80:927-930.

APHA. 1975. Method 208 D. Total Nonfilterable Residue Dried at 103-105 C (Total Suspended Matter). *In:* Standard methods for the examination of water and wastewater, 14th Edition. American Public Health Association. Washington, D.C: 1193pp.

Becker, D.S., and Bigham, G.N. 1995. Distribution of mercury in the aquatic food web of Onondaga Lake, New York. *Water Air Soil Poll*. 80:563-571.

Benoit, J.M., C.C. Gilmour, A. Heyes, R.P. Mason and C.L. Miller. 2002. Geochemical and biological controls over methylmercury production and degradation in aquatic ecosystems. *In:* Biogeochemistry of environmentally important trace elements. American Chemical Society Publ., ACS Symposium Series 835, pp. 262-297.

Benoit, JM., Gilmour, CC., Mason, RP., Riedel, GS. and Riedel, GF. 1998. Behavior of mercury in the Patuxent estuary. *Biogeochem.* 40:249-265.

Bodaly, R.A., St. Louis, V.L., Paterson, M.J., Fudge, R.J.P., Hall, B.D., Rosenberg, D.M., and Rudd, J.W.M. 1997. Bioaccumulation of mercury in the aquatic food chain in newly flooded areas. *In* Metal ions in biological systems. Vol. 34. Mercury and its effects on environment and biology. Sigel, A. and Sigel, H. (eds). Marcel Dekker, Inc., New York, pp. 259-287.

Bodaly, R.A., Rudd, J.W.M, and Fudge, R.J.P. 1993. Mercury concentrations in fish related to size of remote Canadian Shield lakes. *Can. J. Fish. Aquat. Sci*. 50:980-987.

Bodaly, R.A., Hecky, R.E., and Fudge, R.J.P. 1984. Increases in fish mercury levels in lakes flooded by the Churchill River diversion, northern Manitoba. *Can. J. Fish. Aquat. Sci.* 41:682-691.

Bloom, N.S. 1992. Mercury and methylmercury in individual zooplankton: Implications for bioaccumulation. *Limnol. Oceanogr*. 37(6):1313-1318.

Bloom, N.S. 1989. Determination of picogram levels of methylmercury by aqueous phase ethylation, followed by cryogenic gas chromatography with cold vapor atomic fluorescence detection. *Can..J. Fish. Aqua. Sci.* 46:1131-1140.

Bloom, N.S., and Fitzgerald, W.F. 1988. Determination of volatile species at the picogram level by low temperature gas chromatography with cold vapor atomic fluorescence detection. *Analytical Chimica Acta* 208:151-161.

Bloom, N.S. and Crecelius, E.A. 1983. Determination of mercury in seawater and subnanogram per liter levels. *Mar. Chem.* 14:49-59.

Castro, M.S., McLaughlin, E.N., Davis, S.L., and Morgan II, R. 2002. Total Mercury Concentrations in Lakes and Fish of Western Maryland, USA. *Arch. Environ. Contam. Toxicol.* 42: 454-462.

Cope, W.G., Wiener, J.G., and Rada, R.G. 1990. Mercury accumulation in yellow perch in Wisconsin seepage lakes: Relation to lake characteristics. *Environ. Toxicol. Chem.* 9:931-940.

Choi, M.H., Cech, J.J. and Lagunas-Solar, M.C. 1998. Bioavailability of methylmercury to Sacramento blackfish (*Orthodon microlepidotus*): dissolved organic carbon effects. *Environ. Toxicol. Chem.* 17: 695-701.

Choi, B.H. 1990. Effects of Methylmercury on the Developing Brain. In Suzuki T, Imura N, Clarkson TW, eds, Advances in Mercury Toxicology, Plenum, New York, NY, USA, pp 315-337.

Downs, S.G., MacLeod, C.L. and Lester, J.N. 1998. Mercury in precipitation and its relation to bioaccumulation in fish: A literature review. *Water Air Soil Poll.* 108:149-187.

Driscoll, C.T., Blette, V., Yan, C., Schofield, C.L., Munson, R., and Holsapple, J. 1995. The role of dissolved organic carbon in the chemistry and bioavailability of mercury in remote Adirondack Lakes. *Water Air Soil Poll.* 80:499-508.

Dyrssen, D. and Wedborg, M. 1991. The sulfur-mercury (II) system in natural waters. *Water Air Soil Poll*. 56:507-519.

Elser, H.J. 1962. Growth rates of Maryland's freshwater fish. Nat. Res. Inst. Univ. of M.D. Ref. No.62-13.

French, K.J., Scruton, D.A, Anderson, M.R. and Schneider, D.C. 1999. Influence of Physical and Chemcial Characteristics on Mercury in Aquatic Sediments. *Water Air Soil Poll.* 110: 347-362.

Gilmour, C.C. 1999. A preliminary survey of size-specific mercury concentrations in game fish from Maryland fresh and estuarine waters. *Chesapeake Bay and Watershed Programs: Monitoring and Non-Tidal Assessment CBWP-MANTA-AD-98-9.*

Gilmour, C.C., Henry, E.A., and Mitchell, R. 1992. Sulfate stimulation of mercury methylation in freshwater sediments. *Environ. Sci. Technol.* 26:2281-2287.

Grieb TM, Driscoll CT, Gloss SP, Schofield CL, Bowie GL, Porcella DB 1990. Factors Affecting Mercury Accumulation in Fish in the Upper Michigan Peninsula. *Environ. Toxicol Chem.* 9:919-930.

Hakanson, L, Nilson, A. and Andersson, T. 1988. Mercury in fish in Swedish Lakes. *Environmental Pollution* 49:145-162.

Hanten, R.P., Neumann, R.M., Ward, S.M., Carley, R.J., Perkins, C.R and Pirrie, R. 1998. Relationships between concentrations of mercury in largemouth bass and physial and chemical characteristics of Connecticut lakes. *Trans. Americ. Fish. Soci.* 127: 807-818.

Hecky, R.E., Ramsey, D.J, Bodaly, R.A., and Strange, N.E. 1991. Increased methylmercury contamination in fish in newly formed freshwater reservoirs. *In* T. Suzuki, N. Imura, and T.W. Clarksons (eds) Advances in mercury toxicology. Plenum Press, New York, NY.

Heyes, A. and Gilmour, C.C. 1999. The biogeochemical controls on mercury methylation across ecosystems. ASLO Aquatic Science Meeting, Santa Fe, NM.

Horvat, M., Liang, L., and Bloom, N.S. 1993. Comparison of distillation with other current isolation methods for the determination of methylmercury compounds in low level environmental samples *Analytica Chimica Acta* 282:153-168.

Hoyer, M., Burke, J., and Keeler, G., 1995. Atmospheric sources, transport and deposition of mercury in Michigan two years of event precipitation. *Water Air Soil Poll.* 80: 199-208.

Horwitz, R.J., Ruppel, B., Wisniewski, S., Kiry, P., Her, M., and Gilmour, C. 1995. Mercury concentrations in freshwater fishes in New Jersey. *Water Air Soil Poll.* 80:885-888.

Hudson, R. J. M., Gherini, S.A., Watras, C.J. and Porcella, D.P. 1994. Modeling the biogeochemical cycle of mercury in lakes: The mercury cycling model (MCM) and its application to the MTL study lakes. *In* C. J. Watras and J.W. Huckabee (eds), Mercury Pollution: Integration and synthesis. Lewis Publishers. Boa Raton, FL, pp. 473-526.

Hueter, R.E., Fong, W.G., Henderson, G., French, M.F, and Manire C.A. 1995. Methylmercury concentration in shark muscle by size and distribution of sharks in Florida coastal waters. *Water Air Soil Poll.* 80:893-899.

Hultberg, H., Iverfeldt, A. and Lee, Y-H. 1994. Methylmercury input/output and accumulation in forested catchments and critical loads for lakes in southwestern Sweden. *In* C. J. Watras and J.W. Huckabee (eds), Mercury Pollution: Integration and synthesis. Lewis Publishers. Boa Raton, FL, pp. 313-322.

Johnston, T.A., Bodaly, R.A., and Mathias, J.A. 1991. Predicting fish mercury levels from physical characteristics of boreal reservoirs. *Can. J. Fish. Aquat. Sci*. 48:1468-1480.

Klotz, A. and Johnson, J. 2000. Performance Report, Annual (2000) and Five Year (1996-2000), USFWS Federal Aid Grant F-48-R10, Survey and Management of Maryland Freshwater Fisheries Resources, MDNR Fisheries Service, Freshwater Fisheries Division.

Lange, T.R., Royals, H.E., and Connor, L.L. 1993. Influence of water chemistry on mercury concentration in largemouth bass from Florida lakes. *Trans. Am. Fish. Soc.* 122:74-84.

Lawrence, A.L., and Mason, R.P. 2001. Factors controlling the bioaccumulation of mercury and methylmercury by the estuarine amphipod *Leptocheirus plumulosus*. *Environ. Poll*. 111:217-231.

Lawson, N.M., and Mason, R.P. 2001. Concentration of mercury, methylmercury, cadmium, led, arsenic, and selenium in the rain and stream water of two contrasting watersheds in Western Maryland. *Wat. Res.* 35(17): 4039-4052.

Lawson, N.M., Mason, R.P. and Laporte, J-M. 2001. The fate and transport of mercury, methylmercury, and other trace metals in Chesapeake Bay tributaries. *Wat. Res.* 35(2):501-515.

Lee, Y-H. and Iverfeldt, A. 1991. Measurement of methylmercury and mercury in run-off, lake and rain waters. *Water Air Soil Poll.* 56:309-321.

Lee, Y-H. and Hulteberg, H. 1990. Methylmercury in some Swedish surface waters. *Envir. Tox. Chem.* 9:833-841.

Lindquist O. and others. 1991. Mercury in the Swedish environment- recent research on causes, consequences and corrective methods. *Water Air Soil Poll.* 55:143-177.

MacCrimmon, H.R., Wren, C.D. and Gots, B.L. 1983. Mercury uptake by lake trout, *Salelinus namaycush*, relative to age, growth, and diet in Tadenac Lake with comparative data from other PreCambrian Shield lakes. *Can. J. Fish. Aquat. Sci*. 40:114-120.

Maryland Department of Natural Resources (MDNR), 2000. Annual (2000) and final (1996-2000) performance report. Survey and management of Maryland's fishery resources. Maryland Department of Natural Resources, Fisheries Service, Freshwater Fisheries Division.

Mason, R.P. 2001. The bioaccumulation of mercury, methylmercury and other toxic elements into pelagic and benthic organisms. pp. 127-149. *In*: M.C. Newman, M.H. Robert, and R.C. Hale [eds.], *Coastal and Estuarine Risk Assessment*, CRC/Lewis Publ.

Mason, R.P. 2000. An investigation of the influence of water quality parameters on mercury, methylmercury, arsenic, selenium and cadmium concentrations in fish of Maryland streams. Final Report, Maryland Department of Natural Resources, Chesapeake Bay research and monitoring division.

Mason, R.P., J.-M. Laporte, and S. Andres. 2000. Factors controlling the bioaccumulation of mercury, methylmercury, arsenic, selenium, and cadmium by freshwater invertebrates and fish. *Arch. Environ. Contam. Toxicol.*, 38:283-297.

Mason, R.P., N.M. Lawson, and G.-R. Sheu. 2000b. Annual and seasonal trends in mercury deposition in Maryland. *Atmos. Environ*., 34:1691-1701.

Mason, R.P., and Lawrence, A.L. 1999. Concentration, distribution, and bioavailability of mercury and methylmercury in sediments of Baltimore Harbor and Chesapeake Bay, Maryland, USA. *Envir. Toxic. Chem*. 18:2438-2447.

Mason R.P., Lawson N.M., Lawrence A.L., Lee JG., Leaner JJ and Sheu GR. 1999. Mercury in the Chesapeake Bay. *Mar.Chem.* 65:77-96.

Mason, R.P., Lawson, N.M., and Sullivan, K.A. 1997a. Atmospheric deposition to the Chesapeake Bay watershed: Regional and local sources. *Atmos. Environ.* 31, 3531-3540.

Mason, R.P., Lawson, N.M., and Sullivan, K.A. 1997b. The concentration, speciation and sources of mercury in Chesapeake Bay precipitation. *Atmos. Environ.* 31(21): 3541-3550.

Mason R.P., and Sullivan, K.A. 1997. Mercury in Lake Michigan. *Environ. Sci. Technol.* 31:942-947.

Mason, R.P., Reinfelder, J.R, and Morel F.M. 1996. Uptake, toxicity, and trophic transfer of mercury in a coastal diatom. *Environ. Sci. Technol.* 30(6):1835-1845.

Mason, R.P., Reinfelder, J.R. and Morel, F.M. 1995. Bioaccumulation of mercury and methylmercury. *Water Air Soil Poll.* 80:915-921.

Mason, R.P., Fitzgerald, W.F., and Morel, F.M 1994. The biogeochemical cycling of elemental mercury: anthropogenic influences. *Geochim. Cosmochim. Acta* 58(15): 191-3198.

Mathers, R.A. and Johansen, P.H. 1985. The effect of feeding ecology on mercury accumulation in walleye (*Stizostion vitreum*) and pike (*Esox lucius*) in lake Simcoe. Can. J. Zool. 62:2006-2012.

Meili, M. 1994. Mercury in lakes and rivers. *In:* Watras, C.J., Huckabee, J.W. (eds.). Mercury Pollution-integration and Synthesis. Lewis Publication, Boca Raton, FL, pp.21-49.

Pfaff, J.D., Brockhoff, C.A. and O'Dell, J.W. 1991. USEPA Test Method no. 300.0. The determination of inorganic anions in water by ion chromatography. Environmental Monitoring and Systems Laboratory, Cincinnati, OH 45268.

Rasmussen, PE. 1994. Current Methods of estimating atmospheric mercury fluxes in remote areas. *Environ. Sci. Technol.* 28: 2233-2241.

Richman, L.A., Wren, C.D. and Stokes, P.M. 1988. Facts and fallacies concerning mercury uptake by fish in acid stressed lakes. *Water Air Soil Poll.* 37:465-473.

Rodgers, D.W. 1994. You are what you eat and a little bit more: Bioenergetics-based models of methylmercury accumulation in fish revisited. *In*: C.J. Watras and J.W. Huckabel (eds). Mercury Pollution: Integration and Synthesis. Lewis Publishers, NY. Pp 427-439.

Rodgers, D.W. and Beamish, F.W.H. 1983. Water quality modifies uptake of waterborne methylmercury by rainbow trout, *Salmo gairdneri. Can. J. Fish. Aquat. Sci.* 40:824-828.

Rolfhus, KR and Fitzgerald WF. 1995. Linkages between atmospheric mercury deposition and the methylmercury content of marine fish. *Water, Air, Soil Poll* 80:291-297.

Rose, J., Hutcheson, M.S., West, C.R., Pancorbo, O., Hulme, K., Cooperman, A., DeCesare, G. Isaac, R. and Screpetis, A. 1999. Fish Mercury Distribution in Massachusetts, USA Lakes. *Environ. Toxicol. Chem.* 18(7):1370-1379.

Rudd, J.W.M. 1995. Sources of Methylmercury to Freshwater Ecosystems: A Review. *Water, Air, Soil Poll.* 80: 697-713.

Simonin, H.A., Gloss, S.P., Driscoll, C.T., Schofield, C.L, Kretser, W.A., Karcher, R.W. and Symula J. 1994. Mercury in Yellow Perch from Adirondack Drainage Lakes (New York, U.S.). *In*: Watras, C.J., Huckabee, J.W. (eds). Mercury Pollution-Integration and Synthesis. Lewis Publications, Boca Raton, FL, pp. 457-469.

Snodgrass, J.W., Jagoe, C.H., Bryan Jr., A.L., Brant, H.A., and Burger, J. 2000. Effects of trophic status and wetland morphology, hydroperiod, and water chemistry on mercury concentrations in fish. *Can. J. Fish. Aquat.Sci*. 57:171-180.

Somers, K.M., and Jackson, D.A. 1993. Adjusting mercury concentration for fish-size covariation: a multivariate alternative to bivariate regression. *Can. J. Fish. Aqua. Sci.* 50:2388-2396.

Sonesten, L. 2001. Mercury content in roach (*Rutilus rutilus* L.) in circumneutral lakes – effects of catchment area and water chemistry. *Environ. Poll.* 112: 471-481.

St. Lois, V.L., Rudd, J.W.M, Kelly, C.A., Beaty, K.G. Bloom, N.S., and Flett, R.J. 1994. *Can. J. Fish. Aquat. Sci*. 51:1065-1076.

Stafford, C.P., and Haines. T.A. 1997. Mercury concentrations in Maine sport fishes. *Trans. Amer. Fish. Soci.* 126:144-152.

Stumm, W. and Morgan, JJ. 1996. Trace metals: cycling, regulation, and biological role. *In*: Schnoor J., Zehnder A. (eds) Aquatic chemistry: chemical equilibria and rates in natural waters, 3d ed. John Wiley & Sons, Inc., New York, pp 614-671.

Sugimura, Y. and Suzuki, Y. 1988. A high temperature catalytic oxidation method for the determination of non-volatile dissolved organic carbon in seawater by direct injection of a liquid sample. *Mar. Chem.* 24:105-131.

Spry, D.J., and Wiener, J.G. 1991. Metal bioavailability and toxicity to fish in low-alkalinity lakes: A critical review. *Environ. Pollut*. 71:243-304.

Suns, K., and Hitchin, G. 1990. Interrelationships between mercury levels in yearling yellow perch, fish condition and water quality. *Water Air Soil Poll*. 650:255-265.

Sveinsdottir, A.Y. 2002. Methylmercury in largemouth bass (*Micropterus salmoides*) and forage fish from Maryland reservoirs and factors influencing uptake. Masters Thesis. University of Maryland, Chesapeake Biological Laboratory.

U.S Environmental Protection Agency (USEPA). February. 2001. Mercury: General information. http://www.epa.gov/mercury/information.htm.

U.S. Environmental Protection Agency (USEPA). 1999. Office of Water. EPA-823—99-016.

U.S. Environmental Protection Agency (USEPA). 1998. EPA Methods and Guidance for Analysis of Water: CD ROM. Government Institutes, Washington DC, USA.

U.S. Environmental Protection Agency (USEPA). 1995. Method 1631: Mercury in water by oxidation, purge and trap and cold vapor atomic fluorescence spectrometry.

U.S. Environmental Protection Agency (USEPA). 1995. National listing of fish consumption advisories. USEPA, EPA-823-F-95-004, Office of Water, Washington, DC, USA.

U.S. Environmental Protection Agency (USEPA). 1987. Section 11.0 Determination of chloride, nitrate, and sulfate by ion chromatography in Handbook of methods for acid deposition studies: Laboratory analysis for surface water chemistry. United States Environmental Protection Agency, Office of Research and Development. Washington D.C. Report No. EPA 600/4-87-026. September 1987. 336pp.

U.S. Environmental Protection Agency (USEPA). 1979. Method No. 160.2 in Methods for chemical analysis of water and wastes. United States Environmental Protection Agency, Office of Research and Development. Cincinnati, Ohio. Report No. EPA-600/4-79-020. March 1979. 460pp.

U.S Federal Drug Administration (USFDA). February, 2002. Consumer advisory: An Important message for pregnant women and women of childbearing age who may become pregnant about the risks of mercury in fish. http://vm.cfsan.fda.gov/~dms/admehg.html

U.S. Geological Service (USGS). Feb. 2001. http://md.water.usgs.gov/

Watras, C.J., Back, R.C., Halvorsen, S., Hudson, R.J.M., Morrison, K.A. and Wente, S.P. 1998. Bioaccumulation of mercury in pelagic freshwater food webs. *Sci. Tot. Environ.* 219:183-208.

Watras, C.J., Morrison, K.A. and Bloom, N.S. 1995a. Chemical correlates of Hg and methyl-Hg in Northern Wisconsin lake waters under ice-cover. *Water, Air, Soil Poll.* 84:253-267.

Watras, C.J., Morrison, K.A. and Host, J.D. 1995b. Concentration of mercury species in relationship to other site-specific factors in the surface waters of northern Wisconsin lakes. *Limnol. Oceanogr.* 40(3):556-565.

Watras, C.J., and Bloom, N.S. 1992. Mercury and methylmercury individual zooplankton: Implications of bioaccumulation. *Limnol. Oceanogr*. 37(6):1313-1318.

Ward, S.M and Neumann, R.M. 1999. Seasonal Variations in Concentrations of Mercury in Axial Muscle Tissue of Largemouth Bass. *N. Amer. J. Fish. Manage.* 19:89-96.

Weiss, J., Trip, L and Mahaffey, KR. 1999. Methylmercury: A New Look at the Risks. *Public Health Reports* 114: 397-401.

Wetzel, R.G. 1983. The Phosphorus Cycle. *In*: Limnology, 2nd ed. Saunders College Publishing, NY, pp. 255-257.

Wiener, J.G., Martini, R.E., Sheffy, T.B., and Glass, G.E. 1990. Factors influencing mercury concentrations in walleyes in northern Wisconsin lakes. *Trans. Am. Fish. Soci.* 115:862-870.

World Health Organization. 1990. Environmental Health Criteria 101: Mercury I. Geneva. 144 pp.

Wren, C.D., Scheider, W.A., Wales, D.L., Muncaster, B.W. and Gray, I.M. 1991. Relation between mercury concentrations in walleye (*Stizostedion vitreum vitreum*) and northern pike (*Esox lucius*) in Ontario lakes and influence of environmental factors. *Can. J. Fish. Aquat. Sci.* 48: 132-139.

Wren, C.D., and MacCrimmon, H.R. 1983. Mercury levels in sunfish, *Lepomis gibbosus*, relative to pH and other environmental variables of Precambrian Shield lakes. *Can. J. Fish. Aquat. Sci.* 40:1737-1744.

Xun, L., Campbell, N.E.R., and Rudd, J.W.M. 1987. Measurements of specific rates of net methyl mercury production in the water column and surface sediments of acidified and circumneutral lakes. *Can. J. Fish. Aquat. Sci*. 44:750-757.

APPENDIX I: Weight (g), length (mm) and methylmercury conc. (ng/g) in largemouth bass in Maryland reservoirs.

Sample ID	Collection Date	Total Weight (g)	Total Length (mm)	MeHg concentration (ng/g)
Deep Creek Lake				
1	6/30/00	462.5	328	204.8
2	6/30/00	452.2	320	312.4
3	6/30/00	466.4	324	210.6
4	6/30/00	550.5	332	234.0
5	6/30/00	592.7	348	264.3
6	6/30/00	804.3	394	271.3
7	6/30/00	629.5	362	259.2
8	6/30/00	853.7	397	530.1
9	6/30/00	832.1	398	513.3
10	6/30/00	958.3	400	380.2
11	6/30/00	985.7	413	273.1
12	6/30/00	1084.6	420	255.0
13	6/30/00	1297.6	453	451.0
Broadford Lake				
1	6/28/00	496.01	332	348.2
2	6/28/00	358.65	303	138.0
3	6/28/00	360.55	312	20.0
4	6/28/00	520.7	338	119.1
5	6/28/00	398.2	332	388.4
6	6/28/00	525.55	333	148.8
7	6/28/00	753.12	386	233.8
8	6/28/00	651.99	361	209.7
9	6/28/00	738.55	390	616.3
10	6/28/00	781.97	385	230.7
11	6/28/00	1172.91	426	445.8
12	6/24/02	738.29	394	431.13
13	6/24/02	919.63	419	343.68
14	6/24/02	703.06	385	592.14
15	6/24/02	858.29	420	355.79
Savage				
1	6/19/01	527.4	345	623.32
2	6/19/01	519.9	344	341.18
3	6/19/01	401.9	342	174.81
4	6/19/01	365.3	306	218.36
5	6/19/01	512.2	306	396.80
6	6/19/01	599.5	381	460.35
7	6/19/01	623.1	365	914.3
8	6/19/01	632.9	360	973.3
9	6/24/02	564.05	370	111.23
10	6/24/02	641.91	364	101.88
11	6/24/02	901.14	410	847.22

APPENDIX I CONTINUED.

Sample ID	Collection Date	Total Weight (g)	Total Length (mm)	MeHg concentration (ng/g)
Savage Cont.				
12	6/24/02	1024.67	425	647.84
Piney (Frostburg)				
1	6/18/01	398.6	305	263.55
2	6/18/01	404.8	305	401.65
3	6/18/01	358.5	305	441.59
4	6/18/01	541.5	350	653.04
5	6/18/01	423.8	315	577.42
6	6/18/01	849.8	380	782.97
7	6/18/01	962.0	400	895.77
8	6/18/01	669.2	370	546.90
9	6/18/01	775.7	370	815.60
10	6/18/01	982.8	385	557.56
11	6/18/01	1536.5	465	967.54
12	6/18/01	1001.7	410	434.57
13	6/18/01	1028.8	420	675.50
14	6/18/01	872.2	415	443.81
15	6/18/01	900.0	408	770.65
Rocky Gap				
1	6/29/00	436.2	313	92.2
2	6/29/00	366.6	305	65.5
3	6/29/00	448.3	322	71.4
4	6/29/00	422.2	313	76.6
5	6/29/00	555.2	351	197.7
6	6/29/00	650.6	358	139.2
7	6/29/00	669.4	372	113.4
8	6/29/00	613.1	364	124.0
9	6/29/00	663.6	370	58.1
10	6/29/00	692.9	368	135.9
11	6/29/00	752.8	385	167.3
12	6/29/00	883.8	396	38.1
13	6/29/00	1024.5	419	119.1
Big Pool				
1	6/19/01	282.1	290	73.84
2	6/19/01	324.1	311	348.33
3	6/19/01	224.1	283	348.12
4	6/19/01	382.6	318	319.35
5	6/19/01	396.8	318	234.51
Potomac #4				
1	8/3/00	565	367	93.3
2	8/3/00	303.31	294	168.1

APPENDIX I CONTINUED.

Sample ID	Collection Date	Total Weight (g)	Total Length (mm)	MeHg concentration (ng/g)
Clopper				
1	5/16/01	1373.2	446	330.16
2	5/16/01	1096.1	425	144.79
3	5/16/01	981.2	412	213.60
4	5/16/01	1027.8	406	168.97
5	5/16/01	803.3	400	100.48
6	5/16/01	839.8	398	179.59
7	5/16/01	841.5	398	219.25
8	5/16/01	827.5	389	384.53
9	5/16/01	781.7	369	197.44
10	5/16/01	773.7	360	238.93
11	5/16/01	575.6	355	222.35
12	5/16/01	561.7	348	98.54
13	5/16/01	409.6	333	108.01
14	5/16/01	481.1	324	108.53
15	5/16/01	312.3	311	246.32
Tridelphia				
1	7/12/00	1636.34	485	166.7
2	7/12/00	858.92	401	97.7
3	7/12/00	1208.17	440	209.5
4	7/12/00	1090.33	421	155.2
5	7/12/00	1440.3	453	426.1
6	7/12/00	1038.22	432	415.6
7	7/12/00	844.44	385	222.8
8	7/12/00	399.22	311	86.3
9	7/12/00	779.5	378	141.4
10	7/12/00	517.7	328	114.6
11	7/19/00	822.15	386	234.21
12	7/19/00	818.76	379	161.14
13	7/19/00	573.04	319	94.64
14	7/19/00	418.98	306	185.25
Piney Run				
1	6/18/01	1315.1	450	228.80
2	6/18/01	1160.0	412	144.41
3	6/18/01	1003.5	410	123.91
4	6/18/01	1001.6	406	129.26
5	6/18/01	941.8	399	214.14
6	6/18/01	754.6	380	236.32
7	6/18/01	786.1	379	196.83
8	6/18/01	857.3	395	285.20
9	6/18/01	749.6	394	125.54
10	6/18/01	686.5	351	117.31
11	6/18/01	696.8	347	109.58

APPENDIX I CONTINUED.

Sample ID	Collection Date	Total Weight (g)	Total Length (mm)	MeHg concentration (ng/g)
Piney Run				
12	6/18/01	416.6	311	147.35
13	6/18/01	376.8	307	68.93
14	6/18/01	378.3	307	87.66
Duckett				
1	4/15/02	945.68	408	623.57
3	4/15/02	1432.30	445	231.19
4	4/15/02	1753.96	468	867.51
5	4/15/02	415.55	312	187.77
6	4/15/02	829.12	380	163.98
7	4/15/02	1013.65	405	29.69
8	4/15/02	785.23	378	313.61
9	4/15/02	1544.11	459	593.76
10	4/15/02	1880.00	480	537.29
11	4/15/02	759.47	380	139.57
12	4/15/02	876.69	400	454.04
13	4/15/02	501.70	333	38.03
14	4/15/02	515.60	344	66.51
15	4/15/02	486.56	332	85.64
Liberty				
1	7/20/00	965.3	422	314.0
2	7/20/00	1345.0	472	282.0
3	7/20/00	912.9	390	297.7
4	7/20/00	904.8	394	219.3
5	7/20/00	328.8	295	126.4
6	7/20/00	540.8	331	202.3
7	7/20/00	1208.4	406	502.9
8	7/20/00	824.4	380	153.9
9	7/20/00	618.4	353	198.4
10	7/20/00	737.0	375	225.8
11	4/29/02	1165.5	437	224.05
12	4/29/02	1051.22	435	549.24
13	4/29/02	1125.11	428	708.64
14	4/29/02	653.52	360	354.77
15	4/29/02	486.21	333	449.11
16	4/29/02	424.49	317	67.45

APPENDIX I CONTINUED.

Sample ID	Collection Date	Total Weight (g)	Total Length (mm)	MeHg concentration (ng/g)
St. Mary's Lake				
1	4/30/02	327.66	306	412.53
2	4/30/02	308.06	305	564.76
3	4/30/02	308.06	308	531.82
5	5/6/02	649.60	381	335.59
6	5/6/02	2100.00	510	1291.44
7	5/6/02	2688.00	563	1514.48
8	5/6/02	789.60	387	1081.78
9	5/6/02	380.15	324	353.56
10	5/6/02	251.27	276	681.51
11	6/17/02	576.80	386	1051.85
12	6/17/02	711.20	356	833.68
Prettyboy				
1	5/14/01	709.5	380	469.33
2	5/14/01	539.4	375	258.14
3	5/14/01	887.2	405	362.24
4	5/14/01	897.7	401	523.19
5	5/14/01	762.3	366	185.99
6	5/14/01	966.2	410	284.62
7	5/14/01	941.7	417	401.34
8	5/14/01	1152.8	460	647.54
9	5/14/01	1433.7	450	722.37
10	5/14/01	1116.6	428	335.85
11	5/14/01	349.6	295	216.67
12	5/14/01	360.9	301	210.10
13	5/14/01	597.0	345	329.65
14	5/14/01	315.6	296	113.49
15	5/14/01	365.4	302	165.98
Lake Lariat				
1	5/15/01	1917.9	521	1430.91
2	5/15/01	1619.3	487	1261.01
4	5/15/01	518.8	347	608.02
5	5/15/01	1089.9	423	404.74
6	5/15/01	2175.7	541	2076.50
7	5/15/01	497.3	341	731.41

APPENDIX I CONTINUED.

Sample ID	Collection Date	Total Weight (g)	Total Length (mm)	MeHg concentration (ng/g)
Centennial Lake				
1	7/29/02	368.49	322	94.19
2	7/29/02	495.02	340	161.88
3	7/29/02	358.12	321	61.93
4	7/29/02	458.94	337	9.52
5	7/29/02	465.29	336	88.34
6	7/29/02	580.31	344	88.51
7	7/29/02	225.87	306	184.84
Loch Raven				
2	6/20/01	758.0	369	162.3
3	6/20/01	789.2	386	462.0
4	6/20/01	911.0	420	497.6
5	6/20/01	559.1	347	145.8
6	6/20/01	382.2	305	241.1
7	6/20/01	375.0	307	258.4
8	6/20/01	463.3	320	164.5
9	6/20/01	399.9	312	172.1
10	6/20/01	387.0	308	210.2
11	5/6/02	703.77	390	290.44
12	5/6/02	691.42	396	61.68
13	5/6/02	1004.85	420	595.24
14	5/6/02	1040.00	431	430.57
15	5/6/02	1198.25	455	267.65
16	5/6/02	1185.32	455	824.42
Conowingo				
1	7/12/00	939.06	406	202.4
2	7/12/00	577.73	370	123.6
3	7/12/00	818.7	391	78.7
4	7/12/00	574.55	357	52.2
6	7/12/00	442.3	336	118.8
7	7/12/00	566.75	339	124.1
8	7/12/00	617.78	366	144.1
9	7/12/00	346.66	305	56.1
10	8/16/00	382.18	315	104.8
11	8/16/00	381.29	309	45.4
12	8/16/00	1542.3	445	215.5
13	8/16/00	1184.0	426	100.6
14	8/16/00	1033.6	425	120.2

APPENDIX I CONTINUED.

Sample ID	Collection Date	Total Weight (g)	Total Length (mm)	MeHg concentration (ng/g)
Tuckahoe				
1	7/18/00	434.5	323	482.8
2	7/18/00	546.3	343	327.7
3	7/18/00	423.2	310	116.3
4	7/18/00	555.2	333	184.6
5	7/18/00	805.4	370	565.4
7	7/18/00	495.3	321	229.7
8	7/18/00	696.3	366	149.7
9	7/18/00	1183.9	420	96.4
10	7/18/00	1675.1	485	724.1
11	8/1/00	1338.1	437	546.8
12	8/1/00	1509.9	479	576.4
13	8/1/00	1691.2	491	508.0
14	8/1/00	632.5	367	156.1
Johnson's Pond				
1	8/15/00	1476.8	479	350.5
2	8/15/00	1480.0	460	355.5
3	8/15/00	1132.8	437	471.0
4	8/15/00	1873.2	506	500.2
5	8/15/00	1044.2	435	196.1
6	8/15/00	816.6	380	142.9
8	8/15/00	458.8	326	97.1
9	8/15/00	736.0	363	149.9
10	8/15/00	696.8	376	31.9
11	8/15/00	625.0	377	96.7
12	8/15/00	446.8	333	67.0
13	8/15/00	629.7	350	146.7
14	8/15/00	583.0	349	147.9
15	8/15/00	539.3	341	113.8

APPENDIX II: Mercury, Methylmercury and Water Chemistry of Maryland Reservoirs.

IN= sample from inflow to reservoir
MID= sample from middle of reservoir
OUT= sample from outflow from reservoir
SURF= sample from surface water
MIDDLE= sample from mid depth of reservoir
BOTT= sample from bottom of reservoir

Ancillary measurements from Liberty, Prettyboy and Loch Raven are from depth profiles.

Whole = Unfiltered water
Dissolved = Filtered water
L= Under detection limits
DP= Depth Profile
*[1] Hg from Castro et al., 2000.
*[2] Hg and MeHg data from Lawson et al., 2000.
SG=Surface Grabs

Reservoir	Sample Type	Location	HgT (ng/L) Whole	HgT (ng/L) Dissolved	MeHg (pg/L) Whole	MeHg (pg/L) Dissolved
Deep Creek Lake	DP	SURF	2.85	0.64	374.6	279.1
		MID	1.99	0.59	411.9	290.8
		BOTT	1.01	0.59	361.3	262.2
Broadford Lake	SG	IN	1.22	1.00	168.7	137.1
		MID	1.42	0.98	150.8	96.9
Savage	SG	IN	1.23	1.28	135.7	132.1
		MID	0.43	0.07	70.7	70.7
		OUT	2.19	0.47	110.0	15.6
Piney (Frostburg)	DP	SURF	1.70	0.59	351.5	265.2
		MIDDLE	3.86	2.22	322.4	259.2
Rocky Gap*[1]	DP	SURF	0.40	n/a	137.9	24.1
		MID	n/a	n/a	120.8	48.8
		BOTT	n/a	n/a	70.1	69.9
Potomac #4	SG	IN	n/a	0.88	193.0	156.9
		MID	1.48	1.47	286.5	262.7
Clopper	SG	IN	20.38	15.89	428.1	157.3
		MID	12.06	15.27	110.1	62.5
		OUT	18.53	22.99	140.9	62.8
Tridelphia	SG	IN	3.06	3.58	70.5	724.3
		MID	3.99	1.10	110.8	5.2
		OUT	0.97	1.20	46.6	147.6
Piney Run	SG	IN	1.29	0.33	488.7	434.7
		MID	2.08	1.61	186.1	75.0
Duckett	SG	IN	7.44	15.28	130.7	71.8
		MID	19.30	23.78	81.6	43.0
		OUT	19.43	n/a	132.3	34.6

APPENDIX II CONTINUED.

Reservoir	Sample Type	Location	HgT (ng/L) Whole	HgT (ng/L) Dissolved	MeHg (pg/L) Whole	MeHg (pg/L) Dissolved
Liberty	SG	IN	0.67	3.10	103.1	81.0
		MID	4.54	1.44	82.1	48.4
		OUT	0.75	1.12	7.5	95.9
St. Mary's Lake	SG	IN	2.85	0.72	94.3	110.0
		MID	1.49	0.19	286.5	151.8
Prettyboy	SG	IN	5.14	1.96	n/a	81.0
		MID	2.74	2.92	59.8	25.2
		OUT	3.96	7.21	33.9	67.9
Lake Lariat	SG	IN	2.79	2.79	133.2	17.5
		MID	2.05	1.40	113.3	15.4
Loch Raven	SG	IN	7.76	3.02	285.6	213.9
		MID	4.12	5.07	89.6	84.8
		OUT	3.39	8.81	208.9	182.5
Conowingo*[2]	SG	IN	n/a	3.55	n/a	118.6
		MID	n/a	n/a	n/a	n/a
		OUT	n/a	n/a	n/a	n/a
Tuckahoe	SG	IN	4.33	3.31	297.9	172.6
		MID	3.62	1.34	29.3	187.4
		OUT	4.30	2.54	49.6	49.6
Johnson's Pond	SG	IN	3.82	3.08	148.1	105.8
		MID	1.52	0.07	152.5	95.4
		OUT	4.19	1.97	168.2	140.2

APPENDIX II CONTINUED.

Reservoir	Sample Type	Location	pH	DOC	PN	PC	NO_2
				(mg/L)			
Deep Creek Lake	DP	SURF	7.0	3.09	0.063	L	0.002
		MID	6.9	3.06	0.069	L	0.002
		BOTT	6.9	2.67	0.037	L	0.103
Broadford Lake	SG	IN	6.2	3.03	0.069	0.585	0.001
		MID	7.3	4.77	0.277	1.980	0.000
Savage	SG	IN	7.4	2.72	0.082	0.961	0.003
		MID	7.6	2.18	0.034	0.309	0.003
		OUT	6.7	2.24	0.011	0.0638*	0.002
Piney (Frostburg)	DP	SURF	7.2	4.76	0.090	L	0.013
		MIDDLE	7.2	5.21	0.161	1.050	0.010
Rocky Gap*[1]	DP	SURF	6.6		0.026	L	0.001
		MID	6.7	4.07	0.037	L	0.001
		BOTT	7.9	3.59	0.029	L	0.002
Potomac #4	SG	IN	7.0	3.15	0.074	0.572	0.009
		MID	7.4	1.65	0.660	0.601	0.008
Clopper	SG	IN	7.5	4.43	0.246	2.780	0.504
		MID	7.5	4.4	0.103	L	0.456
		OUT	7.6	4.09	0.073	L	0.470
Tridelphia	SG	IN	n/a	n/a	n/a	n/a	n/a
		MID	n/a	n/a	n/a	n/a	n/a
		OUT	n/a	n/a	n/a	n/a	n/a
Piney Run	SG	IN	7.9	3.18	0.070	0.581	0.011
		MID	7.9	3.07	0.078	0.631	0.011
Duckett	SG	IN	7.5	2.89	0.294	1.900	1.410
		MID	7.6	3.18	0.106	0.662	1.300
		OUT	7.5	2.97	0.173	0.885	1.090

APPENDIX II CONTINUED.

Reservoir	Sample Type	Location	pH	DOC	PN	PC	NO_2
				(mg/L)			
Liberty	SG	IN	8.0	2.78	0.061	L	0.005
		MID	7.7	2.63	0.074	L	0.018
		OUT	7.1	2.11	0.041	L	0.018
St. Mary's Lake	SG	IN	6.7	6.37	0.189	3.030	0.002
		MID	6.9	6.65	0.143	1.170	0.002
Prettyboy	SG	IN	7.7	1.34	0.033	0.678	0.002
		MID	7.7	2.02	0.096	0.811	0.018
		OUT	7.4	2.49	0.162	1.290	0.017
Lake Lariat	SG	IN	7.7	3.99	0.192	2.060	0.020
		MID	7.9	4.23	0.227	2.210	0.006
Loch Raven	SG	IN	9.8	1.46	0.012	0.209	0.004
		MID	8.5	2.54	0.238	2.770	0.002
		OUT	7.8	2.41	0.104	0.581	0.001
Conowingo*[2]	SG	IN	8.0	n/a	n/a	n/a	n/a
		MID	8.1	n/a	n/a	n/a	n/a
		OUT	8.2	n/a	n/a	n/a	n/a
Tuckahoe	SG	IN	7.3	5.76	0.079	0.928	0.001
		MID	7.3	4.85	0.134	1.290	0.017
		OUT	7.4	4.84	0.055	0.616	0.015
Johnson's Pond	SG	IN	8.0	6.08	0.034	0.667	0.029
		MID	8.2	3.72	0.041	0.673	0.038
		OUT	7.3	8.06	0.107	1.200	0.092

APPENDIX II CONTINUED.

Reservoir	Sample Type	Location	NH_4	NO_3	PO_4	TSS	CL	SO_4
			(mg/L)					
Deep Creek Lake	DP	SURF	0.011	0.007	0.016	2.4	5.45	12.00
		MID	0.013	0.035	0.028	2.4	6.23	13.30
		BOTT	0.212	0.028	0.023	2.4	8.08	15.70
Broadford Lake	SG	IN	0.027	1.139	0.003	n/a	29.1	8.00
		MID	0.034	1.130	0.003	3.7	52.4	8.80
Savage	SG	IN	0.010	0.467	0.002	2.8	22.8	11.70
		MID	0.009	0.696	0.001	12.7	16.0	12.00
		OUT	0.008	L	0.649	2.4	20.6	13.20
Piney (Frostburg)	DP	SURF	0.009	0.751	0.020	0.7	26.9	6.94
		MIDDLE	0.015	0.684	0.025	1.1	26.5	7.49
Rocky Gap*[1]	DP	SURF	0.009	0.004	0.010	2.4	4.3	11.90
		MID	0.020	0.003	0.022	2.4	2.45	9.28
		BOTT	0.139	0.040	0.017	2.4	3.38	10.60
Potomac #4	SG	IN	0.040	0.792	0.018	6.0	19.5	37.90
		MID	0.033	0.777	0.014	10.0	16.1	35.70
Clopper	SG	IN	0.121	L	0.003	6.8	128	6.40
		MID	0.048	L	0.005	6.0	138	6.20
		OUT	0.088	L	0.002	2.4	141.3	5.90
Tridelphia	SG	IN	n/a	n/a	n/a	n/a	n/a	n/a
		MID	n/a	n/a	n/a	n/a	n/a	n/a
		OUT	n/a	n/a	n/a	n/a	n/a	n/a
Piney Run	SG	IN	0.040	0.585	0.002	2.4	27.0	6.82
		MID	0.036	0.576	0.001	2.4	28.9	7.00
Duckett	SG	IN	0.040	L	0.003	13.6	129.3	5.70
		MID	0.011	L	0.002	11.2	26.7	7.00
		OUT	0.113	L	0.004	6.5	28.1	5.40

APPENDIX II CONTINUED.

Reservoir	Sample Type	Location	NH_4	NO_3	PO_4	TSS	CL	SO_4
						(mg/L)		
Liberty	SG	IN	0.008	1.805	0.052	2.4	1.80	9.26
		MID	0.053	1.572	0.030	2.4	1.80	7.23
		OUT	0.009	1.637	0.932	2.4	1.60	7.23
St. Mary's Lake	SG	IN	0.056	6.368	0.002	18.7	9.3	2.62
		MID	0.003	6.648	0.001	5.0	8.7	7.42
Prettyboy	SG	IN	0.007	2.738	0.007	19.0	36.39	2.93
		MID	0.021	2.163	0.008	14.3	29.87	3.70
		OUT	0.020	1.893	0.018	12.4	25.24	3.67
Lake Lariat	SG	IN	0.153	0.711	0.001	8.4	18.8	13.87
		MID	0.121	0.341	0.001	10.8	22.6	10.48
Loch Raven	SG	IN	0.011	3.166	0.015	39.4	55.74	4.10
		MID	0.023	1.928	0.015	39.4	76.43	5.51
		OUT	0.026	1.919	0.017	5.0	79.24	5.49
Conowingo*[2]	SG	IN	n/a	n/a	n/a	n/a	n/a	n/a
		MID	n/a	n/a	n/a	n/a	n/a	n/a
		OUT	n/a	n/a	n/a	n/a	n/a	n/a
Tuckahoe	SG	IN	0.022	1.079	0.033	4.5	25.17	7.76
		MID	0.174	3.323	0.030	8.0	13.41	6.67
		OUT	0.070	3.745	0.028	2.4	10.71	6.19
Johnson's Pond	SG	IN	0.029	2.701	0.005	3.5	14.58	4.38
		MID	0.038	2.322	0.004	5.4	36.98	3.47
		OUT	0.092	2.188	0.008	8.3	15.38	4.88

APPENDIX III: Concentrations of methylmercury in the whole body of crayfish and forage fish from some of the reservoirs sampled in this study.

Reservoir	**Average Length (mm)**	**Std. Dev**	**MeHg conc. (ng/g wet weight)**	**# Crayfish**
Piney (Frostburg)	83.2	11.8	44.6	5
Clopper	11.1	2.1	32.4	4
Tridelphia	138.2	36.3	28.0	8
Liberty	7.8	2.7	15.4	2
Prettyboy	8.6	1.9	9.6	2
Loch Raven	7.0	2.1	17.7	2
Tuckahoe	12.5	0.7	7.2	2
Average	**38.3**		**22.1**	
Std. Dev	**51.9**		**13.5**	

APPENDIX III CONTINUED:

Forage Fish				
Reservoir	**Fish**	**Average Length (mm)**	**Average MeHg (ng/g)**	**Average HgT (ng/g)**
Deep Creek Lake				
	Blue Gill	6,09	**69,67**	**134,94**
	Yellow Perch	5,59	**25,63**	**45,44**
Piney (Frostburg)				
	Blue Gill	3,23	**20,41**	**63,27**
	Yellow Perch	7,69	**39,70**	**77,67**
Rocky Gap				
	Blue Gill	N/A	**9,41**	**19,50**
Liberty Reservoir				
	Blue Gill	105,00	**51,57**	**52,73**
	Golden Shiner Lg	186,50	**30,62**	**39,51**
	Golden Shiner Sm	94,20	**45,98**	**155,85**
	White Sucker	137,50	**46,27**	**129,12**
Prettyboy				
	Blue Gill	59,70	**22,05**	**48,94**
	Black Crappie	N/A	**82,48**	**164,86**
	Yellow Perch	136,60	**44,43**	**62,81**
	White Sucker	216,00	**47,49**	**52,20**
Loch Raven				
	Blue Gill	8,43	**44,95**	**64,05**
	White Perch	11,88	**108,17**	**94,92**
	Silverside	6,52	**24,36**	**42,85**
Tukahoe				
	Blue Gill	8,27	**48,55**	**77,43**
	Pumpkin Seed	10,83	**32,32**	**63,13**
Johnson's Pond				
	Blue Gill	8,08	**17,75**	**9,10**
	average	59,54	42,73	73,59
	std. Dev	71,96	23,93	43,68

APPENDIX III CONTINUED:

Other Fish			
Reservoir	**Fish**	**Average Length (mm)**	**Average MeHg (ng/g)**
Deep Creek Lake	Blue Gill I	163,60	60,17
	Blue Gill II	170,25	52,05
	Blue Gill III	176,67	51,19
Broadford Lake	Blue Gill I	164,40	52,94
	Blue Gill II	173,50	58,48
	Blue Gill III	169,33	57,64
Savage	Blue Gill I	141,3	234,5
Piney (Frostburg)	Blue Gill I	139,0	80,2
	Blue Gill II	187,8	91,4
	Blue Gill III	203,0	393,6
Rocky Gap	Blue Gill I	143,00	47,64
	Blue Gill II	150,00	52,68
	Blue Gill III	174,50	60,71
Potamoc #4	Blue Gill I	280,50	164,18
	Smallmouth bass II	196,00	17,88
	Smallmouth bass III	220,33	203,48
Clopper	Blue Gill I	157,0	36,0
	Blue Gill II	148,0	26,8
	Crappie I	368,4	287,41
	Crappie II	239,0	212,26
	Crappie III	176,8	60,18
Tridelphia	Blue Gill I	193,20	58,41
	Blue Gill III	225,33	91,42
	Crappie I	218,43	81,92
	Crappie II	239,20	100,09
	Crappie III	245,50	95,57
Piney Run	Crappie I	68,4	47,28
	Crappie II	134,4	67,28
	Crappie III	268,2	42,32
Liberty	Crappie I	262,75	113,37
	Crappie III	291,00	120,37
Prettyboy	Blue Gill I	210,0	60,8
	Blue Gill II	168,2	41,6
	Blue Gill III	135,6	49,69

APPENDIX III CONTINUED:

Lake Lariet	Blue Gill II	176,4	225,00
	Blue Gill III	138,0	210,94
	Crappie I	197,9	38,72
	Crappie II	226,0	147,24
	Crappie III	268,3	525,59
Loch Raven	Blue Gill I	136,2	61,1
	Blue Gill II	172,2	113,0
Conowingo	Blue Gill I	152,50	22,36
	Blue Gill II	172,00	18,54
	Crappie III	269,00	91,24
	Smallmouth bass III	439,00	150,57
Tuckahoe	Crappie I	235,67	106,86
	Crappie II	257,00	118,08
	Crappie III	287,00	161,22
Johnson's Pond	Blue Gill I	163,50	56,10
	Blue Gill II	197,00	99,52
	Blue Gill III	214,50	133,31
Size classes: Blue Gill: I = 127-150 mm; II = 151-202 mm; III = >203 mm. Crappie: I = 152-210 mm, II = 210 -254 mm, III > 255 mm; Smallmouth bass: I = 305-329 mm, II = 330-380 mm, III = >381 mm.			