

A Survey of Size-Specific Mercury Concentrations in Game Fish from Maryland Fresh and Estuarine Waters

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Abstract. A survey of size-specific mercury (Hg) concentrations in game fish from a subset of Maryland fresh and estuarine waters was conducted, in which Hg concentrations in 112 fish from seven freshwater impoundments and three tidal and four estuarine locations in Chesapeake Bay and its tributaries were measured. Striped bass (*Morone saxatilis*) was the most intensively examined species. Of the fish examined, the largest freshwater sportfish contained the highest Hg concentrations. Striped bass and largemouth bass (*Micropterus salmoides*) from Chesapeake Bay and its tributaries contained less Hg at the same size than the same species in fresh waters. Large striped bass, chain pickerel (*Esox niger*), and walleye (*Stizostedion vitreum*) from Deep Creek Lake and Liberty Reservoir exceeded the FDA action level of 1 mg Hg/kg. Striped bass, largemouth bass, and white crappie (*Pomoxis annularis*) in other impoundments equaled or exceeded a common advisory level of 0.5 mg Hg/kg. Large differences in size-normalized Hg concentrations among lakes and particularly between fresh and salt waters highlight the large differences in MeHg production and bioaccumulation among ecosystems. This work indicates that a more comprehensive study of Hg in Maryland fish is warranted to protect human and wildlife health.

In the United States, more fish consumption advisories are posted because of elevated mercury (Hg) concentrations than because of any other contaminant. In 1998, 40 states had Hg-based fish consumption advisories for one or more water bodies (US EPA 1999). Ten states posted consumption advisories for all freshwater lakes and/or rivers, and five posted all coastal waters. Human use of mercury has increased the cycling of this volatile element through the atmosphere since preindustrial times, resulting in elevated atmospheric inputs of mercury to lakes and their watersheds (Mason *et al.* 1994). Many scientists believe that increased deposition contributes to the high mercury concentrations found in fish today (*e.g.*, US EPA 1997). Mercury deposited to aquatic ecosystems can be converted to methylmercury (MeHg) through the action of

bacteria in sediments and other anaerobic habitats (reviewed in Mason and Benoit 2000; Gilmour and Henry 1991). It is MeHg that bioaccumulates in food webs.

Mercury concentrations in fish are functions of feeding habits and food chain structure (reviewed by Wiener and Spry 1996), along with ecosystem characteristics that affect Hg methylation rates and Hg loading rates. Production rates of MeHg among ecosystems vary as much or more as atmospheric Hg deposition, so that MeHg bioaccumulation in fish depends not only on how much Hg enters the ecosystem but also on the ability of an ecosystem to convert that Hg to MeHg. Among water bodies, acidity, dissolved organic carbon (DOC), the ratio of watershed area to lake area, and fraction of wetland area appear to be the most important variables in predicting Hg in fish. Acid deposition, and more specifically deposition of sulfuric acid, can stimulate the microorganisms (sulfate-reducing bacteria) that produce MeHg within sediments (Gilmour *et al.* 1992), and pH may also affect MeHg bioaccumulation (Watras *et al.* 1995). Ecosystems with high DOC tend to have high percentages of wetland area, and these are places where microbial Hg methylation is very rapid (*e.g.*, St. Louis *et al.* 1994; Gilmour *et al.* 1998). DOC may also enhance MeHg transport (Hurley *et al.* 1995) and prevent photochemical degradation of MeHg (Sellers *et al.* 1996). Lake size and temperature affect MeHg bioaccumulation, with more bioaccumulation in smaller warmer lakes (Bodaly *et al.* 1993) where MeHg production is enhanced. Reservoir formation stimulates microbial Hg methylation after flooding (Kelly *et al.* 1997), an effect which persists for a maximum of 30–35 years (Bodaly *et al.* 1997). Marine and estuarine ecosystems appear less sensitive to Hg methylation than fresh waters, potentially because sulfide in sediment inhibits the production of MeHg (Choi and Bartha 1994; Benoit *et al.* 1998, 1999).

Within the United States, Hg-based consumption advisories are common, often statewide, in the upper Midwest, New England, and the Southeast (US EPA 1999). The mid-Atlantic states (with the exception of New Jersey, which has a statewide ban in fresh waters) currently have few or no advisories despite relatively high levels of Hg deposition. Wet deposition of Hg in the lower Chesapeake Bay watershed ranges from 10 to 25 $\mu\text{g Hg/m}^2$ year, depending on distance from sources (Mason *et al.* 1997). This rate is similar to deposition in south Florida, a region with very high levels of Hg in freshwater fish (Ware *et*

al. 1990) and higher than deposition in the Midwest and western United States, where wet deposition fluxes range from 3 to 10 $\mu\text{g}/\text{m}^2$ year (Mason *et al.* 1997). In addition to high Hg deposition rates, some of Maryland's fresh waters have features that could make them susceptible to high levels of MeHg production and bioaccumulation. Most Maryland fresh waters are impoundments (although most were impounded decades ago), and some are acidified by acid deposition and acid-mine drainage (*e.g.*, Ferrier and Biedka 1985). Many water bodies in the coastal plain are poorly buffered and high in DOC (*e.g.*, Knapp and Saunders 1988).

There are no Hg-based fish consumption advisories in Maryland. Although both the State of Maryland (*e.g.*, Maryland Department of the Environment 1988) and the U.S. Fish and Wildlife Service (*e.g.*, Schmidt and Brumbaugh 1990) have examined Hg concentrations in Maryland fish, these data sets are of limited use in assessing Hg contamination because Hg was not examined with regard to fish size. There is an extensive body of literature documenting a positive relationship between Hg concentrations and fish size (Wiener and Spry 1996). Furthermore, both the 1988 MDE and the 1990 FWS studies targeted estuarine fish, whereas freshwater fish are more likely to accumulate MeHg. A small study of Hg in walleye and chain pickerel from four Maryland reservoirs was conducted for MDE in 1992 (Versar 1994), in which individual fish of known size were analyzed. Walleye of roughly 40 cm from two of the reservoirs examined exceeded 0.5 mg Hg/kg.

The objective of the study presented here was to make a preliminary estimate of size-specific mercury concentrations in game fish from a subset of Maryland fresh and estuarine waters. The study was conducted to help assess the need for a more comprehensive survey of Hg in Maryland fish.

Materials and Methods

Study Design and Sampling Locations

Mercury concentrations in fish from seven freshwater, three tidal, and four estuarine locations within Maryland were measured. A total of 112 fish were analyzed, with striped bass (*Morone saxatilis*), most from the Chesapeake Bay and estuarine Potomac River, comprising the largest group ($n = 47$). The Potomac is a large tributary of the Chesapeake Bay. Most freshwater fish were taken from Deep Creek Lake and Liberty, Cash, and Piney Run Reservoirs. Piney Run and Liberty Reservoirs are impoundments of branches of the Patapsco River in Carroll County. Cash Lake is an impoundment of a branch of the Patuxent, in Prince George's County near the Patuxent Wildlife Research Center. Deep Creek Lake in Garrett County is the largest lake in Maryland (3,900 acres). It is an impoundment (constructed in 1924) of the Youghiogheny River and is affected by both acid deposition and acid mine drainage. Deep Creek is sensitive to acid inputs because of the relatively poor buffering capacity of the sediment and watershed. Lake alkalinity in the early 1980s was <10 mg/L CaCO_3 , although pH remained between 6 and 7 (Ferrier and Biedka 1985). These reservoirs represent three geologic provinces within Maryland: Cash Lake, St. Mary's Lake, and Lake Lariat lie in the Coastal Plain; Liberty and Piney Run Reservoirs in the Piedmont; and Deep Creek in the Appalachian Plateau. Like water bodies in the Appalachian Plateau, streams and impoundments in the coastal plain, especially in southern Maryland, are sensitive to acid deposition because of poorly buffered soils (Knapp and Saunders 1988). For example, St. Mary's

Lake, which is a highly colored lake in St. Mary's county, exhibits large pH swings (4.5–9.5) on seasonal cycles (personal observation). Soils in the Piedmont are well buffered.

Fish were collected between 1993 and 1995. Many fish were provided by the Maryland Department of Natural Resources (MD DNR) through their routine sampling programs or through requests to private anglers. Other fish were taken by private anglers or purchased from a waterman. Fish provided by MD DNR were frozen, either as whole fish or fillets. Other fish were filleted fresh. All samples were frozen as quickly as possible.

Analysis of Hg in Fish

Total Hg concentrations were measured in skinless fillets and are reported on a wet weight basis. Nearly all (95–99%) of the Hg accumulated in the upper food web is MeHg, even for nonpiscivorous fish species (Bloom 1992) and in heavily Hg contaminated systems (*e.g.*, Southworth *et al.* 1995). Fillet portions were partially thawed and the cleanest inner section subsampled using an acid-washed plastic knife. Between 0.5 and 1 g of chopped fillet were digested with 5 ml of 30:75 H_2SO_4 : HNO_3 . All acids used in digestion or cleaning were screened for Hg content. Samples were capped with marbles and predigested 1 h at room temperature. The temperature was increased slowly to a boil, and digestion continued until samples were almost clear. After cooling, the samples were brought up to the 50 ml with low-Hg deionized water and 0.5 ml of BrCl was added. A small aliquot of digestate, usually 10–100 μl , was then analyzed for total Hg as below.

Total Hg analysis was performed by cold-vapor atomic fluorescence (CVAF), with preconcentration of digested samples on gold traps (Bloom 1992). For each set of digestions (about 15 samples), two SRMs, two spikes, two duplicates, and two blanks were carried through the entire analysis. Three standard reference materials for Hg in tissue were used during analysis of fish, DORM1 (NRC, dogfish muscle), TORT1 (NRC, lobster hepatopancreas), and 1566a (NIST, oyster tissue), and they generally fell within certified ranges. The average RPD for duplicates across all fish analyses in this study was 15.1 ± 18.7 ($n = 15$). Recovery of a 100-ng Hg spike to 0.5 g of fish tissue averaged $96.5 \pm 12.8\%$ ($n = 8$). Detection limits were determined by the value of the appropriate blank and averaged about 1 ng/g. Our lab takes part in numerous interlaboratory calibrations to maintain a high level of quality assurance (*e.g.*, Bloom *et al.* 1995).

Results and Discussion

Presentation of Data

By regressing fish length or weight against Hg content, the level of Hg contamination in Maryland fish can be compared among the water bodies and with other regions. These size:Hg relationships can also be used to predict the average size above which Hg content may exceed tissue levels that are used in setting consumption advisories. For all species, fish weight was a better predictor of Hg concentration than was fish length, and weight was used where data were available. Log transformation of weight, length, and Hg content allowed analyses that met assumptions of normality.

In this paper, fish tissue Hg concentrations are plotted in comparison with 1.0 and 0.5 mg Hg/kg, fish tissue Hg levels used by many U.S. states in making decisions about consumption advisories. Of the 33 states that had Hg advisories for fish

in 1994, 13 used the FDA action level of 1.0 mg Hg/kg as the basis for consumption advisories, and 20 used methods that resulted in advisories at lower fish tissue Hg concentrations (US EPA 1995).

Striped Bass

Three separate collections of striped bass in the mid and upper Chesapeake and in the estuarine Potomac River were examined, along with a small number of fish from two reservoirs (Figure 1). The Chesapeake and Potomac fish were collected in the oligo- and mesohaline reaches. None of the striped bass caught in the Chesapeake Bay or estuarine Potomac River, which ranged up to 5.5 kg (or about 12 lbs) in size, exceeded 1.0 mg/kg; three fish exceeded 0.5 mg/kg. All of the reservoir fish examined exceeded 0.5 mg/kg, and two equaled or exceeded 1.0 mg/kg.

The individual weight:Hg relationships for Potomac ($n = 14$), and upper Bay fish ($n = 17$), were significant at $p < 0.05$, but the mid-bay ($n = 10$) and reservoirs ($n = 5$) data sets were not (Figure 1). The lack of a significant size:Hg relationship for the reservoir fish is not surprising given the small sample size and the fact that the fish were taken from two water bodies. Although the fish taken from the reservoirs were larger than the fish examined from the bay and therefore expected to contain proportionally higher Hg concentrations, analysis of covariance in Hg by size and site (Somers and Jackson 1993) showed that the reservoir striped bass had significantly higher Hg concentrations, when adjusted to body weight, than did striped bass from each of the estuarine sites ($p < 0.04$ for all three comparisons). There were much smaller differences in Hg accumulation in striped bass among collection sites in the estuary. Size-adjusted mercury levels were slightly lower in fish caught in mid-Chesapeake Bay in 1993 than in fish caught in the Potomac ($p < 0.001$) and upper bay ($p < 0.002$) in 1994. Length-to-weight ratios were the same for fish taken from each of the estuarine sites (data not shown).

The fairly large data set for striped bass in Chesapeake Bay and one of its tributaries, the Potomac, shows with some confidence that fish below 8 kg do not generally exceed 0.5 mg Hg/kg. However, higher levels of Hg in a few larger striped bass from reservoirs suggest that a more complete analysis of Hg in striped bass from Maryland reservoirs is needed.

Fish in Tidal Fresh and Oligohaline Waters

Mercury concentrations in a variety of species from the tidal freshwater and oligohaline reaches of the Patuxent and Potomac Rivers were measured. The expected general increasing trend in Hg concentration with fish size can be seen for all species (Figure 2) but is especially apparent in the larger piscivorous species like largemouth bass (*Micropterus salmoides*). Of the species examined in these rivers, brown bullhead (*Ameiurus nebulosa*) contained the least Hg for their size and showed no significant relationship between Hg concentration and weight. Omnivorous fish, which are lower on the food chain than piscivores of the same size, often contain lower Hg concentrations (Wiener and Spry 1996). A few white perch and

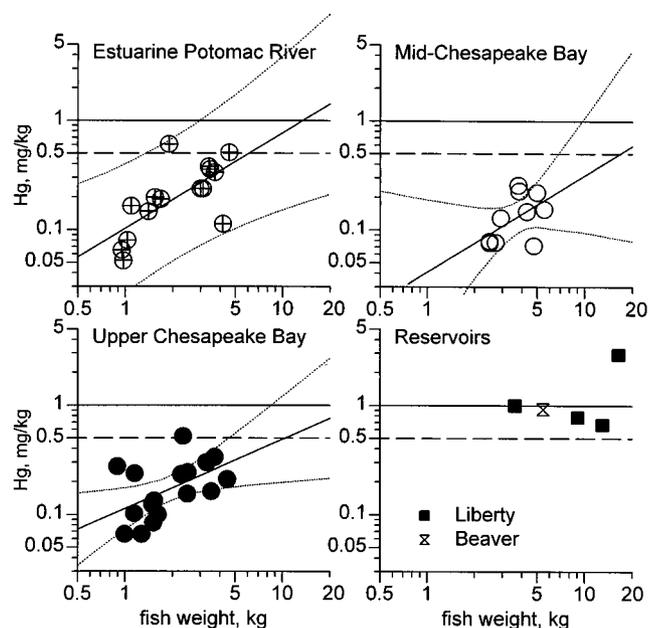


Fig. 1. Relationship between Hg concentration in muscle tissue and total fish weight for Maryland striped bass by sampling location. The line at 1 mg Hg/kg is the FDA action level; 0.5 mg Hg/kg is a common tissue advisory level among U.S. states. Linear regressions of each data set are shown when significant at $p < 0.05$, and dotted lines represent 95% confidence intervals around the mean

croaker in the 0.2–0.4 kg range contained 0.05–0.15 mg Hg/kg, although there was one high outlier at 0.3 kg and 0.4 mg Hg/kg. Yellow perch (*Perca flavescens*) over a somewhat wider size range appeared to increase in Hg with size, but fish up to 0.5 kg did not exceed 0.2 mg Hg/kg.

The data set for largemouth bass from the tidal freshwater Potomac River was large and wide enough in size range to give a significant relationship between weight and Hg concentration ($p < 0.01$; Figure 3). Based on the regression, only fish above 4 kg would exceed 0.5 mg/kg. Two largemouth bass from the tidal freshwater Patuxent River were examined, and they fit roughly into the same range of size and Hg concentration as Potomac fish, an adjacent tributary of Chesapeake Bay.

Fish in Maryland Fresh Waters

Of the fish examined in this study, the largest freshwater sport fish contained the highest levels of Hg. In addition to striped bass from Liberty and Beaver Reservoirs, mercury levels in eight other species from six freshwater lakes were examined (Figure 4). Some of the large piscivores from Deep Creek Lake (chain pickerel) and Liberty Reservoir (striped bass and walleye) equaled or exceeded 1 mg Hg/kg fish tissue. A few individual fish from Beaver Run (striped bass), Cash Lake (white crappie), and St. Mary's Lake (largemouth bass) equaled or exceeded 0.5 mg Hg/kg fish tissue. Where available, weight was a better predictor of Hg concentration than fish length. However, weight data were not provided for many of the freshwater fish. For most fish species in most lakes, Hg

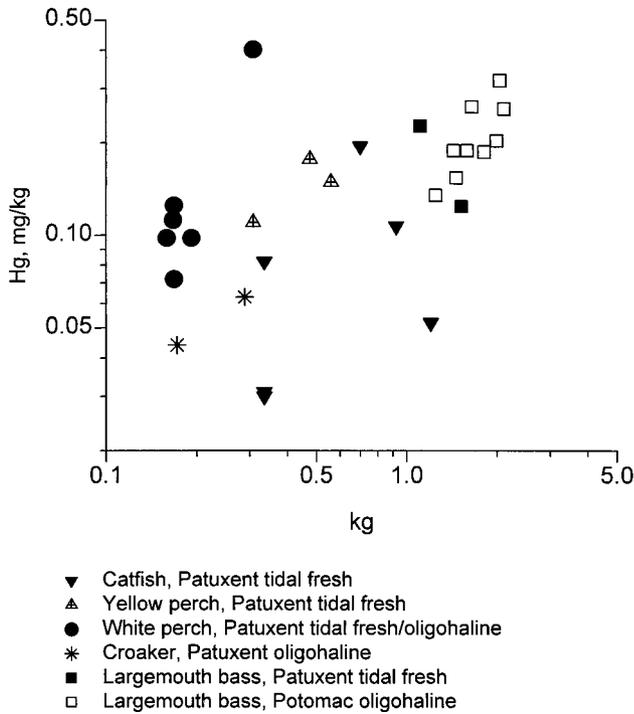


Fig. 2. Mercury-to-weight relationships for all species examined in the oligohaline and tidal Patuxent and Potomac Rivers

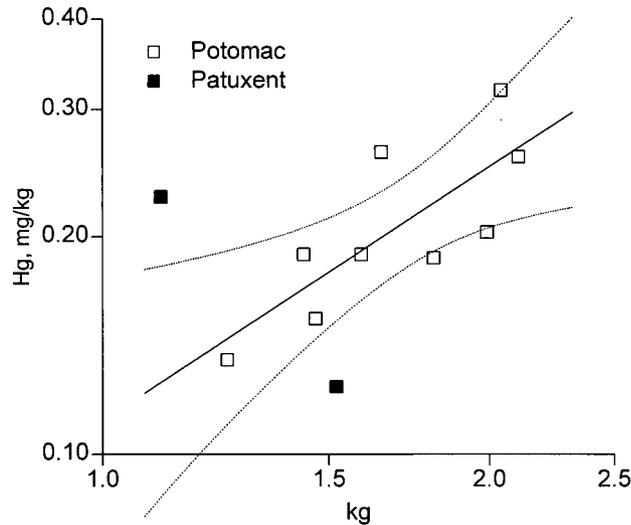


Fig. 3. Mercury-to-weight relationships for largemouth bass from the tidal freshwater Patuxent and Potomac Rivers, with linear regression ($p < 0.01$) and 95% confidence interval about the mean for Potomac River fish only

concentrations increased with the length of fish. Mercury bioaccumulation in fish varied among water bodies, as measured by differences in the Hg to length relationships within species among lakes.

Chain pickerel from Deep Creek Lake contained the highest Hg concentrations per unit size of the fish examined. In a large

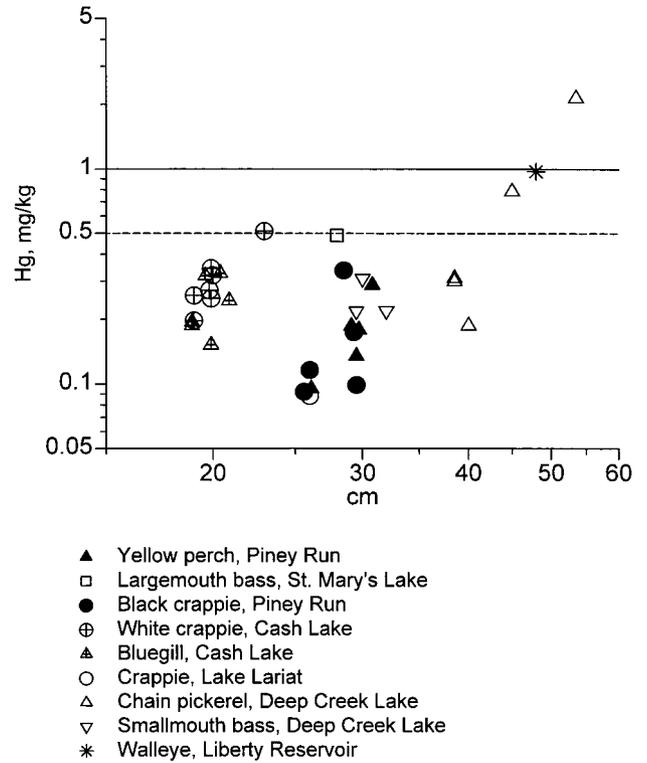


Fig. 4. Relationship between Hg concentration and length for all Maryland freshwater sport fish examined (except striped bass and walleye)

study of Hg in fish in New Jersey, chain pickerel also stood out as having high Hg:size ratios (Horwitz *et al.* 1995). Pickerel from Deep Creek contained Hg concentrations at the upper end of the range found in New Jersey pickerel of the same size, comparable to pickerel in high-DOC, low-pH lakes in the New Jersey Pine Barrens. Collection of enough fish over a sufficient size range gave a significant regression for Hg against both weight and length (Figure 5). Data collected for this study were similar to data collected on Deep Creek pickerel in 1992 (Versar 1994). Regression of the combined data sets (which were not significantly different) suggests that pickerel over 44 cm would exceed 0.5 mg Hg/kg and over 48 cm (or about 0.7 kg) would exceed 1 mg Hg/kg. Chain pickerel from Deep Creek Lake appear to contain more Hg than do pickerel from nearby Herrington Manor Lake (Versar 1994; data not shown). Walleye of roughly 40–50 cm from Deep Creek Lake and Liberty and Savage Reservoirs exceed 0.5 mg Hg/Kg and appear to contain about the same levels of Hg per unit weight, whereas walleye from Jennings Randolph contain less (Figure 6). Smallmouth bass (*Micropterus dolomieu*) averaging 30 cm from Deep Creek Lake contained about 0.3 mg Hg/kg (Figure 4). No larger bass were examined. Deep Creek Lake has many characteristics that make it a candidate for elevated MeHg production and bioaccumulation (acid deposition and acid mine drainage, high atmospheric Hg deposition, reservoir), and it is a heavily used recreational fishery. Deep Creek Lake should become a focus of monitoring efforts for Hg in Maryland fish.

Another reservoir of potential concern is Liberty Reservoir, a recreational fishery northwest of Baltimore. Most of the

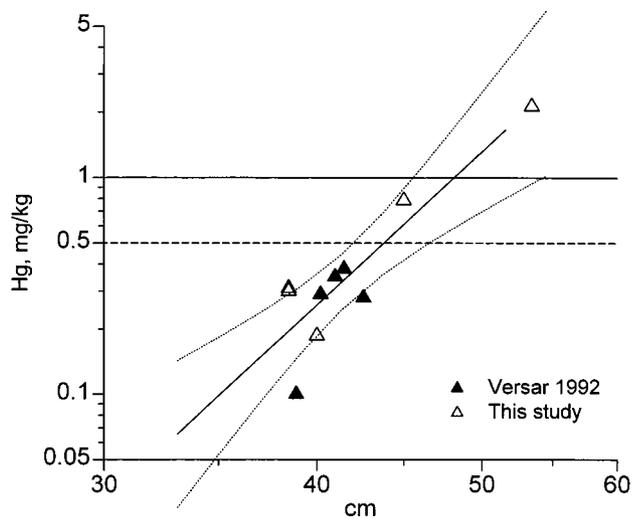


Fig. 5. Mercury concentrations in chain pickerel in Deep Creek Lake, including data from this study and from Versar (1994). The data sets were not significantly different. Regression of both data sets together gave $p < 0.01$. Dotted lines represent 95% confidence intervals around the mean

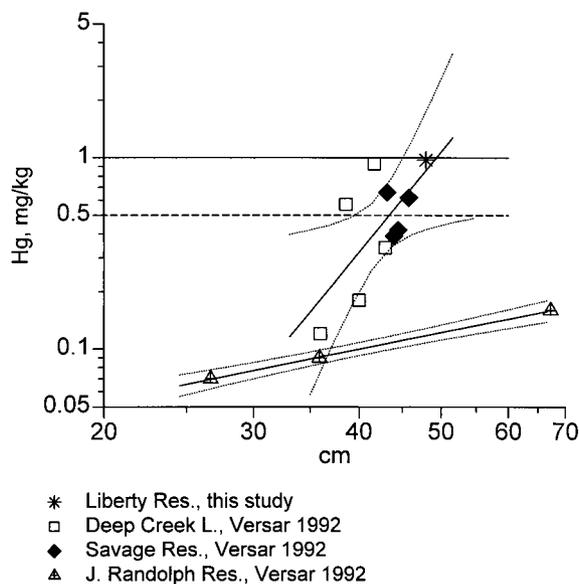


Fig. 6. Relationship between Hg concentration and fish length in walleye from four Maryland reservoirs, including data from Versar (1994) and from this study. Analysis of variance showed no site to site differences among Deep Creek L. and Savage and Liberty Reservoirs; they are regressed together. The slope of the regression was significantly lower for fish from Jennings Randolph Reservoir. Dotted lines represent 95% confidence intervals around the means. Both regressions are significant at $p < 0.05$

freshwater striped bass examined came from this lake, and all exceeded 0.5 mg Hg/kg (Figure 1). The one walleye examined from Liberty Reservoir was 48 cm long and contained 0.98 mg Hg/kg, similar to walleye in Deep Creek Lake (Figure 6). This value is at the upper end of the distribution of Hg concentra-

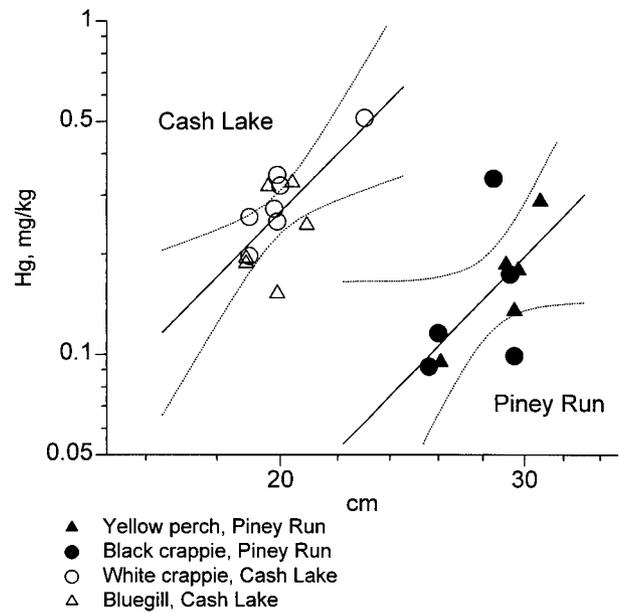


Fig. 7. Comparison of Hg levels in panfish between two lakes, Cash Lake and Piney Run. For each lake, the regression lines and their 95% confidence intervals are based on both species shown. The regressions of Hg concentration on both weight and length were significant for Cash Lake white crappie and Piney Run yellow perch and for the combined panfish data sets for each lake ($p < 0.01$)

tions in similar size walleye from Wisconsin lakes (Wiener *et al.* 1990) and Ontario lakes (Ontario Ministry of the Environment 1997), both areas with generally high Hg levels in fish.

Large differences in size-normalized Hg concentrations in panfish examined in two impoundments, Cash Lake in Prince Georges County and Piney Run Reservoir in Carroll County, highlight the large differences in MeHg production and bioaccumulation among ecosystems (Figure 7). Small sportfish (white crappie, *Pomoxis annularis*, and bluegill, *Lepomis macrochirus*) in Cash Lake contained significantly more Hg per unit weight than did small sportfish (black crappie, *Pomoxis nigromaculatus*, and yellow perch) in Piney Run Reservoir. Mercury levels in yellow perch taken from Piney Run were not significantly different than Hg levels in yellow perch taken from the estuarine Patuxent River (Table 1) and appear lower per unit size than perch from lakes in Maine and Michigan, many of which are influenced by either low pH or high DOC, water quality factors that favor MeHg production and bioaccumulation. The lakes are in different geographic provinces, Cash Lake in the coastal plain and Piney run in the Piedmont. Perhaps the higher Hg levels in Cash Lake fish reflect the poor buffering capacity of the coastal plains soils. However, a number of factors, such as catchment size and lake chemistry, affect Hg methylation and accumulation. Mercury levels in top predators in Cash Lake should be examined.

Largemouth bass also showed differences among water bodies (Table 2), with higher levels in fish from the fresh waters examined, Deep Creek and St. Mary's Lakes, than Maryland estuarine waters. Table 2 lists Hg concentrations in largemouth and smallmouth bass measured in this study, in comparison with fish of roughly comparable size from lakes in other

Table 1. Comparison of Hg concentrations in yellow perch in this report with selected North American lakes

Location	Mean length (cm)	Mean Hg (mg/kg)	Citation
Patuxent River, MD	29	0.15	This report
Piney Run Reservoir, MD	29	0.18	This report
Maine lakes	23	0.28	Stafford and Haines 1997
Michigan UP lakes (seepage)	30	0.35	Grieb <i>et al.</i> 1990
Michigan UP lakes (drainage)	30	1.0	Grieb <i>et al.</i> 1990

Table 2. Comparison of Hg concentrations in like-sized largemouth (LM) and smallmouth (SM) bass among North American lakes

Location	LM or SM	n	Mean length (cm)	Mean Hg (mg/kg)	Citation
Potomac River, MD	LM	9	44	0.16	This report
Patuxent River, MD	LM	2	42	0.18	This report
Deep Creek Lake, MD	SM	3	31	0.25	This report
Lake Tohopekaliga, FL	LM	many	30	0.27	Lange <i>et al.</i> 1994
St. Mary's Lake, MD	LM	1	28	0.49	This report
Ontario lakes	LM	14	30	0.46	Wiener and Spry 1996
Ontario lakes	SM	72	30	0.53	Wiener and Spry 1996
Maine lakes	LM	15	31	0.56	Stafford and Haines 1997
Maine lakes	SM	56	32	0.66	Stafford and Haines 1997
Florida lakes	LM	25	36	0.76	Lange <i>et al.</i> 1993

regions. Note that the mean size of the tidal largemouth bass examined was larger than the mean size of freshwater fish in this study, and most of the freshwater data sets shown in Table 2. Many of the lakes for which data are given in the table are affected by acid deposition, depressed pH, or high DOC. Bass from both St. Mary's and Deep Creek showed similar size to Hg ratios as bass from lakes in other states where consumption advisories are posted, and bass from the Patuxent and Potomac Rivers contained less Hg at larger sizes. St. Mary's Lake is a poorly buffered, high-DOC lake. A more careful examination of bass from both Deep Creek and St. Mary's Lake should be considered. Both lakes have substantial recreational fisheries.

Summary

This survey highlights the large differences in MeHg production and bioaccumulation between water bodies within a region. For example, 30-cm smallmouth bass from Deep Creek Lake averaged about 0.25 mg Hg/kg, and a largemouth bass from St. Mary's Lake contained double that value, and estuarine largemouth bass were predicted to contain less than 0.08 mg Hg/kg at 30 cm. Thus, size-specific Hg concentrations in *Micropterus* vary by at least a factor of five among Maryland water bodies. These differences are probably not entirely due to differences in Hg deposition. Mercury deposition rates vary by perhaps a factor of two to three across Maryland and are highest near urban areas (Mason *et al.* 1997). Mercury concentrations in fish did not follow this pattern. For example, chain pickerel in western Maryland reservoirs contained some of the highest Hg concentrations observed in this study, while large piscivores from Chesapeake Bay, downwind of the Baltimore-

Washington industrial corridor, contained relatively low Hg burdens. Perhaps the most obvious differences are those between fish in fresh and estuarine waters. We have previously speculated that MeHg production in estuarine sediments is limited by sulfide (Gilmour and Henry 1991; Benoit *et al.* 1998). There are also significant differences in MeHg accumulation among lakes. Differences in Hg in fish are functions of differences in the catchment size and morphology, food web structure, and especially in the net rate of MeHg production between the ecosystems.

The experiences of other U.S. states and other countries suggest that certain types of water bodies in the state of Maryland will contain fish that exceed common consumption advisories for Hg. The limited data presented here support that hypothesis. The results of this survey are not meant to be a statistically rigorous assessment of Hg levels in Maryland fish, nor does this study provide sufficient information to make decisions about consumption advisories in Maryland. Further assessment of Hg levels in Maryland sportfish is needed to protect human and wildlife health.

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