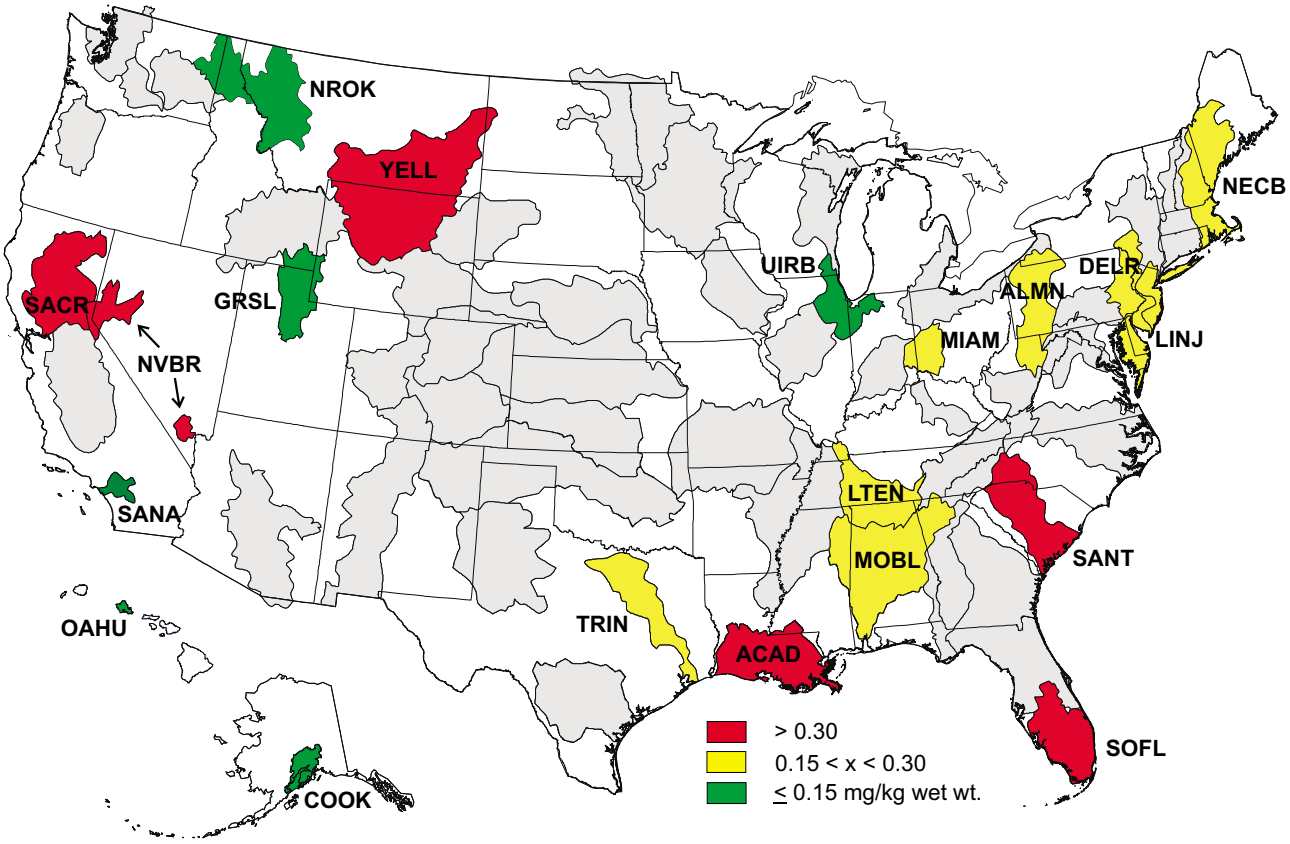
A National Pilot Study of Mercury Contamination of Aquatic Ecosystems Along Multiple Gradients: Bioaccumulation in Fish

Biological Science Report
USGS/BRD/BSR–2001-0009



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Front cover: The U.S. map shows 20 USGS National Water Quality Assessment (NAWQA) basins sampled in this study, categorized by geometric mean of mercury concentration in fish fillet samples. Basins shaded in gray delineate NAWQA study units not sampled.

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Biological Science Report
USGS/BRD/BSR–2001-0009
September 2001

by

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A National Pilot Study of Mercury Contamination of Aquatic Ecosystems Along Multiple Gradients: Bioaccumulation in Fish

Abstract: Water, sediment, and fish were sampled in the summer and fall of 1998 at 106 sites from 20 U.S. watershed basins to examine relations of mercury (Hg) and methylmercury (MeHg) in aquatic ecosystems. Bioaccumulation of Hg in fish from these basins was evaluated in relation to species, Hg and MeHg in surficial sediment and water, and watershed characteristics. Bioaccumulation was strongly (positively) correlated with MeHg in water ($r = 0.63$, $\rho < 0.001$) but only moderately with the MeHg in sediment ($r = 0.33$, $\rho < 0.001$) or total Hg in water ($r = 0.28$, $\rho < 0.01$). Of the other measured parameters, pH, DOC, sulfate, sediment LOI, and the percent wetlands of each basin were also significantly correlated with Hg bioaccumulation in fish. The best model for predicting Hg bioaccumulation included MeHg in water, pH of the water, % wetlands in the basin, and the AVS content of the sediment. These four variables accounted for 45% of the variability of the fish fillet Hg concentration normalized (divided) by total length; however, the majority was described by MeHg in water. A MeHg water concentration of 0.12 ng/L was on average, associated with a fish fillet Hg concentration of 0.3 mg/kg wet weight for an age-3 fish when all species were considered. For age-3 largemouth bass, a MeHg water concentration of 0.058 ng/L was associated with the 0.3 mg/kg fillet concentration. Based on rankings for Hg in sediment, water, and fish, sampling sites from the following five study basins had the greatest Hg contamination: Nevada Basin and Range, South Florida Basin, Sacramento River Basin (California), Santee River Basin and Coastal Drainages (South Carolina), and the Long Island and New Jersey Coastal Drainages. A sampling and analysis strategy based on this pilot study is planned for all USGS NAWQA study units over the next decade.

Key Words: Mercury, methylmercury, bioaccumulation, fish, water, sediment

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INTRODUCTION

Methylmercury (MeHg) is a potent neurotoxin that is among the most widespread contaminants affecting our Nation's aquatic ecosystems. Human fish-consumption advisories for Hg in fish have been issued in more than 40 states and account for more than eighty percent of all such advisories in the Nation (USEPA, 1998). While the threat to humans is very real, there is potentially a more serious threat to piscivorous wildlife, which consume relatively large quanitities of fish (Wiener and Spry, 1995). In Part One of this study (Krabbenhoft and others, 1999), Hg methylation efficiency was evaluated in sediment and water from watersheds that were sampled concurrently with the fish described by this report. Among other findings, MeHg production efficiency was highest in Eastern coastal basins containing high wetland densities. Nationwide, MeHg production was highest in sub-basins characterized as mixed agriculture and forested. In this report, relationships among Hg and MeHg in sediment and water are compared with bioaccumulation in fish axial muscle (the dominant repository for MeHg in fish) for 20 basins nationwide. The importance of total Hg load, methylation efficiency, MeHg in sediments and water, and selected watershed characteristics in determining the bioaccumulation of Hg in fish is assessed.

The USGS National Mercury Pilot Study

This study was conducted jointly by the National Water Quality Assessment (NAWQA) program, Toxic Substances Hydrology program, Wisconsin District Mercury Laboratory (WDML), and Columbia Environmental Research Center (CERC) of the U.S. Geological Survey. The overall objective was to identify ecosystem characteristics that favor the production and bioaccumulation of MeHg and to compare bioaccumulation rates on a national basis. Bioaccumulation of Hg in fish is a complex function of total Hg load, methylation efficiency, fish size, and the food chain dynamics in a given water body (Kidd and others, 1995). Consequently, aquatic ecosystems with high loads may have only moderate bioaccumulation in fish if methylation efficiency is low and conversely, significant bioaccumulation may result when loading is low if methylation efficiency is high. Regional- and national-scale fish surveys have been conducted in the past for Hg and other bioaccumulative contaminants. However, this is the first national-scale study in which water, sediment, and fish were sampled together with low-level Hg speciation analysis.

EXPERIMENTAL SECTION

Study Design

The major design characterics of this study were (1) national scope, (2) sampling of water, sediment, and predator fish muscle, (3) consistent use of ultra-trace clean sampling methods, (4) ultra-trace total and methyl-mercury analytical procedures, and (5) analysis of all routine water and sediment quality parameters. Sampling was conducted from June to October, 1998 at 3 to 8 sites from 20 of the 59 study units (front cover and Table 1) of the USGS NAWQA program (http://water.usgs.gov/nawqa). Nationally, these basins span the dominant east-to-west mercury deposition gradient (USEPA, 1997) and represent a wide range of environmental settings. Individual study basin teams were asked to choose sites within a basin which spanned gradients of wetland density, surface water pH, sulfate, total organic carbon, and suspected or known Hg loading. Most sampling sites were on streams. Some of the sites had high Hg loading from known Hg point sources.

Field crews were asked to focus on largemouth bass (*Micropterus salmoides*) or other black bass (*Micropterus sp.*) of age 3 years (estimated from regional growth rate data, when available) and to collect five individuals per site for compositing. Although collection objectives were not always met, all fish samples submitted were analyzed. In some cases, fish were analyzed individually to avoid creating composites of multiple species or ages. Axial muscle (fillet) was targeted for Hg analysis because it is generally the dominant and most stable repository in fish (Goldstein and others, 1996). Only total Hg was determined in the fish because virtually all of the Hg in the fillet is present as MeHg (Bloom, 1992). Black bass were targeted as ubiquitous predator fish which could facilitate inter-basin comparisons. Also, they might be expected to correlate well with localized sediment and water conditions because they normally inhabit relatively small ranges as compared to nomadic predators such as walleye (*Stizostedion vitreum*) or white bass (*Morone chrysops*), (Carlander, 1977). However, if black bass species were expected to be absent at one or more sites within a basin, samplers were advised to collect a predator species common to the entire basin so that gradients within each basin could be examined.

Age-3 fish were targeted because: (1) they should be relatively plentiful and of reasonable size for sampling fillets, (2) differences in Hg accumulation between males and females of the same size should be small, i.e., sexual growth dimorphism for largemouth bass is reportedly minimal up to this age (Lange and

Table 1. Basins, number of sites, and number of fish samples analyzed.

Abbrev.	Study Basin Name	no. sites	no. samples[a]
ACAD	Acadian-Ponchartrain Basin	5	5
ALMN	Allegheny and Monongahela River Basins	5	6
COOK	Cook Inlet Basin (Alaska)	4	6
DELR	Delaware River Basin	9	12
GRSL	Great Salt Lake Basins	2	4
LINJ	Long Island and N.J. Coast Drainages	4	13
LTEN	Lower Tennessee River Basin	3	8
MIAM	Miami River Basin (Ohio)	7	13
MOBL	Mobile River and Tributaries	7	15
NECB	New England Coastal Basin	5	5
NROK	Northern Rockies Intermontane Basins	2	2
NVBR	Nevada Basin and Range	1	8
OAHU	Oahu Island	6	6
SACR	Sacramento River Basin	5	11
SANA	Santa Ana River Basin	4	4
SANT	Santee River Basin and Coastal Drainages	5	5
SOFL	South Florida Basin	2	6
TRIN	Trinity River Basin	5	18
UIRB	Upper Illinois River Basin	5	6
YELL	Yellowstone River Basin	5	6

[a] Samples per basin, each consisting of either a homogenized composite or an individual depending on size and species submitted.

others, 1994) and (3) age-3 fish should be old enough to exhibit significant bioaccumulation of Hg. Older fish generally bioaccumulate higher concentrations of Hg but would be more difficult to capture in a consistent age class. Also, concentrations of Hg in older fish might be less representative of recent conditions contributing to the observed concentrations of Hg in the sediment and water.

Sample Collection and Preparation

Sampling and analysis of sediment and water was described in Part One of this study (Krabbenhoft and others, 1999). Of note, surface water was not filtered and sediments were taken from the top 2 to 4 cm. Ultra-trace protocols were followed for the sampling of water for Hg. Fish samples were collected by the most efficient means available, usually by electroshocking or gill-netting. Each fish was rinsed in stream water, measured for length and weight, double bagged in zip-seal plastic, and placed on dry ice as soon as possible. Samples from the ACAD and SANT basins were filleted by field crews, who also determined the ages of their specimens. Most samples were shipped within 48 hours of collection, but some were stored frozen for 1 to 2 weeks before shipment. Once received by CERC, they were stored at –20 °C for 3 to 6 months before the fillets were prepared for analysis.

Before removing fillets, individual fish from each site were thawed at room temperature for 1 to 2 hours, depending on size. Several scales were removed from behind the gill cover for aging. Each fish was then rinsed with laboratory-grade deionized (DI) water (> 10 Mohm-cm) and placed on a polypropylene cutting board situated in a polypropylene bin. Fillet knives with either ceramic (ZrO) or titanium-aluminum alloy (for larger fish) blades were used to remove a skinless, boneless (belly flap removed) fillet from the left side of each specimen. Each fillet (including those removed by field crews) was chopped into 2-cm square sections, placed in a heavy-duty polyethylene zip-seal freezer bag, rinsed twice with ultra-pure (UP) water (18 Mohm-cm), drained thoroughly and returned to the freezer. Between samples, the knife and cutting board were scrubbed with tap water and detergent, then rinsed with DI water, 1% (v/v) ultra-pure nitric acid, and HPLC-grade methanol. Equipment and cleaning procedures used for the few samples filleted by field crews were not necessarily as described above. However, we assume that potential surface contamination of all fillets was minimized by rinsing twice with UP water before

analysis.

For each site, fillets of the same species were composited for all fish of a similar size (assumed to be of similar age). However, for many sites the sampled fish varied greatly in size and apparent age, consequently some individuals were analyzed separately to avoid compositing fish of differing ages. For a few sites, the fish collected were too small (< 50 g) to conveniently remove a fillet; these specimens were chopped and processed either whole or whole, less heads. After all samples were filleted, the frozen muscle sections were placed into an acid-washed borosilicate glass jar and lyophilized (freeze-dried) to a constant dry weight at -5°C with a vacuum of about 100 mtorr. Lyophilization facilitates sample manipulation and storage but does not cause loss of biologically-incorporated Hg from tissue (LaFleur, 1973; Lasorsa and Allen-Gil, 1995). The dry product was briefly blended with a hand-held high-speed bio-homogenizer having a polystyrene bowl fitted with a stainless steel cutting blade. The base and blade assembly were washed between samples with detergent and hot water, rinsed with UP water, and dried with filtered compressed air. A representative portion of each homogenate was immediately transferred to a borosilicate glass vial fitted with a telfon-lined cap for storage in a desiccator.

Hg Determination

Tissue samples were digested before determination of Hg with microwave heating in sealable tetra-fluorinated ethylene (TFE) pressure vessels. Five mL of HNO_3 and 0.5 mL of HCl (each sub-boiled in quartz and stored in a TFE bottle) were added to a 0.5-g dry tissue sample and the vessel was sealed and placed overnight in a water bath at 70°C. The vessel was cooled, vented, then heated with a 3-step microwave program. After cooling, 1 mL of ultra-pure 30% H_20_2 was added, the vessel was sealed, and the 3-step heating program was repeated. The vessel was again cooled and the liquid contents were quantitatively transferred and diluted to 100 mL with 1% (v/v) HCl in an acid-cleaned polyethylene bottle. After briefly mixing, a 30-mL portion of the digestate was immediately transferred to a borosilicate glass tube and capped until analysis.

Analysis for Hg was conducted by cold-vapor atomic absorption spectrophotometry with flow injection sample introduction and stannous chloride reduction. Standards used for calibration included solutions containing 0.0, 5.0, 10.0, and 20.0 ng Hg/mL. Quality assurance samples analyzed included method (digestion) blanks, reference tissues, replicate samples, pre-digestion spikes (MeHg), post-digestion spikes (Hg^{2+}), and calibration and blank verification solutions. Sample results were blank-corrected based on the mean of three method blanks processed with each digestion set.

Age Determination

Age was estimated by scale analysis (Jearld, 1983) except for fish from SOFL and ACAD study units, for which sagittal otiliths were analysed (Porak and others, 1988). Scales were soaked in 70% ethanol to clean debris, increase transparency and soften them for flattening. Annuli were determined by two separate readers with the aid of a microfiche reader. A third reader was used in the case of discrepancies. All scale samples (about 10 per fish) were examined for each fish; the scale with the clearest markings was then used by all readers. Because samples were collected in late summer and fall, numerical ages of individuals that were spring spawners were assigned increments of 0.5 yr, whereas fall spawners (e.g., Salmonidae) were assigned increments of whole years. For example, a largemouth bass determined to be age-0 or age-1 was assigned a value of 0.5 or 1.5 yr, respectively. Due to resource limitations, not all individuals for composite samples were aged. Rather, the average for two representative individuals was used to estimate the age for each composite. Consequently, the age assigned for each composite was either a whole number or an increment of 0.5.

Statistical Analysis and Modelling

Linear regression and correlation analysis was conducted using the following variables: Hg concentration (µg/g wet wt.) in fish (Hg_{fish}), fish age (yr), fish length (m), fish weight (kg), methylmercury concentration (ng/g dry wt.) in sediment ($MeHg_{sed}$), total mercury concentration (ng/g dry wt.) in sediment ($HgTot_{sed}$), methylmercury concentration (ng/L) in water ($MeHg_{water}$), total mercury concentration (ng/L) in water ($HgTot_{water}$), percent methyl mercury in sediment, acid-volatile sulfide (AVS) in sediment (µmol/g dry wt.), sediment percent loss on ignition (LOI - an estimate of organic matter), dissolved organic carbon (DOC) in water (mg/L), sulfate concentration in water (wat_{sulf}), water pH (wat_{pH}), and % wetlands (of basin). We examined both Hg_{fish} and Hg_{fish} divided by weight, length, or age, as the dependent variable. Statistical analysis did not include results for fish determined to be less than one year of age, nor the results from the NBVR basin because of the extraordinarily high concentrations. For the multiple regression, residuals and partial plots were used to determine whether transformations were needed to meet the assumptions

of a good regression model (Helsel and Hirsch, 1992). The natural logarithm was used for Hg_{fish}, $MeHg_{water}$, $HgTot_{water}$, $HgTot_{sed}$, and $MeHg_{sed}$, and a cube root transformation for AVS, to obtain approximately normally-distributed residuals with constant variance. Several reasonable candidate regression models were selected using the adjusted R^2 (coefficient of determination) statistic. From this list, the one with the best predictive power (lowest PRESS statistic) was chosen. For principal components analysis (PCA), all data were processed using SIMCA-P (ver 8.0, Umetrics AB). The models and principal components were evaluated using pattern recognition of score (sample) plots and loading (variable) plots. As with the regression models, NVBR data was excluded from the analysis. Additionally, a subset consisting of sites where largemouth bass (LMB) were collected was modeled and evaluated in the same manner as the complete data set. For PCA only, the full data set and the LMB subset were also analyzed with the three size variables (age, length, weight) excluded so that influences of the remaining independent variables on Hg_{fish} could be more clearly examined.

Quality Assurance

Quality control results for the fish determinations of Hg were as follows: recovery of pre-digestion spikes of MeHg averaged 102.3 % (s.d. 4.2, n = 16); mean recovery for post digestion spikes of Hg^{2+} was 98.5 % (s.d. 5.1, n = 25). The measured values for three reference fish samples were in good agreement with certified or control ranges (Table 2). Precision for triplicate determinations (including digestion and analysis) averaged 2.6 % relative standard deviation (RSD) and ranged from 0.6 to 7.5% RSD (n = 8). The method detection limit, calculated for each of the three digestion blocks based on three times the pooled standard deviation of the method blanks (three per block) and a low-level sample analyzed in triplicate, ranged from 0.004 to 0.020 µg/g wet wt. Overall, eight of the nine method blanks were near or below the instrument detection limit (about 0.05 µg/L). The one elevated blank (0.3 µg/L) resulted in a higher reporting method detection limit (0.020 µg/g wet wt.) for samples determined in the same digestion block. However, virtually all sample concentrations were considerably above this highest blank. Consequently, the potential error associated with this worst-case blank was relatively small and it would affect the accuracy of only the very lowest sample concentrations. Overall, the results for quality assurance samples indicated good accuracy and precision for the study samples.

RESULTS AND DISCUSSION

Complete sample site information, species, weights, lengths, ages, and mercury concentrations in fish samples are listed in the Appendix; summary statistics are given in Table 3. For fish greater than 0.5 yr in age, the arithmetic means for total length, weight, and age were as follows: all species – 260 mm, 324 g, 3.2 yr; largemouth bass – 280 mm, 420 g, 3.2 yr; smallmouth bass – 261 mm, 299 g, 3.2 yr. For all fish samples (n = 159) the following statistics for Hg concentration (µg/g wet wt.) were obtained: arithmetic mean, 0.478; geometric mean, 0.218; median, 0.206; minimum, 0.018; maximum, 5.84. For largemouth bass (n = 50) these same statistics were 0.510, 0.329, 0.292, 0.045 and 4.22; for smallmouth bass (n = 37) the values were 0.244, 0.195, 0.205, 0.042 and 1.05, respectively. In comparison, the nationwide geometric mean for 27 largemouth bass composites sampled as part of the 1984 National Contaminant Biomonitoring Program (NCBP, Schmitt and Brumbaugh, 1990) is estimated at 0.31 µg/g wet weight for the fillet based on a conversion equation from whole fish (Goldstein and others, 1996). Thus, in general the national mean concentration of Hg in largemouth bass from our study (0.33) is similar to samples collected 15 years previously for the NCBP. Of course, this comparison does not account for possible differences in age or size of the samples, or differences in watershed coverage.

Table 2. Measured mercury concentrations (µg/g dry wt.) for fish reference tissues (n =3 for each).

MATERIAL I. D.	Fish Matrix (Common Name)	Measured Mean (std.dev.)	Certified or Control Range
CERC STB	Whole Striped Bass	2.21 (0.01)	2.26 ± 0.51
NIST RM-50	Albacore Tuna Fillet	0.99 (0.04)	0.95 ± 0.10
NRCC Dorm-1	Dogfish Fillet	0.90 (0.06)	0.80 ± 0.07

Table 3. Summary statistics for mercury concentrations (µg/g wet wt.) in fish fillet samples.

Statistic	All Samples (n = 159)	Largemouth Bass (n = 50)	Smallmouth Bass (n = 37)
Mean	0.478	0.510	0.244
Median	0.206	0.292	0.205
Geometric Mean	0.218	0.329	0.194
Minimum	0.018	0.045	0.042
Maximum	5.84	4.22	1.05

Mean Fish Concentration by Basin

The geometric means of fish Hg concentrations for each of the 20 study unit basins are presented in Figure 1. Because Hg concentrations in fish from a given body of water are usually a function of size or age (Wiener and others, 1990, Lange and others, 1994) and various sizes and species of fish were collected, the geometric means for the Hg concentrations normalized (divided) by weight, length, and age are also presented in Table 4. All age-0 fish (n = 7) were excluded for this comparison due to the high relative uncertainty associated with the assignment of a fractional age of less than one. For comparing Hg in fish between waterbodies, it would be preferable to conduct an analysis of covariance between concentration and size and adjust each treatment (site) mean to a uniform size variable (Sorenson and others, 1990; Lange and others, 1993). However, for most of our sites we had insufficient observations for this approach. As indicated in Table 4, NVBR, SOFL, SANT, and SACR basins consistently ranked high by any of four measures of Hg bioaccumulation in fish. The YELL, ACAD, NECB, and LINJ basins also ranked high or moderately high by these measures. The extraordinarily high concentration for NVBR fish compared with the other basins (Figure 1) is striking. Indeed, sections of the Carson River Basin are reported to be among the most severely Hg-contaminated in the world (Bonzongo and others, 1996). However, our data from this basin as a whole are greatly skewed relative to the other basins because all samples came from one severely contaminated site (Lahontan Reservoir). Similarly, fish samples from the SOFL unit were limited to two rather contaminated sites, which probably yielded a somewhat elevated mean for that study unit basin, although Hg contamination there is widespread. Means among the other 18 basins varied by a factor of about 20. The wide variety of fish species that were sampled probably factored into this range. Whereas over 65% of our samples were either largemouth-, smallmouth-, spotted-, or white bass, at least 16 other species were also represented. Also, sampling sites were selected to represent a gradient of environmental conditions and Hg levels, but they do not necessarily represent a systematic coverage of each entire basin. Nevertheless, the rankings listed in Table 4 give a reasonable guide as to the relative Hg contamination in fish among the basins sampled.

Ranking of basins for fish concentrations normalized by either length or age tended to mirror the rankings for non-adjusted concentrations. However, normalization by weight yielded some differences in rankings when compared to rankings by other means. For example, fish from the NECB and COOK study units ranked much higher for weight-normalized data as compared to length- or age-normalized data. However, samples from each of these two study units were quite small in size and were of a species that was uncommon for the data set (mixed sunfish and dolly varden, respectively). Also, the fillet data for some of the samples from these two study units were actually estimated from whole-body analysis based on a conversion equation derived from larger species (Goldstein and others, 1996). In the case of length-adjusted data, the largest specimens might have been favored due to the fact that as most fish age they have diminished increases in length relative to weight (Carlander, 1977). On the other hand, normalization by weight may have favored fish samples that were very small because regression equations for Hg concentration with fish weight tend to have a higher positive y-intercept for Hg contaminated systems relative to length-normalized data (Lange and others, 1994). These combined factors might explain why small sunfish from the NECB ranked highest by the weight-normalized criteria, but ranked fourth or lower by other measures (Table 4). Regardless of the fish ranking method chosen, the MeHg concentrations

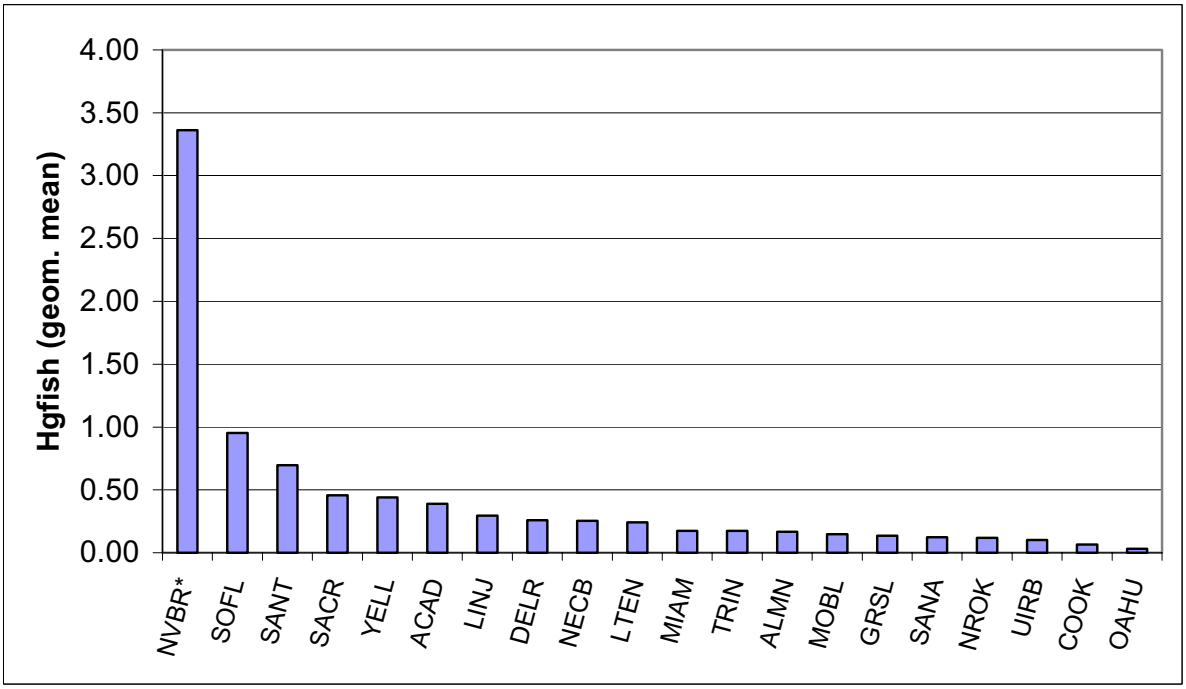

Figure 1. Geometric mean of Hg concentration in fish fillet samples collected for each of the 20 basins. *Samples from NVBR represent only one site (Lahontan Reservoir).

in both the sediment and water from the NECB basin were relatively high (Krabbenhoft and others, 1999), therefore, a high ranking for the fish is not surprising. But the comparison of bioaccumulation for NECB samples with the other basins must be viewed with caution because sunfish do not bioaccumulate Hg as rapidly as larger predator species and the fillet concentrations were estimated from the whole body measurements. In any event, a statewide Hg advisory is presently in effect for Massachusetts (where all of the NECB samples were collected).

Basins of Concern for Human Health

Our study was not designed to address the safety of consuming fish from the various participating study units, or to assess Hg exposure risks to fish and wildlife. Most, if not all of the basins of concern have already been identified by state and other federal agencies. Assessing the toxicological significance of Hg concentrations in fish with respect to populations of fish and fish-consuming wildlife is a complex matter that would be difficult to address from our data considering the the limited number of samples collected for each basin. Sensitivity to MeHg exposure can vary greatly among species and the rate of accumulation in fish apparently affects the toxicity (Wiener and Spry, 1996). Furthermore, the concentration of Hg in tissues other than the axial muscle, such as the brain or in the eggs, would generally be more useful for assessing potential impacts on fish (Wiener and Spry, 1996), whereas whole-body concentrations might be more meaningful for assessing impacts to piscivorous waterfowl and mammals (Wolfe and others, 1998). In order to address human health risks from fish consumption, we would have targeted fish older than 3 years, which typically contain higher concentrations of Hg and are more commonly sought by anglers. But because human health issues invariably arise when nation-wide fish residue data is examined, this aspect is briefly addressed.

Fish from our study that had Hg concentrations above 0.50 µg/g wet weight are listed in Table 5. Historically, 0.50 µg/g was a commonly reported human health advisory Hg fish concentration applicable to consumers of “high risk” categories, e.g., children, expectant mothers, and sub-populations whose diets include large percentages of fish (U.S. EPA, 1998). Recently, the U.S. EPA reduced the human health Hg fish criteria to 0.30 µg/g wet weight (U.S. EPA, 2001). Our results document that Hg contamination in U.S. freshwater fish is a widespread problem. One or more samples from nine of the 20 basins exceeded the 0.50 ug/g wet weight criteria and 15 of the basins had at least one sample above the 0.30 criteria. State fish

Table 4. Ranking of basins by geometric mean mercury concentration of fish fillet (µg/g wet wt) : unadjusted, or normalized by age, length, and weight.

Rank	Basin and geometric mean concentration (µg/g wet)[a]			
	unadjusted	÷ age (yr)[b]	÷ length (m)	÷ weight (kg)
1	NVBR[c] (3.34)	NVBR[c] (0.86)	NVBR[c] (9.2)	NECB[d] (8.7)
2	SOFL (0.95)	SOFL (0.30)	SOFL (3.1)	NVBR (4.8)
3	SANT (0.70)	SACR (0.15)	SANT (2.6)	SOFL (2.6)
4	SACR (0.46)	NECB[d] (0.12)	NECB[d] (2.3)	LINJ (2.5)
5	YELL (0.44)	YELL (0.12)	SACR (1.5)	SANT (2.2)
6	ACAD (0.39)	ACAD (0.12)	ACAD (1.5)	COOK[e] (2.1)
7	LINJ (0.29)	LINJ (0.12)	LINJ (1.3)	LTEN (1.7)
8	DELR (0.26)	LTEN (0.10)	YELL (1.2)	ACAD (1.5)
9	NECB[d] (0.25)	DELR (0.08)	LTEN (1.1)	SANA (1.2)
10	LTEN (0.24)	ALMN (0.06)	DELR (0.9)	SACR (1.1)
11	MIAM (0.17)	SANA (0.06)	COOK[e] (0.8)	DELR (0.9)
12	TRIN (0.17)	MIAM (0.05)	MIAM (0.7)	MOBL (0.9)
13	ALMN (0.17)	TRIN (0.05)	SANA (0.7)	YELL (0.8)
14	MOBL (0.15)	MOBL (0.05)	TRIN (0.6)	MIAM (0.8)
15	GRSL (0.13)	GRSL (0.04)	ALMN (0.6)	UIRB (0.8)
16	SANA (0.12)	UIRB (0.04)	MOBL (0.6)	TRIN (0.6)
17	NROK (0.12)	NROK (0.03)	UIRB (0.5)	ALMN (0.6)
18	UIRB (0.10)	COOK[e] (0.03)	GRSL (0.4)	GRSL (0.4)
19	COOK[e] (0.07)	OAHU (0.01)	NROK (0.4)	NROK (0.3)
20	OAHU (0.03)	-----	OAHU (0.2)	OAHU (0.2)

[a] Excludes age-0 fish.
[b] No age data available for SANT.
[c] All samples from one site (Lahanton Reservoir).
[d] Estimate for fillet based on log [muscle] = 0.35 + 0.92*log[whole body] (Goldstein and others, 1996).
[e] Dolly varden only; fillet concentration estimated for 2 of 3 samples.

consumption advisories for Hg are currently in effect for most sites in the nine basins that exceeded the 0.50 criteria; exceptions include the SACR, MIAM, and MOBL basins. In addition, five states not included in our study (VT, MI, NH, IN, MO) currently have statewide advisories in effect for consumption of one or more species of freshwater fish due to Hg. Two other states not covered (MN and WI) have Hg advisories on a large number of water bodies.

Sources and Factors Enhancing Bioaccumulation of Hg in Fish

Among the primary basins of concern, the geographic and land-use categories varied greatly for the sub-basins from which fish above advisory concentrations were sampled. However, fish from our study with concentrations above 0.50 µg/g were most commonly from coastal or lowland primary basins, e.g. SOFL, SANT, LINJ, ACAD, SACR, MOBL. Presumably, the relatively high percentages of wetlands in these lowland basins enhance methylation rates and in turn, bioaccumulation rates of Hg in fish (St. Louis and others, 1994; Hurley, 1995: Krabbenhoft and others, 1999). The sources of Hg among these basins varies widely.

In the NVBR and SACR basins, nearby cinnabar (HgS) deposits and elemental Hg in streambeds resulting from past gold mining amalgamation practices continue to contribute to elevated Hg concentrations in fish (Bonzango and others, 1996; Domagalski, 1998). In the YELL basin, elevated Hg concentrations in reservoir fish have been associated with coal and phosphate deposits of Wyoming and Montana (May and McKinney, 1981). In a detailed study of that basin, it was concluded that elevated Hg in reservoir fish resulted primarily from weathering of soils and rocks upstream and that reservoirs furthest upstream

Table 5. Fish samples with Hg fillet concentrations greater than 0.50 µg/g wet wt (-- = no data).

Study Unit	Site Name	Species (no. of indiv.)	Mean Wet Weight (g)	Hg conc. µg/g wet wt.	Advisory[a] In Effect?
NVBR	Lahontan Reservoir, NV	White Bass (8)	694	3.36	Yes
SACR	Sacramento Slough nr. Knights Landing, CA	Largemouth Bass (1)	1471	2.17	No[b]
SOFL	Water Conservation District 3A15, FL	Largemouth Bass (3)	788	2.15	Yes
SANT	N. Fork Edisto R. nr. Fairview Crossroad, SC	Largemouth Bass (1)	907	1.82	Yes
SACR	Bear River @ Hwy 70, CA	Largemouth Bass (1)	518	1.21	No[b]
SACR	Bear River @ Hwy 70, CA	Smallmouth Bass (1)	467	1.10	No[b]
LINJ	Great Egg Harbor @ Sicklerville, NJ	Chain Pickerel (2)	172	0.91	Yes[c]
ACAD	Bogue Falaya R. @ Covington, LA	Largemouth Bass (8)	--	0.83	Yes[d]
ACAD	Tangipahoa R. @ Robert, LA	Largemouth Bass (8)	--	0.77	Yes[d]
YELL	Shoshone River, @ mouth nr. Kane, WY	Walleye (5)	817	0.70	Yes
YELL	Bighorn Lake @ Hwy14A, WY	Walleye (5)	896	0.68	Yes[e]
YELL	Bighorn River nr. Kane, WY	Walleye (5)	452	0.66	No
YELL	Shoshone River @ mouth nr. Kane, WY	Walleye (1)	1444	0.66	No
SACR	Sacramento Slough nr. Knights Landing, CA	Largemouth Bass (1)	1156	0.65	No
MOBL	Satilpa Creek nr. Coffeeville, AL	Spotted Bass (2)	140	0.65	No
LINJ	Great Egg Harbor @ Sicklerville, NJ	Largemouth Bass (1)	49	0.65	Yes
SANT	N. Fork Edisto River nr. Branchville, SC	Largemouth Bass (1)	--	0.63	Yes
MOBL	Satilpa Creek nr. Coffeeville, AL	Largemouth Bass (1)	92	0.62	No
LINJ	Great Egg Harbor @ Sicklerville, NJ	Chain Pickerel (5)	84	0.59	Yes
SANT	S. Fork Edisto River @ Springfield, SC	Largemouth Bass (1)	--	0.58	Yes
SANT	S. Fork Edisto River nr. Canaan, SC	Largemouth Bass (1)	--	0.55	Yes
SOFL	Water Conservation District U3	Largemouth Bass (3)	254	0.55	Yes
SACR	Bear River @ Hwy 70, CA	Smallmouth Bass (1)	150	0.54	No[b]
MIAM	E. Fork L. Miami R. nr Williamsburg, OH	Smallmouth Bass (1)	608	0.51	No

[a]Source: USEPA, 1998
[b]Advisory by state of California pending (J. Domagalaski, pers. commun., April 1999)
[c]Statewide advisory for bass and pickerel in New Jersey.
[d] Statewide monitoring program for Hg in fish currently in progress.
[e]Advisory in effect for state of Montana but not Wyoming (April 1999).

exhibited higher bioaccumulation rates because of greater susceptibility to flood events (Phillips and others, 1984). Flooding results in greater scouring of Hg from soils but more importantly methylation of waterborne Hg is enhanced when terrestial vegetation is inundated for extended periods of time (which increases the dissolved organic carbon in the water) as is the case when new reservoirs are filled (Bodaly and others, 1997).

There may be localized point sources of Hg in the ACAD basin associated with Hg-charged manometers used with natural gas wells; however, this is primarily a problem in northeast Louisiana (Facemire and others, 1995). The state of Louisiana and the USGS have been engaged in a statewide assessment of Hg contamination in fish since 1993 (Louisiana Department of Environmental Quality, 1999). Selected water bodies in the SANT and MOBL basins may still be impacted by past uses of mercury in the chloralkali and paper mill industries (May and McKinney, 1981). However, widespread elevation of Hg in fish from the SANT basin has generally not been traceable to specific industrial or wastewater discharges (South Carolina Department of Health and Environmental Control, 1999).

Overlaying the numerous point sources is a broader contribution from atmospheric deposition of mercury originating primarily from emissions associated with waste incineration and coal combustion (Hanisch, 1998). Atmospheric deposition rates of mercury are generally greatest in Florida and the northeastern U.S. (USEPA, 1997). However, many lakes in the upper Midwest have also apparently been affected by atmospheric deposition and those with low buffering capacity (and depressed pH) are likely to have high Hg accumulations in fish (Sorenson and others, 1990; Wiener and others, 1990). There is currently debate as to how localized the effects of atmospheric deposition of Hg are with respect to the emission sources and ultimately, if proposed U.S. atmospheric emission controls for Hg would be effective in reducing Hg burdens in fish (Hanisch, 1998). An index of atmospheric Hg accumulation (AHA) developed in part one of this study indicated that among the study units examined, the NVBR, NROK, GRSL, COOK, and SACR basins are most influenced by ground-based Hg sources, whereas the ALMN, DELR, LINJ, SANT, and SOFL basins are most influenced by atmospheric sources of Hg.

Ranking of Basins by Hg in Fish, Sediment, and Water

The six basins with the greatest contamination, as determined by ranking of individual sites for selected fish, sediment, and water Hg measures, are presented in Table 6. Only the highest ranking site from each study unit was considered for this comparison, i.e., for some criteria multiple sites from one study unit may have ranked above sites from other study units, but only one site from each study unit is presented. The seven Hg criteria evaluated included 1) concentration (µg/g wet) of Hg in fish (Hg_{fish}), 2) Hg_{fish} divided by fish age (yr), 3) Hg_{fish} divided by fish length (m), 4) methylmercury concentration (ng/g dry wt.) in sediment ($MeHg_{sed}$), 5) total mercury concentration (ng/g dry wt.) in sediment ($HgTot_{sed}$), 6) methylmercury concentration (ng/L) in water ($MeHg_{water}$), and 7) total mercury concentration (ng/L) in water ($HgTot_{water}$). For the ranking of sites based on Hg_{fish} normalized by age, age-0 fish (n = 7) were excluded due to the large relative error in asssigning a fractional age.

Based on the three fish criteria the following study units had samples that ranked at least twice in the top six: NVBR, SOFL, SACR, LINJ, and SANT (Table 6). Basins with sites ranking once in the top six included ACAD, YELL, MOBL, and NECB. Study unit basins having a site that ranked in the top six for both sediment and fish criteria included NVBR, SOFL, NECB, SANT, and LINJ. Conversely, basins with a site in the top six for sediment criteria but not for fish included: GRSL, COOK, and OAHU. The NROK basin contained a sampling site that was among the highest for HgTot and MeHg in sediment (S. Fork Coeur d'Alene R), but fish were not collected there because impacts of mining activity has made them scarce. Despite relatively high HgTot and MeHg in sediments at the Weber R. (GRSL) site, mountain whitefish (*Prosopium williamsoni*) collected there were below the median Hg concentration (0.206) for all samples in this study. However, this species feeds primarily on aquatic insects (Carlander, 1977) and therefore may not bioaccumulate Hg as rapidly as piscivorous fishes. The Deshka R. (COOK) and Lake Wilson (OAHU) sites were notable because despite having elevated concentrations of Hg in the sediments, each was among the very lowest for the concentration of Hg in fish (albeit for sculpin and tilapia, respectively). The Deshka R. site had an unusually high concentration of MeHg in the sediment (5.1 ng/g) considering the HgTot was only 21 ng/g, whereas sediment from the Lake Wilson site was relatively high in HgTot but only moderate in MeHg.

Basins having a site ranking high for Hg in water as well as for fish included NVBR, SOFL, SANT, SACR, and LINJ. Essentially all of the highest ranking sites for $MeHg_{water}$ also ranked high for Hg_{fish}. However,

Table 6. Top six study units based on ranking of individual sites according to various Hg criteria. Values for fish are means of up to 7 individuals; sediment and water data are single samples from each site. Only the highest site from each study unit was considered in the overall rankings for each criteria.

Criteria	#1	#2	#3	#4	#5	#6
1. Hg_{fish}	NVBR	SOFL	SACR	SANT	LINJ	ACAD
(ug/g wet wt.)	3.36	2.15	1.80	1.80	0.82	0.79
Site	Lahontan Res.	WC3-A15	Sacramento Sl.	N.Fk. Edisto R.	Gr. Egg Hbr.	Bogue Falaya R.
2. Hg_{fish}/ age[a]	NVBR	SOFL	SACR	LINJ	YELL	MOBL
(ug/g wet / yr)	0.86	0.66	0.40	0.38	0.25	0.22
Site	Lahontan Res.	WC3-A15	Sacramento Sl.	Gr. Egg Hbr.	Big Horn R.	Satilpa Cr.
3. Hg_{fish}/ length	NVBR	SOFL	SANT	LINJ	SACR	NECB
(ug/g wet / m)	9.2	6.0	4.4	3.9	3.6	3.1
Site	Lahontan Res.	WC3-A15	N. Fk. Edisto R.	Gr. Egg Hbr.	Sacramento Sl.	Ipswich R.
4. $MeHg_{sed}$	LINJ	NECB	NROK	SOFL	SANT	COOK
(ng/g)	10.9	9.9	8.2	7.8	6.8	5.1
Site	Swan R.	Ipswich R.	Coeur d' Alene R.	WC3-A15	N. Fk. Edisto R.	Deshka R.
5. $HgTot_{sed}$	NROK	NVBR	NECB	GRSL	OAHU	SOFL
(ng/g)	4520	4130[b]	2480	1040	300	288
Site	Coeur d' Alene R.	Carson R.	Neponset R.	Weber R.	Lake Wilson	WC3-A15
6. $MeHg_{water}$	SANT	NVBR	SOFL	ACAD	NECB	LINJ
(ng/L)	1.5	1.3[b]	0.61	0.46	0.44	0.34
Site	N. Fk. Edisto R.	Carson R.	WC2-U3	Bayou Lacassine	Ipswich R.	Gr. Egg Hbr.
7. $HgTot_{water}$	NVBR	OAHU	GRSL	SACR	SANA	LINJ
(ng/L)	656[b]	24	22	18	15	12
Site	Carson R.	Nuuanu Res.	Weber R.	Bear R.	Santa Ana R.	Gr. Egg Hbr.

[a]excludes age-0 fish due to high relative error in assigning fractional age. No age data available for SANT unit.
[b]based on average of sites immediately above and below Lahontan Reservoir.

three basins had sites that ranked high for $HgTot_{water}$ but not for Hg_{fish}. These included OAHU (Nuunanu Res.), SANA (Santa Ana R.), and GRSL (Weber R.).

Correlation of Water and Sediment Parameters with Hg Bioaccumulation

A summary of the correlation of each measured variable (transformed as necessary to meet linearity requirements) with length-normalized mercury concentrations in fish is given in Table 7. Based on the initial analysis of our data, weight and length were about equally correlated with Hg_{fish} concentrations. But because of possible anomalies previously mentioned for rankings normalized by weight, we normalized by length for the correlation analysis. The correlation for age was significantly lower than for either length or weight, perhaps because the sample age was by design confined to a relatively narrow range and the resolution was limited to 0.5 units. Had our dataset consisted of samples of more widely varying ages, perhaps age would have been more useful. As indicated in Table 7, all of the measured parameters except $HgTot_{sed}$ and AVS_{sed} were significantly correlated with Hg bioaccumulation. The relative strength for correlation of bioaccumulation in fish with measures of Hg in water and

Table 7. Correlation of measured parameters with bioaccumulation of Hg in fish.

		correlation (r) with $\log_e(Hg_{fish}/length)$	
parameter	transformation	all species	largemouth bass
$MeHg_{water}$	$\log_e$	0.623 ***	0.712***
$MeHg_{sed}$	None	0.332***	0.596***
$HgTot_{water}$	$\log_e$	0.277**	0.453**
$HgTot_{sed}$	$\log_e$	ns	ns
DOC_{water}	$\log_e$	0.331***	ns
pH_{water}	None	-0.371***	-0.496**
$sulfate_{water}$	$\log_e$	-0.339***	-0.685***
LOI_{sed}	None	0.250**	0.420**
AVS_{sed}	cube root	ns	ns
% wetland	None	0.413***	0.523***

*** = $p < 0.001$; ** = $p < 0.01$; ns = no significant correlation ($p > 0.05$)

sediment was: $MeHg_{water} > MeHg_{sed} > HgTot_{water} >> HgTot_{sed}$ (no significant correlation). The correlation between $MeHg_{water}$ and $MeHg_{sed}$ was significant but relatively weak ($r = 0.306$, $\rho = .0009$). With the exception of DOC, all variables that were correlated with bioaccumulation exhibited stronger relationships for largemouth bass than for all species combined.

The positive relationship of Hg bioaccumulation with % wetlands, DOC, and sediment LOI (a proxy for organic matter) is widely documented (St. Louis and others, 1994, McMurtry and others, 1989, Mason and Lawrence, 1999). Higher MeHg production is generally associated with increases in organic matter in either the water column or in sediments because of enhanced microbial activity. But whereas low to moderate levels of DOC usually correlate with enhanced methylation rates of Hg (especially if the DOC is from terrestrial sources), high levels may act to reduce methylation and bioaccumulation (Winfrey and Rudd, 1990, Grieb and others, 1990, Driscoll and others, 1995). For reasons that are unclear, our Hg_{fish} data exhibited a significant positive correlation with DOC for all species combined, but not for largemouth bass. The bioacumulation of mercury in fish usually increases in waters of low pH (Wiener and others, 1990, Cope and others, 1990, Grieb and others, 1990, McMurtry and others, 1990) and the results from our nationwide sampling was no exception. In addition to other factors, lower water pH enhances Hg methylation and reduces loss of volatile Hg species from the water column by evasion (Winfrey and Rudd, 1990). Our data also exhibited a highly significant, negative correlation of Hg bioaccumulation with sulfate. The relationship was similar when either sites affected by mining, agriculture, or both were excluded from the analysis (ag and mining activity are often associated with elevated concentrations of sulfate in nearby drainges). At this time, the reason for the inverse relationship is unclear. Elevated levels of sulfate might be expected to correlate positively with Hg bioaccumulation rates because of the presence of sulfate in acidic precipitation (which has been associated with increased Hg bioaccumulation) and because of the role of sulfate-reducing bacteria that are directly involved in the methylation of mercury in water.

However, others have reported both positive and negative correlations of bioaccumulation with sulfate, depending on the watershed type (Grieb and others, 1990) and in general, most studies suggest that sulfate is not a determinant variable in the production of MeHg (Winfrey and Rudd, 1990).

Principal Components Analysis (PCA)

Factor loadings from the two most significant principal components of the PCA are presented graphically as vectors in Figures 2 (all species) and 3 (largemouth bass). Principal components 1 and 2 accounted for 44% and 54% of the variability for all species and for largemouth bass, respectively. From these plots, it is clear that a measure of fish size was the dominant variable influencing factor 2 (vectors in the positive y-direction) whereas the variables associated with the presence of organic matter (DOC, $MeHg_{water}$, LOI, % wetland, and $MeHg_{sed}$) each had a similar level of influence on factor 1 (vectors in positive x-direction). For largemouth bass only, there was a third statistically significant factor (not plotted in Figure 3) that accounted for an additional 14% of the variability. Therefore, a second PCA was conducted for largemouth bass only, but with the measures of length, weight, and age excluded, to allow for convenient examination of the first and third principal components. We expect that the exclusion of size factors may only be useful for data of a single fish species that are of relatively uniform size/age, as was the case for the largemouth bass in this study. With this plot (Figure 4), the importance of MeHg, especially in the water, on the fish Hg concentration is readily apparent because the Hg_{fish} and $MeHg_{water}$ appear as nearly identical vectors. In agreement with the simple regression analysis for largemouth bass (Table 7), DOC appears to correlate less strongly with the Hg_{fish} than many of the other parameters. It also appears that along with pH, sulfate, and AVS, DOC "counteracts" the influence of $TotHg_{water}$ on Hg_{fish} (vectors are opposite in the y-direction). However, the influence of DOC for the largemouth bass subset was probably greatly affected by the sites in the SOFL basin, which had the highest DOC values. And in fact, data for the SOFL sites fall near or outside the 95% confidence ellipse (interval) for PCA site score plots, indicating the SOFL sites were unique with respect to the variables analyzed in comparison to the other basins sampled.

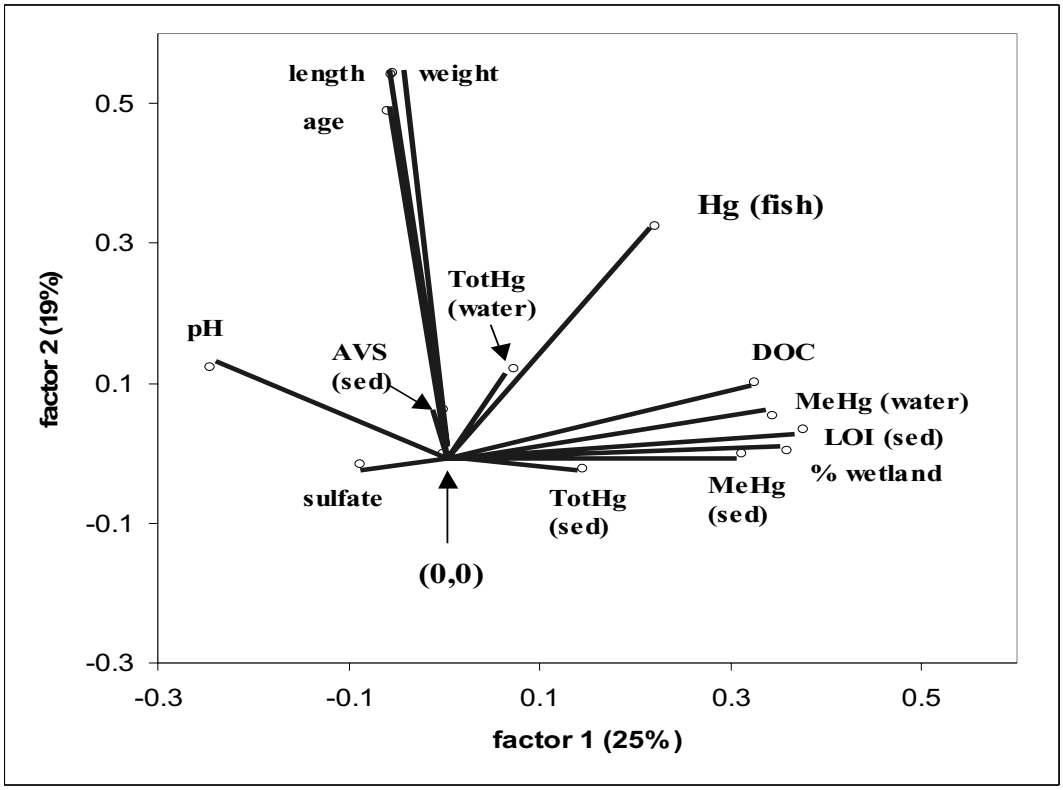

Figure 2. Principal Components Analysis - Factor Loadings for Hg Concentration in Fish (all species).

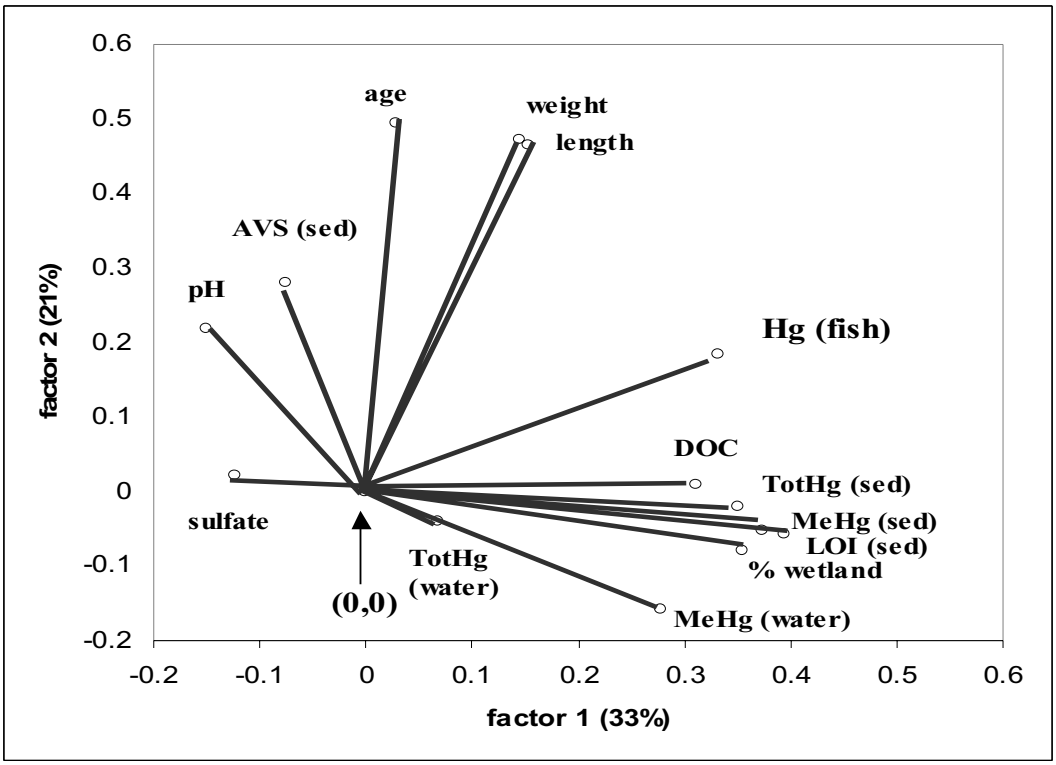

Figure 3. Principal Components Analysis - Factor Loadings for Hg Concentration in Largemouth Bass.

Model for the Bioaccumulation of Hg in Fish

An overall regression model for the bioaccumulation of Hg in fish was developed with the following candidate variables: fish weight (kg), fish total length (mm), fish age (yr), $MeHg_{water}$ (ng/L) , $HgTot_{water}$ (ng/L), $MeHg_{sed}$ (ng/g dry) $HgTot_{sed}$ (ng/g dry), DOC (mg/L), LOI (% dry wt), sediment AVS (µmol/g dry), water pH, and percent wetlands of each basin. Variables were transformed when necessary to meet the assumptions of linear regression. We also determined stable isotope ratios (C and N) in the fillets as a potential measure of trophic position (Kendall and others, 2000). However, the results were difficult to interpret (e.g., stable isotope ratios for the sediments would have been useful for adjusting for basin source differences) and consequently that data is not included in this report. For all fish combined, the following 4-variable model was deemed most useful based on the highest adjusted R^2 (44.6%) and lowest PRESS statistic (48.6):

$$\log_e(\text{Hg/length}) = -3.55 + 0.408 \log_e(\text{MeHg}_{water}) + 0.021\ (\%\text{wetland}) - 0.269\ \text{pH} - 0.121\ (\text{AVS})^{1/3}$$

For our samples the $MeHg_{water}$ was by far the most useful predictor of Hg bioaccumulation "rate" (assuming fish length increases approximately linearly with time); it accounted for 30 of the 45% of the variability described by the model. Essentially, all of the other non-mercury parameters were measured because they were expected to influence the production MeHg. As it was correlated with several other of the explanatory variables, $MeHg_{sed}$ added no new explanatory power to the above regression equation. Many of our sampling sites were flowing (non-stratifying) waters in which the sediment-water boundary might be expected to be the most important Hg-methylation zone for the water body (Krabbenhoft and Gilmour, 1998). Also, because sediments act as a sink for most contaminants, the MeHg load in the sediment might be expected to provide a better indicator of the long term exposure for the age-3 fish that were targeted in our study. Poor correlation of Hg in fish with HgTot in sediments has been widely documented (Wiener and others, 1984; Harrison and Klaverkamp, 1990; Sorensen and others, 1990). However, relatively few studies have been conducted that directly compared MeHg in sediments with Hg in

fish. Our results suggest that on a national basis, MeHg in water is a much better predictor of concentrations in fish than is MeHg in the sediment, and that other sediment-related variables (e.g., AVS, %wetlands) probably replicate any ability of MeHg in sediment to predict Hg in fish. However, limnologic sampling conditions may greatly affect this relationship.

Presumably, fish tend to bioaccumulate a large percentage of their Hg burden during warmer months when rates of feeding and microbial methylation of Hg are highest. In waters that stratify, mixing of anoxic hypolimnetic water during fall "turnover" can result in rapid increases of mercury accumulation in zooplankton and young-of-year fishes (Slotten and others, 1995). Also, hypolimnetic tailrace water below stratified reservoirs may become enriched in MeHg during early fall (Canavan and others, 2000). We collected samples when water levels were generally at low flow - a condition that tends to favor an increase in concentrations of MeHg in the water. Because of these seasonal factors, our samples might have exhibited improved correlations with methylmercury concentrations in water relative to that in the sediment. Calculations presented by Mason and Lawrence (1999) suggest that both the sediment and water column can be significant contributors of MeHg to fish via food chain pathways. However, the greater importance of MeHg in the water relative to the sediment for our model indicates that MeHg production in surficial sediments may be of minimal consequence in some water bodies, perhaps because it is not effectively transported to the water column. Fluxes of MeHg from sediments to the water column are reported to be considerable only under anoxic conditions (Gill and others, 1999) because apparently, MeHg generated at the redox boundary is effectively trapped by sorption to solids in oxic sediment surface layers (Gagnon, and others, 1996). Therefore, for flowing, non-stratifying waters like most that were sampled in our study, effective transfer of MeHg to the water column from sediments might only occur for sediments that are prone to resuspension from wind or other factors (Bloom, and others, 1999).

The positive coefficient for % wetlands and the negative coefficient for pH with our model is in agreement with most other studies. Wetlands can be significant sources of MeHg and their presence can promote

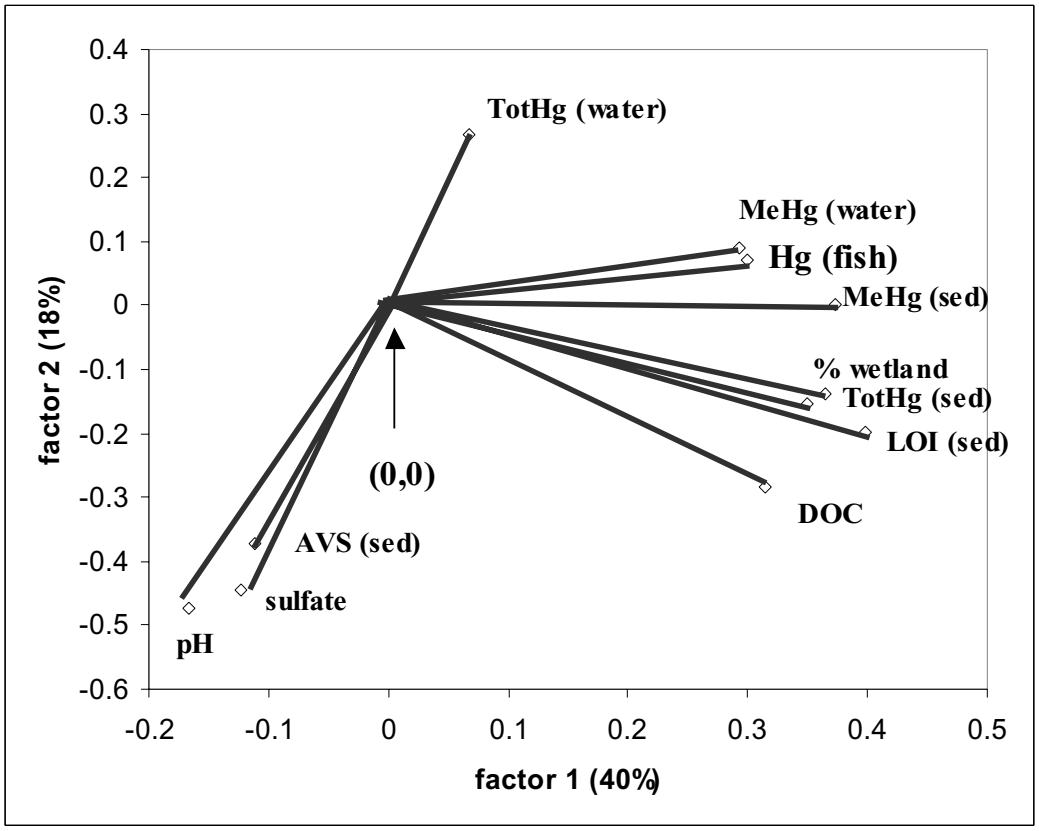

Figure 4. Principal Components Analysis - Factor Loadings for Hg Concentration in Largemouth Bass, excluding weight, length, and age.

MeHg production because, in addition to other factors, they provide DOC (and enhance microbial activity) to the watershed (Rudd, 1995). Bioaccumulation of Hg in fish usually increases in waters of low pH, primarily because the water column retains more Hg under acidic conditions (Winfrey and Rudd, 1990). A minor, but statistically significant factor with our bioaccumulation model, was the sediment acid-volatile sulfide (AVS), which exhibited a negative coefficient. Such a result might seem contradictory, because wetland sediments usually contain considerable AVS, yet Hg bioaccumulation rates are positively correlated with the presence of wetlands. It may be that highly anaeorbic wetlands, which typically contain very high AVS in the sediments, do not contribute to increased bioaccumulation of Hg in fish in the same way that more aerobic wetlands do. Also, it is possible that some of the MeHg in the water originated considerably upstream from our sampling sites. As discussed in Part One of this report, the methylation efficiency of Hg was found to decrease in sediments containing very high AVS, presumably because of the strong affinity of sulfides for Hg, which make it less available. Perhaps the small, but negative coefficient of AVS (as the cube root) for our model accounts for decreased methylation efficiency, or a net removal of MeHg from water, at very high AVS concentrations.

Regression plots of length-normalized fish concentrations for all species and for largemouth bass versus MeHg concentration in water are indicated in Figures 5 and 6, respectively. Based on these data, a water MeHg concentration of 0.12 ng/L was associated with a fish fillet Hg concentration of 0.30 µg/g for an age-3 fish when all species are considered. For age-3 largemouth bass a water MeHg concentration of 0.058 ng/L was, on average, associated with the 0.30 µg/g fillet concentration. In addition to fish species and age, other factors that might affect this relationship include seasonal and hydrologic conditions during water sampling, and prey availability and dietary pathways for the sampled fish. Although the diet is generally the dominant vector for bioaccumulation of MeHg in predator fish, the strong correlation with MeHg in water indicates that the water column was the primary source (at the base of the food chain) for the majority of our sites. It is important to note that our water samples were not filtered for the determination of Hg, although most were collected during low flow and consequently were relatively low in suspended solids. Additional studies are needed to define the limitations in estimating fish concentrations from MeHg concentrations in water, and to determine the sampling conditions that maximize the predictive power of water analyses.

Relation of Hg in Fish, Sediment, and Water to Land Use Patterns

All of the study basins were heterogeneous with respect to land cover and use. For the purposes of this

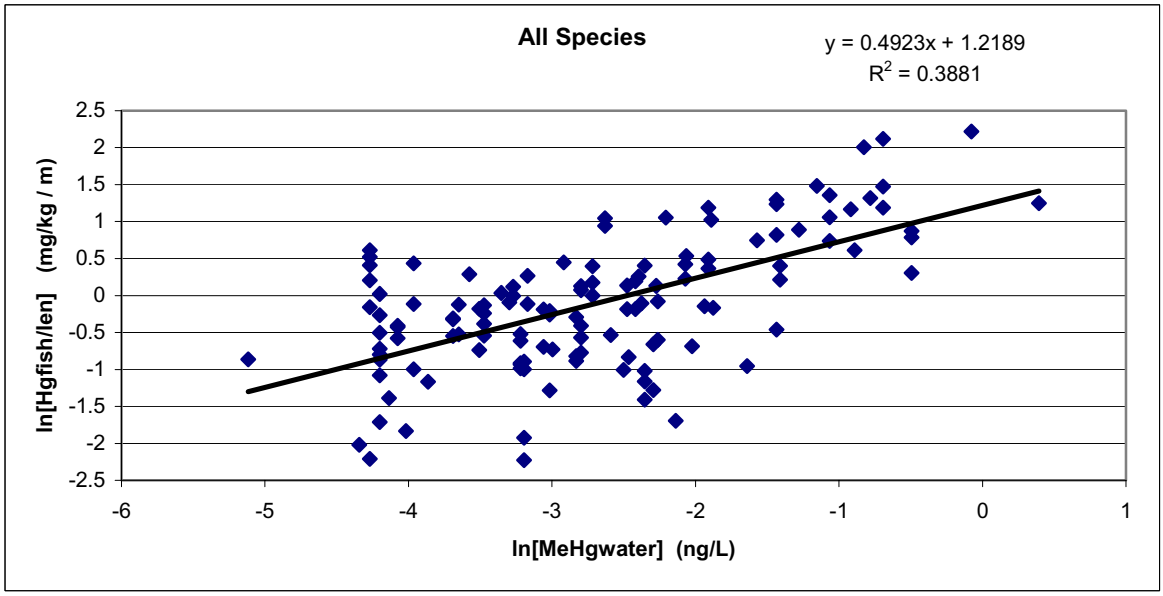

Figure 5. Length-normalized concentration of Hg in fish (all species) as a function of MeHg concentration in water. Excludes NVBR basin.

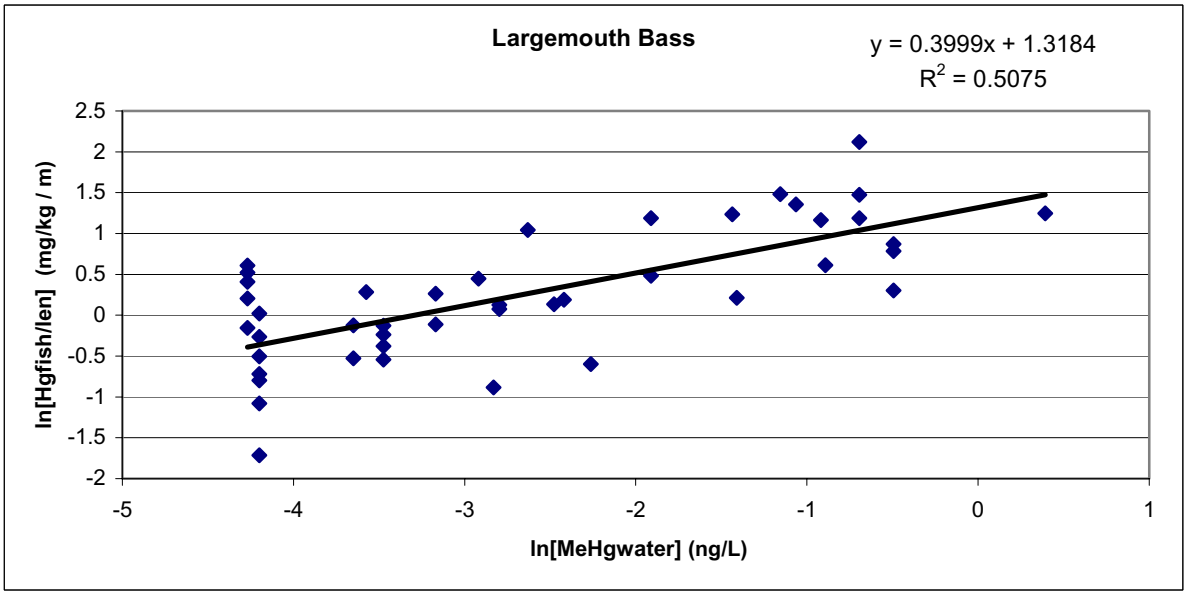

Figure 6. Length-normalized concentration of Hg in largemouth bass as a function of MeHg concentration in water.

analysis, the sub-basins above each of the sampling sites were categorized into one of the five following broad classes: agriculture dominant (Ag.); mixed agriculture and forest (A/F); background or reference site (Bkg.); current or abandonded mining activities near site (Mine); and urban or industrial activity near sampling site (Urb.). The land-use categories assigned to individual sites are listed in Part One of this report. The geometric means for each of the five Hg parameters and five land use catogories are depicted in Figure 7. Rankings for concentrations of Hg in fish were as follows: A/F >> Mine >Ag > Urb.$\simeq$ Bkd. Forested watersheds have been previously identified with high Hg methylation rates (Hurley and others, 1995). Also, A/F basins from our study often contained higher percentages of wetlands, which overall, contribute to higher methylation efficiency. Sub-basins characterized as mining-impacted ranked highest for HgTot in both sediment and water, and second in MeHg in water. As previously discussed, several of the mining basins sampled had past gold-mining operations where Hg amalgamation practices were used. Thus, although the methylation rates are not necessarily high in many of these basins, the overwhelming loads of HgTot in the watershed can yield relatively high concentrations of MeHg in the water and Hg in fish. Surprisingly, sub-basins categorized as urban had a comparatively low mean for Hg_{fish} but ranked fairly high for the HgTot and MeHg in sediment. At first glance, one might suspect that this discrepancy was due to a species bias, i.e., fewer top predators present in urban areas. But in fact, most of the fish sampled from urban sub-basins were either largemouth- or smallmouth bass. Interestingly, the correlation between $MeHg_{water}$ and $MeHg_{sed}$ was highly significant for urban sites ($r = 0.606$, $\rho = .0002$) but it was not significant for any other land-use category. The factors affecting the bioaccumulation of Hg in fish from urban settings deserves additional research.

The bioaccumulation factor (BAF) for MeHg in water to Hg in fish muscle (assumed to be all MeHg) is plotted in Figure 8 for each land-use category (and the SOFL basin) for either largemouth bass or all species combined. The SOFL unit was considered separately because of its unique character and because most other sites were flowing streams. There are no clear differences for BAFs among land-uses for all species combined. However, a trend opposite of that observed for fish concentrations (Figure 7) is apparent for BAFs for largemouth bass alone, i.e., the land-use categories with the highest fish and water concentrations (A/F and Mine) have the lowest BAFs, and vice-versa. Perhaps most striking is the comparatively low BAF for SOFL, where MeHg concentrations in water are generally high. A similar trend was observed for BAFs normalized by fish length, which indicates that overall, there was not a remarkable bias due to size differences of fish collected among land-use categories. The inverse relationship of BAF with the concentration of MeHg in water suggests that lower concentrations of MeHg in water are more efficiently biotransferred than are higher

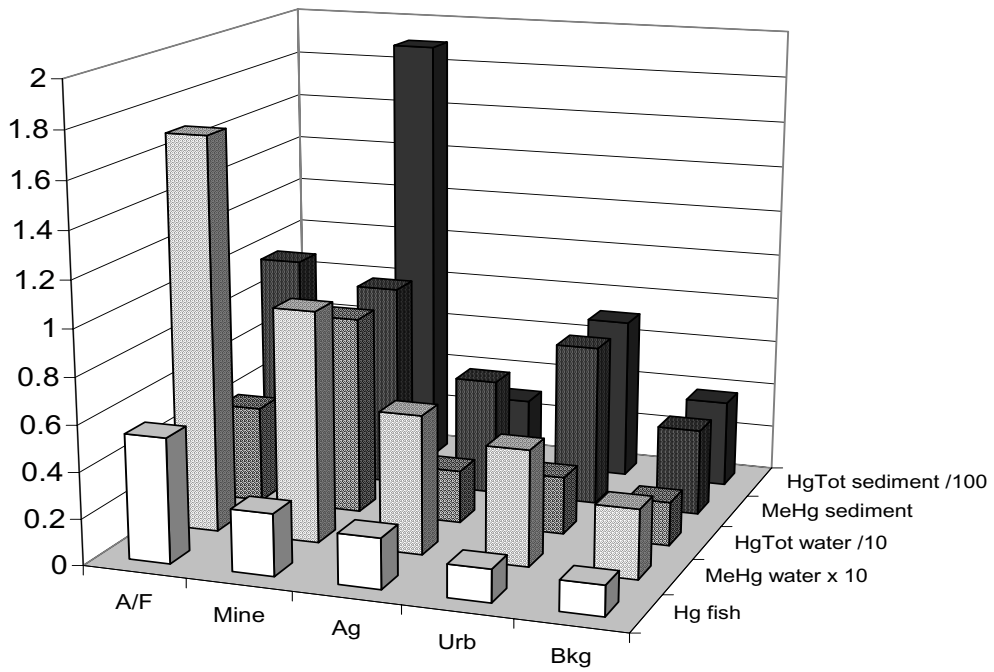

Figure 7. Geometric mean of Hg or MeHg in fish, water, and sediment for each land use category (see text for land use definitions). Number of observations = 13, 42, 23, 15, and 34, for A/F, Mine, Ag., Bkg, and Urb, respectively. Excludes South Florida basin.

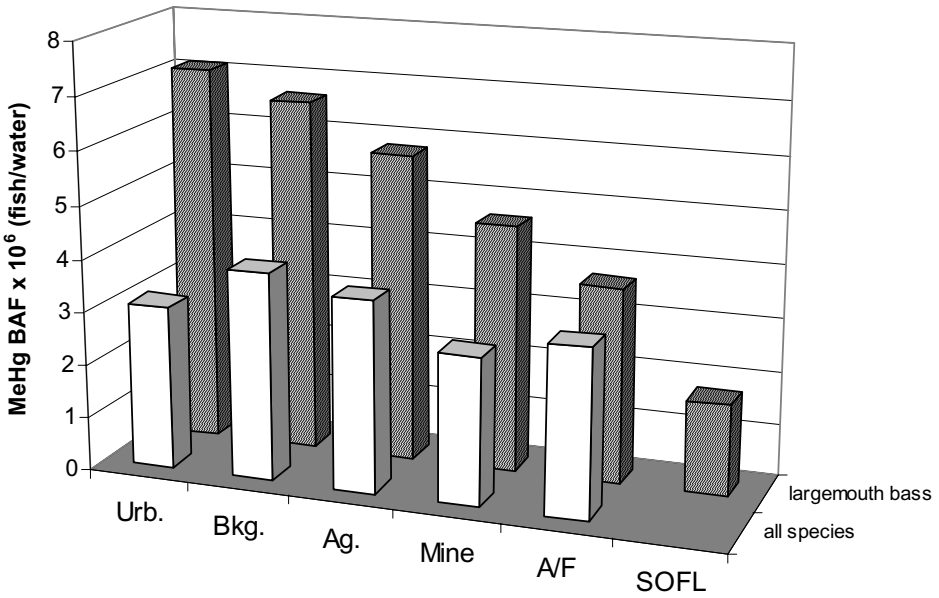

Figure 8. Geometric mean of MeHg bioconcentration factor (MeHgfish / MeHgwater) for all species and for largemouth bass, for each land use category.

concentrations. This might result from lower assimilation (or greater elimination) by fish at higher MeHg concentrations. Or perhaps water quality factors that increase the MeHg production rate (like high DOC) also serve to reduce the relative bioavailablity and ultimately the BAF because of increased complexation of MeHg. These results warrant further investigation. But regardless of the water quality or type, high concentrations of MeHg in water will generally produce high concentrations in fish.

SUMMARY

Mercury contamination of waterways is a widespread problem and sources and accumulation rates in fish vary among basins. One or more fish fillet samples from nine of 20 basins examined in this pilot study had Hg concentrations above the historical advisory level of 0.50 µg/g wet weight and 15 of the basins had at least one sample above EPA's 2001 criteria of 0.30 µg/g wet weight. Based on rankings of selected water, sediment, and fish criteria, sampling sites from the following five basins exhibited the greatest Hg contamination: Nevada Basin and Range, South Florida, Sacramento Basin, Santee Basin and Coastal Drainages, and the Long Island and New Jersey Coast Drainages. The concentrations of Hg in fish were correlated strongly with MeHg in water, but only moderately with MeHg in sediment or HgTot in water. There was no correlation between Hg in fish and total Hg in sediment. The concentration of MeHg in water was by far the most useful variable for predicting the Hg bioaccumulation in fish. However, percent wetlands (+), pH of water (-) , and sediment AVS (-) also contributed significantly to the model. Based on our data, a MeHg water concentration of 0.12 ng/L was on average, associated with a fish fillet Hg concentration of 0.30 µg/g wet weight for an age-3 fish when all species were considered. For age-3 largemouth bass a MeHg water concentration of 0.058 ng/L was associated with the 0.30 µg/g fillet concentration. Additional studies are needed to define the limitations in estimating fish concentrations from MeHg concentrations in water, and to determine the sampling conditions that maximize the predictive power of water analyses.

Based on ranking criteria, sub-basins categorized as mixed agriculture/forest and mining-impacted had the most consistent contamination of Hg in all three sample matrices (water, sediment, and fish). The greatest discrepancy for rankings of Hg in fish and in sediments was for urban watersheds, where sediments often ranked high but fish usually ranked low. The bioaccumulation factor ($MeHg_{fish}$ / $MeHg_{water}$) was lowest for land-use categories having the highest concentrations of MeHg in water, which indicates that low concentrations of MeHg in water are more efficiently biotransferred than higher concentrations. Nevertheless, high concentrations of MeHg in water will generally produce high concentrations in fish. A sampling and analysis strategy based on this pilot study is planned for all USGS NAWQA study units over the next decade. We expect those results to provide a comprehensive national characterization of mercury contamination and bioaccumulation in our aquatic ecosystems.

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The authors are indebted to all NAWQA study-basin teams who participated in the study and demonstrated great diligence, commitment, and enthusiam in the collection of the samples. We thank Jude Smith and coworkers with the U.S. Fish and Wildlife Service (Albuquerque, NM) for conducting age determinations and Dr. Ted Lange (Florida Fish and Game) for providing Hg measures in fish from South Florida. We acknowledge our colleagues Jesse Arms, Mike Walther, and Mark Olson for technical assistance, Robin Lipkin and Cory Stephens for assistance with manuscript preparation, and Chris Schmitt and Denny Buckler for helpful review comments.

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Appendix. Concentrations of mercury in fish collected from USGS NAWQA Hg-Pilot Study basins, June through October, 1998. No. fish indicates number of fish in each composite sample. Sample species with [a] superscript were analyzed as whole-body less head; those with [b] superscript were analyzed as whole body. All others are fillet tissue. See text for assignment of age. CERC = Columbia Environmental Research Center; -- = no data.

CERC #	S.U.	*Site*	*Species*	No. Fish	Mean Weight (g)	Mean Total Length (mm)	Mean Age (yr)	µg Hg / g dry	µg Hg / g wet
18221-1	ALMN	Clarion R. @ Ridgeway, PA	Smallmouth Bass	2	745	363	4.5	1.495	0.327
18222-1	ALMN	Dunkard Cr. @ Shannopin, PA	Smallmouth Bass	4	130	223	3.0	1.102	0.204
18223-1	ALMN	Youghiogheny R. @ Sutersville, PA	Smallmouth Bass	1	292	290	2.5	0.439	0.072
18224-1	ALMN	Allegheny R. @ New Kensington, PA	Smallmouth Bass	3	108	206	1.5	0.449	0.087
18225-1	ALMN	Tenmile Cr. nr. Amity, PA	Smallmouth Bass	2	430	312	4.0	1.517	0.288
18226-1	ALMN	Tenmile Cr. nr. Amity, PA	Largemouth Bass	1	592	329	3.5	0.941	0.181
18227-1	LINJ	Muddy Run @ Centerton, NJ	Chain Pickerel	2	307	345	4.0	0.781	0.159
18227-2	LINJ	Muddy Run @ Centerton, NJ	Chain Pickerel	4	10	120	0.5	0.475	0.100
18228-1	LINJ	Muddy Run @ Centerton, NJ	Largemouth Bass	2	362	295	3.0	1.760	0.334
18228-2	LINJ	Muddy Run @ Centerton, NJ	Largemouth Bass	2	110	192	3.0	1.088	0.207
18228-3	LINJ	Muddy Run @ Centerton, NJ	Largemouth Bass[a]	1	21	119	0.5	0.567	0.132
18229-1	LINJ	Passaic R. @ Millington, NJ	Redfin Pickerel	2	81	229	2.5	1.811	0.340
18230-0	LINJ	Passaic R. @ Millington, NJ	Redfin Pickerel[b]	4	12	112	0.5	0.624	0.125
18231-1	LINJ	Passaic R. @ Millington, NJ	Largemouth Bass	2	179	212	2.5	1.286	0.262
18232-1	LINJ	Passaic R. @ Millington, NJ	Smallmouth Bass	1	25	118	0.5	1.522	0.276
18233-1	LINJ	Swan R. @ E. Patchague, NY	Redfin Pickerel	7	36	166	2.0	0.460	0.073
18234-1	LINJ	Great Egg Harbor @ Sicklerville, NJ	Chain Pickerel	2	172	284	3.0	4.511	0.816
18234-2	LINJ	Great Egg Harbor @ Sicklerville, NJ	Chain Pickerel	5	84	237	1.5	2.911	0.495
18235-1	LINJ	Great Egg Harbor @ Sicklerville, NJ	Largemouth Bass	1	49	148	1.5	3.212	0.575
18236-1	UIRB	Des Plains R. @ Russell, IL	Bowfin	1	617	337	3.5	1.014	0.174
18237-1	UIRB	Nippersink Cr. Abv. Wonder Lake, IL	Smallmouth Bass	2	84	180	1.5	0.330	0.066
18238-1	UIRB	Salt Cr. @ Western Springs, IL	Smallmouth Bass	3	70	175	2.0	0.489	0.088
18239-1	UIRB	Pitner Ditch nr. LaCrosse, IN	Largemouth Bass	3	154	219	3.0	0.707	0.129
18240-1	UIRB	Mukwanago R. @ Mukwanago, WI	Rock Bass	4	62	143	2.0	0.575	0.107
18241-1	UIRB	Mukwanago R. @ Mukwanago, WI	Largemouth Bass	1	114	182	2.5	0.412	0.075
18242-1	LTEN	Sequatchie R. nr. Whitwell, TN	Largemouth Bass	1	228	250	3.5	2.275	0.421
18242-2	LTEN	Sequatchie R. nr. Whitwell, TN	Largemouth Bass	3	114	214	2.5	1.449	0.262
18243-1	LTEN	Buffalo R. nr. Flatwoods, TN	Largemouth Bass	1	351	282	3.5	1.256	0.241
18244-1	LTEN	Buffalo R. nr. Flatwoods, TN	Largemouth Bass	1	132	212	2.5	2.143	0.390
18245-1	LTEN	Buffalo R. nr. Flatwoods, TN	Largemouth Bass	1	35	144	1.5	1.398	0.217
18246-1	LTEN	Indian Cr. nr. Madison, AL	Largemouth Bass	2	288	280	2.5	0.871	0.163
18247-1	LTEN	Indian Cr. nr. Madison, AL	Largemouth Bass	1	470	324	3.5	1.325	0.256
18248-1	LTEN	Indian Cr. nr. Madison, AL	Largemouth Bass	2	38	148	1.5	0.720	0.130
18249-0	COOK	S. Fk. Campbell Cr. nr. Anchorage, AK	Slimy Sculpin[b]	5	23	--	--	0.292	0.069
18250-1	COOK	S. Fk. Campbell Cr. nr. Anchorage, AK	Dolly Varden	5	141	233	--	0.429	0.086
18251-0	COOK	Chester Cr @ Artic Blvd, Anchorage, AK	Slimy Sculpin[b]	5	20	--	--	0.152	0.036
18252-1	COOK	Costello Cr. nr. Colorado & Denali, AK	Dolly Varden[b]	1	35	158	--	0.164	0.036
18252-2	COOK	Costello Cr. nr. Colorado & Denali, AK	Dolly Varden[b]	4	11	114	--	0.101	0.021
18253-0	COOK	Deshka R. NR. Willow, AK	Slimy Sculpin[b]	10	35	--	--	0.246	0.060
18254-0	NECB	Stillwater R. NR. Sterling, MA	Mixed Sunfish[b]	5	12	84	--	0.796	0.173
18255-0	NECB	Neponset R. @ Norwood, MA	Mixed Sunfish[b]	4	50	127	3.5	0.457	0.116

CERC #	S.U.	*Site*	*Species*	No. Fish	Mean Weight (g)	Mean Total Length (mm)	Mean age (yr)	µg Hg / g dry	µg Hg / g wet
18256-0	NECB	Aberjona R. @ Winchester, MA	Mixed Sunfish[b]	5	23	94	--	0.236	0.055
18257-0	NECB	Saugus R. @ Ironworks@ Saugus, MA	Mixed Sunfish[b]	4	19	100	--	0.486	0.107
18258-0	NECB	Ipswich R. @ S. Middleton, MA	Mixed Sunfish[b]	5	33	114	2.5	1.423	0.349
18259-0	DELR	Schuylkill R. @ Philadelphia, PA	Smallmouth Bass	5	317	308	4.1	1.616	0.323
18260-0	DELR	Delaware R. @ Trenton, NJ	Smallmouth Bass	5	749	371	4.3	1.439	0.288
18261-0	DELR	Delaware R. @ Port Jervis, NY	Smallmouth Bass	4	294	276	3.5	1.654	0.331
18262-0	DELR	Lehigh R. @ Glendon, PA	Smallmouth Bass	5	338	297	3.5	0.648	0.130
18263-1	DELR	Racoon Cr. @ Swedesboro, NJ	Chain Pickerel	1	330	372	3.5	1.521	0.287
18263-2	DELR	Racoon Cr. @ Swedesboro, NJ	Chain Pickerel	1	105	254	0.5	0.687	0.126
18264-1	DELR	Little Lehigh Cr. @ E. Texas, PA	Brown Trout	3	141	233	--	0.148	0.031
18265-1	DELR	Hay Cr. nr. Birdsboro, PA	Smallmouth Bass	1	557	335	3.5	1.587	0.333
18265-2	DELR	Hay Cr. nr. Birdsboro, PA	Smallmouth Bass	2	221	258	2.5	1.457	0.290
18266-1	DELR	Manatawny Cr. nr. Pottstown, PA	Smallmouth Bass	3	206	253	3.5	0.883	0.169
18267-1	DELR	Tulpehocken Cr. nr. Bernville, PA	Smallmouth Bass	1	354	283	3.5	1.113	0.236
18268-1	DELR	L. Neshaminy Cr. nr. Neshaminy, PA	Smallmouth Bass	2	180	236	3.0	1.398	0.268
18269-1	SACR	Colusa Basin Drain , CA	Largemouth Bass	4	644	343	4.8	1.945	0.393
18270-1	SACR	Sacramento Slough nr. Knights Landing	Largemouth Bass	1	1471	550	4.5	10.734	1.803
18270-2	SACR	Sacramento Slough nr. Knights Landing	Largemouth Bass	1	1156	390	5.5	3.214	0.633
18270-3	SACR	Sacramento Slough nr. Knights Landing	Smallmouth Bass	1	675	324	4.5	2.220	0.466
18271-1	SACR	Cottonwood Cr. nr. Cottonwood, CA	Smallmouth Bass	1	401	315	3.5	1.264	0.264
18271-2	SACR	Cottonwood Cr. nr. Cottonwood, CA	Smallmouth Bass	5	115	193	1.5	0.438	0.092
18272-1	SACR	Bear River @ Hwy 70 nr. Rio Oso, CA	Largemouth Bass	1	518	325	3.5	5.983	1.119
18272-2	SACR	Bear River @ Hwy 70 nr. Rio Oso, CA	Smallmouth Bass	1	467	288	3.5	5.442	1.050
18273-1	SACR	Bear River @ Hwy 70 nr. Rio Oso, CA	Smallmouth Bass	1	150	220	2.5	2.695	0.499
18274-1	SACR	Bear River @ Hwy 70 nr. Rio Oso, CA	Mountain Whitefish	1	549	390	2.0	1.376	0.246
18275-1	SACR	Putah Cr. nr. Davis, CA	Largemouth Bass	5	62	158	1.5	1.320	0.247
18276-1	SANA	South Fork Santa Ana River, CA	Brown Trout	6	90	194	1.5	0.743	0.127
18277-1	SANA	Santa Ana R. @ Hammer Rd.	Largemouth Bass	2	54	158	2.0	0.743	0.140
18278-1	SANA	Santa Ana R. blw. Prado Dam	Mixed Sunfish	10	53	136	2.0	0.685	0.118
18279-1	SANA	Mill Cr. @ Chino - Corona Rd.	Common Carp	5	393	311	3.0	0.694	0.114
18280-1	NVBR	Lahontan Res., NV	White Bass	1	435	310	3.5	9.381	1.895
18280-2	NVBR	Lahontan Res., NV	White Bass	1	435	325	2.5	29.06	5.696
18280-3	NVBR	Lahontan Res., NV	White Bass	1	490	334	3.5	32.61	5.837
18280-4	NVBR	Lahontan Res., NV	White Bass	1	615	350	3.5	7.387	1.418
18280-5	NVBR	Lahontan Res., NV	White Bass	1	437	335	3.5	16.42	3.219
18280-6	NVBR	Lahontan Res., NV	White Bass	1	883	380	4.5	13.34	2.881
18280-7	NVBR	Lahontan Res., NV	White Bass	1	879	392	4.5	16.01	3.634
18280-8	NVBR	Lahontan Res., NV	White Bass	1	1171	472	5.5	24.90	5.154
18281-1	YELL	Bighorn R. @ Kane, WY	Walleye	5	452	373	2.5	3.289	0.635
18282-1	YELL	Tongue R. @ State Line nr. Decker	Smallmouth Bass	1	299	270	3.5	0.743	0.153
18283-1	YELL	Yellowstone R. @ Sidney, MT	Sauger	2	176	294	3.0	1.287	0.250
18284-1	YELL	Bighorn Lake @ Hwy 14A Causeway	Walleye	5	896	453	4.0	3.382	0.676
18285-1	YELL	Shoshone R. @ Mouth nr. Kane, WY	Walleye	1	1444	535	5.5	3.249	0.669
18285-2	YELL	Shoshone R. @ Mouth nr. Kane, WY	Walleye	5	817	437	4.5	3.452	0.666
18286-1	ACAD	Tangipahoa R. @ Robert, LA	Largemouth Bass	8	562	335	4.2	3.789	0.743
18287-1	ACAD	Turtle Bayou nr. Bayou Penchant, LA	Largemouth Bass	8	387	292	2.5	0.999	0.206
18288-1	ACAD	Bayou Teche nr. St. Martinville, LA	Largemouth Bass	8	334	298	3.1	1.320	0.260

CERC #	S.U.	*Site*	*Species*	No. Fish	Mean Weight (g)	Mean Total Length (mm)	Mean age (yr)	µg Hg / g dry	wet
18289-1	ACAD	Bogue Falaya R. @ Covington, LA	Largemouth Bass	8	392	306	3.5	4.098	0.791
18290-1	ACAD	Bayou Segnette nr. Barataria, LA	Largemouth Bass	8	487	326	2.8	1.027	0.209
18291-1	GRSL	Cub R. nr. Richmond, UT	Largemouth Bass	2	136	204	4.5	1.369	0.271
18292-1	GRSL	Weber R. nr. Coalville, UT	Mountain Whitefish	1	833	391	4.0	0.628	0.141
18292-2	GRSL	Weber R. nr. Coalville, UT	Mountain Whitefish	2	545	374	4.0	0.535	0.117
18292-3	GRSL	Weber R. nr. Coalville, UT	Mountain Whitefish	2	317	300	3.0	0.364	0.073
18293-1	TRIN	Trinity R. below Dallas, TX	Bluegill Sunfish[b]	2	19	95	1.5	0.412	0.064
18294-1	TRIN	Trinity R. below Dallas, TX	Bluegill Sunfish[b]	1	38	120	2.5	0.494	0.079
18295-1	TRIN	Trinity R. below Dallas, TX	Mixed Sunfish	2	77	162	2.5	0.431	0.064
18296-1	TRIN	Trinity R. below Dallas, TX	Flathead Catfish	1	1087	470	--	0.910	0.159
18297-1	TRIN	White Rock Lake Dallas TX	Largemouth Bass	1	91	186	0.5	0.270	0.045
18298-1	TRIN	White Rock Lake Dallas TX	Largemouth Bass	2	341	282	3.5	0.252	0.051
18299-1	TRIN	White Rock Lake Dallas TX	Largemouth Bass	1	663	345	3.5	0.586	0.117
18300-1	TRIN	Lake Livingston, TX	Largemouth Bass	1	1305	412	4.5	1.179	0.249
18300-2	TRIN	Lake Livingston, TX	Largemouth Bass	1	849	366	4.5	1.876	0.373
18300-3	TRIN	Lake Livingston, TX	Largemouth Bass	2	526	324	5.0	1.226	0.249
18300-4	TRIN	Lake Livingston, TX	Largemouth Bass	2	306	278	4.0	0.645	0.125
18301-1	TRIN	Clear Cr. nr. Sanger, TX	Spotted Bass	1	40	150	0.5	1.411	0.258
18302-1	TRIN	Clear Cr. nr. Sanger, TX	Spotted Bass	1	87	180	0.5	0.861	0.157
18303-1	TRIN	Clear Cr. nr. Sanger, TX	Spotted Bass	1	142	228	3.5	1.450	0.299
18304-1	TRIN	Clear Cr. nr. Sanger, TX	Channel Catfish	1	372	380	--	2.245	0.341
18304-2	TRIN	Clear Cr. nr. Sanger, TX	Channel Catfish	2	114	248	--	0.841	0.133
18305-1	TRIN	Trintity R. nr. Crockett, TX	Largemouth Bass	1	311	260	3.5	0.631	0.127
18306-1	TRIN	Trintity R. nr. Crockett, TX	Flathead Catfish	3	191	262	--	0.716	0.110
18307-1	NROK	Clark Fork at Turah, MT	Mountain Whitefish	5	551	356	4.5	0.641	0.155
18308-1	NROK	Clark Fork at St. Regis, MT	Mountain Whitefish	5	264	294	3.5	0.436	0.092
18309-1	MIAM	Little Miami R. @ Milford, OH	Smallmouth Bass	3	55	153	1.5	0.244	0.042
18310-1	MIAM	Little Miami R. @ Milford, OH	Smallmouth Bass	1	698	340	5.5	1.393	0.276
18311-1	MIAM	Whitewater R. nr. Nultown, IN	Smallmouth Bass	1	80	187	2.5	0.792	0.135
18312-1	MIAM	Whitewater R. nr. Nultown , IN	Smallmouth Bass	1	129	220	2.5	0.701	0.128
18313-1	MIAM	Whitewater R. nr. Nultown, IN	Smallmouth Bass	2	257	270	4.0	1.038	0.198
18314-1	MIAM	E. Fork, L. Miami R. Williamsburg, OH	Smallmouth Bass	1	107	205	2.5	1.108	0.205
18315-1	MIAM	E. Fork, L. Miami R. Williamsburg, OH	Smallmouth Bass	2	386	302	3.5	1.859	0.359
18316-1	MIAM	E. Fork, L. Miami R. Williamsburg, OH	Smallmouth Bass	1	608	350	4.5	2.524	0.520
18317-1	MIAM	Stillwater R. @ Union, OH	Smallmouth Bass	3	130	217	2.5	1.074	0.180
18318-1	MIAM	Stillwater R. @ Union, OH	Smallmouth Bass	1	284	275	3.5	0.706	0.138
18319-1	MIAM	Great Miami R. nr. Tipp City, OH	Smallmouth Bass	4	178	242	3.5	0.649	0.117
00012-1	MIAM	Mad R. @ Hwy 41	Smallmouth Bass	1	518	325	5.5	1.355	0.271
00013-1	MIAM	Great Miami R. @ Hamilton	Smallmouth Bass	2	345	292	3.5	0.565	0.113
18523-1	OAHU	Kawainui Canal, HI	Tilapia	5	127	188	4.0	0.157	0.030
18524-1	OAHU	Ala Wai Canal, HI	Tilapia	5	124	179	2.0	0.115	0.020
18525-1	OAHU	Hoomaluhia Reservoir, HI	Tilapia	5	49	133	3.0	0.116	0.018
18526-1	OAHU	Nuuanu Reservoir, HI	Tilapia	5	249	226	4.0	0.317	0.063
18527-1	OAHU	Waikele Stream, HI	Smallmouth Bass[a]	1	32	135	0.5	0.189	0.050
18528-1	OAHU	S. Fork Lake Wilson, HI	Tilapia	5	302	250	4.5	0.237	0.046
18529-1	MOBL	Shades Cr., AL	Longear Sunfish	8	29	118	3.0	0.718	0.122
18530-1	MOBL	Alabama R. @ Claiborne, AL	Black Crappie	2	210	245	3.0	0.810	0.146
18530-2	MOBL	Alabama R. @ Claiborne, AL	Black Crappie	1	94	180	2.5	0.411	0.067

CERC #	S.U.	*Site*	*Species*	No. Fish	Mean Weight (g)	Mean Total Length (mm)	Mean age (yr)	µg Hg / g dry	wet
18531-1	MOBL	Alabama R. @ Claiborne, AL	Spotted Bass	2	754	365	3.5	0.837	0.147
18531-2	MOBL	Alabama R. @ Claiborne, AL	Spotted Bass	2	262	275	3.5	0.789	0.149
18531-3	MOBL	Alabama R. @ Claiborne, AL	Spotted Bass	2	158	240	2.5	0.486	0.094
18532-1	MOBL	Satilpa Cr near Coffeeville, AL	Largemouth Bass	1	92	194	2.5	3.078	0.551
18532-2	MOBL	Satilpa Cr near Coffeeville, AL	Spotted Bass	1	140	235	3.5	3.213	0.601
18533-1	MOBL	Town Cr at Tupelo, MS	Channel Catfish[a]	10	32	160	--	0.359	0.073
18534-1	MOBL	Tombigbee R. nr. Coffeeville Lock, AL	Largemouth Bass	2	588	350	4.5	1.572	0.313
18534-2	MOBL	Tombigbee R. nr. Coffeeville Lock, AL	Largemouth Bass	4	252	269	3.0	1.761	0.351
18535-1	MOBL	Coosa River at Rome, GA	Black Crappie	1	453	304	3.5	0.653	0.125
18535-2	MOBL	Coosa River at Rome, GA	Black Crappie	1	266	249	2.5	0.153	0.027
18535-3	MOBL	Coosa River at Rome, GA	Black Crappie	1	140	204	2.5	0.163	0.030
18536-1	MOBL	Chickasaw Cr., AL	Bluegill Sunfish[a]	2	31	118	--	1.108	0.248
00001-1	SANT	N. Fork Edisto R, nr. Fairview crossroad	Largemouth Bass	1	907	410	--	9.015	1.803
00002-1	SANT	N. Fork Edisto R. nr. Branchville, SC	Largemouth Bass	1	84	180	--	3.130	0.626
00003-1	SANT	South Edisto R. nr Springfield, SC	Largemouth Bass	1	517	310	--	2.860	0.572
00004-1	SANT	S. Fork Edisto R. nr Canaan, SC	Largemouth Bass	1	83	170	--	2.725	0.545
00005-1	SANT	Saluda R., SC	Largemouth Bass	1	900	390	--	2.355	0.471
00006-1	SOFL	Water Conservation 3A15, FL	Largemouth Bass	1	1694	507	4.8	21.1	4.22
00007-1	SOFL	Water Conservation 3A15, FL	Largemouth Bass	1	205	250	1.8	4.10	0.82
00008-1	SOFL	Water Conservation 3A15, FL	Largemouth Bass	1	466	325	2.8	7.10	1.42
00009-1	SOFL	Water Conservation U3, FL	Largemouth Bass	1	307	301	3.8	3.30	0.66
00010-1	SOFL	Water Conservation U3, FL	Largemouth Bass	1	227	273	2.8	1.85	0.37
00011-1	SOFL	Water Conservation U3, FL	Largemouth Bass	1	229	264	3.8	3.15	0.63

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13. ABSTRACT (*Maximum 200 words*)
Bioaccumulation of mercury (Hg) in fish sampled at 106 sites from 20 U.S. watershed basins was evaluated in relation to species, Hg and methylmercury (MeHg) in surficial sediment and water, and watershed characteristics. Bioaccumulation of Hg was strongly (positively) correlated with MeHg in water but only moderately with the MeHg in sediment or total Hg in water. The best model for predicting Hg bioaccumulation included MeHg in water, pH of the water, % wetlands in the basin, and the AVS content of the sediment; however, MeHg in water explained most of the model variability. Based on our data, a MeHg water concentration of 0.12 ng/L was on average, associated with a fish fillet Hg concentration of 0.3 mg/kg wet weight for an age-3 fish when all species were considered. For age-3 largemouth bass, a MeHg water concentration of 0.058 ng/L was associated with the 0.3 mg/kg fillet concentration. Based on rankings for Hg in sediment, water, and fish, sampling sites from the following five study basins had the greatest Hg contamination: Nevada Basin and Range, South Florida Basin, Sacramento River Basin (California), Santee River Basin and Coastal Drainages (South Carolina), and the Long Island and New Jersey Coastal Drainages.

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