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Long-term Changes in Mercury Concentrations in Fish from the Middle Savannah River

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Abstract

Total mercury levels were measured in largemouth bass (*Micropterus salmoides*), "sunfishes" (*Lepomis* spp)., and "catfish" (primarily *Ameiurus* spp.) from 1971 to 2004 in the middle reaches of the Savannah River, which drains the coastal plain of the southeastern U.S. Mercury levels were highest in 1971 but declined over the next ten years due to the mitigation of point sources of industrial pollution. Mercury levels began to increase in the 1980s as a possible consequence of mercury inputs from tributaries and associated wetlands where mercury concentrations were significantly elevated in water and fish. Mercury levels in Savannah River fish decreased sharply in 2001-2003 coincident with a severe drought in the Savannah River basin, but returned to previous levels in 2004 with the resumption of normal precipitation. Regression models showed that mercury levels in Savannah River fish changed significantly over time and were affected by river discharge. Despite temporal changes, there was little overall difference in Savannah River fish tissue mercury levels between 1971and 2004.

Key words: Fish, mercury, Savannah River, long-term monitoring, temporal trends, long-term data

Introduction

Important anthropogenic sources of mercury to aquatic environments include the discharge of mercury containing effluents and the atmospheric release of mercury that is eventually deposited on surface waters and watersheds. Mercury that enters aquatic ecosystems bioaccumulates in fish and other aquatic organisms, which are consumed by humans and wildlife with potentially deleterious effects (Eisler, 1987; Clarkson, 1990). Environmental regulations can alleviate this problem, but improvements may be slowed by lags in the implementation of remedial technologies and the continued and possibly episodic release of mercury to aquatic ecosystems from source pools in soils and sediments (Mason et al., 1994; Lorey and Driscoll, 1999; DiCosty et al., 2006). Therefore, long-term monitoring data is needed to determine the efficacy of regulations and discriminate short-term from secular trends in mercury contamination. Long-term monitoring has demonstrated decreases in mercury in some organisms as a result of the abatement of point sources of mercury pollution (Francesconi et al., 1997; Sager, 2002) and is needed to document the effects of regulations that control the atmospheric release of mercury.

Soil cores from the Florida Everglades in the southeastern United States show relatively high mercury accumulation rates after 1985 that likely resulted from global or regional atmospheric deposition (Rood et al., 1995). Mercury deposition rates have also been relatively high elsewhere in the southeastern United States (EPA, 1997; NADP, 2005). Exacerbating this problem are wetland habitats on the southeastern coastal plain characterized by warm temperatures, low pH, high concentrations of dissolved organic matter, and low oxygen concentrations that favor the methylating bacteria that convert inorganic mercury into more bioaccumulative methylmercury (Gilmour and Henry, 1991; Regnell, 1994; Francis et al., 1998). These and other factors have contributed to relatively high mercury levels in fish from some southeastern environments (EPA, 1999).

The Savannah River is a major river that drains the southeastern United States Piedmont and coastal plain. Some portions of the Savannah River have fish consumption advisories as do many other coastal plain rivers in the region because of relatively high mercury levels in fish. Mercury levels in fish tissue have been measured in the middle reaches of the Savannah River and several tributaries since 1971 as part of an environmental monitoring program conducted by the Savannah River Site (SRS), a Department of Energy facility in South Carolina. These data can be used to assess long-term changes in mercury contamination in Savannah River fish, identify factors that have affected contamination levels, and develop a baseline for evaluating the effects of possible future changes in mercury loading on mercury concentrations in fish. The objective of this paper is to describe changes in mercury concentrations in fish from the middle Savannah River between 1971 and 2004 and identify factors that influenced these changes.

Materials and Methods

The Savannah River is formed by the confluence of the Tugaloo and Seneca Rivers in northeast Georgia. It flows southeast through the Piedmont and Coastal Plain to the Atlantic Ocean,creating the border between the states of South Carolina and Georgia. It also constitutes the southwestern border of the SRS, an 800-km^2 nuclear materials production site established in 1951 near Aiken, South Carolina, USA. The SRS conducts an extensive environmental monitoring program to assess the possible movement of radiological and nonradiological contaminants into the surrounding environment. By 1971 this program had expanded to include the routine analysis of fish samples for mercury. Collection sites were located in the middle reaches of the Savannah River near river kilometers (RKs) 193, 225, and 258 prior to 1992 and RKs 191, 208, 228, 243, 245, 253, and 302 thereafter (Figure 1). Collection sites were also located in four Savannah River tributaries located on the SRS: Upper Three Runs, Fourmile Branch, Steel Creek, and Lower Three Runs. Fourmile Branch may have received mercury contamination from industrial seepage basins located near its headwaters, and a small tributary of Upper Three Runs received groundwater with low concentrations of mercury from a groundwater air stripping facility located approximately 6.5 km from its confluence with Upper Three Runs. However, aqueous total and methylmercury levels in Fourmile Branch and Upper Three Runs were not exceptional compared with Steel Creek, Lower Three Runs, and other tributaries without point sources of mercury contamination (Paller et al., 2004).

Sites were usually sampled yearly to every few years using fish traps, angling, and/or electrofishing. Fish were separated into largemouth bass (*Micropterus salmoides*), sunfishes (*Lepomis* spp.), and catfishes (*Ameiurus spp. and Ictalurus punctatus*); and only fish of edible size were collected (approximately $\geq$ 30 cm, 15 cm, and 30 cm total length, respectively). The numbers of fish collected varied and will be discussed in detail later. Muscle samples from individual whole fish were analyzed prior to 1992; and composite muscle samples from five fish were analyzed afterward. Samples were analyzed for total mercury by cold vapor atomic absorption spectrometry following tissue homogenization and digestion after 1992 and by wet digestion and flameless atomic absorption spectrophotometry in earlier years. Detection limits were $\leq$ 0.1 mg/kg for most samples but were occasionally as high as 0.3 mg/kg.

Annual monitoring reports recorded mercury concentrations for each individual fish after 1991or annual averages (plus maxima and sample sizes) for all fish from each taxonomic group from each site prior to this. These data were used to calculate annual arithmetic means over all Savannah River sites for each of the three taxonomic groups (weighted by sample size for years in which only site means rather than individual fish measurements were available). One half of the detection limit was substituted for values under the detection limit (approximately 11% of the data) when calculating averages. Second order polynomial regression models were used to analyze long-term trends for each taxonomic group. The dependent variable in each model was the average annual fish tissue mercury concentration, and the independent variables were year and Savannah River discharge (measured near Jackson, SC, USGS http://waterdata.usgs.gov/nwis/sw). Polynomial models were used because of the relatively complex relationships between the dependent and independent variables as discussed in detail later. A Durban-Watson D statistic was computed for the residuals from each model to determine if serial correlation was significant. Similarities in time trends among taxonomic groups were assessed with Pearson correlation coefficients. Matched-pairs t-tests were used to identify significant differences between mercury concentrations in fish from the Savannah River and fish from the tributary creeks. Separate t-tests were conducted for each taxonomic group and stream. The matched pairs for each test consisted of the average mercury concentration in fish from the river and the average mercury concentration in fish from the creek for each year in which collections were obtained from both sources. Statistical analyses were conducted with SYSTAT (SYSTAT Software Inc., 2002).

Results

Fish sample sizes from the Savannah River varied prior to 1991, ranging from zero during some years to as many as 188 individuals (catfish during 1974) (Figure 2). Sample sizes became more consistent after 1991, usually ranging from 30-35 individuals for each taxonomic group. Sunfish and catfish were collected during most years except 1977 to 1980 resulting in a fairly complete record for these taxonomic groups. In contrast, largemouth bass were not collected from the Savannah River in substantial numbers until 1985. The tributary creeks were sampled less consistently than the Savannah River; the average number of years each creek was sampled for each taxonomic group was 13.9 (5-19). The number of fish collected from the creeks was also smaller than the number collected from the river, ranging from 2-17 each year for each taxonomic group.

Mercury concentrations in Savannah River catfish and sunfish peaked in the early 1970s and then declined for about ten years (Figure 2), following a reduction in the discharge of mercury from a mercury-cell chlor-alkali plant in Augusta GA (Figure 1) from 5 kg/d to less than 0.1 kg/d in 1970 (Kvartek et al., 1994). Mercury levels in catfish and sunfish stopped decreasing in the early 1980s and around 1990 began a slow, erratic increase that was interrupted during 2000 to 2003, a period of low water level in the Savannah River (USGS http://waterdata.usgs.gov/nwis/sw) resulting from a drought that began in 1999 and ended in April 2003 (Keaton et al., 2005) . Mercury concentrations returned to levels equaling or exceeding those before the drought in 2004. Largemouth bass exhibited some of the same trends as sunfish and catfish, but the largemouth bass data were insufficient to verify patterns before 1985. There was a significant correlation between mercury levels in sunfish and catfish ($r=0.75$, $P<0.001$) and sunfish and largemouth bass ($r=0.68$, $P=0.001$), suggesting similar responses to changes in mercury bioavailability over time. However, the correlation between largemouth bass and catfish was relatively weak ($r=0.38$, $P=0.089$), with the comparatively short period of record for the former being a possible contributing factor.

The 2000-2003 drought reduced the discharge from tributary streams, many of which ceased flowing and some of which were largely dewatered (personal observation, M.H. Paller). Previous research showed that aqueous methylmercury concentrations were higher in these streams than in the Savannah River, and that these streams contributed methylmercury to the Savannah River (Paller et al., 2003). Persistently greater mercury bioavailability in the tributaries was indicated by the fish collection data, which showed that mercury concentrations were significantly higher ($P\leq 0.05$) in fish from the tributaries than in fish from the Savannah River except for largemouth bass in Upper Three Runs and catfish in Fourmile Branch (Figure 3).

Hypotheses suggested by the preceding data were that mercury levels in Savannah River fish varied nonrandomly over time and were affected by hydrological conditions in the Savannah River watershed. These two hypotheses were examined with second order polynomial regression models that tested whether year and Savannah River discharge were significant predictors of annual average mercury levels in fish. Savannah River discharge was the only hydrological metric consistently measured over the entire study period. First and second order terms for year were significant ($P \leq 0.05$) in the catfish and sunfish models, reflecting curvilinear temporal patterns resulting from early decreases in mercury followed by later increases (Table 1, Figure 4). Lack of significance of year in the largemouth bass model likely resulted from the relatively short period of record for this species. First and second order terms were significant ($P \leq 0.05$) for discharge in the sunfish and largemouth bass models, indicating a tendency for mercury levels in fish to be greater during years of intermediate discharge than during years of very low and very high discharge as illustrated by graphing regression residuals (from second order polynomial regressions of mercury on year) against flow (Figure 5). Unexplained variance in the models was relatively high (Table 1). Durban Watson statistics (Table 1) were not significant at $P<0.05$ indicating that the assumption of independence required for accurate statistical testing was met.

Mercury concentrations in Savannah River fish over time can be compared with the current USEPA human health criterion for methylmercury in fish of 0.3 mg/kg. Although total mercury rather than methylmercury was measured in Savannah River fish, most (90% or more) of the mercury in freshwater fish is usually methymercury (Bloom, 1992) suggesting it is not unreasonable to apply the EPA criterion to the Savannah River data. Mercury levels in sunfish exceeded 0.3 mg/kg in the early 1970s, subsequently decreased below this level, and then again exceeded the 0.3 m/l criterion in 2004 (Figure 2). Mercury levels in catfish also exceeded the criterion during the early 1970s, decreased below it during most of the period between 1975 and 1996, and were above the criterion thereafter except during the drought. Concentrations in largemouth bass were consistently above the human health criterion.

Discussion

There are several unmeasured sources of variability that could affect the analysis and interpretation of the Savannah River data. Species combined in the taxonomic groups sunfish and catfish may have differed in mercury body burdens as a result of species-specific physiological, ecological, and behavioral differences that affected mercury uptake and depuration. However, examination of stomach contents (unpublished data) indicated that all fish within each group had generally similar diets and occupied the same trophic level, suggesting generally similar potentials for mercury bioaccumulation. Size and age can also affect mercury concentrations in fish. Neither were typically measured, although some consistency resulted from the requirement that all fish be of edible size. Seasonal changes in fish tissue mercury concentrations constituted another source of variability that could not be evaluated because collection dates (other than year) were usually unavailable. Previous research on Savannah River fish showed that seasonal differences in mercury concentrations were significant (Paller et al., 2004) but smaller than the long-term changes described herein. Variations in sample size and changes in analytical methods also added uncertainty to the analysis. These and possibly other sources of error undoubtedly contributed to the relatively high unexplained variance in the regression models. Monitoring data developed by many individuals over 34 years cannot be expected to possess the same quality as typical research data but remain valuable because of their uniqueness.

We propose that the initial downward trend in fish tissue mercury levels (Figure 4) resulted from the abatement of mercury discharges from the Augusta chlor-akali plant in 1970. Other research has shown decreases in fish tissue mercury levels following the elimination of point sources of mercury pollution, although levels may stay relatively high in benthic organisms as a result of bioavailable mercury remaining in the sediments (Sager, 2002). An environmental half-life for mercury in smelt and sculpin of about 10 years was reported from Lake Ontario (Borgmann and Whittle, 1992), and a comparable environmental half-life was reported for teleost fishes from a marine bay in southwest Australia (Francesconi et al., 1997) following reductions in point source pollution. These rates of mercury reduction in fish tissues are generally similar to the rates observed in Savannah River fish in 1971-1976 following a 98% reduction in point source mercury discharges from the chlor-alkali plant in 1970.

Reasons for the upward trend in fish mercury levels that characterized the latter half of the study period (Figure 4) are more difficult to determine but may be related to mercury inputs from diffuse watershed sources. Sediment cores show relatively high mercury deposition in the Florida Everglades after 1985 (Rood et al., 1995), and it is likely that mercury deposition was also high in the Savannah River basin at this time. Relatively high methylmercury levels in fish from Savannah River tributaries (Figure 3) can be plausibly explained by watershed runoff of atmospherically deposited mercury that was subsequently transformed in the tributaries and associated backwaters where environmental conditions favored methylation. This was suggested by high mercury levels in fish from the tributary streams (Figure 3) and by results from a previous study showing that methylmercury levels were elevated in tributary water and in clams (Corbicula fluminea) residing in the discharge plumes of Savannah River tributary streams (Paller et al., 2003). Tributaries and the floodplain wetlands they drain could have contributed to increased mercury levels in Savannah River fishes by discharging methylmercury that is dissolved and associated with particulate matter into the Savannah River where it entered the food chain and through the migration of relatively contaminated tributary fish into the river as observed with other contaminants (Paller et al., 2005).

These sources of tributary mercury would be expected to diminish during droughts that reduce tributary discharge to the river, reduce the access of fish to the tributaries and floodplain, and dewater wetlands where relatively still water and accumulated organic matter favor methylation. Such conditions could lead to reduced methylmercury in Savannah River fish if they continued long enough and may be responsible for the temporary drop in fish tissue mercury levels observed during 2000-2003. Methylmercury half-lives in most freshwater fishes are under a year (Trudel and Rasmussen, 1997) suggesting that the 2000 through early 2003 low water period in the Savannah River basin would be sufficient for significant declines in fish tissue mercury levels given lower methylmercury availability.

Significant second order terms for Savannah River discharge suggested that high water years as well as low water years were associated with reduced mercury levels in sunfish and largemouth bass. Aqueous total mercury and methylmercury is frequently transported with dissolved and particulate organic matter (Balogh et al., 2003; Maurice-Bourgoin, 2003; Mast et al., 2005), which often peak in concentration on the ascending limb of flood hydrographs (Asselman, 1999; Lawler et al., 2006) and may become diluted during prolonged rainy periods (Johnson et al., 2006). Therefore, mercury levels in fish might be greatest during years of moderate flow that resulted in substantial methylmercury transport from the watershed without strong dilution. The occurrence of significant second order terms for Savannah River discharge in the regression models for sunfish and largemouth bass supports this hypothesis, although this effect was not observed with catfish. Additional research examining the relationship between flood events and aqueous mercury concentrations will be needed to fully understand and verify relationships between floodplain hydrology and mercury availability.

Long-term monitoring data may not provide the detailed information needed to fully understand mechanisms that affect mercury bioavailability, but they provide a unique overview of secular trends and help separate them from fluctuations on shorter time scales. Long-term data from the Savannah River showed that reductions in fish mercury levels in the early 2000s were temporary and probably a result of hydrological factors. In aggregate, the Savannah River data indicate that the control of point sources resulted in relatively rapid decreases in fish tissue mercury levels followed by later increases most likely associated with the mobilization of atmospherically deposited mercury from the Savannah River watershed. As a result, there was little overall difference in fish tissue mercury levels between 1971 and 2004.

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Table 1. Statistical significance of terms in a regression model[a] evaluating the effects of year and flow (Savannah River discharge) on average yearly total mercury levels in three types of fish collected from the Savannah River during 1971-2004.

Model term	Sunfishes		Catfishes		Largemouth bass	
	t	P	t	P	t	P
Constant	3.520	0.002	3.835	0.001	1.114	0.282
Year	-3.517	0.002	-3.840	0.001	-1.118	0.280
$Year^2$	3.515	0.002	3.835	0.001	1.121	0.279
Flow	2.401	0.025	1.652	0.122	2.221	0.041
$Flow^2$	-2.247	0.035	-1.500	0.147	-2.131	0.049
R^2	0.47		0.42		0.28	
Durbin-Watson D statistic	2.061		1.702		1.872	
First order Autocorrelation	-0.039		0.130		-0.008	

[a] Average annual HG (mg/L)=a-b_1(Year)+b_2($Year^2$)+b_3(Flow)-b_4($Flow^2$)

List of Figures.

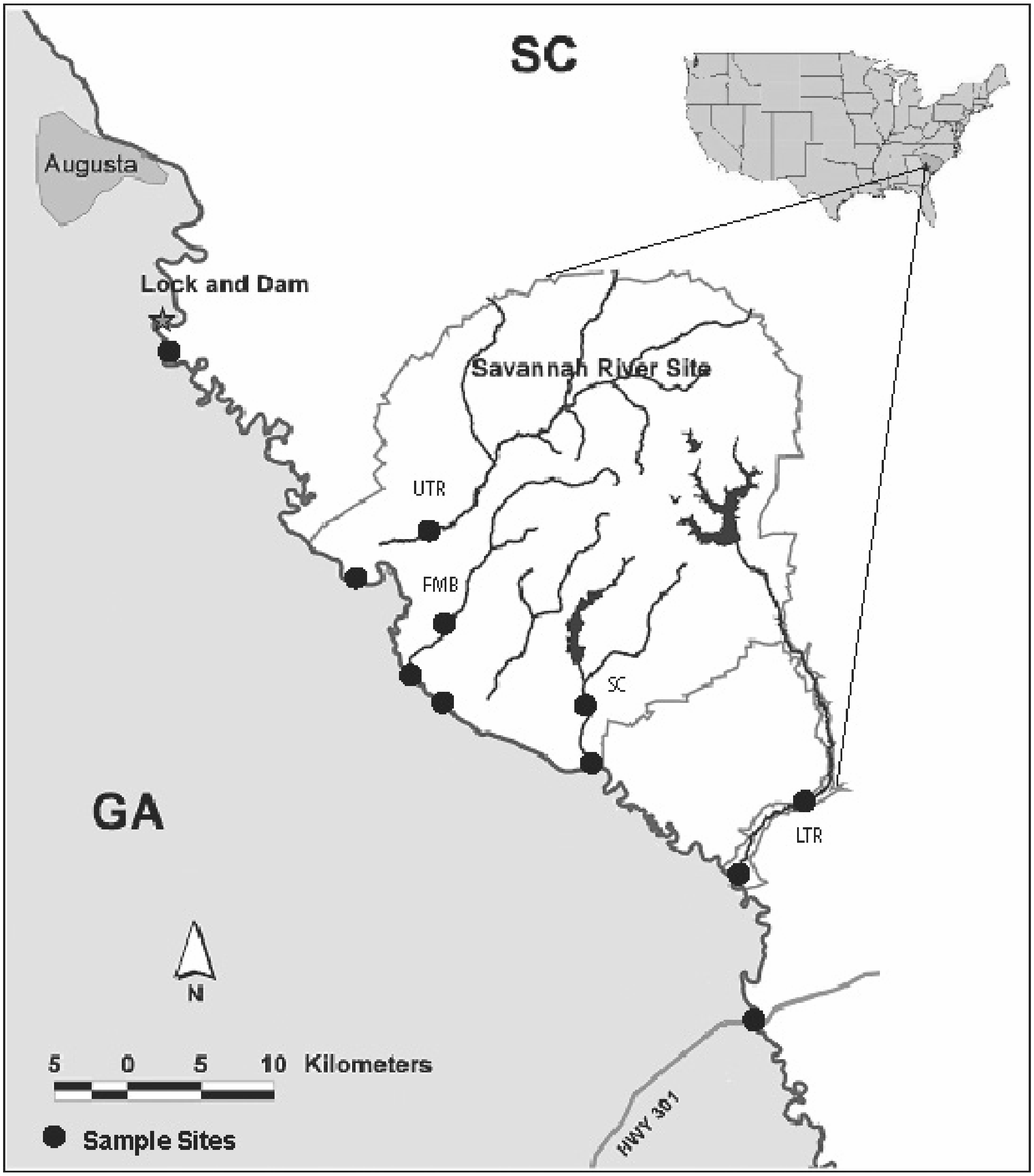
Figure 1. Long-term fish collection sites in the middle Savannah River and Savannah River Site (UTR = Upper Three Runs, FMB = Fourmile Branch, SC = Steel Creek, LTR = Lower Three Runs).

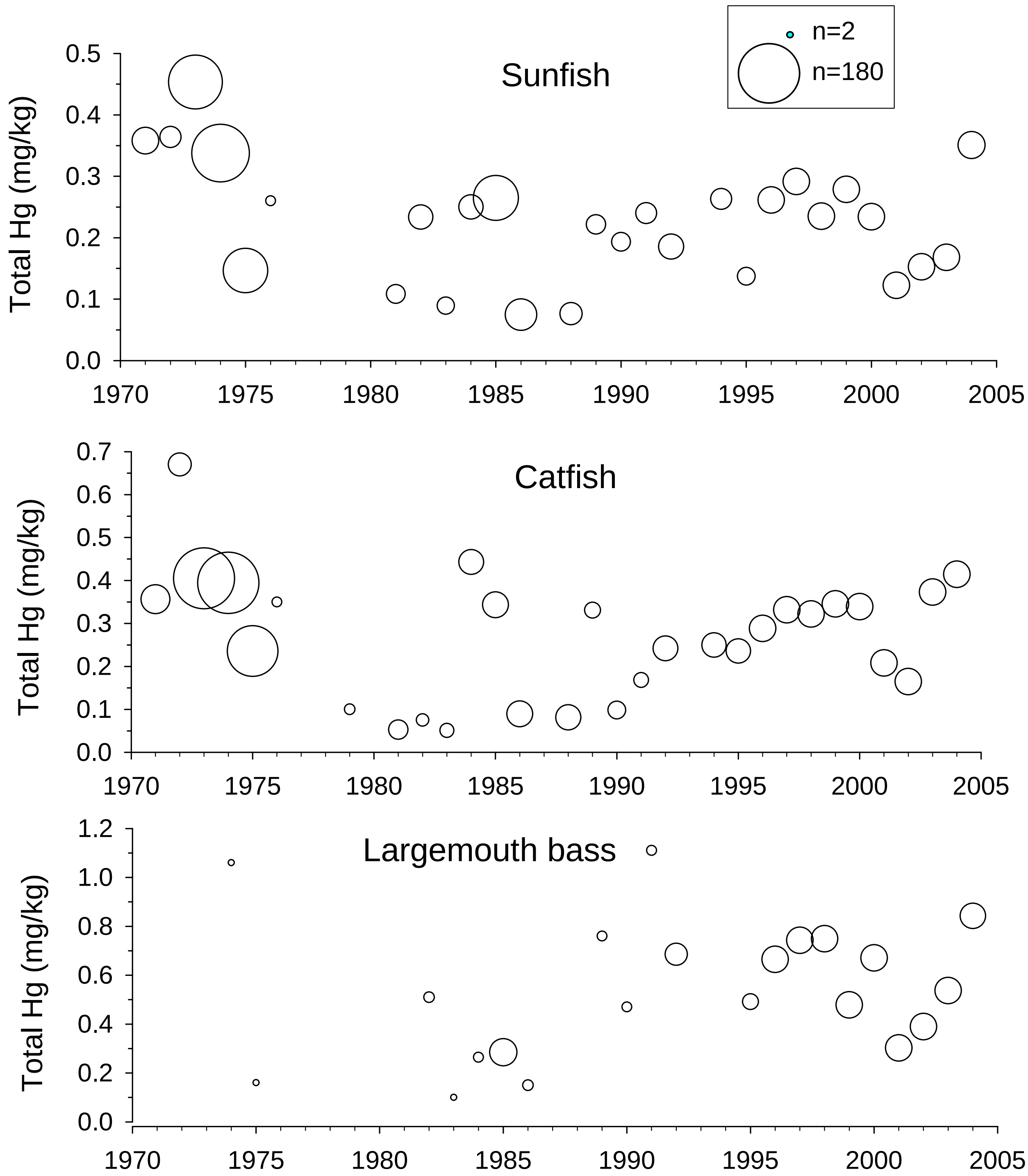
Figure 2. Total mercury in fish collected from the middle Savannah River during 1971-2004.

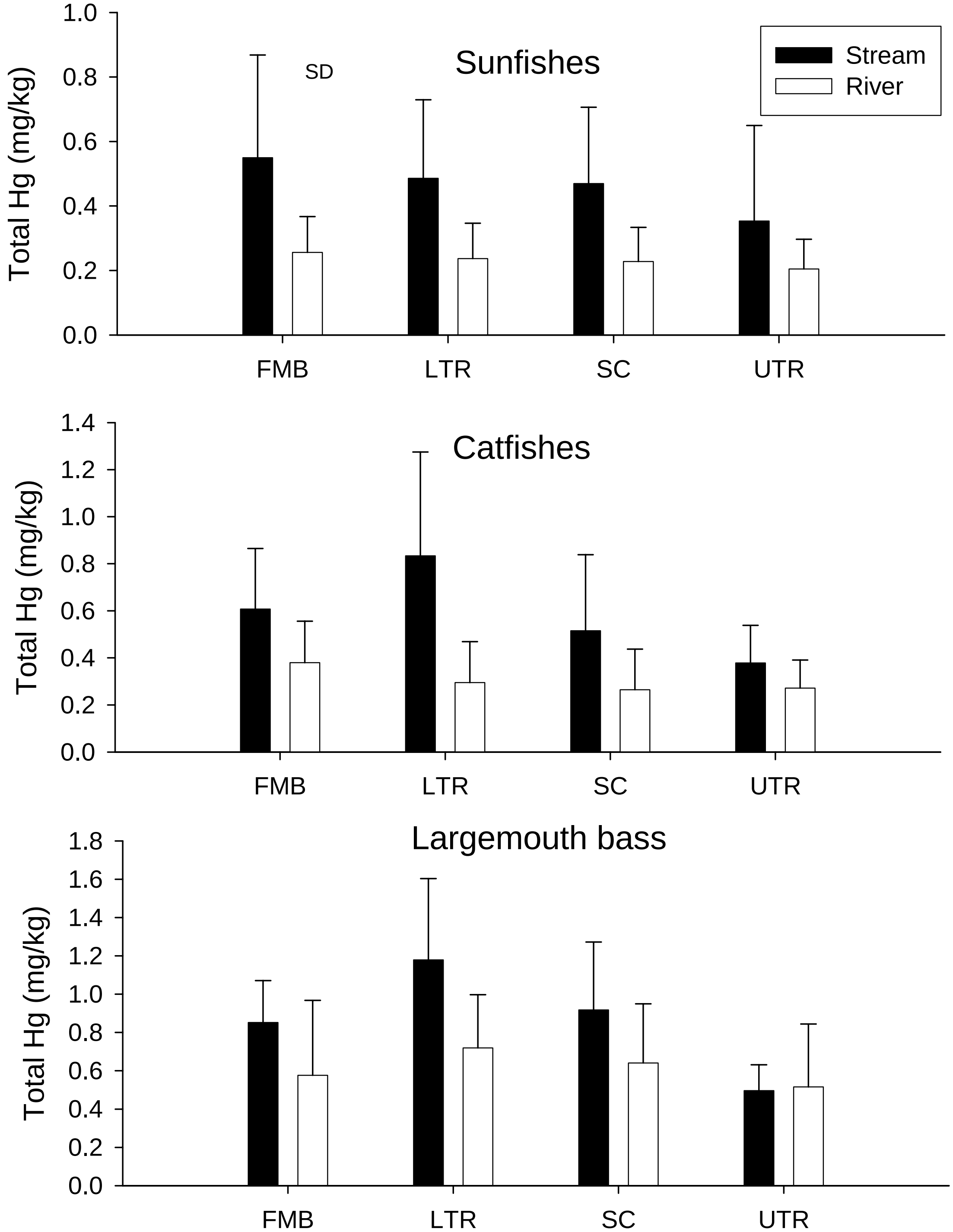
Figure 3. Total mercury in fish from the Savannah River and four Savannah River tributary streams

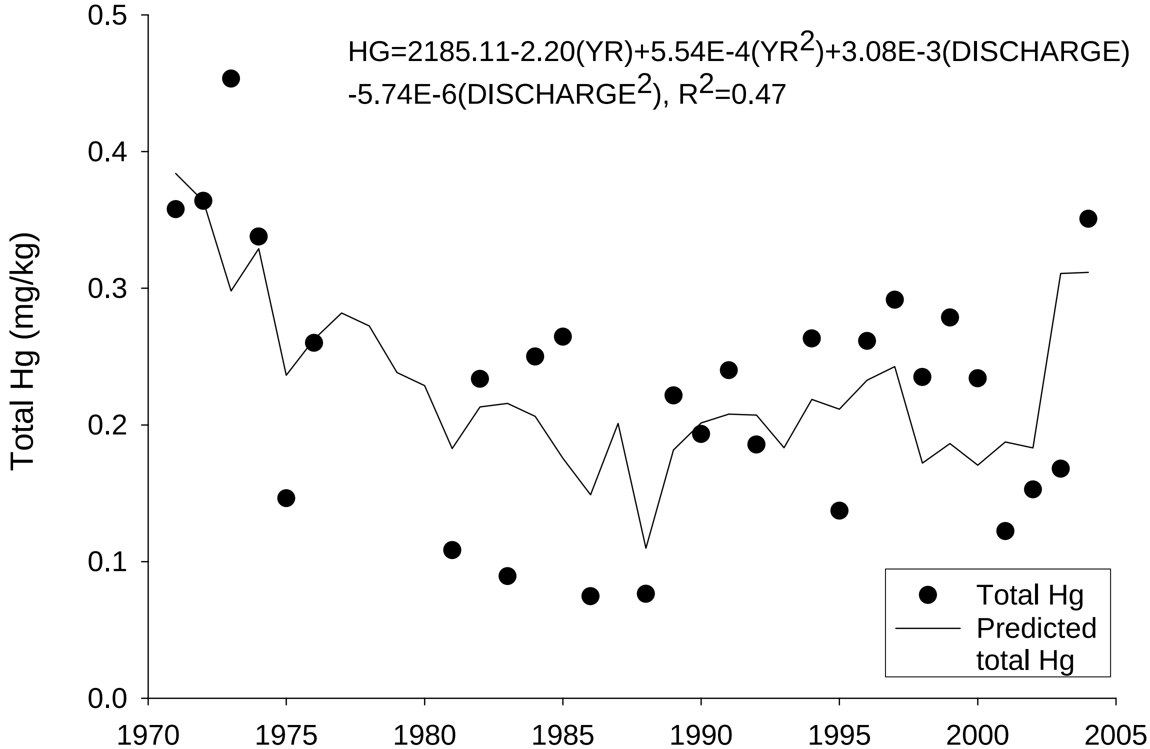
Figure 4. Polynomial regression model predicting total mercury in sunfishes from year and Savannah River discharge (m^3/s)

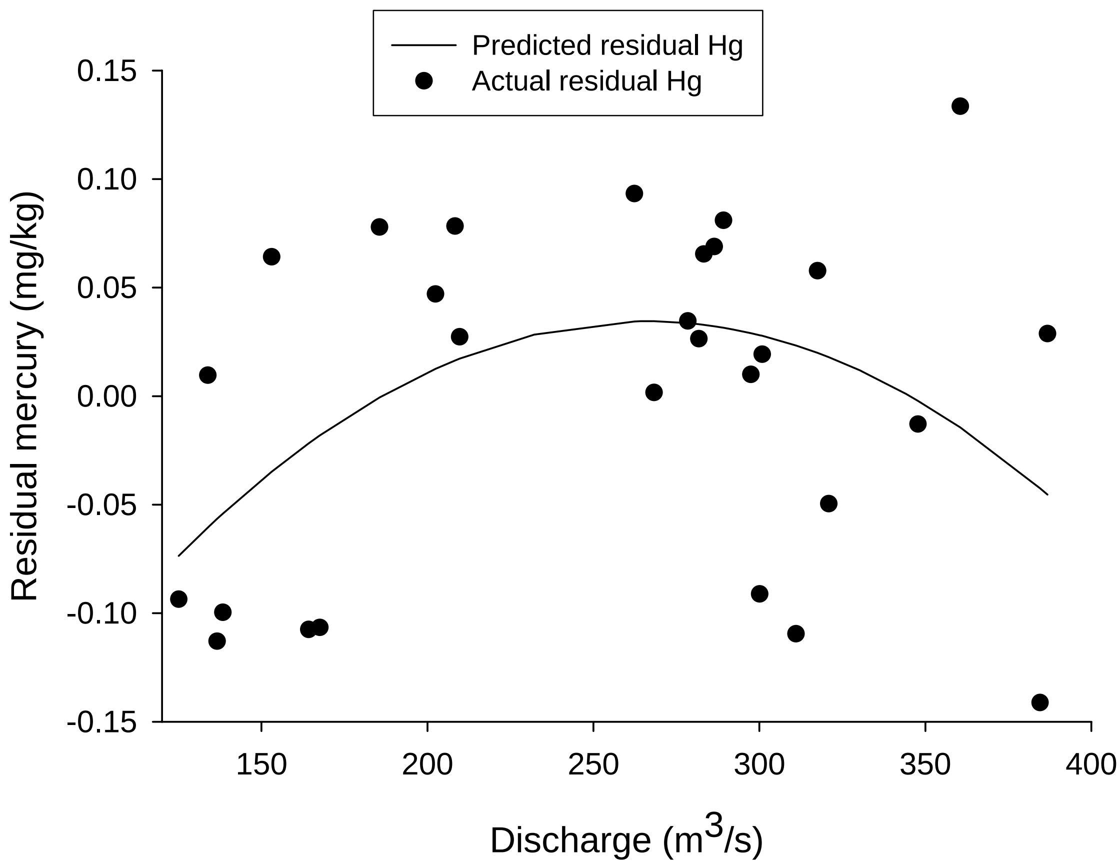
Figure 5. Relationship between Savannah River discharge and residual mercury scores derived from a second order polynomial regression of mercury levels in sunfishes on year (the residuals represent the variation in fish tissue mercury levels stripped of variance associated with year).


SC
Augusta
Lock and Dam
Savannah River Site
UTR
FMB
SC
LTR
GA
N
5 0 5 10 Kilometers
Sample Sites
HWY 301


n=2
n=180
Sunfish
Catfish
Largemouth bass
Total Hg (mg/kg)
0.0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
1.0
1.2
1970
1975
1980
1985
1990
1995
2000
2005


Sunfishes
SD
Stream
River
Total Hg (mg/kg)
1.0
0.8
0.6
0.4
0.2
0.0
FMB
LTR
SC
UTR
Catfishes
Total Hg (mg/kg)
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
FMB
LTR
SC
UTR
Largemouth bass
Total Hg (mg/kg)
1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
FMB
LTR
SC
UTR


HG=2185.11-2.20(YR)+5.54E-4(YR2)+3.08E-3(DISCHARGE)
-5.74E-6(DISCHARGE2), R2=0.47
Total Hg (mg/kg)
0.5
0.4
0.3
0.2
0.1
0.0
1970
1975
1980
1985
1990
1995
2000
2005
Total Hg
Predicted total Hg


Predicted residual Hg
Actual residual Hg
0.15
0.10
0.05
0.00
-0.05
-0.10
-0.15
Residual mercury (mg/kg)
150
200
250
300
350
400
Discharge (m3/s)