Toxic Substances Hydrology Program and
National Water-Quality Assessment Program

Prepared in cooperation with the National Park Service

Comparison of Methylmercury Production and Accumulation in Sediments of the Congaree and Edisto River Basins, South Carolina, 2004–06

Scientific Investigations Report 2009–5021

U.S. Department of the Interior
U.S. Geological Survey

Cover photograph. Early morning in the Congaree National Park, South Carolina.

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Conversion Factors, Datum, and Abbreviations

Multiply	By	To obtain
	Length	
centimeter (cm)	0.3937	inch (in.)
kilometer (km)	0.6214	mile (mi)
	Area	
square kilometer (km^2)	247.1	acre
square kilometer (km^2)	0.3861	square mile (mi^2)
	Volume	
liter (L)	33.82	ounce, fluid (fl. oz)
liter (L)	2.113	pint (pt)
liter (L)	1.057	quart (qt)
liter (L)	0.2642	gallon (gal)
liter (L)	61.02	cubic inch (in^3)

Temperature in degrees Celsius (°C) may be converted to degrees Fahrenheit (°F) as follows:

$$°F=(1.8\times°C)+32$$

Temperature in degrees Fahrenheit (°F) may be converted to degrees Celsius (°C) as follows:

$$°C=(°F-32)/1.8$$

Horizontal coordinate information is referenced to the North American Datum of 1983 (NAD 83).

Concentrations of chemical constituents in water are given either in milligrams per liter (mg/L) or micrograms per liter (µg/L).

Abbreviations

µL/L microliters per liter

ng/g/d nanograms per gram per day

ng/kg nanograms per kilogram

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Abstract

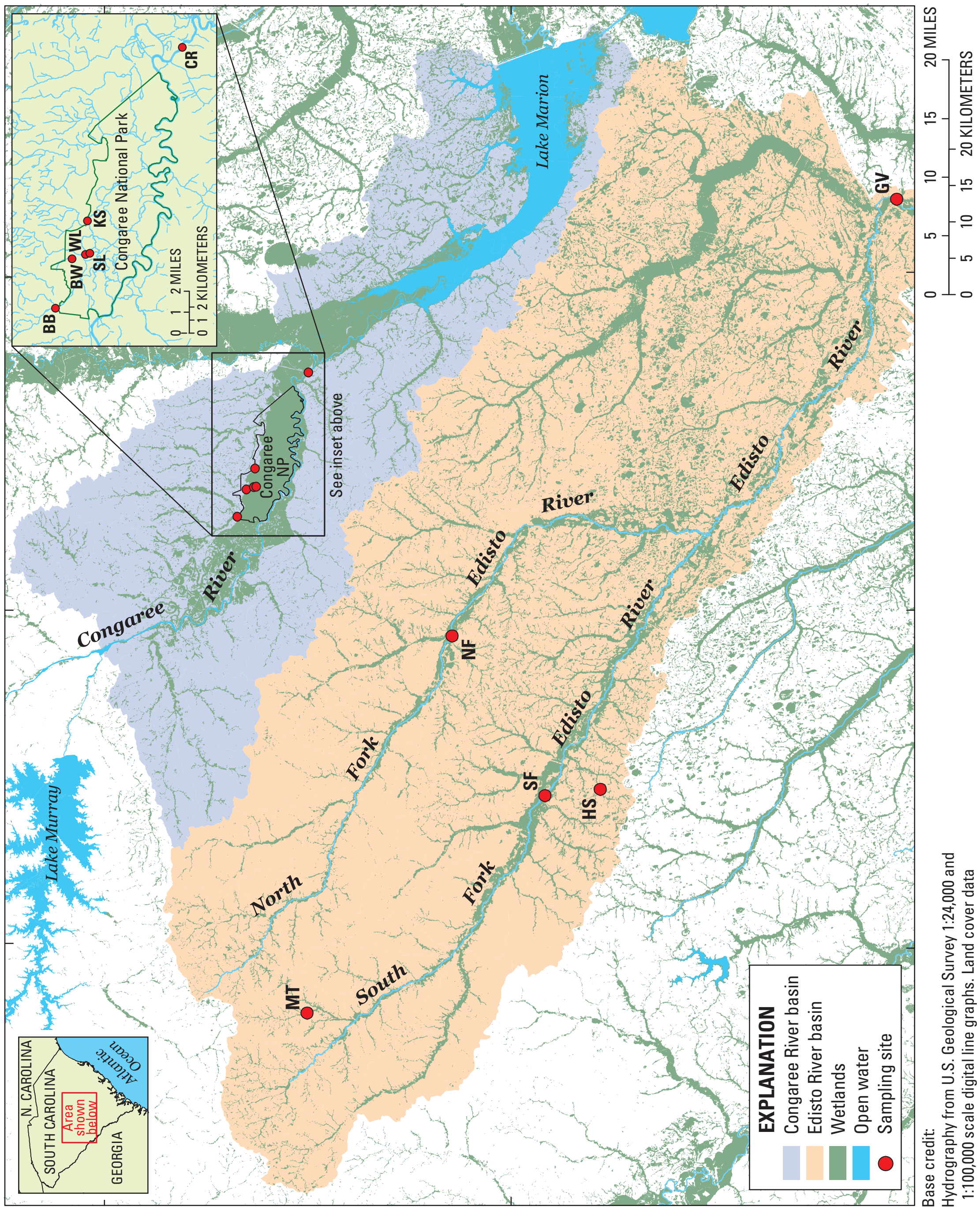
Fish-tissue mercury concentrations (approximately 2 micrograms per gram) in the Edisto River basin of South Carolina are among the highest recorded in the United States. Substantially lower mercury concentrations (approximately 0.2 microgram per gram) are reported in fish from the adjacent (about 30 kilometer) Congaree River basin and the Congaree National Park. In contrast, concentrations of total mercury were statistically higher in sediments from the Congaree River compared with those in sediments from the Edisto River. Furthermore, no statistically significant difference was observed in concentrations of methylmercury or net methylation potential in sediments collected from various Edisto and Congaree hydrologic settings. In both systems, the net methylation potential was low (0–0.17 nanogram per gram per day) for in-stream sediments exposed to continuously flowing water but substantially higher (about 1.8 nanograms per gram per day) in wetland sediments exposed to standing water. These results are not consistent with the hypothesis that differences in fish-tissue mercury between the Edisto and Congaree basins reflect fundamental differences in the potential for each system to methylate mercury. Rather, the significantly higher ratios of methylmercury to total mercury observed in the Edisto system suggest that the net accumulation and(or) preservation of methylmercury are greater in the Edisto system. The marked differences in net methylation potential observed between the wetland and in-stream settings suggest the hypothesis that methylmercury transport from zones of production (wetlands) to points of entry into the food chain (channels) may contribute to the observed differences in fish-tissue mercury concentrations between the two river systems.

Introduction

The bioaccumulation of mercury (Hg) in fish tissue above recommended levels (0.3 microgram per gram [µg/g] Hg) is an important public health concern in many parts of the United States (Krabbenhoft and others, 1999). As of 2004, 44 states, 1 territory, and 2 Native American tribes have listed more than 2,400 fish consumption advisories due to Hg (U.S. Environmental Protection Agency, 2004). In South Carolina, elevated Hg concentrations in fish tissue have resulted in consumption advisories for several river basins within the Coastal Plain Physiographic Province, including the Edisto River basin (South Carolina Department of Health and Environmental Control, 2006). However, the fundamental hydrologic, geochemical, and ecologic processes leading to elevated fish-tissue Hg concentrations remain unclear. This study compared sediment Hg concentrations and sediment net methylation potentials (NMP) in two adjacent river basins in South Carolina that have markedly different fish-tissue Hg concentrations. The comparison was made to improve understanding of the processes leading to Hg accumulation in fish.

The Edisto River (fig. 1) is characterized by warm water temperatures, low-stream gradients, extensive riparian wetlands, and tannic-colored water. Approximately 15 percent of the 4,393 square kilometers (km^2) of surface area in the Edisto River basin is wetlands. Fish-tissue Hg concentrations in the Edisto River basin (typically about 2 µg/g) are the highest in South Carolina and among the highest in the Nation (Krabbenhoft and others, 1999). In contrast, the coastal plain component of the Congaree River basin, which includes the Congaree National Park, is located immediately adjacent to the Edisto River basin (fig. 1) but exhibits significantly lower fish-tissue Hg concentrations (~0.2 µg/g). Approximately 20 percent of the Congaree River basin between the Fall Line and the confluence with the Wateree River is wetland. The Congaree National Park is a 43-mi^2 wetland-dominated system that includes the largest contiguous tract of old-growth bottomland hardwood forest in the United States.

Despite the proximity and the similar wetlands coverage of the two basins, few fish-consumption restrictions apply to the Congaree River, whereas stringent consumption restrictions apply to the Edisto River basin, including no consumption of largemouth bass in parts of the basin (U.S. Environmental Protection Agency, 2004; South Carolina Department of Health and Environmental Control, 2006). These fish-consumption advisories have important economic consequences because of the prevalence of recreational and subsistence fishing in the Edisto River basin. Nor are the health effects of elevated Hg concentrations in fish limited



Base credit:
Hydrography from U.S. Geological Survey 1:24,000 and 1:100,000 scale digital line graphs. Land cover data

Figure 1. Location of the Edisto and Congaree River basins, the Congaree National Park, and Edisto River sampling sites in South Carolina.

to humans. Accumulation of Hg in a number of fish-eating wildlife in South Carolina has been documented and includes the American alligator (*Alligator mississippiensis*; Yanochko and others, 1997) and the bald eagle (*Haliaeetus leucocephalus*; Jagoe and others, 2002). Levels of Hg in the feathers and blood of bald eagles in the Edisto River basin are consistent with the elevated fish-tissue Hg concentrations and are among the highest recorded in South Carolina (Jagoe and others, 2002).

In addition to these ecosystem- and public-health concerns, the differences in fish-tissue Hg concentrations between the Edisto and Congaree Rivers are of scientific interest because of the physical proximity of these systems (fig. 1). This proximity implies that atmospheric loading of Hg, which is the principal source of environmental Hg in South Carolina (U.S. Environmental Protection Agency, 2001), is similar and indicates that observed differences in fish-tissue Hg concentrations are a result of other factors affecting the production, transport, or bioaccumulation of methylmercury (MeHg) (Southworth and others, 2004) rather than differences in the availability of Hg. Thus, a comparison of these systems provides insight into the hydrologic, geochemical, or ecological processes that produce elevated fish-tissue Hg concentrations in surface-water systems.

Because MeHg is the primary form of Hg in fish (Bloom, 1992; Rudd, 1995; Weiner and Spry, 1996; U.S. Environmental Protection Agency, 1997), the concentration of Hg in fish tissues can be attributed to interactions between three conceptual components of the MeHg biocycle: (1) the microbial production and in situ persistence of MeHg; (2) the transport of MeHg from the site and matrix of production to the point of entry into the food web; and (3) the extent of MeHg accumulation within the food web. This study was designed specifically to examine the first of these components and focused on comparing the production and accumulation of MeHg in various hydrologic settings of the Congaree and Edisto River basins. Specifically, relative differences in production and persistence of MeHg were compared by assessing (1) the relative supply of total mercury (Hg_{tot}) in the two systems, (2) the existence within the various hydrologic settings of biogeochemical conditions conducive to Hg methylation, (3) the potential for and relative efficiency of microbial Hg methylation in sediments of each hydrologic setting, and (4) the relative accumulation and persistence of MeHg in each system.

Methods and Materials

This section presents details of the assessment of select hydrogeochemical characteristics and mercury methylation potentials along with applicable statistical approaches used for data interpretation.

Hydrologic Settings

The coastal plain wetland systems associated with the Edisto River, the Congaree River, and the Congaree National Park are complex environments composed of several distinct hydrologic settings. The wide variety of hydrologic settings in these systems makes it challenging to quantify processes that occur on a small scale, such as Hg methylation in sediments, and apply those results to ecosystem-scale phenomena (MeHg accumulation in fish tissue). Results of previous studies have shown that the efficiency of Hg methylation varies with hydrologic settings within surface-water systems (Branfireun, 2004; Stamenkovic and others, 2005; Lambertsson and Nilsson, 2006). In these previous studies, individual hydrologic settings that were considered to be representative of the system as a whole were assessed in order to identify system-wide differences in net methylation. A similar approach was taken in the current study. Sediments from representative and areally important hydrologic settings were sampled to quantify Hg concentrations and to experimentally estimate net methylation potentials. The goal of this effort was to determine if the observed differences in fish-tissue Hg concentrations between the two river basin systems reflected fundamental differences in the production and(or) persistence of MeHg.

In the Edisto River basin (fig. 1), the most prevalent hydrologic settings are surface-water channels, perennially inundated wetlands characterized by standing or slow-flowing water for much of the year, and seasonally inundated wetlands. In the Edisto River basin, all three hydrologic settings are replenished predominantly by ground-water seepage (Zalants, 1990). Two sites characteristic of channel settings—the McTier (MT) site in the upper part of the basin and the Givhans (GV) site in the lower part of the basin (fig. 1)—were selected for study because each site has a gaging station with long-term record of flow conditions (Cooney and others, 2003). Three sites characterized by extensive wetland areas—North Fork (NF), South Fork (SF), and Healing Springs (HS)—also were selected (fig. 1). The Healing Springs site exemplified an area of wetlands fed by perennial ground-water discharge. The North Fork and South Fork study locations were examples of seasonally inundated flood-plain environments.

The Congaree River and associated Congaree National Park have similar hydrologic settings (fig. 1). Three channel settings, the Bannister Bridge (BB) and King Snake (KS) sites within the Congaree National Park (CNP) and the Congaree River (CR) site downstream from the CNP, were selected for study. Ground-water seepage appears to be less substantial in the CNP than in the Edisto River (Zalants, 1990), but one site (Boardwalk [BW]) was selected as an example of a ground-water seepage environment in the CNP. Two additional categories of wetland environments—oxbow lakes and sloughs—are of particular ecological interest in the CNP system and also were assessed. Oxbow lakes of varying dimensions are intermittently flushed and refilled during high-water flood events. Sloughs are shallow channels that transport Congaree River water into the CNP during intermediate and high-flow events, but usually contain standing water when flow in the Congaree River is low. Weston Lake (WL) was selected to represent the oxbow lakes in this system, and one slough site (SL) was selected to represent surface-water channels with intermittent connection to the Congaree River.

Sediments for analyses of in situ Hg_{tot} and MeHg concentrations and for the preparation of microcosm studies were collected from each sampling location during 2004–2006 using standard methods (Radtke, 2005). Briefly, approximately 10 grams (gm) of sediment and 8 milliliters (mL) of water were collected directly into sterile 20-mL glass vials at the sediment–water interface. Each vial was sealed with a Teflon-lined stopper and crimped, leaving a 2-mL internal headspace of air.

Total Mercury and Methylmercury Concentrations in Sediments

Collected sediment vials were immediately frozen and shipped to commercial analytical laboratories for analysis (Frontier Geosciences Inc, Seattle, WA, in 2004 and Brooks Rand LLC, Seattle, WA, in 2005–2006). Duplicate or triplicate sediment samples were analyzed for Hg_{tot} and monomethyl Hg using cold vapor atomic florescence according to U.S. Environmental Protection Agency Methods 1631 and 1630, respectively (U.S. Environmental Protection Agency, 2002, 1998, respectively).

Sedimentary Organic Matter and Redox Conditions

Concentrations of sedimentary organic matter (SOM) present in aquatic sediments have been shown to facilitate Hg methylation (Lambertsson and Nilsson, 2006), and the SOM contents of Congaree and Edisto sediments were measured to compare the two systems. Concentrations of SOM in aquatic sediments collected from the sampling sites were determined by loss-on-ignition at 500 degrees Celsius (ºC). In addition to SOM concentrations, the overall reducing potential of the sediments was assessed by monitoring the accumulation of methane (CH_4) in sediment microcosms incubated under anoxic conditions. The greater the reducing potential of the sediments, the more quickly concentrations of competing electron acceptors (nitrate, Fe(III), and sulfate) become depleted in the vials initiating active methanogenesis. Thus, over a relatively short incubation period, the accumulation of CH_4 in sediment microcosms provides a qualitative comparison of the reducing potential of sediments from different environments. Concentrations of CH_4 in the vial headspace were monitored over time by using gas chromatography with reduction-gas detection (Trace Analytical, Menlo Park, CA). Separation of gases was achieved by using a molecular sieve column (10 centimeters [cm]) packed with Carbosieve II (Supelco) and using nitrogen as the carrier gas. Concentrations of CH_4 were quantified by preparing standard curves using known standards (Scotty). The analytical reproducibility of duplicate standards typically is greater than 97 percent. Concentrations of CH_4 increased continuously over time in the incubating vials, and the reported CH_4 concentrations in the headspace of the duplicate vials were made after 7 weeks of incubation.

Mercury Methylation and Demethylation Potential

Microcosm studies to assess the net Hg methylation and demethylation potential of wetland sediments were conducted as described by Macalady and others (2000). For methylation experiments, live and heat-sterilized control treatments were amended with $HgCl_2$ to a final concentration of 125 milligrams per liter (mg/L). For demethylation experiments, live and heat-sterilized control treatments were amended with MeHg to a final concentration of 25 mg/L. For each Hg and sediment treatment, duplicate heat-sterilized control microcosms were autoclaved three consecutive times for 1 hour over a 24-hour period. For each Hg and sediment combination, initial viable and heat-sterilized control treatments were prepared by freezing the respective sediment microcosms immediately after Hg amendment. Duplicate viable microcosms (final viable) were amended with the appropriate Hg substrate, incubated in the dark for 30 days, and then frozen to end the incubation. This approach involved three sets of controls (initial viable, initial heat-sterilized, and final heat-sterilized) that were compared with the final viable treatments. Concentrations of Hg_{tot} and MeHg in the control and experimental sediment vials were analyzed according to the methods described above.

The differences observed between the various control treatments (initial viable, initial heat-sterilized, and final heat-sterilized) were not statistically significant. For convenience, the final heat-sterilized control treatments were used to quantify net methylation and demethylation activity. In this report, net methylation and demethylation activity is operationally defined as the difference in MeHg concentrations between the final viable treatment and the final heat-sterilized treatment. Because this experimental methodology was carried out over 30 days and conditions in the vial can be expected to change over the incubation period, the results are most suited to comparing the potential for net methylation and demethylation between sediments from different hydrologic settings rather than for estimating in situ rates.

Statistical Tests

The statistical significance ($p < 0.05$) of possible differences between the Edisto and Congaree River basin systems was assessed with regard to (1) Hg_{tot} concentrations, (2) weight percentage of SOM and CH_4 production, (4) net methylation potentials (NMP), (5) the MeHg to Hg_{tot} ratio, and (6) possible differences in NMP between in-stream and wetland settings. The Mann-Whitney U non-parametric test was used for comparing the two populations.

Comparison of Methylmercury Characteristics in Edisto and Congaree River Basin Sediments

This study was designed to examine the general hypothesis that differences in fish-tissue Hg between the Edisto River and Congaree River basins reflect differences in the production and accumulation of MeHg in the sediments of these systems. Specifically, it was hypothesized that differences in the production and accumulation of sediment MeHg between the two basins could reflect differences in (1) the relative supply of Hg_{tot}, (2) the occurrence of hydrologic settings conducive to Hg methylation, (3) the potential for and relative efficiency of microbial Hg methylation, and (4) the extent of accumulation and(or) the environmental persistence of MeHg.

Hypothesis 1, which assumes that the supply of Hg_{tot} is higher in the Edisto River system than in the Congaree River system, is the most straightforward. However, the results of this study show that concentrations of Hg_{tot} were statistically significantly higher ($p = 0.01$) in collected Congaree sediments than in Edisto sediments (fig. 2*A*). This observation, in turn, is not consistent with hypothesis 1. This result is consistent, however, with previous findings that indicate that Hg_{tot} concentrations are a poor predictor of in situ MeHg concentrations (Kelly and Rudd, 1995; Hammerschmidt and Fitzgerald, 2004; Sunderland and others, 2004). There was no statistically significant difference in MeHg concentrations between Congaree and Edisto sediments (fig. 2*B*). However, MeHg/Hg_{tot} ratios were significantly higher ($p = 0.016$) in Edisto sediments compared to Congaree sediments (fig. 2*C*).

The results of this study also do not support hypothesis 2, which assumes that the Congaree River system lacks the geochemical conditions and(or) the hydrologic settings that support Hg methylation. Both the Edisto and Congaree River systems are characterized by wetland hydrologic settings with high organic sediment content and reducing redox conditions that have been shown to promote mercury methylation (St. Louis and others, 1994; Hammerschmidt and Fitgerald, 2004; Lambertsson and Nilsson, 2006). This scenario is illustrated in figure 3, which shows methane production plotted

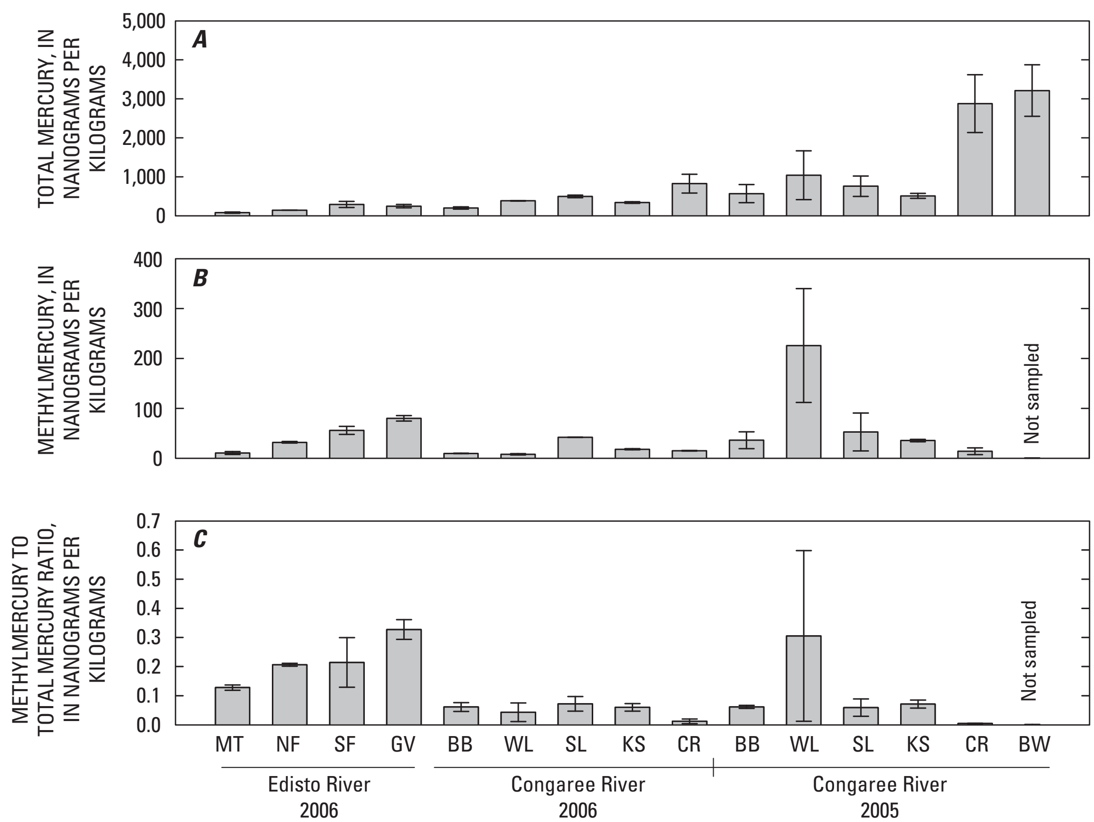


Figure 2. Concentrations of *(A)* total mercury, *(B)* methylmercury, and *(C)* the ratio of methyl to total mercury in sediments of the Edisto River, Congaree National Park, and Congaree River, 2005 and 2006.

against sediment organic matter (SOM) for the Congaree and Edisto Rivers. Statistically, there was no evidence of significant differences between the two systems in either SOM content ($p = 0.5$) or CH_4 production ($p = 0.5$; fig. 3). Furthermore, concentrations of dissolved sulfate (2–10 mg/L) are not statistically different in the river water of these two systems (U.S. Geological Survey, 2008), and there is no evidence that the distribution of the sulfate-reducing conditions that favor Hg methylation (Compeau and Bartha, 1985; King and others, 2000) is fundamentally different between the two systems. The low-sulfate conditions in both systems may serve to poise redox conditions near the interface of sulfate-reduction and methanogenesis, a condition that has been shown to facilitate active Hg methylation (Pak and Bartha, 1998). Moreover, wetlands are more areally extensive in the Congaree River system (20 percent of the basin area) than in the Edisto River system (15 percent of the basin area; fig. 1). The CNP, which has lower bass fish-tissue MeHg concentrations than the Edisto, is more than 90 percent wetlands (fig. 1). Thus, differences in the abundance of high methylation-potential wetland settings in these river basins do not explain the higher fish-tissue Hg concentrations in the Edisto relative to the Congaree.

Hypothesis 3, which assumes that sediments of the Edisto River basin have higher NMPs than sediments of the Congaree River basin, also is not supported by the results of this study. Rather, NMPs were not significantly different ($p = 0.25$) between the two systems (fig. 4). Net methylation potentials were statistically lower in sediments from in-stream sites and higher in sediments from wetland sites characterized by standing water in both the Edisto ($p = 0.036$) and Congaree ($p = 0.013$) systems. This pattern indicates that the primary areas of Hg methylation (wetland sediments) are often physically and hydrologically separated from the primary habitat (in-stream settings) of mid-level trophic (sunfish) and high-level trophic (bass) fish. This, in turn, indicates that the hydrologic transport of MeHg from zones of MeHg production to the point of entry into the food chain may be a factor affecting the accumulation of MeHg in fish tissue.

The results of this study are consistent with hypothesis 4, which assumes that differences exist between the two study areas in the overall accumulation and(or) environmental persistence of MeHg in the sediment environment. These differences are indicated by the statistically significantly higher MeHg to Hg_{tot} ratios (fig. 2*C*) observed in the Edisto system compared with those in the Congaree River and CNP system. MeHg to Mg_{tot} ratios observed in sediments in this study are consistent with the MeHg to Hg_{tot} ratios reported previously in the surface waters of these two systems (Hughes and others, 2000).

In conclusion, the results of this study are not consistent with the general hypothesis that differences in the potential production of MeHg between the Edisto and Congaree systems can explain the observed differences in fish-tissue concentrations. Because of the limited number of samples analyzed and the limited number of hydrologic settings considered, however, this lack of supporting evidence does not unequivocally rule out the hypothesis. Nevertheless, these results indicate that factors other than simply NMP may control the observed differences in MeHg accumulation in the sediments and biota of the two systems.

The observation of higher MeHg to Hg_{tot} ratios in Edisto sediments and in-stream waters indicates (1) that some combination of geochemical and hydrologic factors may limit the actual (as opposed to the potential) extent of Hg methylation under in situ conditions in the Congaree study area, or (2) that the contribution of low-MeHg water originating in the nearby

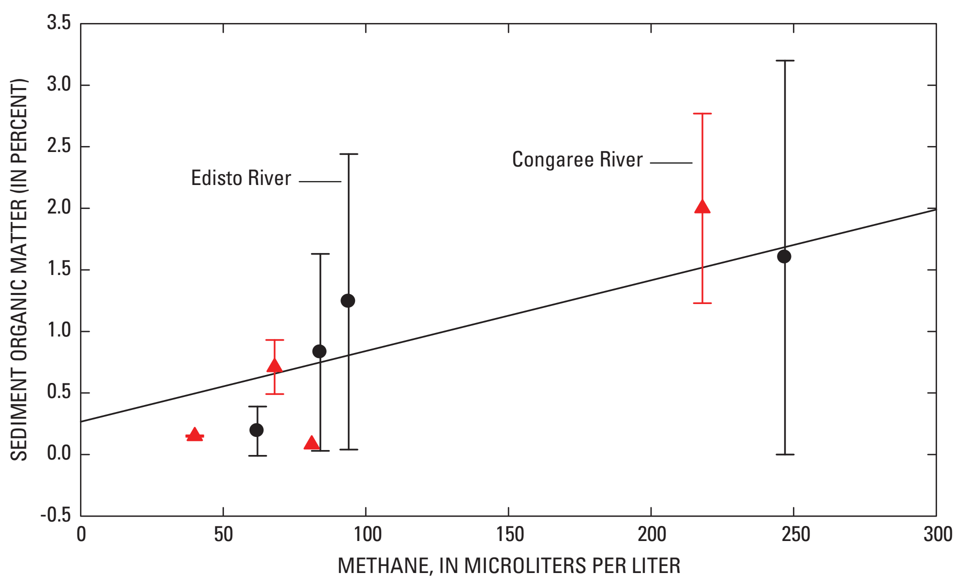


Figure 3. Concentrations of sedimentary organic matter and methane production for sediments collected in the Edisto and Congaree River basins.

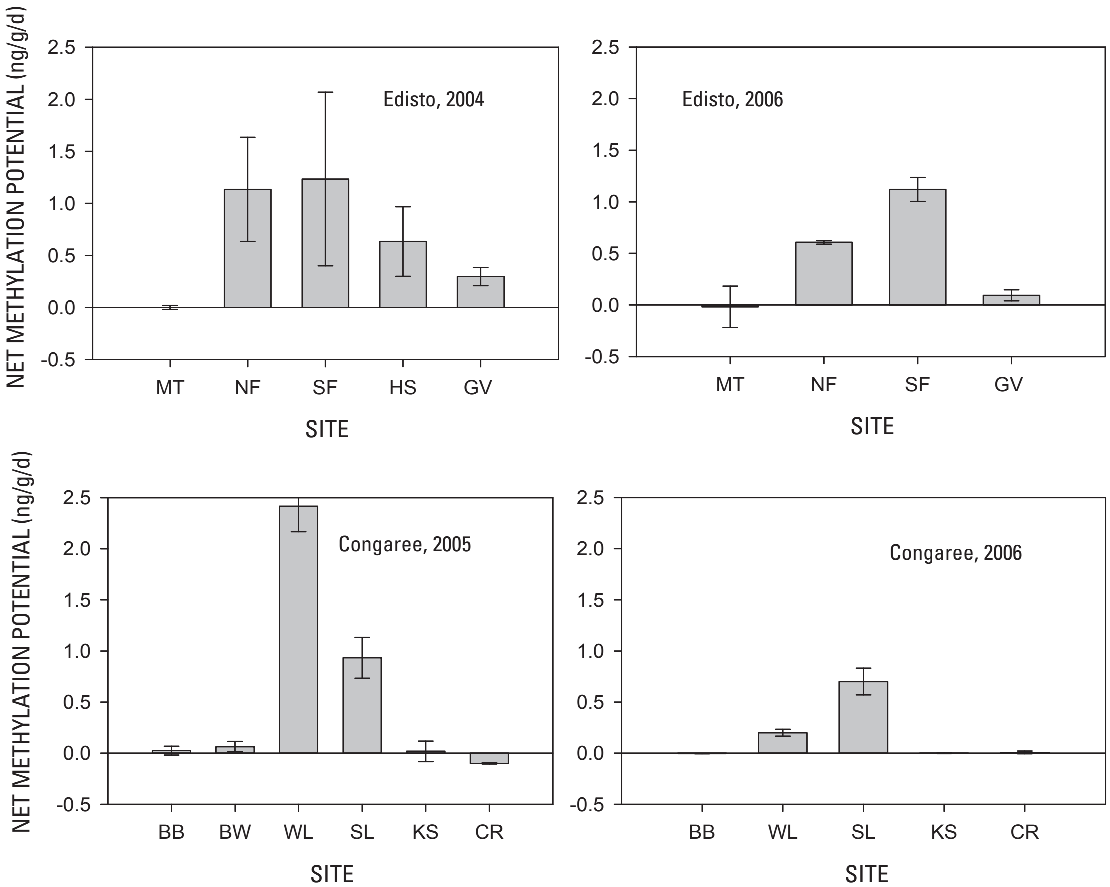


Figure 4. Measured net methylation potential of Edisto and Congaree River sediments. [ng/g/d, nanograms per gram per day]

Piedmont of the Congaree basin dilutes the concentration of MeHg relative to Hg_{tot} in the Congaree River study area as compared to the Edisto study area. Considering the significant differences in hydrology between the Edisto (dominated by local ground-water seepage from the Coastal Plain) and Congaree (dominated by runoff originating in the South Carolina Piedmont; Zalants, 1990), as well as associated differences in Hg content, sediment load, and geochemical characteristics (Patterson and Harvey, 1996; Maluk, 2000), both hypotheses are promising.

Acknowledgments

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