Flood Hydrology and Methylmercury Availability in Coastal Plain Rivers

PAUL M. BRADLEY,*
CELESTE A. JOURNEY,
FRANCIS H. CHAPELLE,
MARK A. LOWERY, AND
PAUL A. CONRADS

U.S. Geological Survey, 720 Gracern Rd, Suite 129, Columbia, South Carolina 29210-7651, United States

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Mercury (Hg) burdens in top-predator fish differ substantially between adjacent South Carolina Coastal Plain river basins with similar wetlands coverage. In the Congaree River, floodwaters frequently originate in the Blue Ridge and Piedmont regions, where wetlands coverage and surface water dissolved methylmercury (MeHg) concentrations are low. Piedmont-driven flood events can lead to downward hydraulic gradients in the Coastal Plain riparian wetland margins, inhibiting MeHg transport from wetland sediments, and decreasing MeHg availability in the Congaree River habitat. In the adjacent Edisto River basin, floodwaters originate only within Coastal Plain sediments, maintaining upward hydraulic gradients even during flood events, promoting MeHg transport to the water column, and enhancing MeHg availability in the Edisto River habitat. These results indicate that flood hydrodynamics contribute to the variability in Hg vulnerability between Coastal Plain rivers and that comprehensive regional assessment of the relationship between flood hydrodynamics and Hg risk in Coastal Plain streams is warranted.

Introduction

Methylmercury (MeHg) is the primary form of mercury (Hg) accumulated in fish (*1*) and wetlands are recognized MeHg source areas (*2*−*6*). Accordingly, the pattern of increasing fish Hg burdens from mountains to Coastal Plain in the Atlantic and Gulf Coast regions of the U.S. has been attributed to corresponding increases in wetland coverages (*7, 8*). In extended river basins, however, the spatial and temporal separation that exists between contributing source areas and downstream aquatic habitats introduces substantial hydrologic variability (*9*), alters geochemical transport efficiencies (*9*), and complicates the relationship between wetlands coverage and fish Hg burdens (*2, 10*).

The Edisto and adjacent Congaree River basins of South Carolina (SC) lie within the Coastal Plain region (*11, 12*), which closely corresponds to the Middle Atlantic Coastal Plain and Southeastern Plains Ecoregions (*13*). Hg burdens in *Micropterus salmoides* (largemouth bass) from the Edisto are consistently higher than in those from the Congaree basin or in the downstream portion of the Santee drainage. Both basins have comparable riparian wetlands coverages and a similar range of sediment Hg methylation potentials (*14*), suggesting that differences in Hg bioaccumulation between the two systems are not due to systematic differences in MeHg production in adjacent wetland/floodplain sediments.

The Edisto basin falls entirely within the Coastal Plain, whereas the Congaree basin is part of the Santee River drainage, which extends from the Atlantic Ocean to the Blue Ridge region of the Carolinas. The Edisto stems from precipitation and groundwater discharge within the Coastal Plain, whereas the Blue Ridge and Piedmont regions are important additional sources of water to the Congaree River basin. Thus, the generally lower *M. salmoides* Hg burdens in the Congaree basin may reflect hydrologic and geochemical impacts of this Blue Ridge/Piedmont contribution (*7, 8*). Consistent with this hypothesis, substantially lower *M. salmoides* Hg burdens are observed in the Blue Ridge/Piedmont-influenced Congaree main channel than in Gills Creek, a Congaree tributary lying entirely in the Coastal Plain.

During 1982−1983, the U.S. Geological Survey (USGS) collected discharge data at a short-term gage station near the downstream margin of the Congaree River (station 02169740) (*15*). A 49% increase in the annual mean discharge was observed between the most upstream Congaree gage (station 02169500, ref *15*) and gage 02169740. The long-term record at the upstream location (02169500) indicates that discharge in the Congaree River in 1982−1983 was in the normal range, falling within the 50−75th percentile range for all observations. Dissolved MeHg concentrations and the associated availability of MeHg in the aquatic habitat are expected to be low in the upper reaches of the Congaree River because of limited wetlands coverages in contributing Piedmont drainages (generally less than 2%, refs *7, 8*. However, the consistently low *M. salmoides* Hg burdens observed in the downstream portions of the basin despite the substantial Coastal Plain contribution to discharge, the 20% wetlands coverage within the basin (*16*), and the demonstrated potential for elevated Hg burdens in Congaree tributaries that fall entirely in the Coastal Plain, suggests that Blue Ridge/Piedmont-derived discharge affects MeHg transport from the wetland margins of the Congaree River to the stream habitat. Hydrologic connectivity between wetland MeHg source areas and adjacent channel habitats is an important control on Hg bioaccumulation (*2, 6, 17*). Floods maximize the area for groundwater/surface water exchange (*2, 9, 18*), but the direction of water and solute transport during flood conditions is dictated by the hydraulic gradient (*19*).

This study assessed the hypothesis that floodwater source location contributes fundamentally to the variation in Hg vulnerability between Coastal Plain streams by influencing the direction of water movement and thus the efficiency of dissolved MeHg transport between wetland source areas and the adjacent stream habitat during flood events. The impact of floodwater source location on flood hydraulic gradients was assessed with stream channel and shallow groundwater level data collected in the Edisto and Congaree River basins. A limited number of filtered MeHg samples were analyzed to illustrate the MeHg transport implications of flood hydraulic gradients.

Coastal Plain Study Sites and Conceptual Model

The Edisto and Congaree River basins have been described previously (*14*). Basin extents and associated wetland coverages are given in Table 1 and in the Supporting Information (SI). Corresponding *M. salmoides* Hg burdens are given in Table 1 and Figure 1. McTier Creek is a headwater tributary (99 km^2 total area) of the South Fork Edisto River, which

* Corresponding author phone: 803-750-6125; fax: 803-750-6181; e-mail: pbradley@usgs.gov.

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TABLE 1. Summary Statistics for Hg Burdens in *Micropterus salmoides* (Largemouth Bass) Collected by the South Carolina Department of Health and Environmental Conservation (SC DHEC) within the Congaree, Santee, and Edisto River Basins During 2001−2007[a]

		wetlands (%)		Hg in *M. salmoides* (μg/g wet weight)			
river	location(s)	basin[b]	contributing drainage[c]	median	minimum[d]	maximum	n[e]
	all	19.4	3.5	0.46	0.05	2.40	60
Congaree	Congaree - main channel	19.4	3.5	0.05	0.05	0.82	40
	Sesquicentennial State Park[f]	6.5	6.5	0.82	0.54	2.40	20
Santee[g]	all	29.5	5.3	0.18	0.05	1.70	326
Edisto	all	20.4	20.4	0.94	0.05	3.10	204

[a] *M. salmoides* Hg data provided courtesy of J. Glover, SC DHEC. Data are available for download from EPA Storet (*26*). [b] Wetland coverage for eponymous basin area only (*16*). [c] Wetlands coverage for eponymous basin area and upstream contributing drainage area (*16*). [d] Minimum reporting limit for SC DHEC data was 0.05 μg/g wet weight. [e] "*n*" is total number of samples collected. [f] Sesquicentennial State Park is located in the headwaters of Gills Creek, a Coastal Plain tributary of the Congaree River. Gills Creek wetlands coverage is 8.4% (*16*). [g] Santee River basin area, downstream of the confluence of the Congaree and Wateree Rivers.

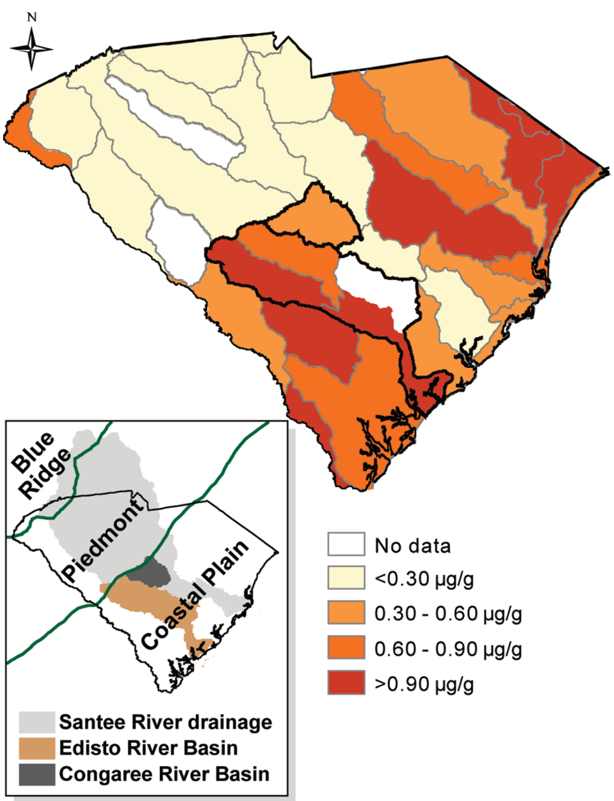

FIGURE 1. Mean *Micropterus salmoides* (largemouth bass) Hg concentrations (μg/g wet weight) in stream basins in South Carolina. Boundaries of physiographic regions are shown in green. Data are for fish collected during 2001−2007 by the South Carolina Department of Health and Environmental Conservation (SC DHEC). Congaree color designation (median = 0.46 μg/g) is based on data from the Congaree main channel (median = 0.05 μg/g) and the Gills Creek tributary (median = 0.82 μg/g). White areas indicate no data collected during 2001−2007.

contains 12% woody/emergent herbaceous wetlands (*16*). Edisto River floods occur throughout the year, primarily during the winter wet season, and are driven by rainfall within the Coastal Plain. The Congaree National Park (CNP) is a riparian-wetland-dominated (>90% by area) flood plain (*16*). Congaree River flooding occurs throughout the year, mainly during the winter wet season, and is primarily due to downstream transport of Blue Ridge/Piedmont floodwater (*20*). Previous results (*14*) demonstrated that MeHg concentrations, MeHg to total Hg ratios, and net Hg-methylation potentials were significantly higher in adjacent wetland/floodplain sediments than in the sandy stream-bed sediments that characterized both streams, indicating that riparian wetland/floodplain sediments are the primary source of MeHg in these basins.

The coarse-grained sandy sediments of the Coastal Plain exhibit efficient vertical recharge and low surface runoff (*21*, *22*), with discharge from the shallow flow system representing 72−100% of the total groundwater discharge to SC Coastal Plain streams (*21*). Under normal to low-flow conditions, the gradient and the direction of shallow groundwater flow are toward the stream channel, with active groundwater/surface water exchange limited to wetlands and channel margins (Figure 2a). Coastal Plain rainfall is expected to recharge groundwater with little to no surface runoff, maintaining the general pattern of groundwater flow toward the stream and resulting in groundwater flooding that favors advective transport of pore-water MeHg to the overlying water column (Figure 2b). Flooding of Coastal Plain reaches by Blue Ridge/Piedmont-derived floodwater may cause a reversal of the hydraulic gradient, infiltration of Blue Ridge/Piedmont floodwater and decreased transport of wetland MeHg to the stream habitat (Figure 2c).

Materials and Methods

Water Level Data Collection. Differences in groundwater/surface water exchange between Coastal Plain floods caused by Blue Ridge/Piedmont-derived floodwaters and those caused by Coastal Plain-derived floodwaters were assessed by monitoring water levels during separate flood events in the Edisto and Congaree River basins. USGS streamgage discharge data are available from the USGS National Water Information System (NWIS) Web site (*15*). Data used in this study are from the McTier Creek (station 02172305; Edisto basin), Cedar Creek (station 02169672; Congaree basin), and Congaree River (station 02169625; Congaree basin) streamgages. Groundwater levels were monitored by pressure transducers in shallow observation wells (2.5 cm diameter PVC) emplaced within the McTier Creek and Congaree River riparian floodplains. McTier Creek wells were 1.5−2.0 m deep with 0.3 m screens. ELB observation wells 1−4 were located approximately 1, 3, 21, and 45 m, respectively, from the edge of the stream (Figure 3A, inset). ALB observation wells 1and 2 were located approximately 3 and 21 m, respectively, from the edge of the stream (Figure 3B, inset). CNP wells were 4.5−8.5 m deep with 1.5 m screens and were located as shown in Figure 4 (insets).

MeHg Sampling and Analysis. Topographic depressions (transiently flooded riparian pools) in the McTier Creek floodplain fill with rising groundwater before an overland surface water connection exists with the adjacent stream

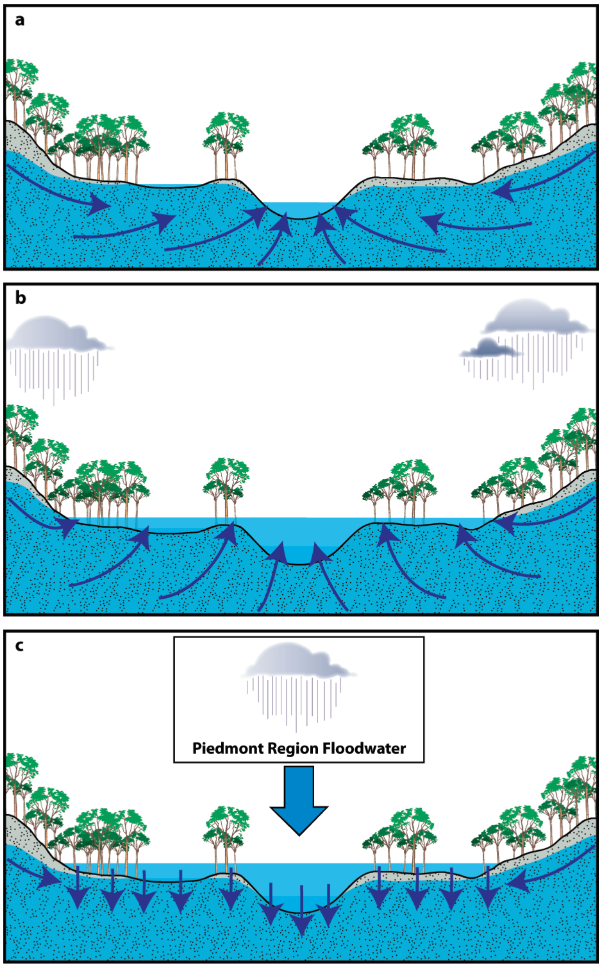

FIGURE 2. Conceptual model of groundwater/surface water hydrodynamics in Coastal Plain river basins under (a) low to normal flow conditions, (b) flood conditions driven by Coastal Plain precipitation, and (c) flood conditions caused by downstream transport of Piedmont floodwater.

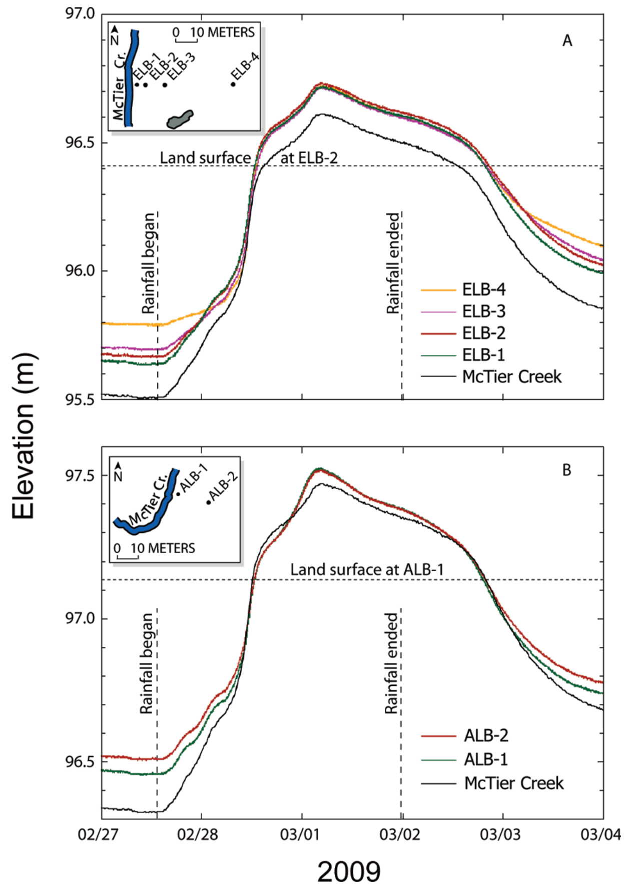

FIGURE 3. Example of water level gradient profiles observed during seasonal flood events at the (A) ELB and (B) ALB groundwater well transects in the McTier Creek sub-basin of the Edisto River basin. Locations of monitoring wells relative to McTier Creek are shown in the insets. The gray area in the ELB inset indicates the approximate extent and location of the transiently flooded riparian pool.

channel. These pools provided a means of assessing the MeHg signature of groundwater discharging through the floodplain sediment. To illustrate the MeHg transport implications of the groundwater flood mechanism, filtered MeHg (FMeHg) concentrations in a riparian pool located next to transect ELB and in the adjacent stream were assessed eight times at the McTier Creek study site during 2007−2009. Additional samples were collected without accompanying water level monitoring at locations throughout the McTier Creek basin to assess the representativeness of the FMeHg data at ELB. To illustrate the comparatively low MeHg signature associated with Blue Ridge/Piedmont-derived floodwater, FMeHg concentrations were assessed during a flood event in November 2009 at single locations in the Saluda and Broad Rivers and four locations in the Congaree River adjacent to and upstream from the CNP. In all cases, USGS ultra trace-level clean-sampling procedures (*23*) were used to collect surface water samples. Processing (*4, 23*) and analysis (*24*) of FMeHg samples were as described.

Results and Discussion

Hydrodynamic Differences in Coastal Plain-Driven and Blue Ridge/Piedmont-Driven Floods. The groundwater flood pattern depicted in Figure 2b was hypothesized to predominate in the Edisto River, because the Edisto is contained entirely in the Coastal Plain. Water level data collected from locations within the McTier Creek basin support this hypothesis. Figure 3A presents groundwater level changes in transect ELB, which is located less than 100 m upstream from the McTier Creek at New Holland streamgage (02172305) and extends inland in increasing numerical order. Prior to flooding the gradient at ELB was approximately 0.3 m from ELB-4 toward McTier Creek. Approximately 4 h after rainfall began, stream and groundwater levels began rising essentially simultaneously, indicating good hydrologic connectivity between the stream and inland groundwater locations. At the onset, peak, and end of flood conditions the groundwater gradient was upward and toward the stream, indicating discharge of groundwater from the sediment to the overlying water column at all well locations. There was no discernible groundwater gradient between well locations during flooding, but a clear gradient was observed from ELB-1 toward the stream throughout flood conditions. A similar pattern was observed at the upstream ALB transect (Figure 3B).

Because the Congaree River is a part of the Santee River drainage and frequently flooded by Blue Ridge/Piedmont-derived water, the flood pattern depicted in Figure 2c was hypothesized to occur. Water level data collected from locations within the CNP during a 2004 flood event support this hypothesis. Precipitation occurred in the SC Piedmont and upper Coastal Plain. Flooding occurred after precipitation ended as the result of downstream transport of Piedmont floodwater. Prior to flooding, the gradient between well RIC-346 and Cedar Creek at station 02169672 was generally low (Figure 4A). Groundwater levels at RIC-346 were approximately 1.5 m below land surface at the onset of flood conditions and more than 2 m below floodwater levels at the

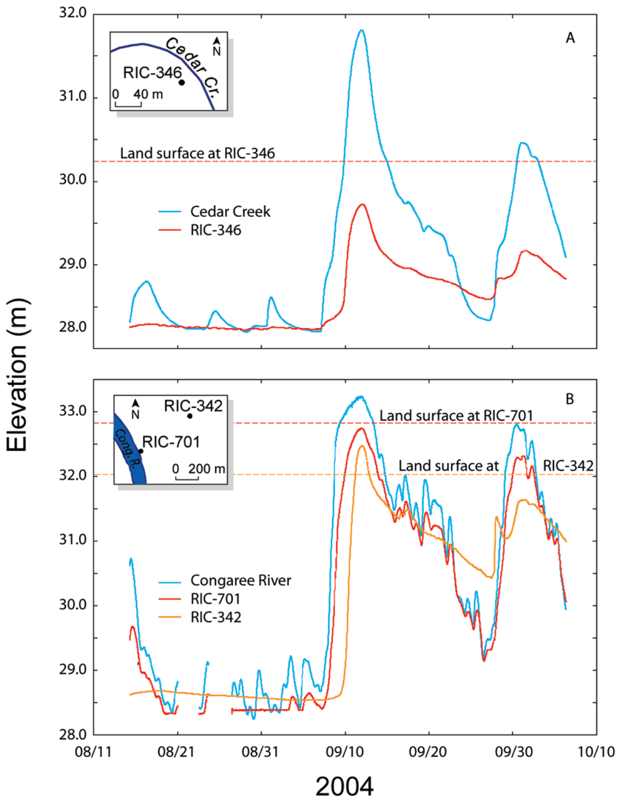

FIGURE 4. Example of water level gradient profiles observed during seasonal flood events at the (A) Cedar Creek and (B) Congaree River locations in the Congaree National Park. Locations of monitoring wells relative to Cedar Creek and the Congaree River are shown in the insets.

peak of flooding, demonstrating a downward (away from the stream) gradient throughout. The rapid rise in groundwater level following the onset of flood conditions indicated vertical infiltration of floodwaters. This pattern was repeated a few days later (Figure 4A).

The water level in the Congaree River began rising rapidly on September 7th (Figure 4B). The levee area at RIC-701 was overtopped on September 9th (Figure 4A). Water levels in RIC-701 rose rapidly as the result of lateral recharge even before flooding occurred, but still were more than 1 m below land surface at the onset of flood conditions. At peak flood, groundwater levels in RIC-701 remained below land surface and approximately 0.5 m below flood levels. Throughout the flood period the gradient remained downward (away from the stream) at RIC-701.

RIC-342 is located inland from the Congaree River and the levee deposits that line the main channel (Figure 4B). Until the river stage overtops the intervening levee system, floodwaters move slowly inland via local breaks in the levee deposits and a network of small drainage features. Immediately after the levee was overtopped on Sept 10th groundwater levels in RIC-342 rose rapidly, indicating the arrival and downward infiltration of floodwater into the shallow subsurface. At peak flood, groundwater levels in RIC-342 remained 0.5 m below flood levels. Throughout the flood period the gradient remained downward (away from the stream) at RIC-342. The hydraulic gradient changed back toward the stream by September 24th, well after flood conditions had ended.

Implications for MeHg Transport in Edisto and Congaree Systems. The hydrologic pattern observed in the McTier Creek sub-basin of the Edisto River system is consistent with continuous discharge of groundwater from the shallow subsurface toward the stream channel habitat even during flood conditions. This type of response favors transport of MeHg from the subsurface source area to the stream channel habitat. To illustrate, a limited number of FMeHg samples were collected from a shallow depression in the McTier Creek floodplain near the ELB water level monitoring transect. These floodplain depressions are inundated by rising groundwater before flooding creates an overland connection to the stream and provide an opportunity to assess MeHg in discharging groundwater prior to the mixing with stream-channel surface water that occurs during flood conditions. This floodplain depression was assessed eight times during 2007–2009 (Figure 5A). On the five occasions that standing water was present, no overland connection to McTier Creek existed and the antecedent conditions of the surrounding floodplain varied from dry (November 2007, March 2008, April 2008), flood recession 1 day prior to sampling (June 2007), and flood recession 3 days prior to sampling (July 2009). In every instance the FMeHg concentrations observed in the pool were greater than were measured in the adjacent stream channel, with concentrations ranging from approximately two times higher in June 2007 to greater than 10 times higher in July 2009. The pattern of discharging groundwater and statistically greater FMeHg concentrations in floodplain locations was observed throughout the McTier Creek basin in 2007–2009 (Figure 5B).

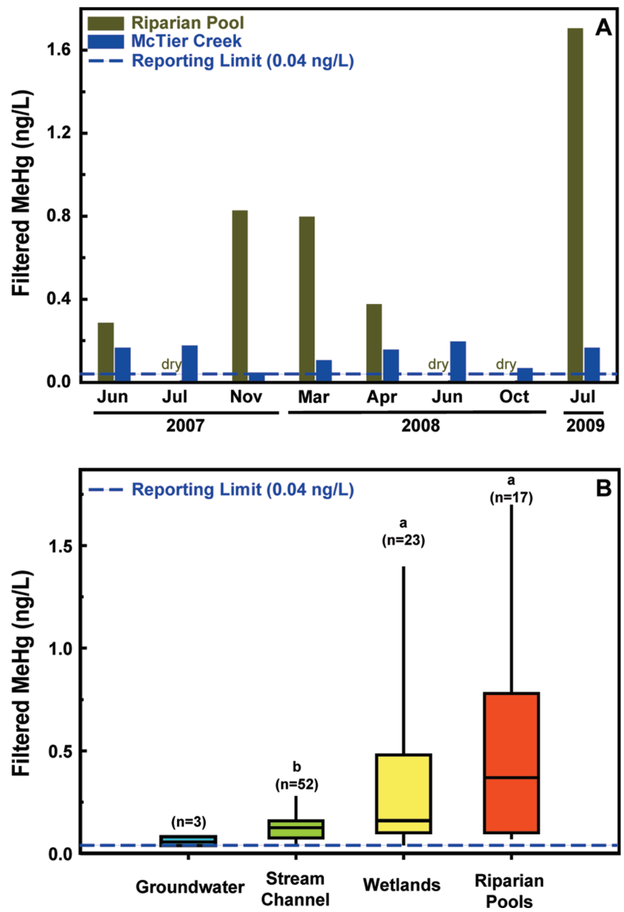

FIGURE 5. Filtered MeHg concentrations (ng/L) observed in the McTier Creek study area during 2007–2009. (A) FMeHg in a transient riparian pool near ELB and in the adjacent McTier Creek channel. "Dry" indicates no standing water. (B) FMeHg in stream channel and in wetland/riparian pool samples throughout the McTier Creek study area. Box indicates the 25–75% quartile range. Center line indicates median. Whiskers indicate the data range. "*n*" is the number of samples. Different letters indicate statistically significant differences ($p < 0.0001$; Kruskal–Wallis and Tukey's HSD).

The November 2007 sample at ELB provides particular insight into the MeHg signature associated with groundwater discharging through the floodplain land surface (Figure 5A). A localized rainfall event the day before sample collection resulted in rising groundwater and streamwater levels. Discharging groundwater partially filled the previously dry depression but did not overtop the floodplain. Dissolved oxygen concentrations, measured in the pool during MeHg sample collection in 2007−2009, ranged from 2 to 5 mg/L, indicating that redox conditions did not favor Hg methylation in the standing water. Thus, because the pool and the surrounding floodplain were dry less than 12 h prior to sample collection, the November 2007 sample reflects the immediate transport of MeHg into the surface water compartment. These results demonstrate that the groundwater flood mechanism can efficiently transfer floodplain MeHg to the surface water compartment.

In contrast, the hydrologic characteristics of the Congaree basin favor low FMeHg concentrations and, consequently, reduced availability for biotic uptake and accumulation in the main channel aquatic habitat. The FMeHg concentrations observed in the Congaree flood in November 2009 were low, consistent with the comparatively low wetlands coverage of the upstream Blue Ridge/Piedmont drainages (*16*) and indicating lower MeHg availability within the Congaree River channel habitat.

Implications for MeHg Bioaccumulation in Coastal Plain States. The results of this study support the hypothesis that characteristically coarse-grained sediments of the SC Coastal Plain favor efficient exchange of water between streams and shallow groundwater systems. This characteristic creates a potential for efficient transport of MeHg from wetland/floodplain source areas to the stream habitat and, consequently, an inherent vulnerability to Hg bioaccumulation in Coastal Plain streams. In this setting, the direction of water movement across the sediment/water interface during high flow periods, in which hydrologic connectivity between the stream and the wetland/floodplain sediments is maximized, appears to be a critical control on MeHg exchange between saturated-sediment source areas and adjacent surface water habitats and, ultimately, on the potential for Hg bioaccumulation. The results of this study indicate that flood events attributable to precipitation within the SC Coastal Plain are driven by groundwater discharge, promoting transfer of MeHg from wetland porewater to the stream habitat. In contrast, Coastal Plain floods caused by external flood events deliver water with low dissolved MeHg concentrations and reverse the direction of groundwater/surface water exchange, inhibiting MeHg transport from the wetland porewater to the stream and reducing MeHg availability in the stream habitat.

This hydrologic context suggests that stream systems, like the Edisto River, are particularly vulnerable to Hg bioaccumulation, because they lie entirely or largely within the Coastal Plain and are primarily subject to groundwater flooding. In contrast, Coastal Plain stream reaches (like the Congaree River) which are subject to both groundwater flood and external floodwater events, are expected to exhibit reduced MeHg availability, depending on the relative frequency of the two mechanisms. Flood events in the Congaree River main channel are predominately due to Blue Ridge/Piedmont floodwaters that exhibit low dissolved MeHg concentrations and lead to low mean Hg burdens in *M. salmoides* in the Congaree and downstream Santee River, consistent with our conceptual model.

Because the Coastal Plain region extends along the Atlantic and Gulf Coasts of the United States from New Jersey to Texas, the results of this limited study may have regional-scale implications for Hg bioaccumulation. The fundamental hydrologic characteristics of the SC Coastal Plain are common in the Coastal Plain region and a similar relationship between flood hydrodynamics and Hg bioaccumulation is expected throughout. In a recent USGS national survey of Hg burdens in high trophic level piscivores (*25*), Scudder et al. reported an elevated incidence of high top predator fish Hg burdens in stream reaches along the Atlantic and Gulf Coasts that corresponded closely to the Coastal Plain region. This observation supports the conclusion that flood hydrodynamics contribute to an increased vulnerability to Hg bioaccumulation in Coastal Plain rivers and indicates that a more comprehensive regional assessment of the relationship between flood hydrodynamics and Hg risk in Coastal Plain streams is warranted.

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Supporting Information Available

Basin maps, FMeHg sampling/analysis details and results, and a conceptual model of McTier Creek groundwater/surface water hydrodynamics. This material is available free of charge via the Internet at http://pubs.acs.org.

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