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Tracking suspended particle transport via radium isotopes (^{226}Ra and ^{228}Ra) through the Apalachicola–Chattahoochee–Flint River system

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A B S T R A C T

Suspended particles in rivers can carry metals, nutrients, and pollutants downstream which can become bioactive in estuaries and coastal marine waters. In river systems with multiple sources of both suspended particles and contamination sources, it is important to assess the hydrologic conditions under which contaminated particles can be delivered to downstream ecosystems. The Apalachicola–Chattahoochee–Flint (ACF) River system in the southeastern United States represents an ideal system to study these hydrologic impacts on particle transport through a heavily-impacted river (the Chattahoochee River) and one much less impacted by anthropogenic activities (the Flint River). We demonstrate here the utility of natural radioisotopes as tracers of suspended particles through the ACF system, where particles contaminated with arsenic (As) and antimony (Sb) have been shown to be contributed from coal-fired power plants along the Chattahoochee River, and have elevated concentrations in the surficial sediments of the Apalachicola Bay Delta. Radium isotopes (^{228}Ra and ^{226}Ra) on suspended particles should vary throughout the different geologic provinces of this river system, allowing differentiation of the relative contributions of the Chattahoochee and Flint Rivers to the suspended load delivered to Lake Seminole, the Apalachicola River, and ultimately to Apalachicola Bay. We also use various geochemical proxies (^{40}K, organic carbon, and calcium) to assess the relative composition of suspended particles (lithogenic, organic, and carbonate fractions, respectively) under a range of hydrologic conditions. During low (base) flow conditions, the Flint River contributed 70% of the suspended particle load to both the Apalachicola River and the bay, whereas the Chattahoochee River became the dominant source during higher discharge, contributing 80% of the suspended load to the Apalachicola River and 62% of the particles entering the estuary. Neither of these hydrologic scenarios, which were moderately low flow regimes, appeared to transport particles contaminated with arsenic and antimony to Apalachicola Bay.

1. Introduction

Rivers contribute the largest flux of freshwater to the coastal ocean. Consequently, considerable attention has been given to the associated constituent input to estuaries and the coastal zone (Hodell et al., 1990; Hope et al., 1994; Kaul and Froelich, 1984; Martin and Meybeck, 1979; Milliman and Meade, 1983; Milliman and Syvitski, 1992). Most heavy metals, phosphorus, pesticides, and many organic compounds are particle-reactive and tend to be transported down-river with particles because of the high surface area available for adsorption (Bonniwell et al., 1999; Horowitz, 1995; Viers et al., 2009). Some of these components are known to desorb upon entering an estuary due to ion exchange processes and thus become more bioactive (Froelich, 1988).

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Two contrasting end-member paradigms have been proposed to explain downstream transport of suspended particles: (1) particles are transported the entire length of a river during a single high discharge event (Cahill et al., 1974; Jobson and Carey, 1989; Partheniades, 1972); or (2) fluvial particles are transported in a series of steps between suspended transport and deposition, often traveling between 150 and 600 m/day and requiring time scales of months to years to transit an entire river length (Verhoff et al., 1979). In order to better understand these mechanisms, a system-wide assessment of the temporal variations in suspended particle behavior is needed.

We focus on the applicability of naturally-occurring radioisotopes ^{226}Ra ($T_{1/2}$: 1600 years), ^{228}Ra ($T_{1/2}$: 5.7 years), and ^{40}K ($T_{1/2}$: 1.25×10^9 years) to examine suspended particle composition and transport behaviors. In freshwater systems, these tracers may be embedded in particle mineral structures, adsorbed onto active surfaces of the particles, or both. However, upon entering high ionic strength waters in an estuary, a fraction of the adsorbed radium may be displaced via ion exchange processes (Martin and Akber, 1999; Nozaki et al., 2001; Peterson et al., 2008). Radium isotopes (^{226}Ra and ^{228}Ra) are derived naturally by decay of primordial radionuclides ^{238}U and ^{232}Th, respectively. The parent isotopes are inherently present in all continental rocks and soils, but their relative activities vary depending on rock type. Sedimentary geologic materials originating from marine deposition (e.g., sandstone and carbonates) contain less ^{238}U and daughters compared to igneous minerals of volcanic and metamorphic origin (Hansen, 1975). A river system transiting different geologic provinces should support different levels of radium isotopes that could serve as conservative tracers within freshwater portions of a river system to track and fingerprint suspended particles.

The Apalachicola–Chattahoochee–Flint (ACF) river system in the southeastern United States (Fig. 1) is a large river basin, heavily influenced by man-made reservoirs, and ideal for studying suspended particle transport. Our initial hypothesis was that the different lithologic and anthropogenic sources within the three rivers of the ACF system might impart chemically-distinct signatures to the suspended particles in each river. These different chemical signatures should then permit quantitative assessment of each river's contribution to the total particle flux to Apalachicola Bay under different flow regimes. Our goal was to establish a set of distinct particle tracers that remain chemically inert during transit and provide unique signatures for the three rivers of the ACF system to ultimately identify the sources of contaminants carried on suspended particles to the estuary. Our approach should be generally applicable to other large river systems.

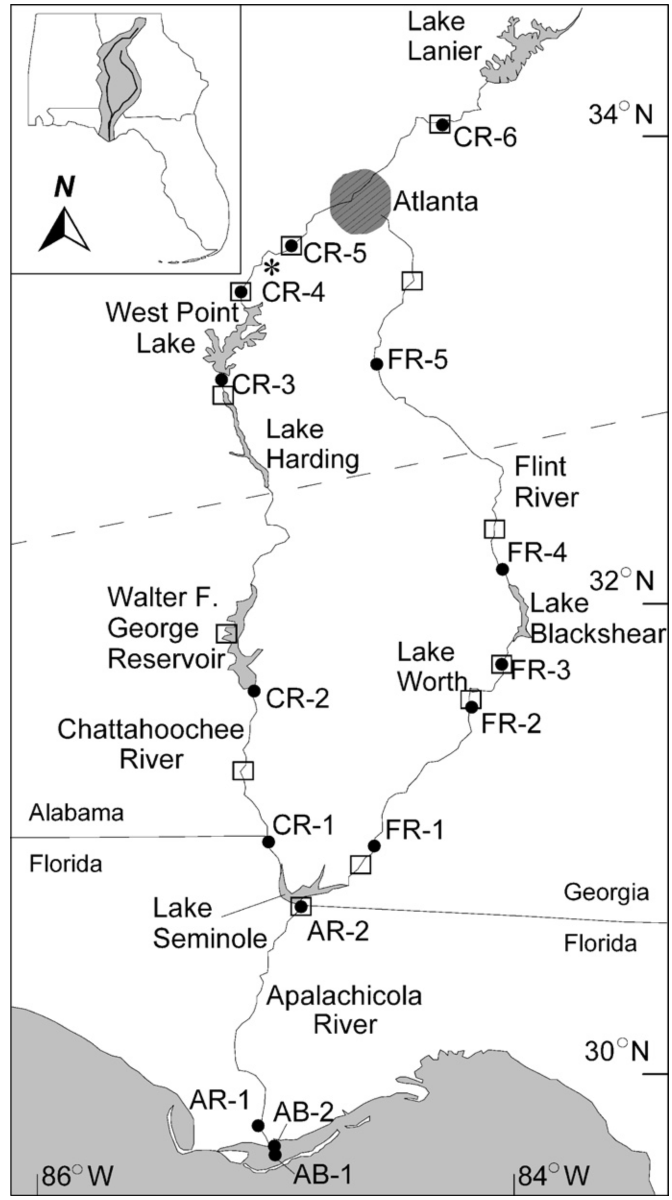

Fig. 1. Map of the Apalachicola–Chattahoochee–Flint River basin through Georgia, Alabama, and Florida (insert). Each sample site is shown (black dots and alphanumeric designations). Major reservoirs (all man-made since 1948) along the river system are highlighted. The fall line where the Piedmont geologic unit meets the coastal plain province is designated by the dashed line. The location of the two CFPPs between CR-4 and CR-5 is indicated by an asterisk. Open squares represent USGS gauging stations (see Table 1).

2. Regional setting

The ACF river basin (Fig. 1) drains 58,000 km^2 of mostly agricultural lands in western and northern Georgia, eastern Alabama, and the Florida Panhandle (Bedosky, 1987). The Chattahoochee and Flint Rivers flow into Lake Seminole at the intersection of the Florida–Georgia–Alabama state lines, from which the Apalachicola River flows southward across the Florida panhandle to the Gulf of Mexico. The Apalachicola River discharges into Apalachicola Bay, a large shallow estuary in northwest Florida separated from the Gulf of Mexico by several barrier islands (Kofoed and Gorsline, 1963). The Chattahoochee and Flint Rivers contribute more than 95% of the water flow to Lake Seminole and about 80% of the water flow to the Apalachicola River (Elder et al., 1988). The dominant source of freshwater to the bay is the Apalachicola River.

The recent 10-year discharges for the Apalachicola River average 640 m^3/s (1988–1998) and 495 m^3/s (1998–2008), with the latter decade influenced by a prolonged drought in the southeastern United States. During high discharge periods, the Chattahoochee River dominates the water flux to the Apalachicola River, whereas the Flint River contributes a greater portion during low discharge due to perennial freshwater springs along its course (Couch et al., 1996; Elder et al., 1988).

The ACF system flows through several geologic provinces. The northern section (above the 'fall line'; see Fig. 1) is composed of the Piedmont and Blue Ridge provinces dominated by crystalline igneous and metamorphic rocks associated with the Appalachian terrain. Below the fall line, the drainage basin is composed primarily of Cretaceous to Recent marine sands and clays in the coastal plain province (Bedosky, 1987). This fall line division is about 425 km upstream from Apalachicola Bay on the Chattahoochee River and about 550 km upstream on the Flint River.