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Tracing historical trends of Hg in the Mississippi River using Hg concentrations and Hg isotopic compositions in a lake sediment core, Lake Whittington, Mississippi, USA

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ABSTRACT

Concentrations and isotopic compositions of mercury (Hg) in a sediment core collected from Lake Whittington, an oxbow lake on the Lower Mississippi River, were used to evaluate historical sources of Hg in the Mississippi River basin. Sediment Hg concentrations in the Lake Whittington core have a large 10–15 y peak centered on the 1960s, with a maximum enrichment factor relative to Hg in the core of 4.8 in 1966. The Hg concentration profile indicates a different Hg source history than seen in most historical reconstructions of Hg loading. The timing of the peak is consistent with large releases of Hg from Oak Ridge National Laboratory (ORNL), primarily in the late 1950s and 1960s. Mercury was used in a lithium isotope separation process by ORNL and an estimated 128 Mg (megagrams) of Hg was discharged to a local stream that flows into the Tennessee River and, eventually, the Mississippi River. Mass balance analyses of Hg concentrations and isotopic compositions in the Lake Whittington core fit a binary mixing model with a Hg-rich upstream source contributing about 70% of the Hg to Lake Whittington at the height of the Hg peak in 1966. This upstream Hg source is isotopically similar to Hg isotope compositions of stream sediment collected downstream near ORNL. It is estimated that about one-half of the Hg released from the ORNL potentially reached the Lower Mississippi River basin in the 1960s, suggesting considerable downstream transport of Hg. It is also possible that upstream urban and industrial sources contributed some proportion of Hg to Lake Whittington in the 1960s and 1970s.

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1. Introduction

The Mississippi River is the third largest river in the world and drains about 41% of the continental United States (Milliman and Meade, 1983). Upstream urban, industrial, and agricultural activities potentially affect water and sediment quality in the Mississippi River and its tributaries. Previous research has reported the presence of various trace element contaminants in the Mississippi River basin (Trefry et al., 1985; Meade, 1995; Horowitz, 2010). Some studies have evaluated trace contaminant histories for the Mississippi River using sediment cores from the Mississippi Delta (Trefry et al., 1985; Santschi et al., 2001; Overton et al., 2004), and a few studies have evaluated Hg in sediment and water collected from the Mississippi River (Beauvais et al., 1995; Anderson and Perry, 1999). Furthermore, a recent study evaluated trace element and organic compound trends in a 242-cm long sediment core collected from Lake Whittington, an oxbow lake on the Lower Mississippi River, which has accumulated Mississippi River sediment since the late 1930s (Van Metre and Horowitz, 2012).

Contamination of Hg to land, air, and water ecosystems has been studied for many years (NAS, 1978; USEPA, 1997; Ullrich et al., 2001). Of the contaminants mentioned in the Clean Air Act of 1972, the U.S. Environmental Protection Agency (USEPA) indicated that Hg has the greatest potential to affect human health (USEPA, 1997). Contamination of Hg has been identified in various lakes, reservoirs, and other waterways worldwide (Hines et al., 2000; Gray et al., 2002; Horvat et al., 2002; Gray and Hines, 2006; Rimondi et al., 2012). In addition, anthropogenic sources of Hg from urban, industrial, and agricultural runoff that potentially affect the Mississippi River have been known for many years (USEPA, 1972; Meade, 1995; Lyons et al., 2006). One Hg source in the Mississippi River basin that has not been directly identified is the Oak Ridge National Laboratory (ORNL) in Tennessee (Fig. 1). Large amounts of elemental Hg were used at ORNL in the 1950s and early 1960s and an estimated 128 (±35) Mg of elemental Hg was discharged downstream from the ORNL primarily in the late 1950s (Brooks and Southworth, 2011). Most of the releases to streams resulted when elemental Hg was washed with nitric acid to prepare it

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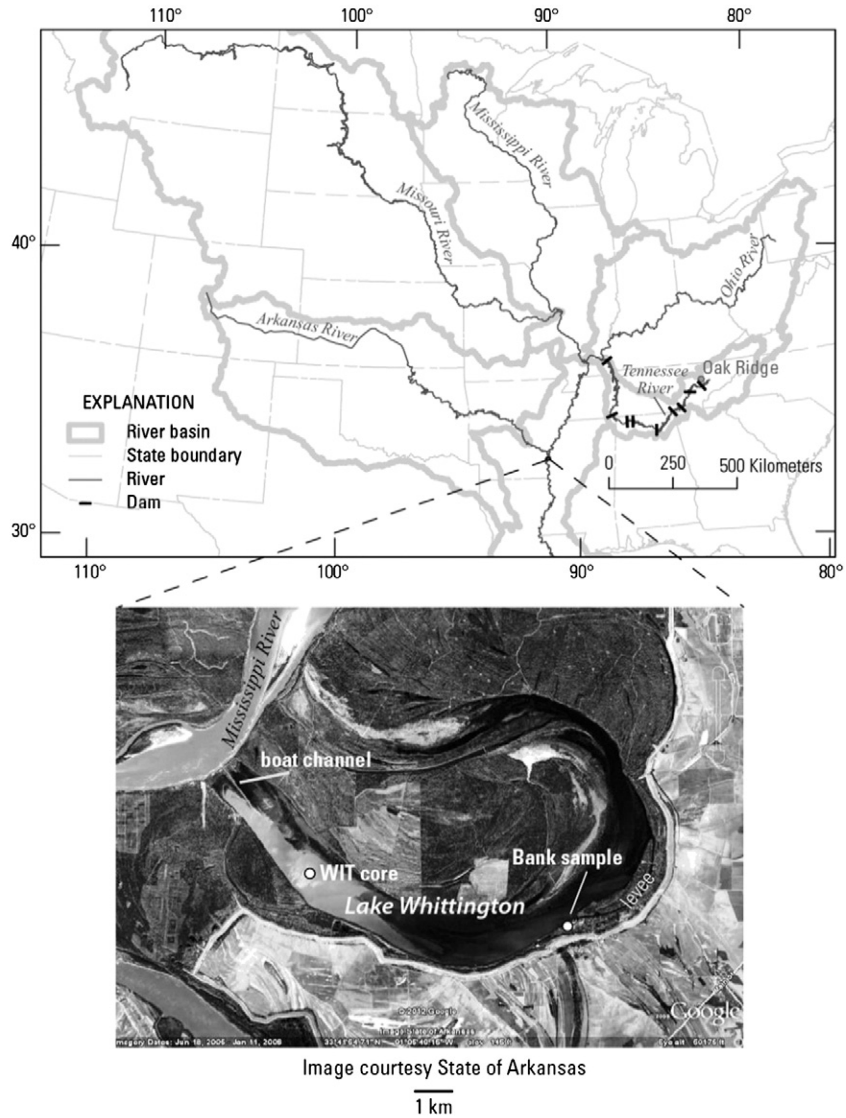

Fig. 1. Location of the study area. The original inset topographic image was from the State of Arkansas that was downloaded from Google Earth.

for use in a process to enrich ^{6}Li, which was used in thermonuclear weapons production (Brooks and Southworth, 2011; Donovan et al., 2014). During this nitric acid washing process, Hg was likely released in rinse water as elemental Hg, dissolved Hg, and other Hg compounds, which flowed into local waterways and eventually farther downstream. Two recent studies using Hg isotopes linked present-day sediment Hg in local and regional waterways to an ORNL source (Bartov et al., 2013; Donovan et al., 2014), although these studies did not evaluate Hg contamination in the Mississippi River.

A few studies have attempted to evaluate historical long-term deposition of Hg in the Upper Mississippi River basin using sediment cores (Balogh et al., 1999; Blumentritt et al., 2013), but no studies have attempted to evaluate long-term deposition of Hg or Hg isotopic compositions in the Lower Mississippi River, which includes inputs from the Ohio River basin, where ORNL is located. Therefore, the objective of this study was to evaluate historical Hg loading to the Lower Mississippi River by determining Hg concentrations and Hg isotopic compositions in a sediment core collected from Lake Whittington. Concentrations of Hg and Hg isotopes were also measured in stream sediment collected downstream from the ORNL to evaluate potential Hg contamination in this area and to characterize this potential source to the Lower Mississippi River.

2. Study area

Lake Whittington was a meander of the Mississippi River until 1937 when the U.S. Army Corps of Engineers dredged a new channel through the neck of the meander to reroute the river bypassing a long oxbow to facilitate commercial traffic; the resulting oxbow lake is Lake Whittington (Bragg, 1977). A boat channel has been maintained between the river and downstream end of Lake Whittington allowing water and sediment inflow into the lake (Fig. 1), and typical spring high flow in the river likely inundates the floodplain and lake depositing additional sediment (Van Metre and Horowitz, 2012). The Arkansas River joins the Mississippi about 10 km upstream from Lake Whittington (Fig. 1). The upstream end of the nearly 30-km long Lake Whittington filled with sediment over time and is now mostly forested wetland. The downstream end of the lake is open water and separated from the Mississippi River by a low natural levee; a boat channel through the levee connects the lake to the Mississippi River. The lake floods periodically, and a plume of suspended sediment extending from the river through the boat channel into the lake is visible in aerial photographs (Fig. 1). The dominant land use in the Mississippi River basin is agriculture. Lake Whittington is isolated from the surrounding agricultural lands by levees, and the predominant source