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Fate of geothermal mercury from Yellowstone National Park in the Madison and Missouri Rivers, USA

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HIGHLIGHTS

- About 7.0 kg/y of geothermal Hg exits Yellowstone National Park via the Madison River.
- 87% of that Hg load lost to sedimentation and volatilization in first downstream lake
- Hg concentrations in water, sediment, and trout are high upstream from lake.
- Hg concentrations in water, sediment, and fish are not elevated below lake.
- Bioaccumulation of Hg in fish correlated with total Hg in water and sediment.

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ABSTRACT

Mercury is a worldwide contaminant derived from natural and anthropogenic sources. River systems play a key role in the transport and fate of Hg because they drain widespread areas affected by aerial Hg deposition, transport Hg away from point sources, and are sites of Hg biogeochemical cycling and bioaccumulation. The Madison and Missouri Rivers provide a natural laboratory for studying the fate and transport of Hg contributed by geothermal discharge in Yellowstone National Park and from the atmosphere for a large drainage basin in Montana and Wyoming, United States of America (USA). Assessing Hg in these rivers also is important because they support fishery-based recreation and irrigated agriculture. During 2002 to 2006, Hg concentrations were measured in water, sediment, and fish from the main stem, 7 tributaries, and 6 lakes. Using these data, the geothermal Hg load to the Madison River and overall fate of Hg along 378 km of the Missouri River system were assessed. Geothermal Hg was the primary source of elevated total Hg concentrations in unfiltered water (6.2–31.2 ng/L), sediment (148–1100 ng/g), and brown and rainbow trout (0.12–1.23 μg total Hg/g wet weight skinless filet) upstream from Hebgen Lake (the uppermost impoundment). Approximately 7.0 kg/y of geothermal Hg was discharged from the park via the Madison River, and an estimated 87% of that load was lost to sedimentation in and volatilization from Hebgen Lake. Consequently, Hg concentrations in water, sediment, and fish from main-stem sites downstream from Hebgen Lake were not elevated and were comparable to concentrations reported for other areas affected solely by atmospheric Hg deposition. Some Hg was sequestered in sediment in the downstream lakes. Bioaccumulation of Hg in fish along the river system was strongly correlated ($r^2 = 0.76–0.86$) with unfiltered total and methyl Hg concentrations in water and total Hg in sediment.

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1. Introduction

The Madison and Missouri Rivers (Fig. 1) are two of the best known trout-fishing streams in the Rocky Mountains yet lie downstream from Hg-rich geothermal springs in Yellowstone National Park (YNP). Mercury is a worldwide contaminant that is derived from natural and anthropogenic sources and is toxic at very low concentrations (Mason et al., 1994; National Research Council, 2000; Fitzgerald and Lamborg, 2007). Understanding the transport and fate of Hg in rivers has important implications for the health of humans, aquatic species, and piscivorous birds and mammals (Ward et al., 2010).

Large river systems play a key role in the transport and fate of Hg because they drain areas affected by wet and dry Hg deposition and provide downstream transport for Hg from point sources. Most detailed studies of downstream transport and fate of Hg have examined rivers affected by industrial or mining activities; the primary Hg source in these rivers was contaminated channel and floodplain sediments that

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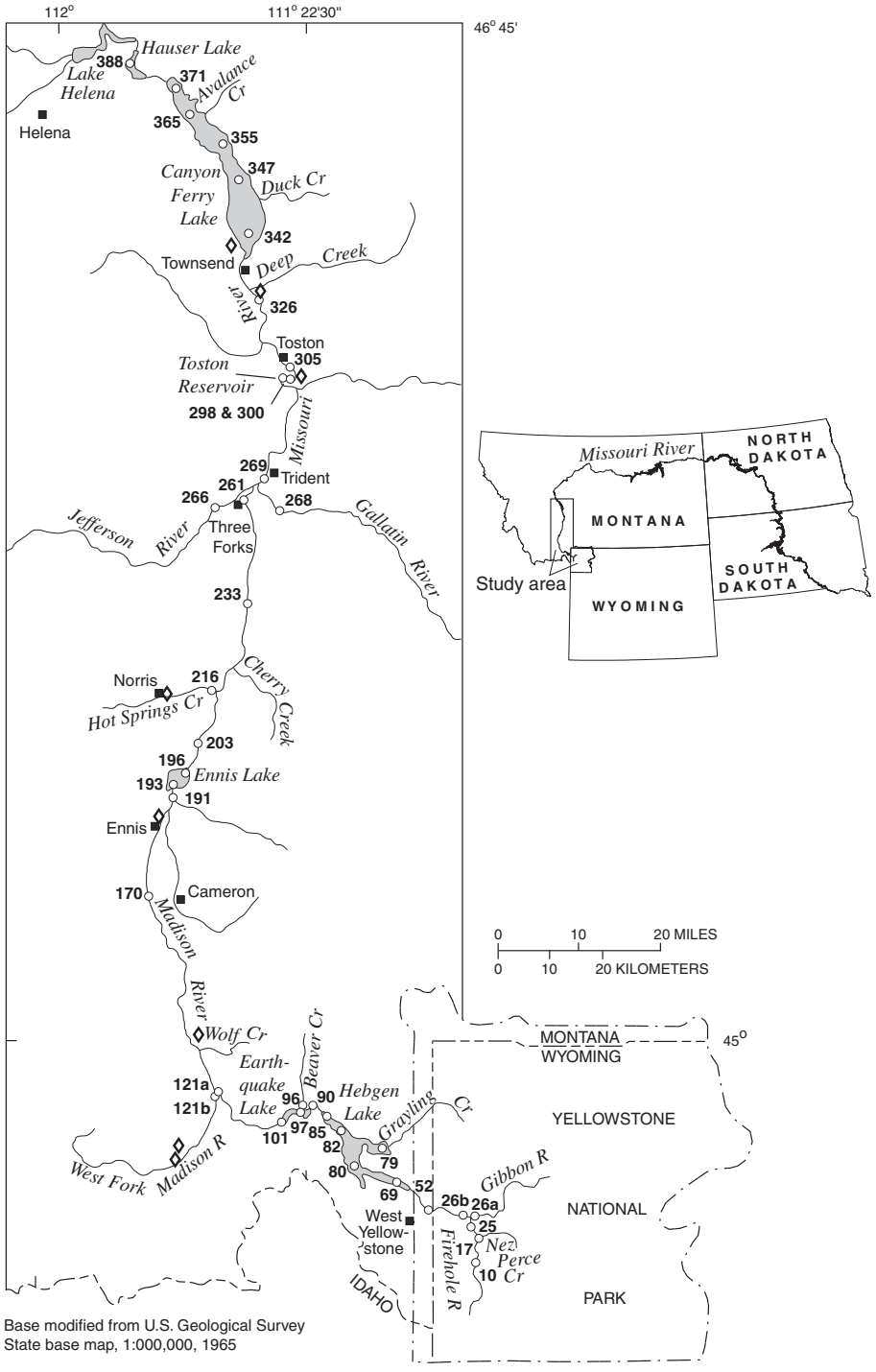

Fig. 1. Location of sampling sites (open circles) and geothermal springs (Sonderegger and Bergantino, 1981) outside of Yellowstone National Park (open diamonds).

were mobilized during high flow (Bonzongo et al., 2006; Domagalski, 1998, 2001; Flanders et al., 2010; Hines et al., 2000; Miller and Lechler, 2003; Ullrich et al., 2007). Other Hg studies have examined rivers in watersheds affected primarily by atmospheric deposition (Leitch et al., 2007) as well as rivers contaminated by waste water and other point sources (Jackson, 1986; Quémerais et al., 1999; Smith et al., 2010). However, mercury contamination and bioaccumulation can be a concern in any river because of atmospheric deposition (Peterson et al., 2007; Seigneur et al., 2004; Ward et al., 2010).

River systems can be the site of active Hg biogeochemical cycling, bioaccumulation, and storage. Factors affecting Hg bioaccumulation in lakes have been well studied and include the presence of forest cover (which can increase local aerial deposition), dissolved organic carbon (which is important for binding with and transporting Hg), and wetlands (which enhance methylation) (Brigham et al., 2009; Chasar et al., 2009; Ward et al., 2010). However, information on Hg contamination and bioaccumulation in river ecosystems is not as abundant as data for lake systems (Smith et al., 2010; Ward et al., 2010). Rivers may be a conduit for Hg from sources and methylation sites in upstream reaches, and perhaps more importantly, in the upstream watershed (Brigham et al., 2009). The primary factor that controls Hg bioaccumulation in fish in rivers is the Hg concentration in