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Use of Stable Mercury Isotopes to Assess Mercury and Methylmercury Transformation and Transport across Critical Interfaces from the Molecular to the Watershed Scale

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Historical and ongoing releases of mercury (Hg) have resulted in a legacy of Hg contamination in streambed sediment, streambanks, and floodplain soils downstream of the Y-12 National Security Complex (Y12), along the flow path of East Fork Poplar Creek (EFPC) near Oak Ridge, Tennessee. Much of the Hg associated with streambed sediments, streambanks, and floodplain soils resides in relatively insoluble fractions, and has thus been considered to have little impact on dissolved total Hg (THg) concentrations. However, recent studies comparing hydrologic discharge and THg flux from Y12 and Lower EFPC suggest that additional dissolved Hg from the hyporheic pore water or groundwater discharge may variably contribute as much as a third of downstream dissolved Hg loads during baseflow conditions. Thus, the over-arching goal of this project is to use natural Hg stable isotope signatures, imparted by molecular-scale reactions, to gain a more comprehensive quantitative and mechanistic understanding of the processes that supply dissolved Hg to surface water, drive observations of watershed-scale Hg fluxes, and result in the bioaccumulation of methylmercury (MeHg).

To achieve this goal, we are combining an intensive multi-seasonal field study with mechanistic laboratory experiments. First, we are coupling the Hg isotopic composition of dissolved Hg in stream water and in critical subsurface ecosystem compartments (i.e., hyporheic zone, riparian floodplains, and groundwater) with hydrologic flux measurements in four gauged reaches of EFPC. This will enable us to establish an isotope mass balance that assesses the relative importance of dissolved Hg contributed to the stream across these critical interfaces.

Second, we are utilizing sequential extraction methods to characterize the isotopic composition of legacy Hg potentially re-mobilized from streambed sediment. This will provide insight into the sources and mechanisms that replenish the supply of dissolved Hg within critical subsurface zones. Third, we are assessing the isotopic composition of MeHg in biota of EFPC, as a step toward identifying the source(s) of bioaccumulative MeHg in the EFPC ecosystem.

Here, we present: (1) Hg concentration and isotopic composition of all surface water, hyporheic pore water, and riparian groundwater samples analyzed to date; (2) isotopic mass balance assessments regarding legacy inputs of dissolved Hg to the stream water of EFPC; (3) sequential extraction results showing potential re-mobilization of recalcitrant legacy Hg from streambed sediment; and (4) measurements of the isotopic composition of MeHg accumulating in biota of EFPC.