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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, stream banks, 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, stream banks, 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 the Lower EFPC suggest that additional dissolved Hg from either the hyporheic zone, riparian wetlands, or groundwater discharge may variably contribute as much as 16-80% of downstream dissolved Hg loads during base flow conditions. Thus, one of the over-arching goals 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 and drive observations of watershed-scale mercury fluxes. To achieve this goal, we are coupling the Hg isotopic composition of dissolved Hg in stream water and in critical subsurface ecosystem compartments (i.e., hyporheic zone, riparian wetlands, 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.

During the first six months of this project we have: (1) initiated monthly base flow surface water sampling to characterize changes in the concentration and isotopic composition of dissolved Hg in each of four gauged reaches of the EFPC; (2) installed infrastructure (semi-permanent piezometers) for sampling hyporheic pore water; and (3) collected the first seasonal sampling of hyporheic pore water and riparian groundwater from five locations along the flow path of the Lower EFPC. Here, we present dissolved Hg concentration and Hg isotopic composition of all surface water, hyporheic pore water, and riparian groundwater samples analyzed to date, and begin to make mass balance assessments regarding legacy inputs of dissolved Hg to the stream water of EFPC.