Poster #21-31

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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BER Program: SBR
Project: University Award

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, 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 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.

During the first half of this project we have: (1) completed more than a full year of ~ monthly baseflow surface water sampling to characterize the seasonal variability in concentration, flux, and isotopic composition of dissolved Hg in each of four gauged reaches of EFPC; (2) installed infrastructure (semi-permanent piezometers) for sampling hyporheic pore water and secured access to groundwater sampling wells in four reaches of Lower EFPC; (3) collected three seasonal sets of high-volume hyporheic pore water and riparian groundwater samples from these four newly instrumented reaches along the flow path; and (4) developed sequential extraction methods for the isotopic analysis of legacy mercury potentially re- mobilized from streambed sediment. Here, we present: dissolved Hg concentration and Hg isotopic composition of all surface water, hyporheic pore water, and riparian groundwater samples analyzed to date; make mass balance assessments regarding legacy inputs of dissolved Hg to the stream water of EFPC; and provide an assessment of our sequential extraction method for the isotopic analysis of legacy mercury sources.