Title: **Using Mercury Stable Isotopes to Identify Sources of Methylmercury to Aquatic Food Webs.**

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Project Abstract:

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. Elevated concentrations of inorganic mercury within this ecosystem have been shown to be associated with elevated concentrations of methylmercury (MeHg) in biota, including benthic invertebrates and fish. However, determining the specific ecosystem source(s) of MeHg that are being contributed to the food web can be challenging. Mercury stable isotope analysis is a powerful tool that allows us to trace biogeochemical pathways and track the transport and fate of sources through complex ecosystems. During previous studies, we have compared measurements of the total mercury isotopic composition of fish in EFPC and the Hinds Creek reference site, and have attempted to use linear regression techniques that utilize the varying percentage of MeHg content (i.e., %MeHg) among organisms to estimate the isotopic composition of MeHg sources within the ecosystem. While this indirect approach succeeded within the reference site, linear regression could not be utilized to estimate the isotopic composition of MeHg within the point-source-contaminated EFPC. Thus, the final objective of our broader ongoing proposed research was to develop a method to directly quantify the MeHg isotopic composition within EFPC biota, which are natural accumulators of MeHg being contributed to the EFPC ecosystem. Here, we successfully developed a novel method for the extraction, isolation, and direct determination of the compound-specific mercury isotopic signature of MeHg in biota. Application of this new method showed that: (i) the isotopic composition of MeHg accumulating in biota was distinct in the upper and lower reaches of EFPC, as well as differing from the reference site; (ii) the isotopic composition of MeHg within the reference site was clearly influenced by precipitation sources, while MeHg accumulating within EFPC biota was not; and (iii) the photochemical processing of MeHg sources within point-source-contaminated watersheds (i.e., EFPC) may differ from those influenced primarily by atmospheric deposition. Overall, these new methods that allow for direct measurement of compound-specific MeHg isotope composition will likely continue to lead to new insights that would otherwise be masked by the assessment of total mercury isotopic composition alone.