# Mercury stable isotope tracing of multiple Mercury sources in the Tennessee River system

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The U.S. Dept. of Energy Y-12 plant near Oak Ridge, TN has input Hg into the East Fork of Poplar Creek (EFPC), which drains into the Clinch/Tennessee River system. On December 22, 2008, a major ash spill at the Tennessee Valley Authority's (TVA) Kingston coal-burning power plant introduced an additional source of Hg into the system. Other sources of Hg include atmospheric deposition and natural background. Variations in Hg stable isotope abundances may enable us to elucidate the sources and fate of Hg as it moves through the system.

Sediment samples from the EFPC, the Clinch River upstream from the Kingston power plant, the Tennessee river system downstream from Kingston, and the Emory River upstream from the plant (nominally uncontaminated), were analyzed using MC-ICP-MS with a double-spike approach. The $^{202}Hg/^{198}Hg$ ratio was measured with a precision of ~0.1‰ and expressed as $\delta^{202}Hg$ values relative to NIST SRM-3133. Odd isotope ($^{199}Hg$ and $^{201}Hg$) anomalies were measured but were not significant.

Preliminary results suggest distinct $\delta^{202}Hg$ 'signatures' influencing the system. Average $\delta^{202}Hg$ values for the Clinch River, Kingston fly-ash, and the Emory River are -0.47‰ ±0.20 (n=3), -1.97‰ ±0.46 (n=4), and -1.04‰ ±0.20 (n=3), respectively. The data suggest the upstream portion of the Clinch River is dominated by a source of Hg with near-zero $\delta^{202}Hg$. This source may be the Y-12 plant, which has near-zero $\delta^{202}Hg$. Downstream of Kingston, sediment $\delta^{202}Hg$ shifts to lower values (-1.41‰± 0.39, n=2) more negative than those of the nominally uncontaminated river sediments and closer to those of the Kingston fly-ash. This suggests Hg from the Kingston spill is evident in the river sediments, though none of them were highly contaminated.

At present, we use the word 'signature' with caution. To move forward in this study, more research is needed to characterize the Hg sources and consider the geochemical transformations that might alter Hg isotope ratios in sediments over time.

# Extraction and reliability of Pb isotopes derived from Fe-Mn bearing phases in deep marine sediments

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Pb isotopes extracted from Fe-Mn oxides in bulk deep sea sediment have been used to study variations in bottom water isotopes over the Holocene and late Pleistocene as they relate to deep ocean circulation patterns and provenance of weathering inputs. The goal of this study is to test a sequential extraction protocol on longer time scales to enable us to study these processes during climatically important time intervals throughout the Cenozoic.

Here we present Pb isotopes, rare earth elements (REE), and major element data from the carbonate, Fe-Mn oxide and residual fractions of ten deep sea samples ranging from the early Eocene to mid-Miocene from Ocean Drilling Program (ODP) Site 1090 in the Cape Basin. Fossil fish teeth were also tested as an archive for seawater Pb isotopes.

Agreement between Pb isotopes recovered from the carbonate and Fe-Mn oxides fractions indicates both archives record the same bottom water isotopic signal. Pb isotopic values for the residual fraction are generally distinct from carbonate and oxide values, indicating minimal detrital contamination in the chemical extractions. In contrast, $^{206}Pb/^{204}Pb$ values from contemporaneous fish teeth tend to be more radiogenic than the oxide and carbonate fractions. The presence of Pb in the mineral structure of bio-apatite, along with radiogenic Pb ingrowth, and open system behavior make fossil fish teeth an unsuitable archive for high resolution Pb isotope studies.

Normalized REE patterns for oxide fractions record a range of Ce anomalies, but other REEs behave more systematically with a prominent middle REE bulge commonly observed in the oxide fraction. The detrital fraction produced characteristic flat REE patterns. Major element data show a linear relationship between Mn and Pb contents in both carbonate and oxide fractions, suggesting that a Mn-bearing phase in addition to Fe-Mn oxides may host seawater Pb isotopes in the sediment.