

## Utilization of mass-dependent and -independent fractionation of Mercury isotopes to understand Mercury cycling and bioaccumulation in aquatic systems

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Mercury is a highly toxic pollutant that has broad societal and scientific significance. Currently the global budget, biogeochemical cycling, and ultimate fate of Hg in the environment are not well understood and new approaches are needed to quantify Hg sources and transformations. Recently we demonstrated experimentally and in natural samples that Hg stable isotopes display two types of isotopic fractionation: 1) mass dependent fractionation (MDF) and 2) mass independent fractionation (MIF) of only the odd isotopes [1].

Natural samples from aquatic ecosystems preserve both MDF and MIF of Hg isotopes. Our initial study focused on fish samples collected from three locations in Lake Michigan [1]. Similar to other isotopic systems used to study food web dynamics and structure (i.e., C and N), the MDF of Hg in fish appears to be related to size and age. The MDF recorded in fish should reflect both the sources of Hg to the fish and the excretion of Hg by the fish. Thus, MDF alone could provide new insights into sources and bioaccumulation of Hg in food webs. In addition, Lake Michigan fish also display MIF signatures. Based on experiments, we have demonstrated that variations in MIF are related to the photochemical cycling of Hg in aquatic ecosystems.

If MDF and MIF in ecosystems can be understood, signatures in fish will likely inform us about the sources and processes transforming Hg and why there are differences in the bioaccumulation of Hg in different ecosystems and populations of fish. This requires sampling of a variety of ecosystems, and the sampling of many ecosystem components. We have expanded our study beyond Lake Michigan to include several smaller lakes. Fish from different lakes display distinct MIF and different relationships between MDF and MIF. The degree of MIF is likely related to the amount of photo-reduction and dissolved organic carbon content. We also sampled food sources and livers of fish to understand processes of isotopic fractionation in the food web.

[1] Bergquist & Blum (2007) *Science* **318**, 417-420.

## <sup>210</sup>Pb-<sup>226</sup>Ra disequilibria: Mantle melting or gas fractionation?

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Disequilibria between nuclides of the U-decay-series have been used to date volcanic eruptions. However, where eruption dates are known, existing disequilibria at the time of eruption can also be used to infer the time since fractionation of the respective isotope pairs by magmatic processes. Most elements, particularly those of the longer lived nuclides are very incompatible and difficult to fractionate magmatically. In many systems, disequilibria require dynamic melting models which rely on the unique ability of these isotopes to be constantly produced by decay. A significant database of (<sup>230</sup>Th/<sup>238</sup>U), (<sup>231</sup>Pa/<sup>235</sup>U), (<sup>226</sup>Ra/<sup>230</sup>Th) data has been amassed, decreasing the timescale of magma formation and ascent further with each isotope pair. Most recently, (<sup>210</sup>Pb/<sup>226</sup>Ra) disequilibria in MORB have been suggested to result from mantle melting [1], requiring magma to move from source to surface in less than 100 years. However, disequilibria between <sup>226</sup>Ra and <sup>210</sup>Pb can also be produced by fractionation of <sup>226</sup>Ra and intermediate isotope <sup>222</sup>Rn during volatile exsolution and transport [2].

A review of published and unpublished (<sup>210</sup>Pb/<sup>226</sup>Ra) data from magmas of different tectonic settings reveals distinct differences; while the global distribution of (<sup>210</sup>Pb/<sup>226</sup>Ra) is normally distributed around unity, magmas from plume and ridge environments have almost exclusively <sup>210</sup>Pb deficits and magmas from arc settings have both <sup>210</sup>Pb deficits and excesses. Furthermore, plume settings have a normal distribution with a median value less than unity (and a single peak of data in unity, due to decay), while arc magmas have a lognormal distribution towards <sup>210</sup>Pb excess with a median value of unity. We interpret this and additional arguments in favour of a young (<sup>210</sup>Pb/<sup>226</sup>Ra) mantle signature in plume environments, which, though potentially also present in arcs, is overprinted by gas-melt fractionation of Ra-Rn. For ocean island basalts to carry such a signature, source to surface transport of magma occurs on a timescale of decades.

[1] Rubin K.H., van der Zander I, Smith M.C. & Bergmanis E.C. (2005) *Nature* **437**, 534-538 [2] Gauthier PJ & Condomines M (1999) *Earth Planet. Sci. Lett.* **172**, 111-126.