Mass-independent fractionation of even mercury isotopes: An enigma

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Preliminary studies have demonstrated both mass-dependent fractionation (MDF) and mass-independent fractionation (MIF) of Hg isotopes in natural samples. To date, more than 100 papers have been published on Hg isotope ratios, which demonstrated the potential of Hg isotopes in tracing the source, processes and the fate of Hg in the atmosphere, biosphere, lithosphere, and hydrosphere. A few special processes such as photochemical reduction of Hg(II) and photodegradation of methylmercury can produce mass-independent fractionation (MIF) of odd Hg isotopes (odd-MIF), which had been largely reported in variable natural samples and laboratory experiments, and was thought to be caused by either nuclear volume effect or magnetic isotope effect. Moreover, recent work reported, unexpectively, intriguing MIF of even Hg isotopes (even-MIF, Δ^{200}Hg up to 1.24‰) in natural samples mainly related to the atmosphere, rendering Hg a “three dimentional” isotope tracing system.

Here, we try to give a tentative review of publications on even Hg isotope anomalies, with a main focus on sample strategies and possible processes and mechanisms triggering even-MIF. Given the fact that Δ^{200}Hg was determined in variable regions with different altitude and latitude in China and in North America, the occurrence of even-MIF is likely a worldwide phenomenon. This is also supported by the positive Δ^{200}Hg (0.22‰) just determined in the tree moss in Sweden. Though Δ^{200}Hg is actually used to refer to the deviation of even Hg isotopes from MDF, other even isotopes are probably subject to the same fractionation. The relationships amongst even isotope anomalies thus need to be fully elucidated. Δ^{200}Hg values were mainly determined in samples related to the atmosphere, implying an upper atmosphere origin of even-MIF. If our conceptual model can hold, even-MIF may serve as a useful indicator of upper atmosphere chemistry. The implication of even-MIF as a possible conservative tracer remains to be largely developed. In fact, ^{200}Hg anomaly is likely related to solar irradiation, air mass move and stratosphere incursion, thus even-MIF could provide additional information about atmospheric chemistry, meteorological condition and even related climate changes. Moreover, the conservative behavior of ^{200}Hg anomaly may also be helpful for better understanding the global biogeochemical cycle of Hg, especially the surface-atmosphere exchange.