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Application of stable mercury isotope fractionation as a new tool to trace contamination sources in the environment

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Analytical advances in the past decade and the recent advent of multicollector inductively coupled plasma mass spectrometry (MC-ICP/MS) have made it possible to measure fractionation of non-traditional elements, where mercury is still one of the more uncommon elements to study. Mercury has seven natural isotopes (^{196}Hg, ^{198}Hg, ^{199}Hg, ^{200}Hg, ^{201}Hg, ^{202}Hg and ^{204}Hg) spanning the mass range from 196 to 204. Because all seven isotopes are stable, any variation measured result from natural fractionation processes.

In our group, we have developed a method for accurate and precise determination of Hg isotope ratios in a wide range of environmental samples. An on-line Hg cold-vapor technique, using stannous chloride as reductant, was coupled to a Thermo-Finnigan *Neptune* MC-ICP/MS. All ratios, ^{198}Hg/^{202}Hg, ^{199}Hg/^{202}Hg, ^{200}Hg/^{202}Hg and ^{201}Hg/^{202}Hg, were corrected for instrumental mass discrimination by simultaneously monitoring ^{205}Tl/^{203}Tl of a standard solution. Finally, variations of the isotope composition of Hg were determined relatively to a standard (Mercury Standard Solutions - SRM 3133 or SRM1641d) using the standard-sample bracketing approach.

This technique was employed to measure isotope ratios in a variety of samples from different locations worldwide. The samples consisted of anthropogenically contaminated sediments, certified reference materials and well characterized sediment cores. All measurements showed mass dependent isotope fractionation and demonstrated the ability of the proposed method to detect significant differences in mercury isotope ratios among samples. Results obtained to date unequivocally demonstrated not only that

Hg isotopes ratios do vary in nature but also that, as a tool, the method allows tracking sources of Hg contamination as well as identifying natural fractionation processes.