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Measurement of Hg isotopic ratios on molecular level using GC-MC-ICP-MS hyphenation

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Isotopic fractionation in the environment can happen due to the following processes: equilibrium chemical processes, non-equilibrium chemical processes, physical processes and biological processes. The study of isotopic signature in aquatic environment is one of the most important tools for the understanding global and local environmental processes on the earth. Main pathways of the Hg migration (Hg biogeochemical cycle) in the environment can be described as followed: (i) deposition from the atmosphere, (ii) methylation in the water column, (iii) reduction to Hg^0; (iv) accumulation in bottom sediments; (v) bioaccumulation in the food chain; and (vi) diffusion and re-suspension of the Hg species. Stable Hg isotopes have been shown to significantly fractionate during these natural processes. The combination of the isotopic and the speciation information opens a new direction to study the fractionation processes at the molecular level. The chemistry of Hg in the environment is highly complex. Therefore, the isotope fractionation measurement can be used as one of the tool to better understand Hg biogeochemical cycling.

Multiple collector – inductively coupled plasma – mass spectrometry (MC-ICP-MS) is a new technique for the measurement of isotopic composition of a wide range of elements at high precision and has a great potential to planetary, earth, ocean, and environmental sciences. This method combines the outstanding ionization efficiency of the ICP source and the use of simultaneous multiple collection to achieve very precise

isotopic measurements for many elements. Hyphenation of gas-chromatography or liquid chromatography with MC-ICP-MS can be used as a powerful tool for the measurement of isotopic fractionation within the different elemental species or in other words at the molecular level.

In this work we have demonstrated the possibility of the measurement of Hg isotopes in molecular species of fish and hair samples. It is known that three main Hg forms in environment are Hg(II), MeHg and Hg^0. All these forms can be separated using GC, and hyphenation of GC with MC-ICP-MS allows the measurement of Hg isotopic ratios. Thallium is used for the mass-bias correction using Russell correlation. NIST-3133 and MeHg STREM standards are recommended to be used as delta-0 reference material to calculate isotopic fractionation for Hg(II) and MeHg species, respectively:

$$\delta^{xxx}Hg = \left(\frac{\left(\frac{^{xxx}Hg}{^{198}Hg}\right)_{sample}}{\left(\frac{^{xxx}Hg}{^{198}Hg}\right)_{SRM3133}} - 1 \right) \times 1000 \text{ per mil}$$

For the first time, this hyphenation was applied to the analysis of real-world samples. After optimization, this coupling offers a precise and accurate method for the measurement of mercury isotopic fractionation in MeHg and Hg(II) species. Also, a necessary calculation approach was developed and a measurement strategy is demonstrated. The developed technique was validated versus two other analytical techniques for the measurement of Hg isotopic ratios, i.e. conventional nebulization (CN) of liquid solution and cold vapor (CV) generation of Hg^0 vapor. The results show mass-dependent and mass-independent fractionation in fish and hair samples.