Mass-independent fractionation of mercury stable isotopes in deep-sea hydrothermal systems

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It has been recognized that mercury (Hg) isotope analysis is an important new tool for identifying Hg source and tracking Hg transformations in the environment. Mass-dependent (MDF) and -independent (MIF) fractionations of Hg isotopes are caused by a wide variety of biogeochemical processes including redox reactions and volatilization. Volcanic activities and its associated hydrothermal activities are the main sources of naturally-emitted Hg in the environment. Several previous studies suggested that the naturally-emitted Hg indicated both MDF and MIF. It was thought that the MDF was caused by a process of liquid-vapor partitioning during ascending and the MIF was caused by photoreduction. It was, however, suggested that both photoreduction and volatilization could cause the MIF, and it has never been distinguish from each other in the hydrothermal systems. In this study, geological samples in active deep-sea hydrothermal systems at Izu-Bonin arc were collected and measured their Hg isotopic compositions. They indicate both MDF and MIF. The $\delta^{202}$Hg values range from -1.0 to 0.5 $\permil$, indicating the liquid-vapor partitioning, whereas the calculated $\Delta^{199}$Hg values are mostly between 0.1 and 0.2 $\permil$, indicating the MIF in the deep-sea hydrothermal systems without photoreduction. The linear relationship between $\Delta^{199}$Hg and $\Delta^{201}$Hg, ranging from 1.6 and 2.0, also indicates the nuclear volume effect. This result suggest that MDF and MIF of Hg isotopic compositions can be utilized to distinguish naturally-emitted Hg from anthropogenic Hg.

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