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Germanium isotopic variations in seafloor hydrothermal systems and sedimentary rocks

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Germanium (Ge) is a trace metalloid in seawater that is of particular interest in marine biogeochemistry because of its strong chemical similarity to silicon. In addition, Ge stable isotope ratios in biogenic silica may have strong potential as paleoceanography proxies. We recently reported Ge-isotope composition of igneous rocks and deep-sea clays and defined a bulk Earth δ74/70Ge value of 0.6‰ (relative to NIST3120a). Based on δ74Ge values of modern marine sponges, we also suggested that Ge-isotope composition of seawater is enriched in heavy isotopes relative to the crust (δ74Ge around 2.2‰.

Here, we report new measurements of Ge isotope composition in hydrothermal fluids and deposits (i.e. Fe oxyhydroxide and sulfides) as well as metalliferous deposits in coal and precambrian iron formations. The samples were analyzed using a continuous flow hydride generation system coupled to a MC-ICP-MS (Thermo-Finnigan Neptune) operating at PSO (Pôle Spectrometrie Océan, Brest). The instrumental mass bias was corrected by either the "double-spike" method or the "standard-sample bracketing" method.

Germanium isotope and Ge/Si ratio systematics were investigated in low temperature hydrothermal vents from Loihi Seamount (Hawaii, USA) and results were compared to high-temperature vents from East Pacific Rise (EPR) at 9-10°N. The study of low temperature hydrothermal fluids at Loihi suggest that both Ge/Si and δ74Ge are fractionated from the host basaltic rock. We obtained a maximum Ge isotope fractionation between Fe-oxyhydroxide and dissolved Ge in the fluid of about -2.7‰ while at EPR, very light Ge isotope signatures of hydrothermal sulfide deposits were obtained with a maximum fraction factor of -6.3‰ between sphalerite and fluid. Both results are consistent with recent experimental and theoretical calculations. Ge isotopes of coal samples and their combustion products were also analyzed in order to investigate the potential use of Ge isotopes as tracers of Ge sources and enrichment mechanisms in coal-hosted Ge deposits. We found that Ge-rich lignite samples have large Ge isotopic fractionation (δ74Ge values range from -2.59‰ to 4.72‰ which suggest that organic matter plays an important role in the transport and precipitation of Ge and its isotope fractionation.

Our results overall suggest that hydrothermal fluids and coal deposits may represent a source of isotopically heavy Ge in the ocean and that this isotope signature may be affected by Ge precipitation in Fe-rich authigenic sink. Additional studies will focus on Ge-isotope systematics in rivers and deep seawater in order to establish a reliable isotope mass balance of Ge in the ocean.