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Hg isotope fractionation during abiotic reduction by the hydroxysulfate green rust 2

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Hg isotope fractionation during biotic reduction using bacterial consortium has recently been conducted and showed a mass dependent Rayleigh fractionation behavior with $\alpha_{202/198}$ ranging from 1.0013 and 1.0020 between oxidized Hg(II) and Hg° produced (Kritee et al., 2007). On the other hand, Fe(II) in hydroxysulfate green rust 2 $(\mathrm{GR2(SO_4^{2-})})$ is also a strong reducing agent for various metals including mercury. (O'Loughlin et al., 2003).

We investigated Hg isotope fractionation during abiotic reduction by $\mathrm{GR2(SO_4^{2-})}$ $(\mathrm{Fe_4^{II}Fe_2^{III}(OH)_{12}SO_4{\bullet}8H_2O})$ synthesized according to Ruby et al. (2005). 20 μg.g^{-1} to 20 ng.g^{-1} Hg(II) (NIST 3133) was added under anoxic conditions at room temperature to different concentrations of $\mathrm{GR2(SO_4^{2-})}$. An aliquot of mercury remaining in the liquid phase was taken up frequently over a period of sixty minutes after shaking or ultrasonic agitation. Hg content remaining in solution is continuously decreasing suggesting an effective reduction. The kinetic of the reduction seems to occur in two steps, as an important decrease of Hg content in solution after two minutes was observed, Hg° produced being out gassed, followed by a smoother and more random decrease during the following minutes.

The isotopic compositions of Hg remaining in solution was measured using cold vapor generation coupled to MC-ICP-MS. Residual Hg(II) was enriched in heavy isotopes suggesting that Hg isotopes were fractionated during abiotic reduction. Results may be modeled by a mass dependent Rayleigh fractionation evolution system with a fractionation factor $\alpha_{202/198}$ ranging from 1.0003 to 1.0010 between Hg(II) and Hg$^{\circ}$ This range of alpha values is certainly a low estimation because liquid mercury droplets on the oxidation products of the green rust solid were observed by TEM, leaving the possibility that Hg$^{\circ}$ was not completely degassed before sampling the remaining Hg(II) in solution with GR2(SO_4^{2-}).

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