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Antimony isotopes variations during stibnite dissolution and secondary mineral precipitation: an experimental study

LALI CARRIÈRE[1], ELÉONORE RESONGLES[2], PIERRE LE PAPE[3], DR. AREEJ ADRA, PHD[3], RÉMI FREYDIER[2], OLEG POKROVSKI[4], MRS. SOPHIE AYRAULT[5] AND CORINNE CASIOT[2]

[1]HydroSciences Montpellier, Université de Montpellier, CNRS
[2]HydroSciences Montpellier, Université de Montpellier, CNRS, IRD
[3]Institut de Minéralogie, de Physique des Matériaux et de Cosmochimie (IMPMC), UMR 7590 Sorbonne Université-CNRS-MNHN-IRD
[4]Géosciences Environnement Toulouse (GET) – Research Institute for Development [IRD]: Toulouse University, CNRS, Toulouse, France
[5]Laboratoire des Sciences du Climat et de l'Environnement, LSCE (CEA-CNRS-UVSQ)

Antimony (Sb), an emerging contaminant mainly emitted from non-exhaust traffic emissions in urban environments, is notably present as reduced Sb(III)-S species in underwater sediments of roadside retention pond, where it can be remobilized into the aqueous phase under certain conditions. Sizable Sb isotopic fractionation has been observed at the sediment/aqueous interface in these environments, but the mechanisms driving these changes remain unclear.

This study aims to assess kinetic and equilibrium Sb isotope fractionation during dissolution of crystalline stibnite (Sb_2S_3) under oxic and anoxic conditions, in the presence of different ligands that can affect mineral dissolution rates and Sb speciation. The isotopic composition of dissolved Sb ($\delta^{123}Sb$) was measured by HG-MC-ICP-MS. The temporal evolution of solid phases and Sb local environment were investigated by XRD, SEM-EDXS and XANES at the Sb *K*-edge.

In oxic dissolution experiments performed under undersaturated stibnite conditions (14 mg/L), the $\delta^{123}Sb$ composition of dissolved Sb matched that of the initial stibnite, irrespective of the dissolution stage or physico-chemical conditions such as pH, temperature and ligands (Cl, citrate). This suggests that stibnite dissolution does not generate kinetic isotopic fractionation under the experimental conditions. In oxic dissolution experiment performed under supersaturated stibnite conditions (2 g/L), three stages were observed : (1) dissolution of stibnite with no fractionation of Sb isotopes between the dissolved Sb and the initial stibnite, (2) dissolution of stibnite accompanied by secondary mineral precipitation, reflecting a transient kinetic isotopic fractionation with a maximum apparent fractionation factor ($\Delta^{123}Sb_{aqueous\text{-}solid}$) of +0.44 ± 0.06 ‰, showing the preferential precipitation of the light Sb isotope, and (3) equilibrium between dissolved Sb and newly formed precipitates, with a progressive decrease of the apparent fractionation factor between the aqueous and the solid phases down to 0 ‰, reflecting isotopic exchange.

Under anoxic conditions supersaturated with stibnite and excess Na_2S, the dissolution occurred without secondary mineral precipitation or oxidation of dissolved Sb(III). The absence of measurable isotopic fractionation confirms that the dissolution does not fractionate Sb isotopes without concomitant processes. This study provides quantitative constrains to model Sb isotope variations in stibnite-rich contaminated environments, with implications for tracing Sb sources and processes.