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Mobility of antimony in samples from diverse environmental compartments of Sb mines in Spain: leaching experiments in oxidizing and reducing conditions

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Antimony (Sb) is a valuable element, exploited for diverse applications including flame retardants, munitions, batteries, glasses, and industry for diodes. However Sb is also a toxic metalloid, often associated with other harmful elements (arsenic, lead, mercury...) in mining sites. The biogeochemical behavior of Sb remains poorly documented, and data must be acquired in order to elaborate solid environmental studies related with Sb mining. Here, the mobility of Sb from solid phases present in former mining sites was assessed through leaching experiments performed in oxidizing or reducing conditions. Five Sb mines located in South-Central Spain were considered: La Nazarena, Accesos, Balanzona, Pilar, and Susana mines. Rock samples were analysed by X-ray diffraction, confirming that the main Sb carrier was stibnite (Sb_2S_3), present in all mines. Surface soils and mine wastes were sampled, together with sediments when ponds or galleries were present. The total Sb concentrations of 18 samples varied from 28 to 221 000 mg kg^{-1} . However, stibnite was only detected in a soil sample from Balanzona mine and tetrahedrite ($(\text{Cu,Fe})_{12}\text{Sb}_4\text{S}_{13}$) in a sediment from the Balanzona mine. The most common Sb secondary minerals were bindheimite ($\text{Pb}_2\text{Sb}_2\text{O}_6\text{O}$), senarmontite (Sb_2O_3), valentinite (Sb_2O_3) and stenhuggarite ($\text{CaFeSb}(\text{AsO}_3)_2\text{O}$). These materials were incubated in slurries at 10 % solids, at 25°C under agitation, either in presence of air or under N_2/H_2 atmosphere. Sb was generally more mobile in oxidizing conditions; however, for 2 samples, mobility was higher in reducing conditions. The highest Sb concentrations in water were in the range 20 to 30 mg L^{-1} . The final percentage of solubilized Sb exceeded 1 % (between 1 and 12 %) for 10 samples. For one sediment sampled in Balanzola mine, final Sb concentrations were close to 20 mg L^{-1} in oxidizing conditions and 10 mg L^{-1} in reducing conditions. Acidification was observed with several samples; however, Sb release was not systematically related with the evolution of pH. The mobility of Sb during leaching might be driven by diverse mechanisms: release of sorbed Sb, abiotic or biotic dissolution of Sb-bearing minerals, including oxidation of Sb sulfides in aerobic conditions, or reductive dissolution of Sb-bearing iron or manganese oxides, and finally release of soluble thio-Sb complexes in anaerobic conditions. Supporting the occurrence of these last mechanisms, final analyses indicated solubilization of Fe and Mn and traces of dissolved sulfide in reducing conditions. Our results, that

showed a higher mobility of Sb in oxidizing conditions, are globally consistent with previous works indicating a higher occurrence of the oxidized form of Sb, i.e. Sb^V, in water streams impacted by mining sites. However, we also observed that non negligible release of Sb can be linked to mechanisms occurring in reducing conditions. Perspectives of this work include the elucidation of the biological processes, directly or indirectly involved in Sb release or immobilisation, in order to better predict the evolution of environmental quality on mining sites and propose remediation strategies.

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