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INVITED CENTENNIAL REVIEW

Uptake and release of arsenic and antimony in alunite-jarosite and beudantite group minerals

KAREN A. HUDSON-EDWARDS[1,*]

[1]Environment & Sustainability Institute and Camborne School of Mines, University of Exeter, Penryn, Cornwall TR10 9FE, U.K.
Orcid 0000-0003-3965-2658

ABSTRACT

Arsenic and antimony are highly toxic to humans, animals, and plants. Incorporation in alunite, jarosite, and beudantite group minerals can immobilize these elements and restrict their bioavailability in acidic, oxidizing environments. This paper reviews research on the magnitude and mechanisms of incorporation of As and Sb in, and release from, alunite, jarosite, and beudantite group minerals in mostly abiotic systems. Arsenate-for-sulfate substitution is observed for all three mineral groups, with the magnitude of incorporation being beudantite (3–8.5 wt% As) > alunite (3.6 wt% As) > natroalunite (2.8 wt%) > jarosite (1.6 wt% As) > natroalunite (1.5 wt% As) > hydroniumalunite (0.034 wt% As). Arsenate substitution is limited by the charge differences between sulfate (–2) and arsenate (–3), deficiencies in B-cations in octahedral sites and for hydroniumalunite, difficulty in substituting protonated H_2O-for-OH^- groups. Substitution of arsenate causes increases in the *c*-axis for alunite and natroalunite, and in the *c*- and *a*-axes for jarosite. The degree of uptake depends on, but limited by, the AsO_4/TO_4 ratio. Aerobic and abiotic As release from alunite and natroalunite is limited, especially between pH 5 and 8. Release of As is also very limited in As-bearing jarosite, natrojarosite, and ammoniumjarosite at pH 8 due to the formation of secondary maghemite, goethite, hematite, and Fe arsenates that resorb the liberated As. Abiotic reductive dissolution of As-bearing jarosite at pH 4, 5.5, and 7 is likewise restricted by the formation of secondary green rust sulfate, goethite, and lepidocrocite that take up the As. Similar processes have been observed for the aerobic dissolution of Pb-As-jarosite (beudantite analog), with secondary Fe oxyhydroxides resorbing the released As at pH 8. Higher amounts of As are released, however, during microbial-driven jarosite dissolution. Natural jarosite has been found to contain up to 5.9 wt% Sb^{5+} substituting for Fe^{3+} in the B-site of the mineral structure. Sb(V) is not released from jarosite at pH 4 during abiotic reductive dissolution, but at pH 5.5 and 7, up to 75% of the mobilized Sb can be structurally incorporated into secondary green rust sulfate, lepidocrocite, or goethite. Further research is needed on the co-incorporation of As, Sb, and other ions in, and the uptake and release of Sb from, alunite, jarosite, and beudantite group minerals, the influence of microbes on these processes and the long-term (>1 yr) stability of these minerals.

Keywords: Arsenic, antimony, alunite, jarosite, beudantite; Invited Centennial article