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As and Sb behaviour post freshwater restoration of the jarosite and Fe(II)-rich ASS/AMD-affected wetlands

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Jarosite is an important scavenger for As [1] and Sb [2] in acid sulfate soils (ASS) and acid mine drainage (AMD) environments. Previous studies suggest that Fe²⁺ can catalyse the rapid dissolution of jarosite that may result in the release of the scavenged metalloids to aquifers and soils [3]. In this study, we subject synthetic jarosite with structurally incorporated As(V)/Sb(V) to Fe²⁺_(aq) (20 mM) for 24 h, under anoxic conditions at pH 4.0, 5.5 and 7.0 to mimic the early, mid and final stages of freshwater restoration of the ASS/AMD-affected environments. We addressed the hypothesis that at higher pH, the Fe²⁺-induced transformation of jarosite can significantly influence the bioavailability of As and Sb through changing both their fractionation and speciation during the mineralogical transformation process. We examined changes in aqueous and solid phase chemistry via selective extractions, X-ray diffraction (XRD), X-ray absorption spectroscopy (XAS) and transmission electron microscopy (TEM). Addition of Fe²⁺, resulted in the formation of a green rust (GR) intermediary in pH 5.5 and 7 treatments prior to the formation of goethite within 24 h, however, no significant mineralogical transformations was observed at pH 4.0 over time. At pH 7.0 and to a lesser extent at pH 5.5, Fe²⁺-induced transformation of jarosite enhanced aqueous-phase concentrations of Sb, while As concentrations in aqueous-phase were negligible in all the treatments and it mostly was repartitioned to the phosphate exchangeable phase in both pH 5.5 and 7 treatments. The results imply that Fe²⁺-induced transformation of jarosite during mid-stages of freshwater restoration and under pH 5.5-7 conditions, can increase Sb bioavailability. In addition, repartitioning of As to the surface of the new host minerals (e.g. goethite, green rust) in the same pH-range may enhance the risk of its mobilization during future mineral transformation processes in Fe²⁺-rich systems.

[1]Johnston, S.G., et al., Arsenic mobilization in a seawater inundated acid sulfate soil. *Environmental Science and Technology*, 2010. 44(6): p. 1968-1973.[2]Tighe, M., et al., The availability and mobility of arsenic and antimony in an acid sulfate soil pasture system. *Science of The Total Environment*, 2013. 463-464(0): p. 151-160 [3]Amstaetter, K., T. Borch, and A. Kappler, Influence of humic acid imposed changes of ferrihydrite aggregation on microbial Fe(III) reduction. *Geochimica et Cosmochimica Acta*, 2012. 85: p. 326-341.

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