

The bioaccessibility of arsenic and lead from sulfidic mine tailings is controlled by contaminant and host speciation

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Arsenic and lead are toxicants with no beneficial biological function. These chalcophile elements are enriched in sulfide ores and can become problematic when deposited as fine particulate matter (PM) in mine tailings. Communities nearby mine wastes in arid and semi-arid regions are potentially exposed to toxic metal(loid)s from fugitive dusts deriving from these impoundments. To assess the relation between potentially lofted PM and human health risk, we studied the relationship between pharmacokinetic bioaccessibility and metal(loid) molecular speciation for tailings from a Superfund site with arsenic and lead as the contaminants of concern by coupling in vitro bioassay (IVBA) with X-ray absorption spectroscopy (XAS). Tailings PM with arsenic and lead up to 59 and 34 mmol kg⁻¹ were reacted with synthetic gastric and lung fluid for 30 s to 100 h to investigate toxic metal(loid) release kinetics. Bioaccessible (BAc) fractions of arsenic and lead were about 10 and 100 times greater in gastric than in lung fluid simulant, respectively, and 10-100% of the maximum gastric BAc from PM₁₀ and PM₁₅₀ occurred within 30 s, with parabolic dissolution highly-reactive PM followed by slower release from less soluble sources. Arsenate within jarosite and sorbed to ferrihydrite, and lead from anglesite, were identified by XAS as the principal contaminant sources in near surface tailings. Analysis of pre- and post-IVBA PM indicated the release of arsenic in lung fluid was principally from arsenic-substituted jarosite, whereas in synthetic gastric fluid arsenic complexed on ferrihydrite surfaces was preferentially released and subsequently repartitioned to jarosite-like coordination at extended exposures. Lead dissolved at 30 s was subsequently repartitioned back to the solid phase as pyromorphite in phosphate rich lung fluid. The bioaccessibility of lead in PM was limited due to robust sequestration in plumbojarosite.

