Arsenic distribution and speciation in mine wastes: developing a proxy for long-term exposure risk

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The historic mining and processing of gold and silver ore deposits throughout the state of California has left a pervasive legacy of arsenic-contaminated wastes posing health and environmental threats. Key to predicting the potential toxicity of arsenic (As) in these materials is its bioaccessibility, commonly defined as the fraction of the solid-phase arsenic that can be solubilized and thus mobilized within the body.

In complex heterogeneous mine wastes, As bioaccessibility is controlled by a variety of physicochemical properties including initial As concentration, particle size, surface area, As speciation, and the spatial distribution of the As within the waste material particles themselves. The last of these is particularly challenging to characterize but holds particular importance for the long-term risk of arsenic exposure as a result of the physical and chemical weathering processes to which mine wastes are exposed.

Combining size-dependent EXAFS studies of As speciation and μXRF spatial mapping with *in vitro* simulated gastric fluid (SGF) extractions can enable the correlation of As speciation, As spatial distribution, and As bioaccessibility with one another. By additionally pulverizing selected size fractions of the mine waste samples, re-measuring As bioaccessibility, and analyzing As speciation pre- and post-extraction, we can both identify and quantify changes in speciation and assess the extent of physical encapsulation of As as a limiting factor to its mobility. Recent findings from this work include:

- Particle grinding increases As bioaccessibility (in some cases by over a factor of 3 relative to unground wastes), although with diminishing effectiveness as particle size decreases
- Arsenic persists as reduced sulfides within particle grains but dominates as As(V) sorbed to iron hydroxides at particle surfaces
- Particle grinding exposes interior (reduced) arsenic phases which are more amenable to dissolution and transformation in SGF than surface-bound species

These results are expected to significantly improve our ability to predict the role that As speciation and distribution play in controlling As bioaccessibility in contaminated mine wastes over time.