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Solid-phase As and Fe speciation of groundwater treatment sludge collected from around the world: Insights for sludge management

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Iron (Fe)-based treatment methods are applied worldwide to remove carcinogenic arsenic (As) from groundwater. These treatment methods can vary in capacity and As removal mechanism, but all generate toxic As-rich Fe oxide sludge. Currently, no sustainable strategies exist for managing this toxic waste.[1] At the same time that As-rich sludge represents a disposal challenge, As-bearing compounds used in alloys and electronics have a critical supply risk in Europe,[2] highlighting a potential As recovery opportunity for groundwater treatment sludge. A critical step to create technologies that can convert As-rich waste to valuable As-bearing compounds is to determine the molecular-scale structure of sludge generated from different Fe-based treatment methods.

In this work, we collected solid As-rich waste residues from several existing treatment plants situated in different continents (i.e., North America, Europe, Asia). The location and type of treatment facility were selected strategically to span a wide range of groundwater composition, plant capacity and As removal mechanism. The sludge samples were characterized comprehensively by synchrotron-based As and Fe K-edge X-ray absorption spectroscopy (XAS) and several macroscopic measurements such as total digestions, thermogravimetric analysis, and BET surface area.

Our results revealed many key similarities in the composition and structure of As-rich sludge, regardless of the influent As concentration (20-300 µg/L), plant capacity (<1 to >100 m^3/h) and design (aeration-filtration; ferric chloride addition; electrocoagulation; electro-Fenton). First, all sludges contained similar As mass fractions (800-1200 mg/kg), suggesting that high As sludge contents can occur even with low influent As levels. Second, the Fe K-edge XAS analysis indicated that all samples consisted dominantly of hydrous ferric oxide (HFO) solids with less structural order than nanoscale 2-line ferrihydrite. Third, the As K-edge XAS analysis showed that As was exclusively present in all samples as As(V) bound to HFO in the binuclear ^2C complex. These similarities suggest that As recovery technologies optimized for one sludge can likely be applied to sludge generated by a variety of different Fe-based groundwater treatment methods.

1. van Genuchten et al. (2022). *Environmental Science & Technology* 56 (19), 14109-14119.
2. Blengini et al. (2020), Study on the EU's list of Critical Raw Materials Final Report.

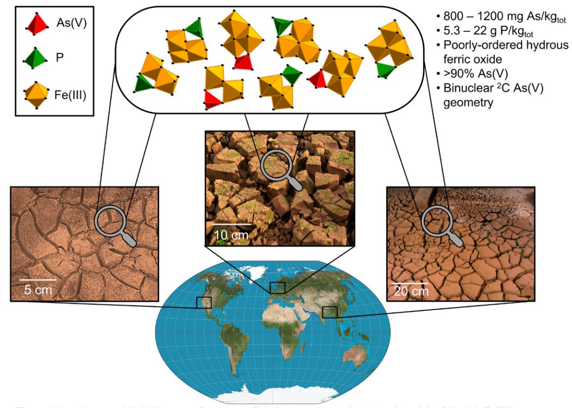

Figure 1: Locations and digital images of a subset of sludge samples and structural models of As-rich Fe(III) (oxyhydr)oxide sludge. In the structural models, which were derived from Fe and As K-edge XAS analysis, Si is not included. The As and P solids ratios are given in mass of As or P per total mass of dried sludge.