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Insights from multi-scale, multi-element synchrotron spectroscopy: Considering the role of sulfur in promoting arsenic mobilization by the river-aquifer interface

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Geogenic arsenic (As) contamination of groundwater poses significant threats to human health, especially in Vietnam. There, the river-groundwater interface is a dynamic biogeochemical hotspot for As. The predominant mechanisms proposed for sustained As release at this interface rely on co-occurrence of continuously replenished labile organic carbon and As-bearing iron(III) oxides combined with advection of As-enriched pore waters [1]. However the generalizability of these mechanisms across other river-groundwater intersections remains unknown. Understanding how micro-scale biogeochemical heterogeneity impacts large-scale As mobilization near river-groundwater interfaces is thus particularly relevant for explaining As distribution changes over time and across various hydrogeologic environments.

This work combines micro and bulk scale X-ray absorption spectroscopy (XAS) towards understanding a two-fold increase in aqueous As observed over twenty years near a field site in northwestern Hanoi, Vietnam along the Red River Delta. Direct measurement of thiolated arsenic across a long well screen at our field site highlights the role of sulfur in promoting As release through formation of thiolated arsenicals. Recent research suggests these species often occur in discrete sediment micro-structures yet can migrate [2]. While segments of the delta characterized by river water advection into the aquifer are well-studied, our core represents an area where groundwater flow dynamics are changing. Sediment subsamples were collected along the 30-m core (every 20-cm) and analyzed for high resolution As, iron, and sulfur XAS at Argonne National Laboratory's Advanced Photon Source. Five subsamples across a 1-m redox gradient identified above the well screen interval were measured on X-ray microprobe. High energy maps (iron, As) were compared to low energy maps (sulfur). Selected points for micro-XANES analysis show spatial correlations between elevated concentrations of As and sulfur at the edges of iron-coated sediments. Micro-XAS insights are compared against those produced from bulk XANES (As, iron) collected along the entire core, and to sediment cores previously characterized nearby [3]. Overall, this research showcases how multi-scale synchrotron analyses can be combined to assist in further elucidating complex biogeochemical environmental contaminant dynamics.

[1] Wallis et.al., 2020 *Nat.Geosci.*
[2] Kumar et.al., 2020 *ES&T*
[3] Berg et.al., 2008 *Chem.Geol.*