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Dynamic arsenic mobilization in groundwater under pumping-induced disturbances: high-resolution spectroscopic observations of arsenic, iron, and sulfur cycling in organic-rich settings

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In South and Southeast Asia, groundwater contamination of arsenic (As) occurs widely: usually via microbial reductive dissolution of As-sorbed iron(III) oxides fueled by dissolved organic matter. Recently, increased groundwater pumping has exacerbated the risk of As contamination. One hypothesis suggests that pumping disturbs organic-rich clay layers and accelerates As release into underlying aquifers. Specifically, peat layers—defined as organic-rich lenses within clay units—are common in deltaic settings in Vietnam. We investigate this mechanism in pumping-disturbed aquifers in the Red River Delta (RRD). We obtained sediment cores from two sites near groundwater cones of depression that were expected to contain organic-rich layers (>5 m) based on previous geological surveys — and importantly where As concentrations have significantly increased compared to two decades ago. Groundwater As rose from 7.0 μg/L to 92.9 μg/L at one site and from 170 μg/L to 295 μg/L at another. To understand the biogeochemical dynamics of As coupled to the iron, sulfur and carbon cycle, we conducted X-ray absorption spectroscopy (XAS) with high-resolution sampling across stratigraphic transition zones and X-ray microprobe mapping. At the first site (clay-dominated), we identified two organic-rich layers. The shallower organic-rich layer was dominated by arsenopyrite and Fe oxide-bound As(III/V) with negligible iron sulfides, whereas the deeper organic-rich layer was enriched in orpiment and Fe oxide-bound As(III). At the second site (sand-dominated), organic-rich layers occurred as multiple thin lenses. The shallow aquifer was oxidized and Fe oxide-bound As(III) dominated. However, within a thicker/deeper organic-rich layer, minor arsenopyrite and As-sulfide phases emerged alongside Fe oxide-bound As(III). Across both cores, distinct As species associated with As sulfide minerals was found in the deeper organic-rich clay layer. Compared with peat investigations in the Mekong Delta [1-2], the As speciation results (e.g., As-associated sulfides) are similar but contrast in terms of purported As mobilization. This suggests that pumping-induced drawdown in the RRD may have shifted redox conditions in the deeper layer and As speciation associated with sulfur cycling caused subsequent As mobilization.

[1] Stuckey et al. 2015, *GCA*
[2] Wang et al. 2018, *ES&T*