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Sedimentary organic matter and iron mineralogy control solid-phase arsenic behavior in the hyporheic zone of the Meghna River, Bangladesh

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The biogeochemical cycling of iron (Fe) within the alluvial aquifers of Bangladesh governs the mobility and bioavailability of arsenic (As). Along the tidally-fluctuating Meghna River, the mixing of oxic river water and reducing groundwater generates dynamic redox conditions which may either cause the precipitation of Fe-oxides under oxic conditions or support the microbially-mediated dissolution of Fe-oxides fueled by labile organic matter under reducing conditions. Here, the sedimentary organic matter (SOM) and Fe mineralogy within the hyporheic zone (HZ) sediments are investigated to determine their control on the solid-phase distribution of As. The molecular properties of the SOM were determined following an alkaline extraction by FTIR, the mineralogy of Fe-oxides were determined by diffuse reflectance, Fe(III) to Fe(II) proportions were estimated by the ferrozine method, and the solid-phase partitioning of As within the sediment was determined by a 6-step sequential extraction process. The FTIR measurements indicate that the bulk SOM content of the riverbank (<1.8 wt%) and an organic-rich shallow silt layer (4.8 wt%) underlying the riverbank sands (~3 m bgl) is comprised of high proportions of amides and polysaccharide moieties indicative of fresh, labile SOM potentially of microbial origin. The ferrozine analysis revealed that Fe(III)/total Fe ratio in the riverbank sediment was ~50%, wherein the Fe(III) was present as amorphous ferrihydrite/goethite as shown by diffuse reflectance measurements. Finally, the sequential extractions revealed average As concentrations of 7.7 mg/kg in the riverbank, of which >80% was distributed among 4 phases: Amorphous Fe-oxides (32%), crystalline Fe-oxides (23%), residual (16%), and organic matter (12%). When groundwater flows towards the river, the labile SOM in the silt layer and riverbank sands contain abundant electron donating moieties which provide electrons for the microbially mediated reduction of As-bearing Fe-oxides and thereby generate dissolved As. Conversely, As is immobilized by the precipitation of Fe-oxides, as suggested by the proportion of As associated with amorphous Fe-oxides (32%), high Fe(III) content (~50%), and the presence of Fe as ferrihydrite. These results highlight the dynamic redox controls on Fe cycling, and ultimately, As mobility within the HZ.