Arsenic removal from natural waters using iron electrocoagulation – kinetics, spectroscopy and modeling

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Geogenic contamination of groundwater resources by arsenic (As) is a public health concern worldwide given the link between As exposure and the development of cancers and skin lesions. The biogeochemistry of iron (Fe) and As are strongly coupled in natural systems, giving rise to many technical treatment options for As removal from drinking water that are mainly based on the formation of Fe(III) precipitates. Iron electrocoagulation (EC), which is being developed as a method for As removal, provides a model redox boundary to investigate the geochemistry of As in Fecontaining waters. In EC, a current is passed through an Fe(0)electrode, which allows for controled oxidative dissolution of Fe(0) to Fe(II). Successive reactions between Fe(II) and O_2/H_2O_2 generate reactive oxidants that can oxidize Fe(II) to Fe(III) and As(III) to As(V). In addition, nanoscale Fe(III) precipitates efficiently sorb As(III) and As(V) at alkaline and acidic pH values, respectively.

In natural waters, other many ions can compete with As for sorption sites on the Fe oxides and/or reactive oxidants. In this study, two surface waters and one groundwater were sampled and filtered at 0.2 μ m. Synthetic (NaCl/HCO₃⁻ electrolyte) and the natural waters were spiked with 10 μ M As(III) or As(V) and either with or without 250 μ M H₂O₂.

Our results show that As removal depends on the initial H₂O₂ concentrations, As oxidation state, and concentration of Ca²⁺ and HCO₃⁻. Arsenic removal was faster for lake- (69 mg L^{-1} HCO₃⁻) > river- (140.7 mg L^{-1} HCO₃⁻) > ground- (179.9 mg L^{-1} HCO₃⁻) > synthetic (122 mg L^{-1} HCO₃⁻) water. These results are consistent with the formation of sub-nanoscale Asrich Fe polymers that cannot be removed by filtration at 0.2 µm at high HCO3⁻ concentrations. In addition, As(III) removal was slower than As(V) removal indicating As(V) sorbs more favorably to the Fe(III) precipitates (7.4-8.6). These trend was confirmed through kinetic modelling of As and Fe oxidation by Fe(IV) and OH[•]. Scanning electron microscopy coupled to energy-dispersive X-ray spectroscopy and X-ray absorption spectroscopy showed different precipitate structures depending on water composition. These results provide insight into the control of co-occuring ion on As removal efficiency in EC systems.