

Fate and transport of arsenic from constructed soil aggregates

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Reduction of As (V) and Fe (III) are commonly the most prominent processes controlling the fate and transport of As in soils and sediments. However, the physical structure of such environments results in biogeochemical heterogeneity that can cloud the operative processes controlling the fate and transport of As. To resolve the role of soil physical structure on the fate and transport of As, we examined As desorption from constructed aggregates by coupling experimental observation and reactive transport simulation. Spherical aggregates were made with As (V)-bearing ferrihydrite-coated sand inoculated with *Shewanella* sp. ANA-3. Single aggregates were placed in a cylindrical reactor and aerated or anoxic solute was initiated around the aggregate. Arsenic desorption and release to advecting water was similar between anoxic and aerated systems for the first 20 d; thereafter, the anoxic advecting solutes increased As release relative to the aerated counterpart. With aerated advecting solutes, Fe (III) remained oxidized, or was oxidized, in the cortex of the aggregate, forming a 'protective' barrier that limited As release to the advective channel. Reactive transport simulations confirm the sharp decrease in oxygen concentration and gradual increase in Fe (II) concentration from the aggregate exterior toward the aggregate interior.

Selenium in shales: Where is it?

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Selenium (Se) is an essential trace element that can reach unacceptable toxic levels of pollution in soils and groundwaters and Se contaminated waters can lead to severe human health effects. In most natural settings, the prime natural source of selenium (Se) is derived from the weathering of shales, yet the weathering pathways or release mechanisms of Se from shales are poorly understood. This is a consequence of the lack of understanding of the distribution and specific associations between Se and the main potential selenium host phases: sulphides, silicates, and organic matter. In addition, a reliable and efficient separation and thus a quantification of the Se concentrations in the various fractions within shales is lacking. Using 9 different shale samples with bulk total [Se] concentrations varying from 1.4 and 5000 mg/Kg, we developed a new method that successfully separates the three fractions for further analyses. The separation is based on a sodium polytungstate heavy liquid sequential separation approach that targets each fraction at different densities (3.0 and 1.6 g/mL for sulphides and silicates and organic matter respectively). In order to quantify the efficiency of the separation approach as well as the [Se] concentration in the separates we have characterised all post separation fractions chemically, mineralogical and spectroscopically using AAS-GF, FTIR, XRD, SEM/EDS, elemental analyses (CS) and EMPA. The results show that we can achieve an extremely good separation. For example, for the sulphide fraction XRD patterns, SEM imaging/spectral evaluations and EMPA confirmed the high purity of the separates (primarily pyrite) and the close association of Se with some of the pyrite fractions (300 to 29000 ppm) which is between 4 and ~ 25 % higher than the corresponding bulk sample. The purity of the separated silicate and organic fractions were confirmed via XRD (clays only), FTIR (both) and elemental analyses (organics) and the Se concentration in a separated organic fraction was only ~0.250 ppm, or 0.1 % of the bulk. This indicates that sulphides are the most important host phase of selenium in shales.

Our data indicate that during weathering the oxidation and dissolution of pyrite from shales most likely controls the release and thus delivery of selenium to groundwaters.