As(V) Reduction to As(III) in the Presence of Chloride in Ambient Conditions

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Arsenic (As) is a geogenic contaminant that affects aquifers worldwide. In aqueous environment, As mainly exists in As(III) or As(V) forms, depending on the prevalent pH, redox potential (E_H) and co-ions. For conditions relevant to As-contaminated aquifers, As(V) precipitation, as pharmacolite ($CaHAsO_4 \cdot 2H_2O_{(s)}$), is thermodynamically favorable in the presence of calcium (Ca). However, this precipitation kinetics is poorly uunderstood; specifically, its variation with the nature of Ca source as well as the presence of co-ions, such as chloride (Cl^-). This study aims to identify whether the nature of Ca-source has an impact on the rate and form of As uptake in the presence of Cl^-. Batch experiments were performed with an initial As(V) concentration of 50 µM in different atmospheric conditions – ambient (open), partially-open and anaerobic (O_2 < 2 mg/L; glove box) – in the presence of 50 µM $CaCl_2 \cdot 2H_2O$ or 1g L^{-1} $CaCO_3$, or both. Control experiments with NaCl and KCl were also performed to investigate dissolved As variation in the absence of Ca . ICP-MS analysis of the solute data showed that the maximum total dissolved As (As_T) uptake occurred in reactors containing both $CaCl_2$ and $CaCO_3$ (~56 % of initial As_T) after 21 days in ambient conditions. It could be due to precipitation of the only supersaturated Ca-As solid, pharmacolite, on $CaCO_3$ surface. Contrary to thermodynamic predictions, reduction of As(V) to As(III) was observed in reactors containing Cl^- in ambient conditions for all Cl^- sources as indicated by solute data from IC-ICP-MS (Fig.1). However, such reduction was not observed in partially-open and anaerobic conditions. XPS analysis on As-reacted solids also suggested the presence of As(III). These analyses suggest the role of dissolved oxygen or dissolved CO_2in As(V) reduction under ambient conditions. W-SEM-EDX analysis of As-reacted solids revealed rod-shaped structures and recorded high As signals. Preliminary microbiological characterization indicated the presence of rod-shaped, gram-negative bacteria in the ambient conditions, which could be responsible for As reduction. Results from macroscopic wet chemical experiments as well as microbial characterization studies will be combined to explain the fate of As in the presence of Cl^-.

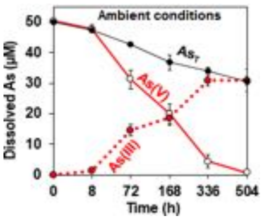

Figure 1: Variation of dissolved arsenic with time in the presence of $CaCl_2$.