Geophysical Research Abstracts,
Vol. 11, EGU2009-7614, 2009
EGU General Assembly 2009

Reductive dissolution of As(V)-Fe oxyhydroxides: an experimental insight at biogeochemical interfaces in soil

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Iron (III) oxides are ubiquitous components of soils, sediments, aquifers and geological materials. Trace metals associate with Fe (III) oxides as adsorbed or co-precipitated species and, consequently the biogeochemical cycles of Fe and trace metals are closely linked. Using a new monitoring tool recently developed, this study was dedicated to understand how do interplay biological and mineralogical (crystallographic and specific surface area) controls in the Fe oxyhydroxide reductive dissolution within soils and which can be the consequences on associated trace metal release. For this purpose, polymer slides covered by synthetic As-spiked ferrihydrite (As-Fh) or As-spiked lepidocrocite (As-Lp) were inserted into an organic-rich wetland soil in non conventional columns system under anaerobic conditions. This technique was developed to allow the insertion of slides into a structured soil without significant disturbance and to avoid the mechanical abrasion of oxides from slides that would occur in an equilibrium batch system under stirring. Slides were recovered after different periods of time to evaluate (i) the impact of (bio)reduction on both Fe-oxide dissolution and secondary mineral precipitation and, (ii) the subsequent effects on As mobility.

XRF analyses of the slides were conducted before and after contact with the soil to determine the amount of Fe and associated As remaining on the slides. Fe(II), acetate, nitrate, sulphate and total metals of the soil solution was followed through time by ion chromatography and ICP-MS measurements. The important bacterial colonization and occurrence of biofilm evidenced by SEM analyses of the slides suggested the presence of biologically mediated processes. As previously shown elsewhere the kinetics of the suspected occurring bacterial reduction differ significantly from abiotic reduction data from literature. The important point is that conversely to what has been observed in published experimental data, the dissolution rates remained here fairly constant through time since the organic matter present in the interacting solution complexed the released Fe(II), which was therefore not able to accumulate onto the bacterial cell surfaces. The organic matter mediated complexation of Fe(II) prevented thus the progressive inhibition of the enzymatic reduction to occur as elsewhere evidenced with other experimental conditions. As expected, the reductive dissolution of the less crystallized ferrihydrite started quicker than that of lepidocrocite. The newly formed minerals were mostly composed of Fe-sulphides. Iron(II) complexation by organic molecules in solution likely prevented formation of secondary Fe(II, III)-rich minerals. The relative proportion of As(III) increased with time on the As-Fh slides, and was combined with a decrease of the Fe/As ratio, suggesting a partial adsorption of As(III) onto minerals. By contrast, for lepidocrocite, the Fe/As ratio increased, suggesting that As(III) was less readsorbed due the lower available site number and the deletion of As adsorption sites on the reduced lepidocrocite surface. Only a weak proportion of As(III) was sequestered by readsorption onto unreduced Fe-oxides and possibly on secondary Fe-sulphide minerals, especially when the iron oxide had a low surface area. Therefore, wetlands and their waterlogged soils could be a non negligible source of As within soils, migrating first through soil solutions and then to the whole hydrosystem.