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Linking Isotope Exchange and Fe(II)-Catalyzed Ligand-Controlled Dissolution of Iron(hydr)oxides

Jagannath Biswakarma^{1,2}, Kyounglim Kang³, Walter D.C. Schenkeveld⁴, Stephan M. Kraemer³, Janet G. Hering^{1,2}, and Stephan J. Hug²

¹ETH, Swiss Federal Institute of Technology Zurich, Switzerland

²Eawag, Swiss Federal Institute of Aquatic Science and Technology, Switzerland (jagannath.biswakarma@eawag.ch)

³Dept. of Environmental Geosciences, University of Vienna, Austria

⁴Faculty of Geosciences, Utrecht University, the Netherlands

Dissolution of iron(oxyhydr)oxides is a key biogeochemical process that affects the cycling and bioavailability of iron (Fe). Recently, we demonstrated that submicromolar concentrations of Fe(II) accelerate dissolution of Fe(III)(hydr)oxides with the synthetic ligands ethylenediaminetetraacetate (EDTA) and hydroxybenzyl ethylenediaminediacetic acid (HBED) and also with the biogenic ligand desferrioxamine-B (DFOB) in anoxic conditions at circumneutral pH. The catalytic effect of Fe(II) was explained by electron transfer (ET) to surface Fe(III) and accelerated detachment of surface Fe(III)-ligand complexes. However, the extent of ET on the mineral surface before and during accelerated dissolution remained unclear. Here we describe the extent of ET by investigating dissolution and isotope exchange with lepidocrocite (Lp) and goethite (Gt) and varying concentrations of Fe(II), ⁵⁷Fe(II), and DFOB. Most experiments were conducted under anoxic conditions at pH 7.0 in bicarbonate-CO₂-buffered suspensions.

Our results show that in anoxic carbonate-buffered suspensions, 1-5 μM Fe(II) increased the rates of Lp dissolution at pH 7.0 by up to 60-fold. The addition of 20 or 50 μM DFOB after ⁵⁷Fe(II) led to accelerated detachment of ⁵⁶Fe(III) from Lp and release of already adsorbed/exchanged ⁵⁷Fe into the solution. A kinetic model considering exchange of charge on the surface between ⁵⁷Fe(II) and ⁵⁶Fe(III), before and during dissolution, was developed to explain the observed results. The rates for ET and isotope exchange before and during accelerated dissolution are very different for Lp and Gt, presumably reflecting the differences in structure and mineralogy.

This study contributes to the quantification of ET from added Fe(II) to the surface of Fe(III)(hydr)oxides and of the acceleration of overall non-reductive dissolution by traces of Fe(II) in anoxic environments. In this presentation, the key findings of the isotope exchange and dissolution studies with Lp and Gt will be presented in order to highlight the importance of interfacial Fe(II)/Fe(III) ET processes occurring at (sub)oxic-anoxic interfaces of soils and sediments.