Influence of surface speciation on the release of dissolved iron and ferrihydrite colloids from porous media

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Colloidal particles play a major role in contaminant transport in the aquatic and subsurface environments. Results from both field and laboratory studies suggest that organic ligands could dissolve Fe(III) oxide and simultaneously mobilized the oxide colloids in situ. We propose that the surface speciation on iron oxide is fundamental to the observed oxide dissolution and particle release. To quantitatively predict the release of dissolved iron and colloidal oxide particles from a porous medium, we conducted the current work with changing citrate concentrations, in an attempt to model the surface speciation. Experiments were conducted in a column flow-through system filled with ferrihydrite-coated quartz. The kinetics of ferrihydrite colloid release was investigated by eluting a citrate solution of respectively 1, 3 and 5 mM at pH 4 and 10 mM NaCl. Effluent samples were collected over time for citrate and iron analyses and pH was measured with time. Dissolved and colloidal iron was separated via ultra-filtration and iron concentrations in the bulk and filtrate were determined using ICP-MS. Ferrihydrite colloids were characterized by micro-electrophoretic mobility measurements. For model calculations, we used MINTEQA2 to determine surface species and PREEQC to simulate the flow through experiment. We observed that as citrate eluted through the column, the effluent pH increased from the influent value of 4 to >7 and returned to 4 when the citrate breakthrough completed. Effluent colloid concentrations and the particle electrophoretic mobility followed the same trend as pH, reaching maximum at the highest observed pH. These results showed that during the initial phase of reaction, colloid release correlated with the variations of surface electric potential, which is a direct result of surface protonation and complexation. Model calculations are undertaken to quantitatively assess the transport of iron and ligand species through the column and to simulate the variations of ferrihydrite surface potential.