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Modelling the release kinetics of adsorbed phosphate during sulfide-induced dissolution of lepidocrocite (γ-FeOOH)

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Excess phosphate (P) is the main cause of eutrophication and has become an environmental problem in Europe. In limnic systems, the dynamics of P and iron (Fe) are often closely coupled. Iron (hydr)oxides, such as lepidocrocite, can effectively remove dissolved P via adsorption. However, when Fe (hydr)oxides with adsorbed P are entering the sediment, they can be exposed to free sulfide (S(-II)). Reaction with S(-II) might lead to a release of P into solution due to sulfide-induced reductive dissolution of Fe (hydr)oxides and formation of iron-sulfides (sulfidation). The recycling of dissolved P can, in turn, exaggerate eutrophication. Although P adsorption onto Fe (hydr)oxides and sulfidation of Fe (hydr)oxides have been studied separately in detail, it is unclear how the rates of P release are coupled to the progress of the sulfidation reaction. This uncertainty is because multiple reactions influencing P release can occur in parallel. Here we describe laboratory experiments combined with mechanistic modelling that helped us gain new insights into the regulation of P release during sulfidation.

The experiments were performed in a flow-through reactor at two different pH levels (7.3 and 8.3). Sulfide was continuously pumped into the reactors containing lepidocrocite with pre-adsorbed P, while P and S(-II) concentrations in the outflow were measured to determine the reaction progress. Results showed that P release was incongruently linked to the sulfidation progress. Ligand exchange between S(-II) and different types of P surface complexes can explain the deviation between the rates of P release and sulfidation. The mechanistic model, developed as an interactive tool in R, helped us identify and parameterize rate expressions for the sulfidation of lepidocrocite and P release that can reproduce the measured outflow concentrations for all experimental conditions. Furthermore, sensitivity analysis was performed to assess the uncertainty of the model parameters. The rate expressions and the dependency of the optimized parameters on pH will be discussed.