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Stabilization of ferrihydrite and lepidocrocite by silica during Fe(II)-catalyzed transformation and the impacts on mineral transformation products

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In soils and sediments, ferric iron (Fe(III)) minerals, such as ferrihydrite and lepidocrocite, are subject to microbial reductive dissolution under anoxic conditions. This leads to the release of dissolved ferrous iron (Fe(II)) which interacts with iron mineral phases, and catalyzes their recrystallization and transformation to more crystalline iron minerals. The mineral transformation process can be hindered by silica (Si), which, in form of silicic acid, is a widespread component of natural soil solutions. However, it is still unclear which mineral phases result from the transformation of ferrihydrite and lepidocrocite at low Si/(Si+Fe) mol ratios (5%, 15%) and Fe(II):Fe(III) mol ratios (0.02, 0.2). In this study, ferrihydrite, Si-ferrihydrite co-precipitates, lepidocrocite and Si-adsorbed lepidocrocite were reacted with isotopically labelled ^{57}Fe(II) at pH 7 for up to four weeks. We sampled the solid phase at six time points to follow mineral transformations by X-ray diffraction and sampled the aqueous phase to analyze dissolved Fe(II) dynamics and its isotope composition in order to follow iron atom exchange between aqueous Fe(II) and solid Fe(III). Our results demonstrate that coprecipitated Si does not hinder iron atom exchange but stabilizes ferrihydrite effectively against mineral transformation. The extent of ferrihydrite transformation depended on the silicon to iron molar ratio, rather than the Fe(II) concentration. While 64-65% of ferrihydrite was transformed in the absence of Si, 44-51% and 0-6% were transformed in the presence of low and high Si, respectively. Electron microscopic images provide evidence for the redistribution of Si in the solid phase during ferrihydrite transformation and for the change in morphology of mineral transformation products. The recrystallization of pure lepidocrocite over four weeks at low Fe(II) concentration is confirmed by iron atom exchange data, demonstrating its stability against Fe(II)-catalyzed mineral transformation. In the presence of adsorbed Si on the lepidocrocite surface, iron atom exchange was strongly reduced. Due to the influence of Si on iron mineral transformation processes, it can also affect the sorption, speciation, and mobility of nutrients and contaminants in redox-affected soil environments.