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Spatial patterns in mineral formation driven by dissimilatory iron reduction of ferrihydrite

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Iron (hydr)oxides are ubiquitous in soils, playing a dominant geochemical role in surface and subsurface environments. Iron is subject to biotic and abiotic redox transformations, and iron cycling depends on a tight interplay between hydrodynamic transport and (bio)geochemical reactions. In structured soils, solutes move preferentially (by advection) through macropores and slowly (by diffusion) into intra-aggregate micropores, leading to the establishment of redox gradients at the aggregate scale. In the present study, artificial soil aggregates, representing systems of intermediate complexity, were used to study the coupling of physical, chemical, and biological processes affecting spatial heterogeneity in the distribution of secondary mineralization products of ferrihydrite reduction, under environmentally relevant geometries.

We used novel aggregate-based flow-through cell experiments and reactive transport modeling to determine mass transfer and biogeochemical redox controls on the iron transformation ranging from micropore- to aggregate-scales. Constructed aggregates were made of ferrihydrite coated-sand, homogeneously inoculated with *Shewanella putrefaciens* strain CN32, a dissimilatory Fe(III) reducer. Lactate was added in the input solution flowing around the aggregate. The spatial and temporal evolution of secondary mineralization products of dissimilatory ferrihydrite reduction under diffusion-controlled conditions was monitored. The secondary mineralization of ferrihydrite were spatially distinct within the aggregates, owing to diffusively controlled supply of lactate and efflux of Fe(II) and bicarbonate. Goethite and/or lepidocrocite were the dominant secondary minerals, with siderite being present in the interior of the aggregate due to the accumulation of Fe(II) and bicarbonate. Under advective flow,

magnetite has been reported to be the dominant products via dissimilatory ferrihydrite reduction. Our findings indicate that diffusion controls on both electron donor supply and metabolite removal have profound impacts on geochemical products and demonstrate the large spatial and temporal variation in biotransformation of iron within soil aggregates characterized by a transition between advective and diffusive transport domains.