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Multicomponent transport and geochemical reactions under evaporative conditions at the soil/atmosphere interface

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The reactive transport of gas components in the subsurface significantly influences key biogeochemical processes. For instance, reactive transport of oxygen in soil influences mineral dissolution/precipitation and control pore water chemistry. The dynamics of such processes is affected by land-atmosphere interactions and controlled by the exchange processes occurring at the soil/atmosphere interface. One notable example is soil water evaporation that is driven by the exchange of water vapor and energy across the soil/atmosphere interface. This process creates a two-phase system in soil pores and induces a non-linear and complex distribution of the fluid phases (i.e., liquid and gaseous phase) and gas components in the individual phases. The spatiotemporal evolution of the fluid phases and the transport of gas components with and across the phases, in turn, exert important controls on key subsurface biogeochemical processes.

In this study, we explore the impact of evaporation on reactive transport of oxygen in soil using well-controlled laboratory experiments and numerical simulations. We performed a set of evaporation experiments in which an initially water saturated, anoxic soil column containing a layer of pyrite is exposed to a low-humidity atmospheric condition. This resulted in the formation of a partially saturated zone, the invasion of a drying front, and the penetration of oxygen into the porous medium, leading to oxidative dissolution of pyrite. In parallel, we also performed similar experiments under fully water-saturated conditions in order to compare the extent of mineral dissolution with and without evaporation. The spatiotemporal distribution of oxygen was measured using a non-invasive optode technique during the experiments and the concentration of dissolved reaction products (i.e., sulfate, iron and pH) was quantified at the end of the experiments. We developed a non-isothermal multiphase and multicomponent reactive transport model and applied the model to quantitatively interpret the experimental datasets and to understand the coupling between fluid displacement, component transport and geochemical processes.