Impacts of evaporation-recharge cycles on chemical weathering in the unsaturated zone

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The distribution and evolution of mineral–water–gas interfacial areas exert a fundamental yet poorly documented control on mineral–fluid reactions in the unsaturated zone. Mineral-gas-water interfaces are subject to frequent alteration in location and magnitude in the unsaturated zone at the Earth's surface due to periodic evaporation and recharge. Evaporation-recharge cycles can alter the physical pore structure of porous media by redistributing mineral particles and inducing efflorescent precipitation. Although physical impacts of wetting and drying cycles have been recognized, their potential influence on the chemical composition of the pore fluid and mineral dissolution-precipitation reactions are not well understood. Here, we explore the impact of changing mineral-water-gas interfaces during periodic evaporation-recharge cycles on element release from clay minerals common at the Earth's surface. A series of flow-through column experiments was conducted that contained quartz/clay mixtures using either kaolinite, illite, or Fe-rich chlorite (ripidolite). Evaporation was promoted in one set of experiments followed by recharge of one pore volume every second day with a dilute acetic acid solution (0.4 mM, pH = 4). A second set of experiments received the same recharge volume and frequency but evaporation was suppressed. The chemical composition of the column effluent was found to differ significantly between columns subjected to repeated wetting and drying compared to wetting alone. Differences in cation concentrations between columns exposed to wetting-drying cycles and those with wetting only could not be attributed to evapo-concentration alone. Neither the release of different elements from the same mineral, or the release of the same element from different minerals was generalizable across experimental conditions. These results suggest that physico-chemical changes induced by wetting-drying cycles influence reaction rates or pathways, and therefore may be an important control on element fluxes and nutrient cycles in the unsaturated zone.