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A reactive transport approach to understanding elemental and isotopic signatures of cave waters and speleothems

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Speleothem carbon isotope (δ^{13}C) and trace element records hold great potential for reconstructing past changes in Critical Zone response to climate change, such as changes in vegetation, soil respiration, carbon stabilization in deep soils, water infiltration rates, and chemical weathering in the epikarst. However, interpreting time-averaged speleothem records requires integration of observational data and simulation studies that provide a temporal bridge between long and short-term processes. We address this requirement through the development of a novel reactive transport modeling approach capable of simulating surface –to-cave elemental and isotopic transformations. Key questions include how the dissolved inorganic carbon content of seepage water and its isotopic signature vary with soil moisture and fluid flow rate and how these signals are preserved and modified with transit through the epikarst.

To do this, we have adapted the reactive transport model CrunchTope for karst systems. This includes modification of the existing isotopic solid solution model implemented in CrunchTope to accommodate the three-isotope carbon system (^{12}C, ^{13}C, ^{14}C) with concurrent radioactive decay of ^{14}C in both fluid and solid phases. To parameterize the model, we use the chemical, isotopic, and mineralogic properties of the soils and host rocks at Blue Spring Cave in Tennessee. Controlled dissolution experiments on Blue Spring host limestone provide the dissolution rates used in the model. Four years of monitoring data from the surface and cave environment at Blue Spring are used to validate our reactive transport model. Monitoring of drip rates demonstrates that some drips are fed by fracture flow from the surface, some are fed by diffuse flow paths, and others exhibit overflow behaviour. For all drip types, $\delta^{13}C_{DIC}$ is inversely correlated with monthly rainfall. Drip water Sr/Ca and Mg/Ca indicate prior carbonate precipitation occurs in the epikarst, but do not appear to reflect cave ventilation.

Initial results suggest that the CrunchTope model reproduces the observed behavior of drip water elemental concentrations, simulating decreasing Ca concentrations, but increasing Mg and Sr concentrations with decreasing flow rate. This indicates the model is producing changes in secondary carbonate precipitation along the flow path that are flow rate dependent, and supports the control of prior carbonate precipitation on drip water Mg/Ca and Sr/Ca and its relation to hydroclimatic change. The model also highlights a nonlinear relationship between flow rate and the preservation of $\delta^{13}C_{DIC}$ signals originating in the soil. Ongoing work involves further improving model parameterization to better match karst systems with the ultimate goal of producing a tool that can be widely used to provide quantitative interpretations of speleothem proxy records from different climate regimes on a variety of temporal scales.