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Monitoring of crystallization pathways of speleothem fabrics sensitive to hydroclimate: The case of Tham Doun Mai, Laos

Kathleen Johnson (1), Silvia Frisia (2), and Andrea Borsato (2)

(1) Dept. of Earth System Science, University of California, Irvine, CA 92697, United States, (2) SELS, The University of Newcastle, Callaghan, New South Wales, 2308, Australia

The isotopic and elemental composition of speleothem calcite are widely utilized as paleoclimate proxies, though interpretation is often complicated since multiple processes, from atmospheric to within cave, can influence the geochemical signals. Speleothem crystal fabrics are potentially more robust markers for paleohydrogical changes, given that they respond immediately to recharge, regardless of the residence time of water in the aquifer and the mixing of waters from diverse hydrological pathways. To improve understanding of the hydrologic controls on speleothem fabrics and to develop more robust interpretations of geochemical proxy data, we have conducted paleoclimate and cave monitoring research at Tham Doun Mai (TDM) cave in Northern Laos since 2010. Stalagmites from TDM present diverse fabrics, which we interpret as the result of complex crystallization pathways driven by the interplay between drip rate, supersaturation with respect to calcite, and the presence of colloids. We performed 30-minute long precipitation experiments in the cave by immersing a carbon coated TEM grid in drip waters from two locations within the cave, a fast (TM10) and a slow drip (TM5). We then conducted a 3-hour experiment by leaving a drop of the same water on top of a carbon-coated TEM grid during the evening after exiting the cave. Finally, we conducted a 2-year calcite precipitation experiment utilizing a glass plate placed under the TM10 drip site. All samples were analyzed on a JEOL 2100 LaB₆ TEM instrument equipped with STEM and EDS systems at the University of Newcastle, Australia. Results from the 30-minute experiment indicate that the fast TM10 drip carries Si-rich colloids, whose morphology is typical of humic substances (HS). Critically, Sr appears to be associated with Si, and therefore speleothem Sr variations may reflect infiltration of particulate matter. The 3-hour experiment also resulted in silicate colloids, with no calcite nucleation or growth observed, but rather quartz and amorphous silica precipitates. The 2-year experiment yielded a porous calcite fabric with Mg substitution for Ca observed within the coherent lattice. The porous fabric likely results from high concentrations of colloidal and particulate impurities, which hinder calcite growth and are likely associated with faster infiltration. Importantly, these porous fabrics appear more susceptible to diagenesis and increased likelihood of carbon isotope enrichment due to associated degassing. Results from the slower TM5 drip, in contrast, show calcite nanocrystals forming within 30-minutes with no presence of colloidal or particulate material and stalagmites from such drips, are therefore less likely to be affected by diagenesis. Our results demonstrate the importance of combining detailed cave monitoring and speleothem microfabric observations with geochemical measurements to generate more robust speleothem records of past hydrologic change.