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Using stable isotopes ($\delta^{18}O$ and δD) of gypsum hydration water to unravel the mode of gypsum speleothem formation in semi-arid caves

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Subaerial gypsum speleothems form during the evaporation of calcium-sulfate-rich solutions in caves. The evaporation of infiltration water is the widely accepted mechanism to explain precipitation of gypsum speleothems; i.e., the dissolution of gypsum host-rock (e.g. Messinian marine gypsum) supplies Ca^{2+} and SO_4^{2-} ions to cave waters and subsequent evaporation leads to gypsum saturation. However, water condensation actively occurs in caves of semi-arid regions and plays a key role in subaerial cave speleogenesis and recharge of aquifers in low-rainfall environments. To date, water condensation in karstic environments has not been considered as an important factor in gypsum speleothem formation.

We collected speleothem samples from the upper passages of Covadura Cave in the gypsum karst of Sorbas (Almeria, SE Spain). This cave is located in a temperate (annual mean temperature of 19.5°C), semi-arid region (<300 mm yr^{-1}) with a karstic network over 100 km in length. We also sampled condensation water in caves, as well as rainwater and infiltration water (dripwater and water from the main karstic springs draining the Sorbas massif). Furthermore, microclimate parameters (air temperature, relative humidity and effective condensation rate) were monitored over an annual cycle. We analyzed stable isotopes ($\delta^{18}O$ and δD) of gypsum hydration water (GHW) to evaluate the relative importance of condensation and infiltration water in the formation of gypsum speleothems. $\delta^{18}O$ and δD of GHW were used to infer the isotopic composition of the solution from which gypsum precipitated in a variety of speleothems (gypsum coralloid, frostwork, stalactites, crystals formed in the sediment, etc.).

The isotopic values of the initial solution in some samples (e.g. gypsum frostwork and coralloid) were similar to those of the condensation water (-3‰ and -10‰ for $\delta^{18}O$ and δD, respectively); this suggests that they formed from condensation water that was previously enriched in calcium sulfate by dissolution of the gypsum host-rock. Other speleothems (e.g. stalactites, gypsum balls and crusts) lie on an evaporation line whose origin intersects the Local Meteoric Water Line at values between the condensation water and the infiltration water (-5 to -3‰ and -30 to -10‰ for $\delta^{18}O$ and δD, respectively). This second group of speleothems formed from evaporation of a mixture of condensation and infiltration water. Finally, crystals grown in the sediment were precipitated from solutions with a mean isotopic composition of the infiltration water (-5‰ and -35‰ for $\delta^{18}O$ and δD, respectively), within a damp clayey matrix saturated in calcium sulfate with no evaporation.

Measurements on microclimate parameters show that effective condensation occurs when the temperature differences between the external air and the internal cave atmosphere are greatest (June to December). In contrast, evaporation becomes the dominant mechanism as the external and internal temperatures converge (January to May). This suggests that most gypsum speleothems in Covadura Cave precipitate during the coldest months by evaporation of condensation water that deposited on the cave surfaces during the warmer period of the year, with varying contributions of infiltration water. This finding has important implications for further studies of gypsum speleothems as paleoenvironmental proxies.