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Recrystallization-induced oxygen isotope changes in inclusion-hosted water of speleothems – paleoclimatological implications

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Stable hydrogen and oxygen isotope data of water trapped in fluid inclusions were collected for recently forming stalagmites and flowstones in order to determine how dripwater compositions are reflected and preserved in the inclusion water compositions. The samples were collected from different cave sites (with temperatures around 10 ± 1 °C) from the central and north-eastern parts of Hungary. Hydrogen isotope compositions were found to reflect dripwater values, whereas the oxygen isotope data were increasingly shifted from the local dripwater compositions with the time elapsed after deposition. The δ18O data are correlated with X-Ray diffraction full width at half maximum values (related to crystal domain size and lattice strain), suggesting that the oxygen isotope shift is related to recrystallization of calcite. Transmission electron microscope analyses detected the presence of nanocrystalline (<50 nm) calcite, whose crystallization to coarser-grained calcite crystals (>200 nm) may have induced re-equilibration between the carbonate and the trapped inclusion water. Additional data indicated that amorphous calcium carbonate (ACC) may have formed as a precursor of nanocrystalline calcite. ACC-calcite transformation followed by Ostwald ripening process provides an explanation for unexpectedly low oxygen isotope compositions in the inclusion water, especially in cold caves where carbonate may form first as an amorphous phase.
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