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Amorphous calcium magnesium carbonates: key precursors in the formation of low-temperature dolomite

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Understanding the formation of dolomite under Earth surface conditions remains a long strongly debated issue in geology and carbonate geochemistry. This challenge has broad implications for paleoenvironmental reconstruction, hydrocarbon reservoir evaluation, carbon capture strategies, and biomineralization studies.

In surface environments, carbonate precipitation is typically dominated by calcite and aragonite. However, the relative scarcity of dolomite in modern settings, contrasting with its abundance in the ancient sedimentary record, together with the difficulty of synthesizing ordered dolomite at near-surface temperatures in laboratory experiments, are some of the main issues of the long-standing "*dolomite problem"*. Despite decades of debate, the physicochemical mechanisms controlling low-*T* formation of dolomite remain incompletely understood. Recent studies have proposed that protodolomite (at low *T*) and structurally ordered dolomite (at high *T*) can form via a non-classical pathway involving an amorphous Ca-Mg carbonate (ACMC) precursor. Although this crystallization mechanism has been reproduced in laboratory experiments through both abiotic and microbially mediated processes, it has not yet been reported in natural environments.

Here, we report the formation of poorly ordered dolomite (protodolomite) and ordered dolomite in close association with ACMC within moonmilk deposits from the Cueva de Nerja (Spain). Our results indicate that (i) these amorphous carbonate phases play a critical role as precursors in the formation of dolomite; and (ii) dolomite crystallization is controlled by the dynamic interaction among fluctuations in drip-water chemistry, cyclic precipitation–dissolution of huntite, which serves as Mg reservoir, and the stabilization and subsequent crystallization of amorphous carbonate phases mediated by dissolved silica and microbially derived organic compounds. This study provides new insights into alternative low-*T* formation pathways for dolomite in sedimentary environments.