Goldschmidt 2023 Abstract
https://doi.org/10.7185/gold2023.18601

Diagenetic alteration of carbonate clumped isotope composition in Himalayan foreland deposits

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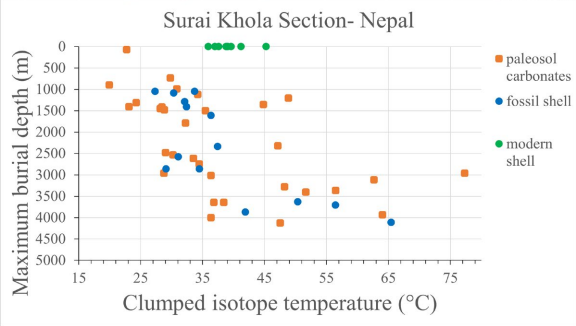

Clumped isotope-derived temperatures of various carbonate phases can be powerful tools for reconstructing past environmental signals such as surface and animal body temperatures, as well as calculating the oxygen isotope composition of ancient waters. However, diagenesis can alter clumped temperatures without affecting the carbon and oxygen isotopic composition of the samples. Such alteration can, in theory, be corrected using theoretical models when a thermal history is known. Here, we utilize a new laser spectroscopic system for clumped isotope analysis to produce a large dataset (201 samples, 621 analyses) of modern and fossil soil carbonates, bivalve shells, tooth enamel, eggshell and carbonate cemented sandstones from Himalayan foreland deposits in Nepal and Pakistan with well-known thermal histories. We use these to 1) establish the conditions required to alter primary clumped isotope composition in these phases, and 2) test whether the two latest theoretical models accurately predict the magnitude of reordering in these phases.

Our results indicate that soil carbonate temperatures were altered by burial to 80-110°C for less than 5Myr. Aragonitic shells appear to reorder at a similar rate to calcitic soil carbonates and eggshells and were not converted to calcite. Tooth enamel samples from Pakistan did not consistently produce realistic body temperatures even in samples that experienced only shallow burial. Carbonate cemented sandstone, thought to cement at shallow depth, produced clumped isotope temperatures of 30-60°C with no coherent depth-temperature pattern, with several samples yielding 70-90°C possibly due to detrital contamination. Based on soil carbonate samples from the section with the best-constrained thermal history (Surai Khola, Nepal), both the theoretical model of Stolper and Eiler [1] and that of Hemingway and Henkes [2] underpredict the degree of alteration in some samples, though the Stolper and Eiler model is closer to the observed values. This could be due to faster reordering mediated by internal water or organic matter in the carbonates, an effect that may not be captured in heating experiments on optical calcite, on which the models currently rely.

[1] Stolper and Eiler (2015) *American Journal of Science* 315, 363–411

[2] Hemmingway and Henkes (2021) *Earth and Planetary Science Letters* 566, 116962