

Continuous reequilibration of C isotopes in natural calcite at 25°C

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The carbon isotope composition in carbonate minerals has been used in the past in order to distinguish processes such as biological activity and respiration.

Recently however, it has been experimentally shown that C isotope signatures in hydrous Mg-carbonates undergo re-equilibration with the ambient solution [1] similar to the behavior of Mg isotopes in Mg-bearing carbonates [2] [3]. In order to test the validity of such findings in CaCO₃ minerals, we conducted a series of experiments where calcite equilibrated with a C rich fluid at 25°C.

Experiment comprised of multiple replicates of glass reactors containing 0.5g of natural calcite (<50μ) powder and ~20 ml of NaHCO₃ or Na₂CO₃ solution. The vials were filled to the top and sealed air tight with Teflon coated caps. During the sampling, complete recovery of the fluid and solid from each reactor at different time steps along 7 months occurred and C isotopes were measured in solid and liquid samples.

Temporal evolution of Ca concentration indicates that initial calcite dissolved and re-precipitated attaining steady-state equilibrium value consistent with calcite solubility at the given T and pH. δC¹³DIC increased systematically over time from an initial value of -4.5‰ to +2.25‰. The corresponding δC¹³_{solid} exhibited a decrease of -0.18±0.03. Δ¹³Ca_{Calcite-DIC} ranged between 6.6 and 4.2. With only a 0.002% of the calcite dissolved, solid exhibits a net change in δC¹³ of 0.18‰ indicating that isotopic signature might not exactly reflect the original formation conditions of the crystal.

[1] Mavromatis *et al* (2015) *Chem. Geol.*, doi:10.1016

[2] Pearce *et al* (2012) *Geochim. Cosmochim. Acta*, **92**, 170-183.

[3] Mavromatis *et al* (2012) *Geochim. Cosmochim. Acta*, **76**, 161–174.