

## SUPPLEMENTAL MATERIALS

### Local Equilibrium Model Details

The following set of commands was compiled using React version 5.0.6 to model local equilibrium (Bethke, 1996) behavior in the terrestrial spring system:

```

temperature = 73.25
1 kg H2O
291.69 mg/l Ca++
3.58 mg/l B(OH)3
.072 mg/l Ba++
1.6 mg/l Sr++
65 mg/l Mg++
balance on Na+
117 mg/l Na+
50.2 mg/l K+
.02 mg/l Fe++
.026 mg/l Mn++
1.09 mg/l As(OH)4-
24 mg/l SiO2(aq)
5.99 mmolar SO4--
.02155 molality HCO3-
5.06 mmolar Cl-
.159 mmolar F-
4.7 umolar NO3-
6.14 pH
.02 mg/l Al+++
slide fugacity of CO2(g) .02388

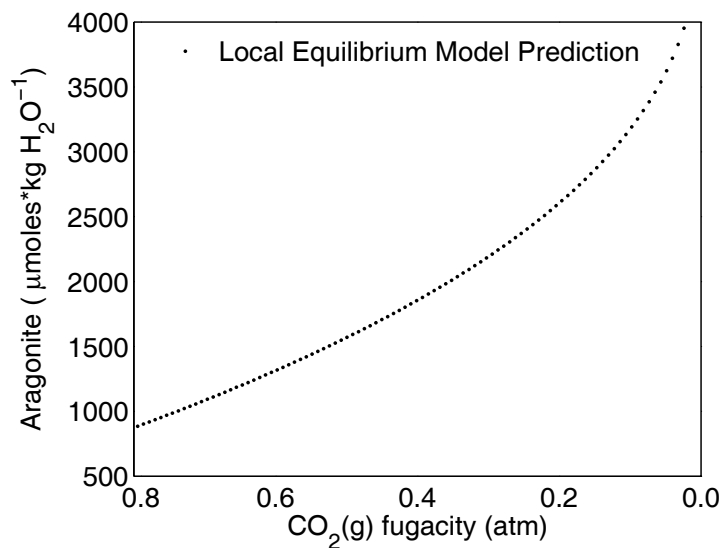
```

```

suppress Dolomite Calcite Dolomite-dis Dolomite-ord
suppress Strontianite Muscovite Fluorite Talc
suppress "Maximum Microcline" Barite Kaolinite Antigorite
suppress Saponite-Mg Beidellit-Mg Beidellit-K Beidellit-Na
suppress Chrysotile Beidellit-Ca Illite Saponite-Ca
suppress Gibbsite Saponite-K Saponite-Na Saponite-H
suppress Phengite Phlogopite Clinocl-14A

```

All supersaturated minerals not known to be present in the portion of Spring AT-3 analyzed in this study were suppressed to facilitate the comparison between model predicted precipitation and our observations of aragonite precipitation in the natural system. Model predicted aragonite precipitation is plotted in Figure S1 as a function of CO<sub>2</sub>(g) fugacity.



**Figure S1.** Local equilibrium model predicted dependence of total aragonite precipitation on the fugacity of  $\text{CO}_2(\text{g})$  in the natural system. Model represents open system precipitation resulting from solely  $\text{CO}_2(\text{g})$  exsolution across the natural system transect (Fig. 1).

TABLE S1. RESULTANT  $k_f \bullet k_c^{-1}$  VALUES CALCULATED USING EQUATION 1 OVER THE MODELED RANGE OF N FOR THE FILTRATION ( $n_f$ ) AND NATURAL CONTROL ( $n_c$ ) TREATMENTS

$n_c$	1.0	1.5	2.0	2.5	3.0	3.5	4.0
$n_f$							
1.0	0.48	0.78	1.3	2.1	3.3	5.4	8.8
1.5	0.30	0.48	0.78	1.3	2.1	3.3	5.4
2.0	0.18	0.30	0.48	0.78	1.3	2.1	3.3
2.5	0.11	0.18	0.30	0.48	0.78	1.3	2.1
3.0	0.070	0.11	0.18	0.30	0.48	0.78	1.3
3.5	0.043	0.070	0.11	0.18	0.30	0.48	0.78
4.0	0.026	0.043	0.070	0.11	0.18	0.30	0.48

### Additional Information Regarding Figure 8

#### *Converting Reactive Surface Area (cm<sup>2</sup>) to Geometric Area (cm<sup>2</sup><sub>geometric</sub>)*

Using the mean natural control precipitation rate – (15.0 mol CaCO<sub>3</sub> • 10<sup>10</sup> • min<sup>-1</sup> • cm<sup>-2</sup>) as an example:

$$\underbrace{\frac{1.50 \cdot 10^{-9} \text{ mol CaCO}_3}{\text{min} \cdot \text{cm}^2}}_{\text{mean precipitation rate}} \cdot \underbrace{964 \text{ cm}^2}_{\text{empirically determined mean reactive surface area of aragonite on a } 38.71 \text{ cm}^2 \text{ Na}_2\text{CO}_3 \text{ precipitation substrate}^*} \cdot \underbrace{\frac{1}{38.71 \text{ cm}^2_{\text{geometric}}}}_{\text{geometric area of a Na}_2\text{CO}_3 \text{ precipitation substrate}} = \underbrace{\frac{3.73 \cdot 10^{-8} \text{ mol CaCO}_3}{\text{min} \cdot \text{cm}^2_{\text{geometric}}}}_{\text{mean precipitation rate (expressed using geometric area)}}$$

\* Please see the Reactive Surface Area Determination section of the text for additional details

#### *Converting Precipitation Rates to Accumulation Rates (cm • yr<sup>-1</sup>)*

$$\frac{3.73 \cdot 10^{-8} \text{ mol CaCO}_3}{\text{min} \cdot \text{cm}^2_{\text{geometric}}} \cdot \frac{34.16 \text{ cm}^3 \text{ CaCO}_3}{\text{mol aragonite}} = \frac{1.27 \cdot 10^{-6} \text{ cm}^3 \text{ CaCO}_3}{\text{min} \cdot \text{cm}^2_{\text{geometric}}} = \beta$$

$$\left[ \beta + \frac{\beta \phi}{(1 - \phi)} \right] \cdot \frac{525960 \text{ min}}{\text{yr}} = \frac{0.74 \text{ cm}}{\text{yr}} \text{ when porosity } (\phi) \text{ is set equal to } 0.1.$$

#### *Assumptions Inherent to Accumulation Rate Calculations*

Modeling accumulation rates in this manner relies upon the assumptions that: (1) no compaction occurs during sediment accumulation; (2) the geometry of the plane upon which CaCO<sub>3</sub> precipitates is invariant over time and can therefore be projected in the vertical direction; and (3) the development of sediment porosity is unaffected by changes in the concentration of microbial biomass on the CaCO<sub>3</sub> mineral surface. Measuring rates of cement accumulation, reconstructing saturation state and quantifying porosity along with compaction requires none of these assumptions and permits CaCO<sub>3</sub> precipitation rates to be determined directly from sedimentary sections.

**Supplemental Materials References Cited**

Bethke, C.M., 1996, *Geochemical Reaction Modeling: Concepts and Applications*: New York, Oxford University Press.