Supporting Information for:

Balance and imbalance in biogeochemical cycles reflect the operation of closed, exchange, and open sets

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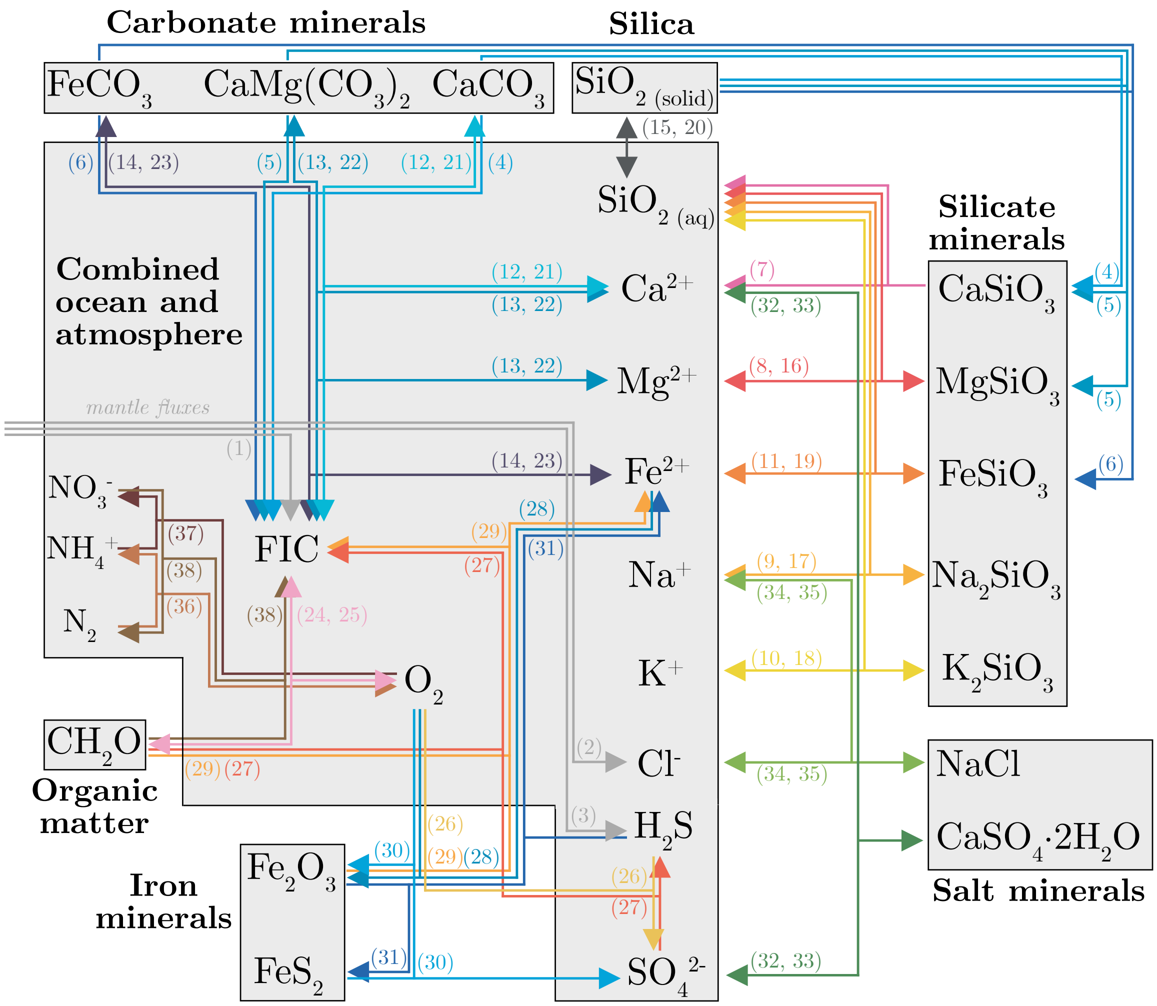
S1: Model architecture



Process list:

1. Mantle CO_2 degassing
2. Mantle HCl degassing
3. Mantle H_2S degassing
4. Calcite metamorphism
5. Dolomite metamorphism
6. Siderite metamorphism
7. Ca-silicate weathering
8. Mg-silicate weathering
9. Na-silicate weathering
10. K-silicate weathering
11. Fe-silicate weathering
12. Calcite weathering
13. Dolomite weathering
14. Siderite weathering
15. Silica weathering
16. Mg-reverse weathering
17. Na-reverse weathering
18. K-reverse weathering
19. Fe-reverse weathering
20. Silica formation
21. Calcite formation
22. Dolomite formation
23. Siderite formation
24. Oxygenic photosynthesis
25. Aerobic respiration
26. Sulfide oxidation
27. Sulfate reduction
28. Ferrous iron oxidation
29. Ferric iron reduction
30. Pyrite oxidation
31. Pyrite formation
32. Gypsum dissolution
33. Gypsum formation
34. Halite dissolution
35. Halite formation
36. Nitrogen fixation
37. Nitrification
38. Denitrification

Fig. S1: Model architecture with reservoirs and labeled biogeochemical fluxes. Double-headed arrows reflect mass fluxes in both directions. ALK are H_2O are omitted for clarity.

S2: Derivation of sets

We used the MATLAB function *null* with the *rational* option to calculate the null spaces of $\boldsymbol{A_{closed}}$, $\boldsymbol{A_{exchange}}$, and $\boldsymbol{A_{open}}$. The default $\boldsymbol{A_{closed}}$ null space returned by MATLAB was a matrix with 38 rows and 18 columns (Figs. S2). This result means that there are 18 non-zero linearly independent vectors $(\vec{\psi}_1 - \vec{\psi}_{18})$ that, when multiplied by $\boldsymbol{A_{closed}}$, generate $\vec{0}$ (with dimensions of 38 x 1). It is acceptable for our purposes that the $\vec{\psi}_i$ vectors are not orthonormal. However, the $\vec{\psi}_i$ vectors contain negative values, which is unphysical because the biogeochemical processes in Table 1 of the main text were defined to proceed only with positive magnitude. As a result, an alternative basis had to be constructed that only contained positive reaction rates. Because linear combinations of the $\vec{\psi}_i$ are also contained within the null space of $\boldsymbol{A_{closed}}$, we combined the initial basis vectors to reach a new basis containing only positive terms. The new basis vectors for the $\boldsymbol{A_{closed}}$ null space define the closed sets (eqs. S1-S18). Of the 18 new basis vectors, 11 were equivalent to $\vec{\psi}_i$ from the original basis and 7 were reached by linear combination of the $\vec{\psi}_i$. The new basis vectors are indicated as $\vec{x}_1$ through $\vec{x}_{18}$, are written out in Fig. 2, and are visualized in Fig. 3.

The default $\boldsymbol{A_{exchange}}$ null space returned by MATLAB was a matrix with 38 rows and 25 columns (Fig. S3). This result means that there are 25 non-zero linearly independent vectors $(\vec{\phi}_1 - \vec{\phi}_{25})$ that, when multiplied by $\boldsymbol{A_{exchange}}$, generate $\vec{0}$ (with dimensions of 15 x 1). As with the $\vec{\psi}_i$, the $\vec{\phi}_i$ contained negative values but were combined to reach a viable basis spanning the same null space. It is important to note that the 18 basis vectors of the $\boldsymbol{A_{closed}}$ null space $(\vec{x}_1 - \vec{x}_{18})$ are also contained within the $\boldsymbol{A_{exchange}}$ null space. As a result, $\vec{x}_1$ through $\vec{x}_{18}$ can be calculated from the $\vec{\phi}_i$ vectors (eqs. S19 – S36). However, 7 additional vectors exist in the null space of

$\boldsymbol{A_{exchange}}$ that were not in the null space of $\boldsymbol{A_{closed}}$. These 7 vectors are the exchange sets, and they can be arbitrarily chosen so long as they are linearly independent of one another and of $\vec{x}_1$ through $\vec{x}_{18}$. We have thus defined 7 additional vectors ($\vec{x}_{19} - \vec{x}_{25}$), as linear combinations of the $\vec{\phi}_i$ (eqs. S37 -S43). These exchange sets are written out in Fig. 2 and visualized in Fig. 4 of the main text. As required, a matrix comprised of $\vec{x}_1$ through $\vec{x}_{25}$ has rank 25.

Five additional exchange reactions were given in Table 2. These reactions were derived from linear combinations of the closed sets ($\vec{x}_1 - \vec{x}_{18}$) and exchange sets ($\vec{x}_{19} - \vec{x}_{25}$) with weightings described by α_i values (Fig. 2). The equations below detail how a subset of these additional reactions are generated from the $\vec{x}_i$ vectors (eqs. S44 - S46).

The default $\boldsymbol{A_{open}}$ null space returned by MATLAB was a matrix with 38 rows and 36 columns (Fig. S4). This result means that there are 36 non-zero linearly independent vectors $\left(\vec{\xi}_1 - \vec{\xi}_{36}\right)$ that, when multiplied by $\boldsymbol{A_{open}}$, generate $\vec{0}$ (with dimensions of 2 x 1). As with the $\vec{\psi}_i$ and the $\vec{\phi}_i$, the $\vec{\xi}_i$ contained negative values but could be combined to reach a viable basis spanning the same null space. Again, note that the 18 basis vectors of the $\boldsymbol{A_{closed}}$ null space ($\vec{x}_1 - \vec{x}_{18}$) and the 7 additional basis vectors of the $\boldsymbol{A_{exchange}}$ null space ($\vec{x}_{19} - \vec{x}_{25}$) are also contained within the $\boldsymbol{A_{open}}$ null space. As a result, $\vec{x}_1$ through $\vec{x}_{25}$ can be calculated from the $\vec{\xi}_i$ (eqs. S47 – S71). However, 11 additional vectors exist in the null space of $\boldsymbol{A_{open}}$ that were not in the null space of $\boldsymbol{A_{exchange}}$. These 11 vectors are the open sets, and they can be arbitrarily chosen as long as they are linearly independent of one another and of $\vec{x}_1$ through $\vec{x}_{25}$. We thus defined 11 additional linearly independent vectors ($\vec{x}_{26} - \vec{x}_{36}$), 7 of which were equivalent to $\vec{\xi}_i$ vectors and 4 of which were linear combinations of $\vec{\xi}_i$ vectors (eqs. S72-S82). The new basis vectors are written out in Fig. 2. As required, a matrix comprised of $\vec{x}_1$ through $\vec{x}_{36}$ has rank 36.

Two additional open reactions were given in Table 2 of the main text. These reactions were derived from linear combinations of the closed sets ($\vec{x}_1 - \vec{x}_{18}$), exchange sets ($\vec{x}_{19} - \vec{x}_{25}$), and open sets ($\vec{x}_{26} - \vec{x}_{36}$) with weightings described by α_i values. The equations below detail the exact α_i values and how these additional reactions are generated from the $\vec{x}_i$ vectors (eqs. S83 – S84). In addition, the reactions corresponding to $\vec{x}_{29}$ through $\vec{x}_{36}$ are provided (eqs. S85 – S92).

$$\overset{\vec{\boldsymbol{x}}}{\begin{bmatrix} x_{\text{Mantle CO2}}^{(\#1)} \\ x_{\text{Mantle HCl}}^{(\#2)} \\ x_{\text{Mantle H2S}}^{(\#3)} \\ x_{\text{Calcite metamorphism}}^{(\#4)} \\ x_{\text{Dolomite metamorphism}}^{(\#5)} \\ x_{\text{Siderite metamorphism}}^{(\#6)} \\ x_{\text{Ca-silicate weathering}}^{(\#7)} \\ x_{\text{Mg-silicate weathering}}^{(\#8)} \\ x_{\text{Na-silicate weathering}}^{(\#9)} \\ x_{\text{K-silicate weathering}}^{(\#10)} \\ x_{\text{Fe-silicate weathering}}^{(\#11)} \\ x_{\text{Calcite weathering}}^{(\#12)} \\ x_{\text{Dolomite weathering}}^{(\#13)} \\ x_{\text{Siderite weathering}}^{(\#14)} \\ x_{\text{Silica weathering}}^{(\#15)} \\ x_{\text{Mg-reverse weathering}}^{(\#16)} \\ x_{\text{Na-reverse weathering}}^{(\#17)} \\ x_{\text{K-reverse weathering}}^{(\#18)} \\ x_{\text{Fe-reverse weathering}}^{(\#19)} \\ x_{\text{Silica formation}}^{(\#20)} \\ x_{\text{Calcite formation}}^{(\#21)} \\ x_{\text{Dolomite formation}}^{(\#22)} \\ x_{\text{Siderite formation}}^{(\#23)} \\ x_{\text{Oxygenic photosynthesis}}^{(\#24)} \\ x_{\text{Aerobic respiration}}^{(\#25)} \\ x_{\text{Sulfide oxidation}}^{(\#26)} \\ x_{\text{Sulfate reduction}}^{(\#27)} \\ x_{\text{Ferrous iron oxidation}}^{(\#28)} \\ x_{\text{Ferric iron reduction}}^{(\#29)} \\ x_{\text{Pyrite oxidation}}^{(\#30)} \\ x_{\text{Pyrite formation}}^{(\#31)} \\ x_{\text{Gypsum dissolution}}^{(\#32)} \\ x_{\text{Gypsum formation}}^{(\#33)} \\ x_{\text{Halite dissolution}}^{(\#34)} \\ x_{\text{Halite formation}}^{(\#35)} \\ x_{\text{Nitrogen fixation}}^{(\#36)} \\ x_{\text{Nitrification}}^{(\#37)} \\ x_{\text{Denitrification}}^{(\#38)} \end{bmatrix}} = \overset{\boldsymbol{A}_{closed}\ \text{null space original basis}}{\begin{array}{c} \begin{array}{cccccccccccccccccc} \vec{\psi}_1 & \vec{\psi}_2 & \vec{\psi}_3 & \vec{\psi}_4 & \vec{\psi}_5 & \vec{\psi}_6 & \vec{\psi}_7 & \vec{\psi}_8 & \vec{\psi}_9 & \vec{\psi}_{10} & \vec{\psi}_{11} & \vec{\psi}_{12} & \vec{\psi}_{13} & \vec{\psi}_{14} & \vec{\psi}_{15} & \vec{\psi}_{16} & \vec{\psi}_{17} & \vec{\psi}_{18} \end{array} \\ \begin{bmatrix} 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 2 & 1 & -1 & 0 & 0 & 0 & 0 & 1 & 0 & -2 & -1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ -1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 1 & 1 & -1 & 0 & 0 & 0 & 0 & 1 & 0 & -1 & -1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ -1 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ -2 & -1 & 1 & 0 & 0 & 0 & 0 & -1 & 1 & 2 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 2 & 1 & 0 & 0 & 0 & 5 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 & -2 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 1/4 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1/4 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 4 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 \end{bmatrix} \end{array}} \begin{bmatrix} {}^{\psi}\omega_1 \\ {}^{\psi}\omega_2 \\ {}^{\psi}\omega_3 \\ {}^{\psi}\omega_4 \\ {}^{\psi}\omega_5 \\ {}^{\psi}\omega_6 \\ {}^{\psi}\omega_7 \\ {}^{\psi}\omega_8 \\ {}^{\psi}\omega_9 \\ {}^{\psi}\omega_{10} \\ {}^{\psi}\omega_{11} \\ {}^{\psi}\omega_{12} \\ {}^{\psi}\omega_{13} \\ {}^{\psi}\omega_{14} \\ {}^{\psi}\omega_{15} \\ {}^{\psi}\omega_{16} \\ {}^{\psi}\omega_{17} \\ {}^{\psi}\omega_{18} \end{bmatrix}$$

Fig. S2: The basis $(\vec{\psi}_i)$ calculated in MATLAB for the null space of $\boldsymbol{A_{closed}}$. Because the biogeochemical processes are defined to run unidirectionally, negative values in the matrix represent unphysical outcomes. The $\vec{\psi}_i$ vectors were used to generate a new physically allowable basis set for the same space ($\vec{x}_1 - \vec{x}_{18}$). The ${}^{\psi}\omega_i$ values refer to the magnitude of each vector.

Defining $\vec{x}_1$ to $\vec{x}_{18}$ with $\vec{\psi}_i$ (closed sets):

$\vec{x}_1 = \vec{\psi}_8 + \vec{\psi}_9$ (eq. S1)

$\vec{x}_2 = 2\vec{\psi}_8 + \vec{\psi}_{10}$ (eq. S2)

$\vec{x}_3 = \vec{\psi}_8 + \vec{\psi}_{11}$ (eq. S3)

$\vec{x}_4 = \vec{\psi}_9$ (eq. S4)

$\vec{x}_5 = \vec{\psi}_1 + \vec{\psi}_{10}$ (eq. S5)

$\vec{x}_6 = \vec{\psi}_2 + \vec{\psi}_{11}$ (eq. S6)

$\vec{x}_7 = \vec{\psi}_4$ (eq. S7)

$\vec{x}_8 = \vec{\psi}_5$ (eq. S8)

$\vec{x}_9 = \vec{\psi}_6$ (eq. S9)

$\vec{x}_{10} = \vec{\psi}_7$ (eq. S10)

$\vec{x}_{11} = \vec{\psi}_{12}$ (eq. S11)

$\vec{x}_{12} = \vec{\psi}_{14}$ (eq. S12)

$\vec{x}_{13} = \vec{\psi}_{13}$ (eq. S13)

$\vec{x}_{14} = 4\vec{\psi}_{15} + 8\vec{\psi}_{13}$ (eq. S14)

$\vec{x}_{15} = \vec{\psi}_{18}$ (eq. S15)

$\vec{x}_{16} = \vec{\psi}_{16}$ (eq. S16)

$\vec{x}_{17} = \vec{\psi}_{17}$ (eq. S17)

$\vec{x}_{18} = \vec{\psi}_3 + \vec{\psi}_8$ (eq. S18)

$\vec{x}$ $\boldsymbol{A}_{\boldsymbol{exchange}}$ null space original basis

$$
\begin{bmatrix}
x_{\text{Mantle CO2}}\,(\#1)\\
x_{\text{Mantle HCl}}\,(\#2)\\
x_{\text{Mantle H2S}}\,(\#3)\\
x_{\text{Calcite metamorphism}}\,(\#4)\\
x_{\text{Dolomite metamorphism}}\,(\#5)\\
x_{\text{Siderite metamorphism}}\,(\#6)\\
x_{\text{Ca-silicate weathering}}\,(\#7)\\
x_{\text{Mg-silicate weathering}}\,(\#8)\\
x_{\text{Na-silicate weathering}}\,(\#9)\\
x_{\text{K-silicate weathering}}\,(\#10)\\
x_{\text{Fe-silicate weathering}}\,(\#11)\\
x_{\text{Calcite weathering}}\,(\#12)\\
x_{\text{Dolomite weathering}}\,(\#13)\\
x_{\text{Siderite weathering}}\,(\#14)\\
x_{\text{Silica weathering}}\,(\#15)\\
x_{\text{Mg-reverse weathering}}\,(\#16)\\
x_{\text{Na-reverse weathering}}\,(\#17)\\
x_{\text{K-reverse weathering}}\,(\#18)\\
x_{\text{Fe-reverse weathering}}\,(\#19)\\
x_{\text{Silica formation}}\,(\#20)\\
x_{\text{Calcite formation}}\,(\#21)\\
x_{\text{Dolomite formation}}\,(\#22)\\
x_{\text{Siderite formation}}\,(\#23)\\
x_{\text{Oxygenic photosynthesis}}\,(\#24)\\
x_{\text{Aerobic respiration}}\,(\#25)\\
x_{\text{Sulfide oxidation}}\,(\#26)\\
x_{\text{Sulfate reduction}}\,(\#27)\\
x_{\text{Ferrous iron oxidation}}\,(\#28)\\
x_{\text{Ferric iron reduction}}\,(\#29)\\
x_{\text{Pyrite oxidation}}\,(\#30)\\
x_{\text{Pyrite formation}}\,(\#31)\\
x_{\text{Gypsum dissolution}}\,(\#32)\\
x_{\text{Gypsum formation}}\,(\#33)\\
x_{\text{Halite dissolution}}\,(\#34)\\
x_{\text{Halite formation}}\,(\#35)\\
x_{\text{Nitrogen fixation}}\,(\#36)\\
x_{\text{Nitrification}}\,(\#37)\\
x_{\text{Denitrification}}\,(\#38)
\end{bmatrix}
=
\begin{array}{c}
\vec{\phi}_1\;\vec{\phi}_2\;\vec{\phi}_3\;\vec{\phi}_4\;\vec{\phi}_5\;\vec{\phi}_6\;\vec{\phi}_7\;\vec{\phi}_8\;\vec{\phi}_9\;\vec{\phi}_{10}\;\vec{\phi}_{11}\;\vec{\phi}_{12}\;\vec{\phi}_{13}\;\vec{\phi}_{14}\;\vec{\phi}_{15}\;\vec{\phi}_{16}\;\vec{\phi}_{17}\;\vec{\phi}_{18}\;\vec{\phi}_{19}\;\vec{\phi}_{20}\;\vec{\phi}_{21}\;\vec{\phi}_{22}\;\vec{\phi}_{23}\;\vec{\phi}_{24}\;\vec{\phi}_{25}\\
\begin{bmatrix}
-1&-2&-1&0&0 & -1&0&0&0&0 & 1&0&0&0&0 & 0&-3&3&-1&1 & -1&1&1/2&-1/2&0\\
0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&-1&1&0\\
0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&-8&2 & -1&1&0&0&0\\
1&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0\\
0&1&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0\\
0&0&1&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0\\
0&0&0&1&1 & -1&0&0&0&0 & 1&0&-1&-1&0 & 0&-4&4&0&1 & 0&0&1/2&-1/2&0\\
0&0&0&-1&0 & 0&1&0&0&0 & 0&0&1&0&0 & 0&0&0&0&0 & 0&0&0&0&0\\
0&0&0&0&0 & 0&0&1&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&-1/2&1/2&0\\
0&0&0&0&0 & 0&0&0&1&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0\\
0&0&0&0&-1 & 0&0&0&0&1 & 0&0&0&1&0 & 0&4&-4&0&-1 & 0&0&0&0&0\\
0&0&0&-2&-1 & 1&0&0&0&0 & -1&1&2&1&0 & 0&4&-4&0&-1 & -1&1&-1/2&1/2&0\\
0&0&0&1&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0\\
0&0&0&0&1 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0\\
0&0&0&0&0 & 1&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0\\
0&0&0&0&0 & 0&1&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0\\
0&0&0&0&0 & 0&0&1&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0\\
0&0&0&0&0 & 0&0&0&1&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0\\
0&0&0&0&0 & 0&0&0&0&1 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0\\
0&0&0&0&0 & 0&0&0&0&0 & 1&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0\\
0&0&0&0&0 & 0&0&0&0&0 & 0&1&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0\\
0&0&0&0&0 & 0&0&0&0&0 & 0&0&1&0&0 & 0&0&0&0&0 & 0&0&0&0&0\\
0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&1&0 & 0&0&0&0&0 & 0&0&0&0&0\\
0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&1 & 2&1&0&-1&0 & -2&2&0&0&5\\
0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&1 & 0&0&0&0&0 & 0&0&0&0&0\\
0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 1&0&0&-8&0 & -1&1&0&0&0\\
0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 1&0&0&0&0 & 0&0&0&0&0\\
0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&1&0&0&0 & 0&0&0&0&0\\
0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&1&0&0 & 0&0&0&0&0\\
0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&1&0 & 0&0&0&0&0\\
0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&1 & 0&0&0&0&0\\
0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 1&0&0&0&0\\
0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&1&0&0&0\\
0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&1&0&0\\
0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&1&0\\
0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&1\\
0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&4\\
0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&0 & 0&0&0&0&1
\end{bmatrix}
\end{array}
\begin{bmatrix}
{}^{\phi}\omega_1\\ {}^{\phi}\omega_2\\ {}^{\phi}\omega_3\\ {}^{\phi}\omega_4\\ {}^{\phi}\omega_5\\ {}^{\phi}\omega_6\\ {}^{\phi}\omega_7\\ {}^{\phi}\omega_8\\ {}^{\phi}\omega_9\\ {}^{\phi}\omega_{10}\\ {}^{\phi}\omega_{11}\\ {}^{\phi}\omega_{12}\\ {}^{\phi}\omega_{13}\\ {}^{\phi}\omega_{14}\\ {}^{\phi}\omega_{15}\\ {}^{\phi}\omega_{16}\\ {}^{\phi}\omega_{17}\\ {}^{\phi}\omega_{18}\\ {}^{\phi}\omega_{19}\\ {}^{\phi}\omega_{20}\\ {}^{\phi}\omega_{21}\\ {}^{\phi}\omega_{22}\\ {}^{\phi}\omega_{23}\\ {}^{\phi}\omega_{24}\\ {}^{\phi}\omega_{25}
\end{bmatrix}
$$

Fig. S3: The basis $(\vec{\phi}_i)$ calculated in MATLAB for the null space of $\boldsymbol{A}_{\boldsymbol{exchange}}$. Because the biogeochemical processes are defined to run unidirectionally, negative values in the matrix represent unphysical outcomes. The $\vec{\phi}_i$ vectors were used to generate a new physically allowable basis set for the same space ($\vec{x}_1 - \vec{x}_{25}$). The ${}^{\phi}\omega_i$ values refer to the magnitude of each vector.

Defining $\vec{x}_1$ to $\vec{x}_{18}$ with $\vec{\phi}_i$ (closed sets):

$$\vec{x}_1 = \vec{\phi}_1 + \vec{\phi}_{11} + \vec{\phi}_{12} \quad \text{(eq. S19)}$$
$$\vec{x}_2 = \vec{\phi}_2 + 2\vec{\phi}_{11} + \vec{\phi}_{13} \quad \text{(eq. S20)}$$
$$\vec{x}_3 = \vec{\phi}_3 + \vec{\phi}_{11} + \vec{\phi}_{14} \quad \text{(eq. S21)}$$
$$\vec{x}_4 = \vec{\phi}_{12} \quad \text{(eq. S22)}$$
$$\vec{x}_5 = \vec{\phi}_4 + \vec{\phi}_{13} \quad \text{(eq. S23)}$$
$$\vec{x}_6 = \vec{\phi}_5 + \vec{\phi}_{14} \quad \text{(eq. S24)}$$
$$\vec{x}_7 = \vec{\phi}_7 \quad \text{(eq. S25)}$$
$$\vec{x}_8 = \vec{\phi}_8 \quad \text{(eq. S26)}$$
$$\vec{x}_9 = \vec{\phi}_9 \quad \text{(eq. S27)}$$
$$\vec{x}_{10} = \vec{\phi}_{10} \quad \text{(eq. S28)}$$
$$\vec{x}_{11} = \vec{\phi}_{15} \quad \text{(eq. S29)}$$
$$\vec{x}_{12} = \vec{\phi}_{17} + \vec{\phi}_{18} \quad \text{(eq. S30)}$$
$$\vec{x}_{13} = \vec{\phi}_{16} \quad \text{(eq. S31)}$$
$$\vec{x}_{14} = 8\vec{\phi}_{16} + \vec{\phi}_{17} + \vec{\phi}_{19} + 4\vec{\phi}_{20} \quad \text{(eq. S32)}$$
$$\vec{x}_{15} = \vec{\phi}_{25} \quad \text{(eq. S33)}$$
$$\vec{x}_{16} = \vec{\phi}_{21} + \vec{\phi}_{22} \quad \text{(eq. S34)}$$
$$\vec{x}_{17} = \vec{\phi}_{23} + \vec{\phi}_{24} \quad \text{(eq. S35)}$$
$$\vec{x}_{18} = \vec{\phi}_6 + \vec{\phi}_{11} \quad \text{(eq. S36)}$$

Defining $\vec{x}_{19}$ to $\vec{x}_{25}$ with $\vec{\phi}_i$ (exchange sets):

$$\vec{x}_{19} = \vec{\phi}_{11} + \vec{\phi}_{12} \quad \text{(eq. S37)}$$
$$\vec{x}_{20} = \vec{\phi}_{11} + 2\vec{\phi}_{24} \quad \text{(eq. S38)}$$
$$\vec{x}_{21} = \vec{\phi}_6 + \vec{\phi}_{10} + \vec{\phi}_{20} \quad \text{(eq. S39)}$$
$$\vec{x}_{22} = \vec{\phi}_4 + \vec{\phi}_7 + 2\vec{\phi}_{12} \quad \text{(eq. S40)}$$
$$\vec{x}_{23} = \vec{\phi}_5 + \vec{\phi}_{10} + \vec{\phi}_{12} \quad \text{(eq. S41)}$$
$$\vec{x}_{24} = 7\vec{\phi}_4 + 7\vec{\phi}_6 + 7\vec{\phi}_7 + \vec{\phi}_{19} + 8\vec{\phi}_{22} \quad \text{(eq. S42)}$$
$$\vec{x}_{25} = \vec{\phi}_5 + 3\vec{\phi}_{11} + \vec{\phi}_{17} \quad \text{(eq. S43)}$$

Calculation of additional exchange set reactions (in reference to main text Table 2):

Fe^{2+}-Mg^{2+} substitution:

$\alpha_6 = 1, \alpha_{10} = 1, \alpha_{22} = 1\,, \alpha_{23} = -1$

$$\vec{x} = \vec{x}_6 + \vec{x}_{10} + \vec{x}_{22} - \vec{x}_{23} \quad \text{(eq. S44)}$$

Redox balance from carbon and iron cycles (alternative):

$\alpha_{10} = 1, \alpha_{22} = 1\,, \alpha_{23} = -1, \alpha_{25} = 1$

$$\vec{x} = \vec{x}_{10} + \vec{x}_{22} - \vec{x}_{23} + \vec{x}_{25} \quad \text{(eq. S45)}$$

Redox balance from sulfur and iron cycles:

$\alpha_4 = -\frac{1}{4}, \alpha_{10} = \frac{45}{4}, \alpha_{12} = \frac{15}{4}, \alpha_{18} = \frac{45}{4}, \alpha_{22} = -\frac{7}{4}, \alpha_{23} = \frac{15}{4}, \alpha_{24} = \frac{1}{4}, \alpha_{25} = -\frac{15}{4}$

$$\vec{x} = -\frac{1}{4}\vec{x}_4 + \frac{45}{4}\vec{x}_{10} + \frac{15}{4}\vec{x}_{12} + \frac{45}{4}\vec{x}_{18} - \frac{7}{4}\vec{x}_{22} + \frac{15}{4}\vec{x}_{23} + \frac{1}{4}\vec{x}_{24} - \frac{15}{4}\vec{x}_{25} \quad \text{(eq. S46)}$$

$\boldsymbol{A}_{open}$ null space original basis

$\vec{\xi}_1 \; \vec{\xi}_2 \; \vec{\xi}_3 \; \vec{\xi}_4 \; \vec{\xi}_5 \; \vec{\xi}_6 \; \vec{\xi}_7 \; \vec{\xi}_8 \; \vec{\xi}_9 \; \vec{\xi}_{10} \; \vec{\xi}_{11} \; \vec{\xi}_{12} \; \vec{\xi}_{13} \; \vec{\xi}_{14} \; \vec{\xi}_{15} \; \vec{\xi}_{16} \; \vec{\xi}_{17} \; \vec{\xi}_{18} \; \vec{\xi}_{19} \; \vec{\xi}_{20} \; \vec{\xi}_{21} \; \vec{\xi}_{22} \; \vec{\xi}_{23} \; \vec{\xi}_{24} \; \vec{\xi}_{25} \; \vec{\xi}_{26} \; \vec{\xi}_{27} \; \vec{\xi}_{28} \; \vec{\xi}_{29} \; \vec{\xi}_{30} \; \vec{\xi}_{31} \; \vec{\xi}_{32} \; \vec{\xi}_{33} \; \vec{\xi}_{34} \; \vec{\xi}_{35} \; \vec{\xi}_{36}$

$$
\vec{\boldsymbol{x}} =
\begin{bmatrix}
x_{\text{Mantle CO2}}^{(\#1)} \\
x_{\text{Mantle HCl}}^{(\#2)} \\
x_{\text{Mantle H2S}}^{(\#3)} \\
x_{\text{Calcite metamorphism}}^{(\#4)} \\
x_{\text{Dolomite metamorphism}}^{(\#5)} \\
x_{\text{Siderite metamorphism}}^{(\#6)} \\
x_{\text{Ca-silicate weathering}}^{(\#7)} \\
x_{\text{Mg-silicate weathering}}^{(\#8)} \\
x_{\text{Na-silicate weathering}}^{(\#9)} \\
x_{\text{K-silicate weathering}}^{(\#10)} \\
x_{\text{Fe-silicate weathering}}^{(\#11)} \\
x_{\text{Calcite weathering}}^{(\#12)} \\
x_{\text{Dolomite weathering}}^{(\#13)} \\
x_{\text{Siderite weathering}}^{(\#14)} \\
x_{\text{Silica weathering}}^{(\#15)} \\
x_{\text{Mg-reverse weathering}}^{(\#16)} \\
x_{\text{Na-reverse weathering}}^{(\#17)} \\
x_{\text{K-reverse weathering}}^{(\#18)} \\
x_{\text{Fe-reverse weathering}}^{(\#19)} \\
x_{\text{Silica formation}}^{(\#20)} \\
x_{\text{Calcite formation}}^{(\#21)} \\
x_{\text{Dolomite formation}}^{(\#22)} \\
x_{\text{Siderite formation}}^{(\#23)} \\
x_{\text{Oxygenic photosynthesis}}^{(\#24)} \\
x_{\text{Aerobic respiration}}^{(\#25)} \\
x_{\text{Sulfide oxidation}}^{(\#26)} \\
x_{\text{Sulfate reduction}}^{(\#27)} \\
x_{\text{Ferrous iron oxidation}}^{(\#28)} \\
x_{\text{Ferric iron reduction}}^{(\#29)} \\
x_{\text{Pyrite oxidation}}^{(\#30)} \\
x_{\text{Pyrite formation}}^{(\#31)} \\
x_{\text{Gypsum dissolution}}^{(\#32)} \\
x_{\text{Gypsum formation}}^{(\#33)} \\
x_{\text{Halite dissolution}}^{(\#34)} \\
x_{\text{Halite formation}}^{(\#35)} \\
x_{\text{Nitrogen fixation}}^{(\#36)} \\
x_{\text{Nitrification}}^{(\#37)} \\
x_{\text{Denitrification}}^{(\#38)}
\end{bmatrix}
=
\begin{bmatrix}
0 & -1 & -2 & -1 & 0 & 0 & 0 & 0 & 0 & -1 & -2 & -1 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 2 & 1 & 1 & -1 & 0 & -2 & 0 & -1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & -5 \\
0 & 0 & 0 & 0 & 2 & 2 & 2 & 2 & 2 & 2 & 4 & 2 & 0 & -2 & -2 & -2 & -2 & 0 & -2 & -4 & -2 & 0 & 0 & -2 & 2 & -8 & 8 & -16 & 2 & 0 & 0 & 0 & 0 & 4 & -2 & 4 \\
1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
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\end{bmatrix}
\begin{bmatrix}
{}^{\xi}\omega_1 \\ {}^{\xi}\omega_2 \\ {}^{\xi}\omega_3 \\ {}^{\xi}\omega_4 \\ {}^{\xi}\omega_5 \\ {}^{\xi}\omega_6 \\ {}^{\xi}\omega_7 \\ {}^{\xi}\omega_8 \\ {}^{\xi}\omega_9 \\ {}^{\xi}\omega_{10} \\ {}^{\xi}\omega_{11} \\ {}^{\xi}\omega_{12} \\ {}^{\xi}\omega_{13} \\ {}^{\xi}\omega_{14} \\ {}^{\xi}\omega_{15} \\ {}^{\xi}\omega_{16} \\ {}^{\xi}\omega_{17} \\ {}^{\xi}\omega_{18} \\ {}^{\xi}\omega_{19} \\ {}^{\xi}\omega_{20} \\ {}^{\xi}\omega_{21} \\ {}^{\xi}\omega_{22} \\ {}^{\xi}\omega_{23} \\ {}^{\xi}\omega_{24} \\ {}^{\xi}\omega_{25} \\ {}^{\xi}\omega_{26} \\ {}^{\xi}\omega_{27} \\ {}^{\xi}\omega_{28} \\ {}^{\xi}\omega_{29} \\ {}^{\xi}\omega_{30} \\ {}^{\xi}\omega_{31} \\ {}^{\xi}\omega_{32} \\ {}^{\xi}\omega_{33} \\ {}^{\xi}\omega_{34} \\ {}^{\xi}\omega_{35} \\ {}^{\xi}\omega_{36}
\end{bmatrix}
$$

Fig. S4: The basis $(\vec{\xi}_i)$ calculated in MATLAB for the null space of $\boldsymbol{A_{open}}$. Because the biogeochemical processes are defined to run unidirectionally, negative values in the matrix represent unphysical outcomes. The $\vec{\xi}_i$ vectors were used to generate a new physically allowable basis set for the same space $(\vec{x}_1 - \vec{x}_{36})$. The ${}^{\xi}\omega_i$ values refer to the magnitude of each vector.

Defining $\vec{x}_1$ to $\vec{x}_{18}$ with $\vec{\xi}_i$ (closed sets):

$$\vec{x}_1 = \vec{\xi}_2 + \vec{\xi}_5 + \vec{\xi}_{18} + \vec{\xi}_{19} \quad \text{(eq. S47)}$$
$$\vec{x}_2 = \vec{\xi}_3 + \vec{\xi}_5 + \vec{\xi}_6 + 2\vec{\xi}_{18} + \vec{\xi}_{20} + \quad \text{(eq. S48)}$$
$$\vec{x}_3 = \vec{\xi}_4 + \vec{\xi}_9 + \vec{\xi}_{18} + \vec{\xi}_{21} \quad \text{(eq. S49)}$$
$$\vec{x}_4 = \vec{\xi}_{10} + \vec{\xi}_{19} \quad \text{(eq. S50)}$$
$$\vec{x}_5 = \vec{\xi}_{11} + \vec{\xi}_{20} \quad \text{(eq. S51)}$$
$$\vec{x}_6 = \vec{\xi}_{12} + \vec{\xi}_{21} \quad \text{(eq. S52)}$$
$$\vec{x}_7 = \vec{\xi}_6 + \vec{\xi}_{14} \quad \text{(eq. S53)}$$
$$\vec{x}_8 = \vec{\xi}_7 + \vec{\xi}_{15} \quad \text{(eq. S54)}$$
$$\vec{x}_9 = \vec{\xi}_8 + \vec{\xi}_{16} \quad \text{(eq. S55)}$$
$$\vec{x}_{10} = \vec{\xi}_9 + \vec{\xi}_{17} \quad \text{(eq. S56)}$$
$$\vec{x}_{11} = \vec{\xi}_{22} + \vec{\xi}_{23} \quad \text{(eq. S57)}$$
$$\vec{x}_{12} = \vec{\xi}_{22} + \vec{\xi}_{26} + \vec{\xi}_{27} \quad \text{(eq. S58)}$$
$$\vec{x}_{13} = 2\vec{\xi}_{22} + \vec{\xi}_{24} + \vec{\xi}_{25} \quad \text{(eq. S59)}$$
$$\vec{x}_{14} = 16\vec{\xi}_{22} + 8\vec{\xi}_{25} + \vec{\xi}_{26} + \vec{\xi}_{28} + 4\vec{\xi}_{29} \quad \text{(eq. S60)}$$
$$\vec{x}_{15} = 5\vec{\xi}_{22} + \vec{\xi}_{34} + 4\vec{\xi}_{35} + \vec{\xi}_{36} \quad \text{(eq. S61)}$$
$$\vec{x}_{16} = \vec{\xi}_{30} + \vec{\xi}_{31} \quad \text{(eq. S62)}$$
$$\vec{x}_{17} = \vec{\xi}_{32} + \vec{\xi}_{33} \quad \text{(eq. S63)}$$
$$\vec{x}_{18} = \vec{\xi}_{13} + \vec{\xi}_{18} \quad \text{(eq. S64)}$$

Defining $\vec{x}_{19}$ to $\vec{x}_{25}$ with $\vec{\xi}_i$ (exchange sets):

$$\vec{x}_{19} = \vec{\xi}_5 + \vec{\xi}_{18} + \vec{\xi}_{19} \quad \text{(eq. S65)}$$
$$\vec{x}_{20} = \vec{\xi}_7 + \vec{\xi}_{18} + 2\vec{\xi}_{33} \quad \text{(eq. S66)}$$
$$\vec{x}_{21} = 2\vec{\xi}_1 + \vec{\xi}_{13} + \vec{\xi}_{17} + \vec{\xi}_{29} \quad \text{(eq. S67)}$$
$$\vec{x}_{22} = \vec{\xi}_5 + \vec{\xi}_{11} + \vec{\xi}_{14} + 2\vec{\xi}_{19} \quad \text{(eq. S68)}$$
$$\vec{x}_{23} = \vec{\xi}_5 + \vec{\xi}_{12} + \vec{\xi}_{17} + \vec{\xi}_{19} \quad \text{(eq. S69)}$$
$$\vec{x}_{24} = \vec{\xi}_{10} + 7\vec{\xi}_{11} + 7\vec{\xi}_{13} + 7\vec{\xi}_{14} + 15\vec{\xi}_{22} + \vec{\xi}_{28} + 8\vec{\xi}_{31} \quad \text{(eq. S70)}$$
$$\vec{x}_{25} = 3\vec{\xi}_9 + \vec{\xi}_{12} + 3\vec{\xi}_{18} + \vec{\xi}_{22} + \vec{\xi}_{26} \quad \text{(eq. S71)}$$

Defining $\vec{x}_{26}$ to $\vec{x}_{36}$ with $\vec{\xi}_i$ (open sets):

$$\vec{x}_{26} = 4\vec{\xi}_{12} + 4\vec{\xi}_{22} + \vec{\xi}_{26} \quad \text{(eq. S72)}$$
$$\vec{x}_{27} = 8\vec{\xi}_5 + \vec{\xi}_{28} \quad \text{(eq. S73)}$$
$$\vec{x}_{28} = \vec{\xi}_{11} + 2\vec{\xi}_{19} \quad \text{(eq. S74)}$$
$$\vec{x}_{29} = \vec{\xi}_1 \quad \text{(eq. S75)}$$
$$\vec{x}_{30} = \vec{\xi}_5 \quad \text{(eq. S76)}$$
$$\vec{x}_{31} = \vec{\xi}_7 \quad \text{(eq. S77)}$$
$$\vec{x}_{32} = \vec{\xi}_8 \quad \text{(eq. S78)}$$
$$\vec{x}_{33} = \vec{\xi}_9 \quad \text{(eq. S79)}$$
$$\vec{x}_{34} = \vec{\xi}_{13} \quad \text{(eq. S80)}$$
$$\vec{x}_{35} = \vec{\xi}_{34} \quad \text{(eq. S81)}$$
$$\vec{x}_{36} = \vec{\xi}_{34} + 2\vec{\xi}_{35} \quad \text{(eq. S82)}$$

Calculation of additional open set reactions (in reference to main text Table 2):

O_2 from siderite (alternative):

$\alpha_{10} = 1,\ \alpha_{18} = 1,\ \alpha_{25} = -\frac{1}{3},\ \alpha_{26} = \frac{1}{3}, \alpha_{34} = -1$

$$\vec{x} = \vec{x}_{10} + \vec{x}_{18} - \frac{1}{3}\vec{x}_{25} + \frac{1}{3}\vec{x}_{26} - \vec{x}_{34} \quad \text{(eq. S83)}$$

O_2 from dolomite:

$\alpha_{10} = 1,\ \alpha_{18} = 1,\ \alpha_{22} = 1,\ \alpha_{23} = -1,\ \alpha_{25} = -\frac{1}{3},\ \alpha_{26} = \frac{1}{3}, \alpha_{34} = -1$

$$\vec{x} = \vec{x}_{10} + \vec{x}_{18} + \vec{x}_{22} - \vec{x}_{23} - \frac{1}{3}\vec{x}_{25} + \frac{1}{3}\vec{x}_{26} - \vec{x}_{34} \quad \text{(eq. S84)}$$

Open set chemical reactions:

$\vec{x}_{29}$: H_2S (mantle) $\rightarrow$ H_2S (eq. S85)

$\vec{x}_{30}$: $2HCl$ (mantle) + $CaSiO_3 \rightarrow Ca^{2+} + SiO_2$ (aq) + $2Cl^- + H_2O$ (eq. S86)

$\vec{x}_{31}$: $2HCl$ (mantle) + $Na_2SiO_3 \rightarrow 2Na^+ + SiO_2$ (aq) + $2Cl^- + H_2O$ (eq. S87)

$\vec{x}_{32}$: $2HCl$ (mantle) + $K_2SiO_3 \rightarrow 2K^+ + SiO_2$ (aq) + $2Cl^- + H_2O$ (eq. S88)

$\vec{x}_{33}$: $2HCl$ (mantle) + $FeSiO_3 \rightarrow Fe^{2+} + SiO_2$ (aq) + $2Cl^- + H_2O$ (eq. S89)

$\vec{x}_{34}$: SiO_2 (solid) $\rightarrow$ SiO_2 (aq) (eq. S90)

$\vec{x}_{35}$: $4HCl$ (mantle) + $6H_2O + 2N_2 \rightarrow 4NH_4^+ + 4Cl^- + 3O_2$ (eq. S91)

$\vec{x}_{36}$: $4H_2O + 2N_2 + O_2 \rightarrow 2NH_4^+ + 2NO_3^-$ (eq. S92)

S3: Gross fluxes and net fluxes

The biogeochemical processes presented in Table 1 include both forward and reverse reactions, such as $CaCO_3$ dissolution (process #12) and $CaCO_3$ precipitation (process #21). However, these two processes could alternatively have been written as a single reaction capable of proceeding in either direction. To implement this change, one would remove the 21st column of the $\boldsymbol{A_{closed}}$ matrix and allow $\vec{x}_i$ at the 12th position to attain both positive and negative values for reaction rate. Let such an updated $\boldsymbol{A_{closed}}$ matrix be called $\boldsymbol{A'_{closed}}$. While $\boldsymbol{A'_{closed}}$ would have the same rank as $\boldsymbol{A_{closed}}$, it would have one fewer columns and thus, by the rank-nullity theorem, one fewer null space dimensions and one fewer closed set. In this case, the closed set that would no longer exist is that which had previously corresponded to the balance of carbonate weathering and carbonate formation ($\vec{x}_4$ in the main text), which would be subsumed within the trivial solution to the equation $\boldsymbol{A'_{closed}}\vec{x} = \vec{0}$. Overall, our implementation of two gross fluxes was thus chosen rather over a single net flux in order to best demonstrates the behavior of closed sets.

S4: Implementation of additional processes

Implementation of additional processes would produce additional closed, exchange, and open sets. For a simple example, accounting for the dissolution and precipitation of the mineral sylvite (KCl) would produce at least one additional closed set and one additional exchange set. The closed set would be KCl dissolution and precipitation, analogous to halite dissolution and precipitation ($\vec{x}_{17}$). The exchange set would be HCl degassing, K-silicate weathering, and sylvite formation, analogous to the halite formation exchange set ($\vec{x}_{20}$), and would follow the equation: $\mathrm{2HCl\ (mantle) + K_2SiO_3 \rightarrow 2KCl + SiO_2\ (solid) + H_2O}$. Relatedly, adding chemical constituents to existing fluxes could modify the existing sets. For example, our current treatment of oxygenic photosynthesis ($\mathrm{CO_2 + H_2O \rightarrow CH_2O + O_2}$) does not resolve any role for the alkalinity fluxes associated with uptake of N and P. Although accounting for such alkalinity fluxes does not matter when considering a closed set such as oxygenic photosynthesis and aerobic respiration, as the flux arrows would simply be rotated in the space of FIC against ALK, the alkalinity component may become more important when considering the role of feedbacks in setting the timescale over which open sets can stabilize pCO_2. Though we chose a set of biogeochemical reactions meant to capture the main dynamics of major biogeochemical cycles, applying this framework to evaluate habitability during any specific period of Earth's History or on other planets may require additional or modified reactions compared to those listed in Table 1.