

GEOLOGIC CARBON CYCLE CONSTRAINTS ON THE TERRESTRIAL HYDROLOGICAL RESPONSE TO HIGHER ATMOSPHERIC CO₂

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Key Points:

1. The silicate weathering negative feedback depends upon runoff to drive recovery from carbon cycle perturbations.
2. Decreases in runoff with warming would lead to runaway greenhouse states.
3. The sensitivity of runoff to warming has a lower-bound of 0%/K as dictated by carbon cycle constraints.

Abstract

How runoff will change as atmospheric CO₂ rises depends upon several difficult to project factors, including CO₂ fertilization, lengthened growing seasons, and vegetation greening. However, geologic records of the hydrological response to past carbon cycle perturbations indicate large increases in runoff with higher CO₂. We demonstrate that the fact that the Earth has remained habitable since life emerged sets a lower-bound on the sensitivity of runoff to CO₂ changes. The recovery of the Earth system from perturbations is attributed to silicate weathering, which transfers CO₂ to the oceans as alkalinity via runoff. Though many factors mediate weathering rates, runoff determines the total flux of silicate-derived cations and hence the removal flux of excess CO₂. Using a carbon cycle model that parameterizes weathering as a function of rock reactivity, runoff, temperature, and soil CO₂, we show that recovery from a perturbation is only possible if the lower-bound for the sensitivity of runoff to atmospheric CO₂ is 0%/K. Using proxy data for the Paleocene-Eocene Thermal Maximum, we find that to match the marine $\delta^{13}\text{C}$ record requires a runoff sensitivity greater than 0%/K and similar to estimates of the modern runoff sensitivity derived from an ensemble of Earth system models. These results suggest that the processes that enhance global runoff are likely to prevail over processes that tend to dampen runoff. In turn, that the Earth has always recovered from perturbations suggests that, though the runoff response is spatially complex, global discharge has never declined in response to warming, despite quite varied paleogeographies.

Plain Language Summary

Runoff—the quantity of water delivered by rivers to the ocean—is likely to change as the climate warms. However, because generation of runoff involves many small-scale processes, it is difficult to forecast how global patterns of runoff will change as atmospheric CO₂ rises. We take advantage of the fact that runoff is the primary mechanism that transfers excess CO₂ from the atmosphere to the ocean for eventual burial as carbonate rock. It is this process that permits the Earth to recover from major perturbations. We find that runoff must increase in a warmer world; if it did not, then atmospheric CO₂ would continually increase in the atmosphere, leading to a runaway greenhouse. This hard constraint places a lower-bound on how runoff must change in response to rising atmospheric CO₂.

1. Introduction

Runoff—the transfer of water from the continents to the oceans and to closed basins—is a critical component of the Earth system. Runoff (q) determines the amount of freshwater available for societal use, transports nutrients from upland landscapes to lowland and marine ecosystems and provides the water necessary to weather rocks that sequester atmospheric CO₂. Despite its importance for the functioning of the Earth system and for societies, how q will change as atmospheric CO₂ rises remains highly uncertain. Part of this uncertainty arises because the hydrological cycle is comprised of a number of different and interacting components—including precipitation (P), bare-soil and vegetation-controlled evaporation (ET), and q —which may respond differently to rising atmospheric CO₂ and temperature (Milly & Dunne, 2016). For some components, there are well grounded theoretical expectations for how they should change. Precipitation globally is thought to increase by approximately 2%/K given constraints on how much energy is available for evaporation (Held & Soden, 2006; Pendergrass & Hartmann, 2014). Potential evapotranspiration (PET) over land should also increase, as rising temperatures drive increases in the saturation vapor deficit. This increase in PET is robustly predicted to be larger than the increase in P (Scheff & Frierson, 2014).

Collectively, these expectations—combined with observations that support them—suggest that much of the land surface will become more arid with warming (Cook et al., 2014; Ficklin & Novick, 2017; Mankin et al., 2019; Milly & Dunne, 2020; Novick et al., 2016; Overpeck & Udall, 2020). However, there remain two critical, linked components of the hydrological cycle for which theoretical expectations and observational constraints are of low-confidence and poor—actual ET and q (Douville et al., 2021). ET determines how much P is returned to the atmosphere and how much is partitioned to q ; as a consequence, ET and P determine surface water availability. Vegetation water use (*i.e.*, transpiration) is thought to be the predominant component of ET (~ 60%) (Good et al., 2015; Schlesinger & Jasechko, 2014; Wei et al., 2017); vegetation additionally impacts another 10-15% of ET due to intercepted P that evaporates directly from the leaf or woody surface (Wang-Erlandsson et al., 2014). Thus, in addition to the physical response to CO₂ increase (P and PET), the global vegetation response to rising CO₂ will further determine the trajectory of surface water availability.

However, projecting how global vegetation will change and its impact on q remains difficult. Though leaf-level transpiration is expected to decrease as CO₂ rises, total leaf area is expected to increase; the competition of these two processes globally will partially determine the total change in q . Satellite observations indicate that the world is overwhelmingly greening and that this greening is leading to greater ET (Forzieri et al., 2020; Winkler et al., 2021; Zhu

et al., 2016). In semi-arid regions, observations indicate this greening and associated increase in ET has led to declining q (Ukkola et al., 2016), and many land-surface models—which are designed to encapsulate a number of specific plant physiological processes—find that increases in leaf area have been substantial enough to reduce q globally (Forzieri et al., 2020; Piao et al., 2007). In contrast, coupled land-atmosphere models find that, while the Earth’s surface greens substantially, the resulting increase in ET fluxes is less than the increase in P (Lemordant et al., 2018; Swann et al., 2016). Though this prediction is spatially heterogeneous (Mankin et al., 2019) and of low-confidence, the mean global average change in q with temperature is predicted to be $\sim 2.9 \pm 1 \text{ \%}/\text{K}$ across the CMIP5 ensemble using the RCP8.5 scenario (Zhang et al., 2014). Much of this increase is driven by widespread model agreement of increasing q in the northern high latitudes (Scheff et al., 2017). However, there remain serious questions about how leaf-level processes and slower vegetation responses (*i.e.*, species compositional changes) are parameterized within these coupled models. As a consequence, there is no firm theoretical expectation nor well-constrained observations to indicate whether global ET will increase or decrease relative to P and, therefore, how globally averaged q will change as atmospheric CO_2 rises.

This disagreement and uncertainty regarding the direction and magnitude of the global q change as CO_2 rises stands in sharp contrast to both geologic observations of the hydrological response to increasing CO_2 and theory regarding how the Earth system has recovered from large carbon cycle perturbations in the geologic past. These recoveries are mediated by the weathering of silicate rocks, which converts atmospheric CO_2 to alkalinity and permanently buries carbon in marine sediments (R. A. Berner & Kothavala, 2001; Penman et al., 2020). Critically, the weathering of rocks and transport of alkalinity to the ocean is thought to be mediated primarily by q (Kump et al., 2000; Li et al., 2022; Maher & Chamberlain, 2014; Otto-Bliesner, 1995; Park et al., 2020), both to supply water to the weathering zone and to transport the products of weathering to the ocean. The fact that life has persisted on Earth for more than 3 Ga indicates that this feedback must be particularly robust, capable of regulating Earth’s climate largely independent of the precise position of the continents, extant flora and fauna, and magnitude and nature of the various climatic perturbations that have befallen Earth over its history (Sagan & Mullen, 1972; Walker et al., 1981). Similarly, a growing body of literature suggests that these past perturbations caused dramatic reorganizations of the terrestrial hydrological cycle, recorded as thick packages of coarse-grained fluvial sediments during peak warming (Chen et al., 2018; Foreman et al., 2012) and enhanced fluxes of nutrients and silica

to the ocean as reflected in widespread marine anoxia and chert deposits during many of these carbon cycle perturbations (Penman, 2016; Them et al., 2017).

Here, we exploit this fundamental negative feedback in the Earth system and its relationship with q to provide a lower bound on how globally q must respond to changes in atmospheric CO₂. To constrain how q must respond to changes in atmospheric CO₂, we employ a version of a geological carbon cycle model that has been modified to represent the explicit role of q in modulating weathered solute concentrations and fluxes. We demonstrate that the operation of this weathering feedback requires that q does not decrease with rising atmospheric CO₂ and, most likely, is positively sensitive to changes in CO₂, such that the flux of weathered material always increases if CO₂ rapidly increases in the atmosphere. We further test this model against geological data from the Paleocene-Eocene Thermal Maximum (PETM), and demonstrate that, to match geological proxy data from the PETM requires that globally averaged q increase with atmospheric CO₂. We conclude by discussing how these constraints on the sensitivity of the hydrological cycle carries profound implications for our understanding of biogeochemical cycles in Earth's past and for Earth's future as CO₂ rises.

2. Background

2.1 Silicate Weathering and Runoff

Silicate weathering is thought to be the primary process that removes CO₂ from the coupled atmosphere-ocean system on geological timescales, neutralizing the input of CO₂ from volcanoes (R. A. Berner & Kothavala, 2001). Silicate weathering is also thought to be sensitive to climate, such that increases in the input flux of CO₂ and the resulting rise in atmospheric CO₂ causes increased silicate weathering fluxes, which removes this excess CO₂ from the Earth's surface (Robert A. Berner & Caldeira, 1997; Zeebe & Caldeira, 2008). Though other Earth surface processes are important for the geological carbon cycle, including the weathering and burial of organic carbon and oxidation/reduction of sulfur (Hilton & West, 2020; Torres et al., 2014), the evidence for these processes being sensitive to climate—and therefore capable of providing a negative feedback on atmospheric CO₂—is less extensive. Further, the reaction of silicate rocks with CO₂ dissolved in water (*i.e.*, carbonic acid) is likely to have been applicable for the entirety of Earth history. Critically, both data and models suggest that Earth system recovery once a perturbation ceases typically occurs within 1 Myr or less (Bowen, 2013; Colbourn et al., 2015; Lenton & Britton, 2006; Uchikawa & Zeebe, 2008).

Equation 1 describes the long-term relationship between total carbon in the ocean-atmosphere system (M ; mol C) and the input and output fluxes of CO_2 (Caves et al., 2016; Kump & Arthur, 1999):

$$\frac{dM}{dt} = I - F_{silw} \quad (\text{Eq. 1})$$

where F_{silw} is the silicate weathering flux [mol C/yr] and I is the sum of the primary remaining input and output fluxes of CO_2 [mol C/yr], including CO_2 from volcanism and solid Earth degassing and organic carbon weathering and burial. On timescales longer than ocean overturning, changes in the atmospheric partial pressure of CO_2 ($p\text{CO}_2$) are proportional to changes in M . The global silicate weathering flux is defined as (White & Blum, 1995):

$$F_{silw} = Q \times [C]_{sil} \quad (\text{Eq. 2})$$

where Q is global discharge [L/yr] and $[C]_{sil}$ is the global average concentration of bicarbonate (HCO_3^-) derived from the weathering of silicate rocks [mol C/L]. Q is related to q via the global land area [m^2] ($Q = q \times \text{area}$), which, for the purposes of our simulations, we assume to be constant. Consequently, for silicate weathering to act as a negative feedback on atmospheric CO_2 , either q , $[C]_{sil}$, or both must be sensitive to climate.

To demonstrate this, we use CH2O-CHOO (the Carbon- H_2O Coupled Hydrological model), a modified version of CLiBeSO-W, which is a geological carbon cycle model that calculates the mass balance of carbon and alkalinity in the ocean-atmosphere system (Caves Rugenstein et al., 2019), and uses equation 2 to calculate F_{silw} . We first treat q and $[C]_{sil}$ as being independently sensitivity to CO_2 -induced global warming (*i.e.*, Δq and $\Delta[C]_{sil}$ have units of %/K) and assume a climate sensitivity of 4K/ CO_2 doubling (Knutti et al., 2017). In these simulations, we inject 1000 Pg C into the atmosphere over 100 years and calculate the amount of time required for atmospheric CO_2 to return to its initial value—termed the recovery time [yr].

As shown in Figure 1, a negative sensitivity in either q or $[C]_{sil}$ with temperature requires a correspondingly larger sensitivity in the other parameter. Recovery from perturbations is fastest when both parameters have a positive sensitivity, but small, positive sensitivities still result in recovery times greater than 1 Ma. Thus, to place constraints on the sensitivity of q to CO_2 requires knowledge of how $[C]_{sil}$ will respond to atmospheric CO_2 .

Recent advances in biogeochemistry, however, have resulted in improved constraints on how $[C]_{\text{sil}}$ is likely to change with climate (Godsey et al., 2009, 2019; Li et al., 2022; Maher & Chamberlain, 2014; West, 2012; Winnick & Maher, 2018). Following previous work, we consider here three parameters that are likely to have the greatest impact on $[C]_{\text{sil}}$ and which are also sensitive to climate: temperature (T), runoff (q), and soil $p\text{CO}_2$.

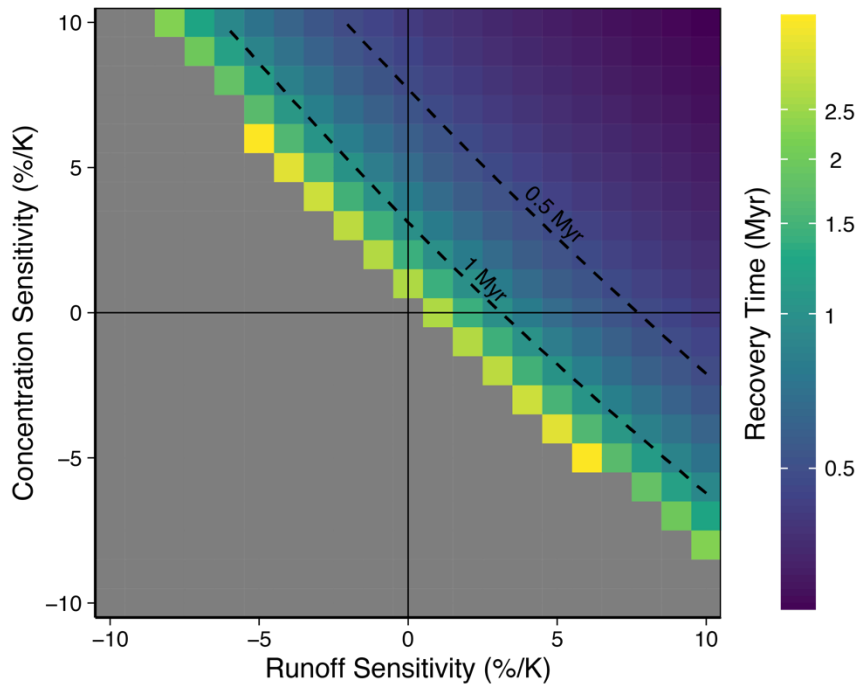


Figure 1: The time required for atmospheric CO_2 to recover to pre-perturbation values (shading in 10^6 yr) as a function of the q -sensitivity (%/K) and $[C]_{\text{sil}}$ -sensitivity (%/K). Gray shading indicates that atmospheric CO_2 does not return to pre-perturbation values within 4 Ma post-perturbation. Solid lines denote the 0% q - and $[C]_{\text{sil}}$ -sensitivities; dashed lines denote the 0.5 and 1 Myr recovery time contours.

To encapsulate these three terms, we use a recently developed reactive-transport model of solute generation from Maher and Chamberlain (2014) to predict $[C]_{\text{sil}}$ (also termed the MAC model (Baum et al., 2022; Graham & Pierrehumbert, 2020). Critically, this model also permits us to explicitly treat the reactivity of the land surface—set by erosion and by the type of exposed lithologies on Earth’s surface (Caves Rugenstein et al., 2019; Ibarra et al., 2016; West, 2012)—and which determines the rate at which silicate weathering occurs. These equations are summarized in the Supporting Information, but are briefly described here. Temperature affects $[C]_{\text{sil}}$ through its impact on net reaction rates, as parameterized by an Arrhenius equation. Runoff affects $[C]_{\text{sil}}$ via dilution, though only in weathering systems that are kinetically limited; whether $[C]_{\text{sil}}$ is kinetically-limited depends upon the reactivity of the

weathering material, which is parameterized as a function of the age of the weathering zone (T_s [yr]). Low weathering zone ages, for instance, are indicative of fast supply of fresh minerals, resulting in higher reaction rates, and consequently reduced kinetic limitation. Many catchments today have concentrations that are invariant to changes in runoff (*i.e.*, chemostatic; Godsey et al., 2009), suggesting that kinetic-limitation may not be widespread. Lastly, soil $p\text{CO}_2$ modifies the maximum equilibrium $[C]_{\text{sil}}$ ($[C]_{\text{sil,eq}}$), and we assume an open weathering zone CO_2 system (Winnick & Maher, 2018), which is a reasonable assumption given that plants and microbes continually supply CO_2 to the weathering zone via root and soil respiration. We assume that soil $p\text{CO}_2$ scales—via a Michaelis-Menton relationship—to changes in atmospheric $p\text{CO}_2$ that increase aboveground gross primary productivity (Volk, 1987). Assuming a linear scaling between soil CO_2 and atmospheric CO_2 does not substantially alter the conclusions presented here. This set of equations permits us to constrain $[C]_{\text{sil}}$ as a function of several climatic parameters (*i.e.*, temperature (T), CO_2 , and q) and the weathering zone reactivity and, therefore, treat q -sensitivity as the main unknown parameter.

2.2 Projected changes in runoff

To compare our CH_2O - CHOO model results with state-of-the-art Earth system model projections of global q change, we interrogate those models that participated in the C4MIP experiments (Table S2) (Arora et al., 2013, 2020). These model versions account for a more elaborate representation of land processes, such as the response of evapotranspiration to warming and rising CO_2 . We calculate the ensemble mean runoff sensitivity to warming ($\%/K$) in the 1pct CO_2 experiments (1% annual increase in atmospheric CO_2) across CMIP5 and CMIP6 models. For each model, we estimate the relative q -sensitivity by linearly regressing the change in q against the change in temperature. We find that, while the spatial distribution of q change is patchy and there is poor model agreement across much of the land surface, the ensemble average global q -sensitivity is $3.1 \pm 0.9\%$, driven by an increase in P and a slight decrease in ET (Figure 2).

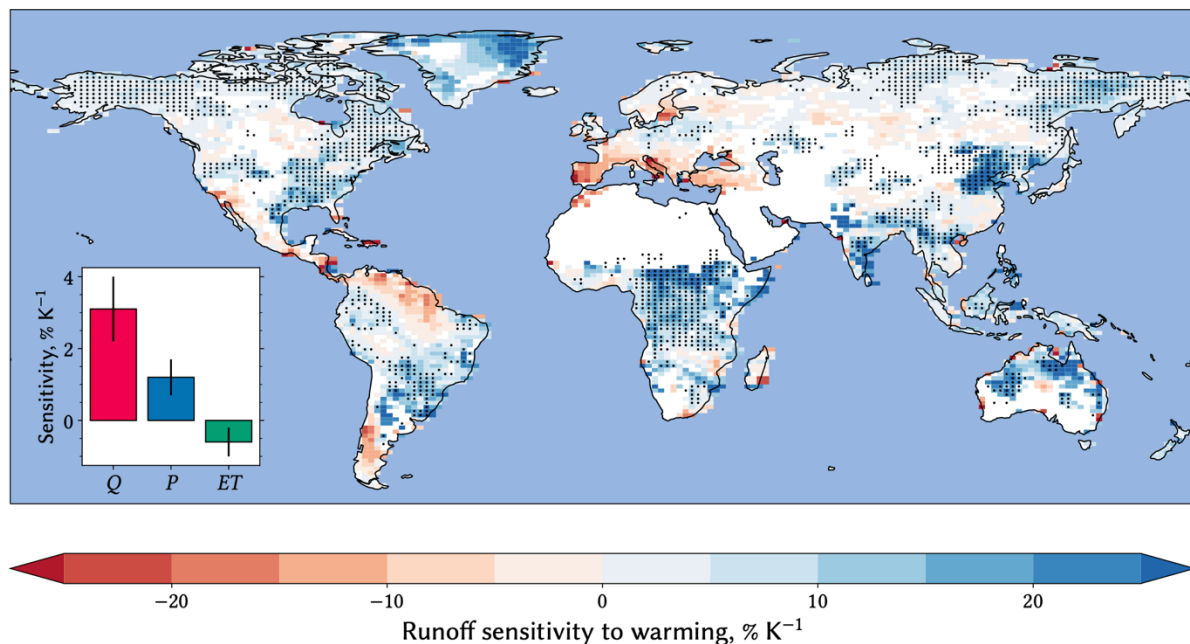


Figure 2: The q -sensitivity ($\%/K$) averaged across the C4MIP models (combined CMIP5 and CMIP6). Stippling indicates where at least 75% of the models (6 of 8) agree on the sign of the runoff sensitivity. Inset shows the globally-averaged q -, P -, and ET -sensitivity ($\%/K$)

3. Carbon Cycle Constraints on Runoff Changes with Warming

To estimate how differences in the sensitivity of runoff with warming (*i.e.*, the q -sensitivity) affect the long-term carbon cycle, we again use CH2O-CHOO and inject 1000 Pg of carbon into the ocean-atmosphere over the span of 100 years and permit long-term carbon cycle processes to restore atmospheric CO_2 to pre-perturbation conditions. Here, we treat the q -sensitivity as an independent variable; however, unlike in the example above, $[C]_{sil}$ is now constrained via its relationship to atmospheric CO_2 through T , q , and soil pCO_2 (Maher & Chamberlain, 2014; Volk, 1987; Winnick & Maher, 2018). We test cases where (1) we vary T_s between 10,000 and 100,000 years (the average weathering zone age today is 20,000; Larsen et al., 2014); (2) $[C]_{sil}$ is constant, following indications that many catchments on short timescales are chemostatic (Godsey et al., 2009; Ibarra et al., 2016); (3) seafloor weathering—which is dependent only upon bottom-water temperatures and not upon q (Brady & Gíslason, 1997; Coogan & Gillis, 2018)—represents 20% of the pre-perturbation weathering flux (Hilton & West, 2020), and; (4) silicate weathering rates are predicted using a different model, from West (2012), which also contains terms for q , T , and land surface reactivity.

Though the precise recovery time [yr] for CO_2 at a given q -sensitivity varies depending upon the value of the weathering parameters, in all cases, the timescale for atmospheric CO_2

to return to pre-perturbation values decreases as the q -sensitivity increases. This is perhaps most easily shown by the case of constant concentrations (blue line, Figure 3). In this case, $[C]_{\text{sil}}$ remains constant as CO_2 changes and is unaffected by changes in q , T , or $p\text{CO}_2$; consequently, recovery from a perturbation is impossible once the q -sensitivity falls below $0\%/K$, given the relationship in Eq. 1. Indeed, even where $[C]_{\text{sil}}$ is constant, recovery timescales surpass 1 Ma for q -sensitivities below $\sim 2.5\%/K$. In all other cases, both q and $[C]_{\text{sil}}$ are permitted to vary as atmospheric CO_2 changes; yet, even in these other experiments, recovery times surpass 1 Ma at or near a q -sensitivity of $0\%/K$. This result arises because, though both q and $[C]_{\text{sil}}$ are linearly related to F_{silw} (Eq. 2), changes in $[C]_{\text{sil}}$ are typically smaller than changes in q as atmospheric CO_2 changes (Figure S1), resulting in a greater influence of q on the ultimate recovery from a C-cycle perturbation. While this supposition is based upon our model of weathering, decreases in $[C]_{\text{sil}}$ do not follow perfect dilution trajectories as q increases, as seen in datasets that reflect short-term (Godsey et al., 2009) and long-term processes (Godsey et al., 2019; Li et al., 2022), further supporting the idea that variations in q —and not in $[C]_{\text{sil}}$ —are the dominant control on F_{silw} .

The critical influence of q can be seen in how different parameterizations affect the recovery time. As weathering zone age increases, the recovery time increases for a given q -sensitivity because $[C]_{\text{sil}}$ becomes insensitive to changes in q , due to the relative decrease in weathering zone reactivity. Similarly, if 20% of the initial, global F_{silw} occurs due to seafloor basalt weathering (green line), recovery times are slightly shorter for a given q -sensitivity, because a portion of $[C]_{\text{sil}}$ —that generated by seafloor weathering—is insensitive to changes in q and only sensitive to changes in T . Below a q -sensitivity of $0\%/K$, seafloor weathering begins to substantially reduce recovery times because T rises far more in response to the carbon cycle perturbation due to the reduction in weathering on land, causing seafloor weathering to become a greater proportion of total F_{silw} . The West (2012) formulation (red line) posits a faster recovery time for most q -sensitivities, largely because F_{silw} is formulated not as a linear combination of q and $[C]_{\text{sil}}$ but rather as linearly dependent upon erosion rate, which is presumed to be independent of climate. This linear sensitivity to erosion rate yields a strong dependence upon land surface reactivity that helps to decouple F_{silw} from changes in q . Nevertheless, as in the model of Maher and Chamberlain, if erosion rate decreases (equivalent to an increase in the weathering zone age), the West (2012) model predicts longer recovery times for equivalent q -sensitivities (Figure S2).

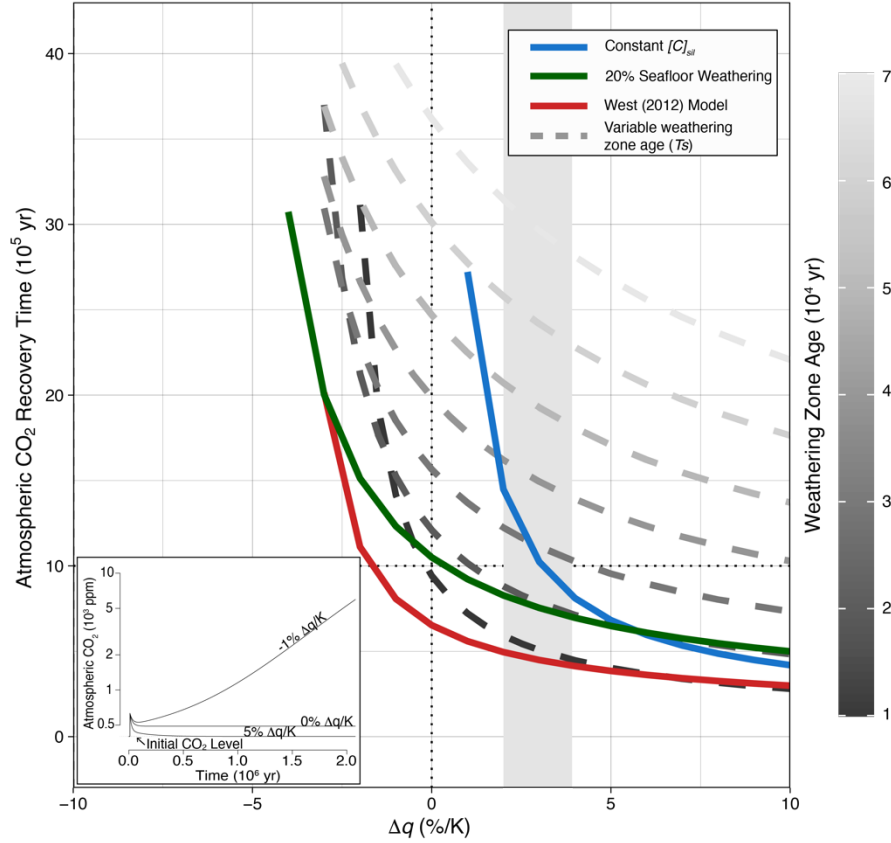


Figure 3: Recovery time [10^5 yr] for atmospheric CO_2 as a function of the q -sensitivity ($\%/K$) given a 1000 Pg C-cycle perturbation over 100 years. Dashed, gray lines use the standard parameters in Table S1 and variable T_s ; blue line assumes globally chemostatic catchments (*i.e.*, $[C]_{\text{sil}}$ is invariant with changes in CO_2); green line assumes that seafloor weathering accounts for 20% of the total alkalinity flux to the ocean (with all other parameters derived from Table S1); red line uses the silicate weathering model of West (2012) using values from Table S1. Vertical gray bar is the range of q -sensitivity from the C4MIP models considered herein. Inset shows atmospheric CO_2 (log-scale) of three representative experiments from the case of invariant $[C]_{\text{sil}}$ (blue line). Given a -1% q -sensitivity, atmospheric CO_2 increases in perpetuity. With 0% q -sensitivity, atmospheric CO_2 stabilizes but does not recover to pre-perturbation conditions. A q -sensitivity of 5% results in recovery to pre-perturbation CO_2 within ~ 700 kyr.

The above analysis suggests that, for the ocean-atmosphere system to recover to pre-perturbation CO_2 within 1 Ma, q -sensitivity must be greater than -2 $\%/K$ for any conceivable combination of weathering parameters; depending upon the precise model and parameterization, this value may be higher. For example, our preferred model of weathering—

using Maher and Chamberlain (2014)—requires a q -sensitivity of > 0 %/K for all reasonable estimates of the global weathering zone age.

However, the above modeling results assume that the Earth's surface can be characterized by single, mean values; such an assumption is likely incorrect. For example, while the C4MIP ensemble global-mean q -sensitivity is 3.1 ± 0.9 %/K (similar to the CMIP5 ensemble mean; Zhang et al., 2014), the spatial variability across large catchments is enormous, spanning -20 to +40 %/K (Dai & Trenberth, 2002; Scheff et al., 2017; Swann et al., 2016) (Figure 2). High, regional q -sensitivity may coincide with particularly reactive rocks (*i.e.*, low weathering zone ages and consequently high $[C]_{\text{sil}}$ -sensitivity) yielding a different CO_2 recovery time due to spatial co-variability in weathering zone reactivity and runoff change. Indeed, consistently high tropical runoff is the basis for theories that posit that exposure of reactive rocks (such as ophiolites or exposure of Large Igneous Province basalts) near the equator is the primary mode of long-term atmospheric CO_2 control (Donnadieu et al., 2004; Macdonald et al., 2019; Park et al., 2020; Swanson-Hysell & Macdonald, 2017).

Consequently, our estimate of the minimum q -sensitivity may be biased high because we do not consider regions with coincident high q -sensitivity and high weathering zone reactivity. To test this possibility, we modify CH2O-CHOO such that two distinct regions contribute toward the global silicate weathering flux: Mountain Region consists of a highly reactive weathering zone ($T_s = 1000$ yr) and contributes 50% of the initial, pre-perturbation global F_{silw} and Lowland Region comprises the rest of the world and is characterized by low weathering zone reactivity ($T_s = 39,000$ yr). Though this example is simplified, the Mountain Region may be considered equivalent to the role of mountains today, which contribute half of the global weathering flux (silicate and carbonate weathering combined) (Larsen et al., 2014). We treat the Mountain Region as having a fixed, positive q -sensitivity and the Lowland Region as having variable q -sensitivity. Globally, the q -sensitivity of mountains more likely tracks changes in P than changes in ET because most precipitation in mountain ranges is partitioned to q . In general, mountain ranges—particularly coastal ones—witness changes in P similar to the zonal mean, such that subtropical ranges see declines in P (and likely q), whereas mid-latitude ranges may see changes in P equal to or greater than 7 %/K (Shi & Durran, 2014). We repeat the experiment above, treating the q -sensitivity of the Low-Land Region as an independent parameter and injecting 1000 Pg C into the ocean-atmosphere system over 100 years. We test two cases: (1) where the Mountain Region has a fixed q -sensitivity of 3%/K and (2) where the Mountain Region has a fixed q -sensitivity of 6%/K.

Even the coincidence of high weathering zone reactivity with high runoff sensitivity still requires that average global runoff sensitivity be $> 0\%/K$ in order for atmospheric CO_2 to recover to pre-perturbation levels within ~ 1 Ma (Figure 4). Though a greater Mountain Region q -sensitivity (6 vs. 3 $\%/K$) results in faster recovery times, the minimum q -sensitivity is still 0 $\%/K$ to recover from a perturbation within 1 Myr. The different relationship between Δq and recovery time and compressed Δq variability in these simulations is due to how Δq is calculated for this experiment; in contrast to the first experiments above, where Δq is prescribed, here 50% of the Earth experiences a fixed 3 or 6 $\%/K$ q -sensitivity. Though we vary the q -sensitivity for the Lowland region (*i.e.*, with low weathering zone reactivity and high T_s), variations in global Δq are damped, given the fixed q -sensitivity in the Mountain Region (*i.e.* with high weathering zone reactivity and low T_s). Further, any decreases in global q when the q -sensitivity of the Lowland Region is $< 0\%/K$ are small (see inset in Figure 2) and global average $[C]_{sil}$ increases due to the effect of rising temperatures that offset the small decrease in q .

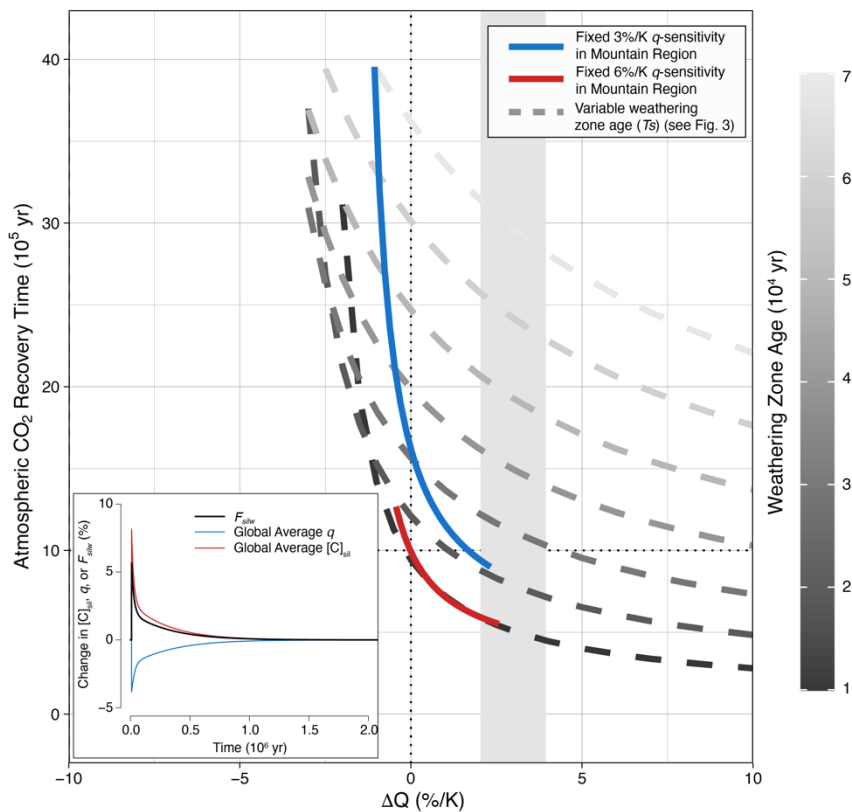


Figure 4: Recovery time [10⁵ y] for atmospheric CO₂ as a function of the average q -sensitivity ($\%/K$) given a 1000 Pg C-cycle perturbation over 100 years and two distinct weathering regions (outlined in text). Dashed lines use the standard parameters in Table S1 with variable weathering zone age (T_s) (see Figure 3); solid, red line assumes a fixed Mountain Region q -

sensitivity of 6 %/K; solid, blue line assumes a fixed Mountain Region q -sensitivity of 3%/K. In both cases, the q -sensitivity of region 2 is permitted to vary between -10 and 10 %/K. Gray shading is the C4MIP estimate of runoff sensitivity. We calculate Δq as the percentage change in global q at the peak of the C-cycle perturbation relative to starting global q against the change in global T between the peak of the perturbation and the initial condition. Inset shows how F_{silw} (black line), global average q (blue line), and global average $[C]_{sil}$ (red line) change during a perturbation when the Lowland Region has a -10 %/K q -sensitivity and the Mountain Region has a low T_s (ie, highly reactive) and a fixed q -sensitivity of 6 %/K.

4. Geologic Constraints on q -sensitivity

We now use a well-studied past large carbon cycle perturbation—the Paleocene-Eocene Thermal Maximum (PETM), ~ 55 Ma—to constrain the q response to a large carbon cycle perturbation. Though the exact cause of the PETM remains debated, a substantial quantity of carbon (5000-20000 Pg C) was released to the ocean-atmosphere system within 3000–10000 years, lowering global marine dissolved inorganic carbon (DIC) $\delta^{13}C$ by 3–4‰ (Cui et al., 2011; Gutjahr et al., 2017; Panchuk et al., 2008; Penman et al., 2016). This negative carbon isotope excursion is thought to have lasted $\sim 170 \pm 30$ ka (Zeebe & Lourens, 2019), at which point the global marine DIC pool had recovered to pre-perturbation $\delta^{13}C$ values. Critically, the recovery from the PETM was likely mediated by silicate weathering—as evidenced by an excess burial of Si across the PETM (Penman, 2016)—and involved large-scale increases in runoff in places (Chen et al., 2018; Foreman et al., 2012). The PETM thus provides an opportunity to test our CH₂O-CHOO model and determine the most likely q -sensitivity necessary to match the duration of the $\delta^{13}C$ negative excursion.

We force CH₂O-CHOO with net carbon emissions and net emissions $\delta^{13}C$ estimates from Gutjahr et al. (2017), which also includes estimates of increased organic carbon burial during the recovery phase (Bowen, 2013; Bowen & Zachos, 2010). As before, we prescribe a wide-range of q -sensitivity estimates that determine how runoff responds to changes in atmospheric CO₂. Because we also do not *a priori* know the average weathering zone age (T_s) at the PETM, we vary this parameter as well, though evidence from a range of carbon cycle models and isotopic data indicates that bulk Earth surface weatherability was lower in the early Eocene, likely reflecting higher T_s (Caves et al., 2016; Froelich & Misra, 2014; Krissansen-Totton & Catling, 2017; van der Ploeg et al., 2018; Vervoort et al., 2021).

Given these constraints, we find that, to match the duration of the PETM negative $\delta^{13}\text{C}$ excursion requires q -sensitivity to be equal to or greater than 0%/K if T_s is similar to today (Figure 5). If, instead, T_s was higher in the late Paleocene, then q -sensitivity must be higher to match the duration of the $\delta^{13}\text{C}$ negative excursion. Though our model lacks a number of critical carbon cycle processes that are thought to modulate the impact of rapid CO_2 emissions (such as seafloor carbonate sediments that buffer against acidic bottom waters), these effects are likely to matter primarily for the initial stages of the perturbation, rather than mediating the long-term removal of CO_2 by silicate weathering (Penman et al., 2020). We note that, while the PETM provides a useful analogue with which to understand how q responds to CO_2 , we do not expect the q -sensitivity at the PETM to exactly match that of today's Earth, due to paleogeographic and floral changes that have occurred during the Cenozoic that would have changed precisely how much precipitation is converted to q as CO_2 rises.

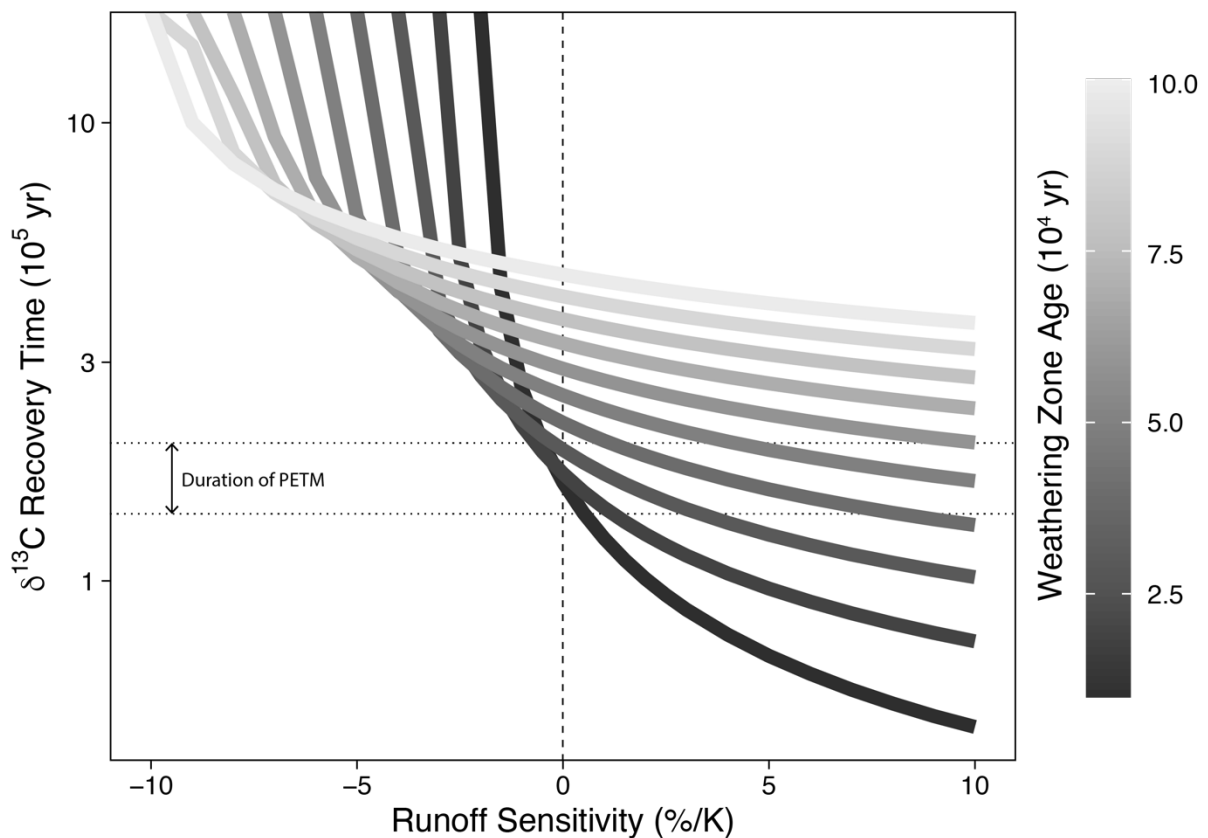


Figure 5: Estimated $\delta^{13}\text{C}$ recovery times (10^5 yr) as a function of the q -sensitivity and the weathering zone age (grayscale). Horizontal dotted lines indicate the estimated $\delta^{13}\text{C}$ recovery time based upon astrochronological tuning of the PETM $\delta^{13}\text{C}$ record (Zeebe & Lourens, 2019).

5. Implications

The geological record—and our understanding of how the Earth recovers from carbon cycle perturbations—provides long-term constraints on how q must respond to increases in atmospheric CO₂. In a broad sense, our results bolster previous work pointing towards the importance of runoff in modulating the long-term carbon cycle and climate (Barron et al., 1989; François & Walker, 1992; Otto-Bliesner, 1995). Critically, our long-term perspective on the runoff response to CO₂ incorporates a number of processes that are thought to affect q but are difficult to constrain in the modern due to temporally limited instrumental records or experiments. For example, remotely sensed products of global discharge are affected by internal variability, which precludes their use currently in assessing how total runoff will respond to rising atmospheric CO₂ (Chandanpurkar et al., 2017, 2021). Experiments that seek to understand how plant responses might affect runoff—such as the Free Air CO₂ Enrichment (FACE) experiments—are relatively short-term and the long-term response of ecosystems may differ from the short-term response due to changes in nutrient demands and supply and non-linear plant responses to increasing CO₂. In this sense, the known response of the long-term carbon cycle—and how this response is linked to q —provides a constraint on how global q must respond to rising CO₂ that avoids the inherent noisiness of the relatively short instrumental record.

Our finding that q -sensitivity must be greater than or equal to 0%/K (when using our preferred model of weathering) holds implications for the processes that control changes in runoff with warming. Though PET should increase faster than P , stomatal conductance changes driven by CO₂ fertilization and increasing vapor pressure deficits may result in a greater fraction of P being partitioned to q (Novick et al., 2016; Roderick et al., 2015; Swann et al., 2016). This effect, however, is complicated by a number of factors that may instead drive greater plant water use, including a longer growing season (Mankin et al., 2019) and widespread greening that increases total leaf area (Zhu et al., 2016). How these factors will interact across the planet to produce a change in global q remains difficult to predict; across much of the Earth's surface, for example, models frequently do not even agree on the sign of the q change (Figure 2). For example, in much of the mid-latitudes, projected ensemble mean runoff changes are negligible, with little model agreement on the sign of the change (Elbaum et al., 2022; Scheff et al., 2017). Our results suggest that the processes on a global scale that tend towards greater q with increasing atmospheric CO₂ and warming—such as reductions in stomatal conductance—will be predominant in a warmer world, though there may be substantial spatial variability. Further, we note that our results likely hold on the millennial or

longer timescale, and, as such and given the potential complexity of the biospheric response to warming, there may be long periods when global runoff may not increase in response to warming.

Our findings also hold implications for understanding how paleogeographic changes may have modified the strength of the silicate weathering negative feedback in the geologic past. General circulation model simulations highlight that the runoff response to warming is spatially complex; such results indicate the precise global q -sensitivity may vary with the position of the continents (Otto-Bliesner, 1995). For example, models frequently predict that subtropical areas should become drier (Burls & Fedorov, 2017; Mankin et al., 2019), suggesting that a world with predominantly sub-tropical continents would perhaps see a minimal increase or even decrease in global discharge. Our results suggest that either there are additional mechanisms that keep global discharge from decreasing or that such a continental configuration has never occurred on Earth during a major carbon cycle perturbation. If such an Earth experienced a major perturbation, our results indicate that atmospheric CO₂ may have never returned to pre-perturbation values.

Though we have attempted to test the robustness of our result by using different parameters and even a different model of weathering (West, 2012), there are a number of hypothesized feedbacks with warming and rising CO₂ that may impact weathering fluxes, but are not—to our knowledge—yet incorporated into any model of weathering. For example, warming is thought to increase the number of extreme precipitation events (Fischer & Knutti, 2016) which is likely to increase q relative to ET as P is rapidly converted to q . Though q increases with these extreme events, much of the water may bypass the weathering zone, limiting their impact on weathering. In turn, extreme events may also increase erosional efficiency (Deal et al., 2017, 2018), increasing the availability of fresh minerals and thereby enhancing silicate weathering by reducing average T_s . Though this process is not represented in our model, we suspect that the impact of extreme events on silicate weathering is likely to be minimal, as lower T_s is offset by a greater quantity of q that bypasses the weathering zone via overland flow or exceptionally fast transit times. Additionally, other carbon fluxes may change with a warming climate, such as erosion and burial of terrestrial organic carbon (Hilton, 2017) or increases in marine organic burial due to widespread anoxia (Lau et al., 2016). However, these mechanisms also rely upon runoff to transport terrestrial organic matter to continental shelves for burial and to delivery sufficient nutrients to drive marine anoxia (Them et al., 2017), suggesting that increases in runoff are also necessary for these hypothesized negative feedbacks on CO₂.

Our work highlights the tight coupling between the hydrological cycle and the long-term carbon cycle and demonstrates that this coupling places a lower-bound on the globally averaged runoff response to warming and rising CO₂. Whereas abundant research has examined the links between the hydrological cycle and the short-term carbon cycle (Forzieri et al., 2020; Lemordant et al., 2018; Novick et al., 2016), our work demonstrates that the operation of the long-term carbon cycle is also critically linked to the hydrological cycle. It is this linkage that has acted to remove excess CO₂ from the ocean-atmosphere system during perturbations, thus maintaining Earth's habitability. Though our knowledge of weathering continues to improve, ongoing model and observational work indicates the centrality of runoff in linking the weathering of silicate rocks to the ultimate burial of carbon as calcium carbonate in the marine environment. Thus, the importance of runoff for weathering indicates that, on long timescales, runoff must increase as CO₂ rises.

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Software Availability Statement:

Model source code used for this research is available in Rugenstein and Winkler (2022) and can be accessed at: <https://doi.org/10.5281/zenodo.7219627>.

References:

- Arora, V. K., Boer, G. J., Friedlingstein, P., Eby, M., Jones, C. D., Christian, J. R., et al. (2013). Carbon-concentration and carbon-climate feedbacks in CMIP5 earth system models. *Journal of Climate*, 26(15), 5289–5314. <https://doi.org/10.1175/JCLI-D-12-00494.1>
- Arora, V. K., Katavouta, A., Williams, R. G., Jones, C. D., Brovkin, V., Friedlingstein, P., et al. (2020). Carbon – concentration and carbon – climate feedbacks in CMIP6 models and their comparison to CMIP5 models. *Biogeosciences*, 17, 4173–4222.
- Barron, E. J., Hay, W. W., & Thompson, S. (1989). The hydrologic cycle: A major variable during earth history. *Palaeogeography, Palaeoclimatology, Palaeoecology*, 75(3), 157–174. [https://doi.org/10.1016/0031-0182\(89\)90175-2](https://doi.org/10.1016/0031-0182(89)90175-2)
- Baum, M., Fu, M., & Bourguet, S. (2022). Sensitive Dependence of Global Climate to Continental Geometry. *Geophysical Research Letters*, 49(11), 1–8. <https://doi.org/10.1029/2022gl098843>
- Berner, R. A., & Kothavala, Z. (2001). GEOCARB III: A revised model of atmospheric CO₂ over Phanerozoic time. *American Journal of Science*, 301, 182–204. Retrieved from <http://ajsonline.org/content/301/2/182.short>
- Berner, Robert A., & Caldeira, K. (1997). The need for mass balance and feedback in the geochemical carbon cycle. *Geology*, 25(10), 955–956. [https://doi.org/10.1130/0091-7613\(1997\)025<0955:TNFMBA>2.3.CO;2](https://doi.org/10.1130/0091-7613(1997)025<0955:TNFMBA>2.3.CO;2)
- Bowen, G. J. (2013). Up in smoke: A role for organic carbon feedbacks in Paleogene hyperthermals. *Global and Planetary Change*, 109, 18–29. <https://doi.org/10.1016/j.gloplacha.2013.07.001>
- Bowen, G. J., & Zachos, J. C. (2010). Rapid carbon sequestration at the termination of the Palaeocene–Eocene Thermal Maximum. *Nature Geoscience*, 3(12), 866–869. <https://doi.org/10.1038/ngeo1014>
- Brady, P. V., & Gíslason, S. R. (1997). Seafloor weathering controls on atmospheric CO₂ and global climate. *Geochimica et Cosmochimica Acta*, 61(5), 965–973. [https://doi.org/10.1016/S0016-7037\(96\)00385-7](https://doi.org/10.1016/S0016-7037(96)00385-7)
- Burls, N. J., & Fedorov, A. V. (2017). Wetter subtropics in a warmer world: Contrasting past and future hydrological cycles. *Proceedings of the National Academy of Sciences*, 114(49), 12888–12893. <https://doi.org/10.1073/pnas.1703421114>
- Caves, J. K., Jost, A. B., Lau, K. V., & Maher, K. (2016). Cenozoic carbon cycle imbalances and a variable silicate weathering feedback. *Earth and Planetary Science Letters*, 450,

152–163. <https://doi.org/10.1016/j.epsl.2016.06.035>

Caves Rugenstein, J. K., Ibarra, D. E., & von Blanckenburg, F. (2019). Neogene cooling driven by land surface reactivity rather than increased weathering fluxes. *Nature*, 571, 99–102. <https://doi.org/10.1038/s41586-019-1332-y>

Chandanpurkar, H. A., Reager, J. T., Famiglietti, J. S., & Syed, T. H. (2017). Satellite- and reanalysis-based mass balance estimates of global continental discharge (1993–2015). *Journal of Climate*, 30(21), 8481–8495. <https://doi.org/10.1175/JCLI-D-16-0708.1>

Chandanpurkar, H. A., Reager, J. T., Famiglietti, J. S., Nerem, R. S., Chambers, D. P., Lo, M. H., et al. (2021). The Seasonality of Global Land and Ocean Mass and the Changing Water Cycle. *Geophysical Research Letters*, 48(7). <https://doi.org/10.1029/2020GL091248>

Chen, C., Guerit, L., Foreman, B. Z., Hassenruck-Gudipati, H. J., Adatte, T., Honegger, L., et al. (2018). Estimating regional flood discharge during Palaeocene-Eocene global warming. *Scientific Reports*, 8(1), 1–8. <https://doi.org/10.1038/s41598-018-31076-3>

Colbourn, G., Ridgwell, A., & Lenton, T. M. (2015). The time scale of the silicate weathering negative feedback on atmospheric CO₂. *Global Biogeochemical Cycles*, 29, 583–596. <https://doi.org/10.1002/2014GB005054>

Coogan, L. A., & Gillis, K. M. (2018). Low-Temperature Alteration of the Seafloor: Impacts on Ocean Chemistry. *Annu. Rev. Earth Planet. Sci*, 46, 21–45. <https://doi.org/10.1146/annurev-earth-082517>

Cook, B. I., Smerdon, J. E., Seager, R., & Coats, S. (2014). Global warming and 21st century drying. *Climate Dynamics*, 43, 2607–2627. <https://doi.org/10.1007/s00382-014-2075-y>

Cui, Y., Kump, L. R., Ridgwell, A. J., Charles, A. J., Junium, C. K., Diefendorf, A. F., et al. (2011). Slow release of fossil carbon during the Palaeocene–Eocene Thermal Maximum. *Nature Geoscience*, 4(7), 481–485. <https://doi.org/10.1038/ngeo1179>

Dai, A., & Trenberth, K. E. (2002). Estimates of Freshwater Discharge from Continents : Latitudinal and Seasonal Variations. *Journal of Hydrometeorology*, 3, 660–687.

Deal, E., Favre, A.-C., & Braun, J. (2017). Rainfall variability in the Himalayan orogen and its relevance to erosion processes. *Water Resources Research*, 53, 4004–4021. <https://doi.org/10.1002/2013WR014979>.Reply

Deal, E., Botter, G., & Braun, J. (2018). Understanding the role of rainfall and hydrology in determining fluvial erosion efficiency. *Journal of Geophysical Research: Earth Surface*. <https://doi.org/10.1002/2017JF004393>

Donnadieu, Y., Godd  ris, Y., Ramstein, G., N  d  lec, A., & Meert, J. (2004). A ‘snowball

573 Earth' climate triggered by continental break-up through changes in runoff. *Nature*,
 574 428(6980), 303–306. <https://doi.org/10.1038/nature02408>
 575 Douville, H., Raghavan, K., Renwick, J., Allan, R. P., Arias, P. A., Barlow, M., et al. (2021).
 576 Water Cycle Changes. In V. Masson-Delmotte, P. Zhai, A. Pirani, S. L. Connors, C.
 577 Péan, S. Berger, et al. (Eds.), *Climate Change 2021: The Physical Science Basis.*
 578 *Contribution of Working Group I to the Sixth Assessment Report of the*
 579 *Intergovernmental Panel on Climate Change* (p. 239). Cambridge University Press.
 580 Elbaum, E., Garfinkel, C. I., Adam, O., Morin, E., Rostkier-Edelstein, D., & Dayan, U.
 581 (2022). Uncertainty in projected changes in precipitation minus evaporation: Dominant
 582 role of dynamic circulation changes and weak role for thermodynamic changes.
 583 *Geophysical Research Letters*. <https://doi.org/10.1029/2022gl097725>
 584 Ficklin, D. L., & Novick, K. A. (2017). Historic and projected changes in vapor pressure
 585 deficit suggest a continental-scale drying of the United States atmosphere. *Journal of*
 586 *Geophysical Research: Atmospheres*, 122, 2061–2079.
 587 <https://doi.org/10.1002/2016JD025855>
 588 Fischer, E. M., & Knutti, R. (2016). Observed heavy precipitation increase confirms theory
 589 and early models. *Nature Climate Change*, 6(11), 986–991.
 590 <https://doi.org/10.1038/nclimate3110>
 591 Foreman, B. Z., Heller, P. L., & Clementz, M. T. (2012). Fluvial response to abrupt global
 592 warming at the Palaeocene/Eocene boundary. *Nature*, 1–4.
 593 <https://doi.org/10.1038/nature11513>
 594 Forzieri, G., Miralles, D. G., Ciais, P., Alkama, R., Ryu, Y., Duveiller, G., et al. (2020).
 595 Increased control of vegetation on global terrestrial energy fluxes. *Nature Climate*
 596 *Change*, 10. <https://doi.org/10.1038/s41558-020-0717-0>
 597 François, L. M., & Walker, J. C. G. (1992). Modelling the Phanerozoic carbon cycle and
 598 climate: constraints from the $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratio of seawater. *American Journal of*
 599 *Science*. <https://doi.org/10.2475/ajs.292.2.81>
 600 Froelich, F., & Misra, S. (2014). Was the late Paleocene-early Eocene hot because Earth was
 601 flat? An ocean lithium isotope view of mountain building, continental weathering,
 602 carbon dioxide, and Earth's Cenozoic climate. *Oceanography*, 27(1), 36–49. Retrieved
 603 from <http://eprints.esc.cam.ac.uk/2999/>
 604 Godsey, S. E., Kirchner, J. W., & Clow, D. W. (2009). Concentration – discharge
 605 relationships reflect chemostatic characteristics of US catchments. *Hydrological*
 606 *Processes*, 23, 1844–1864. <https://doi.org/10.1002/hyp>

- Godsey, S. E., Hartmann, J., & Kirchner, J. W. (2019). Catchment chemostasis revisited: Water quality responds differently to variations in weather and climate. *Hydrological Processes*, 1–14. <https://doi.org/10.1002/hyp.13554>
- Good, S. P., Noone, D., & Bowen, G. (2015). Hydrologic connectivity constrains partitioning of global terrestrial water fluxes. *Science*, 349(6244), 175–177.
- Graham, R. J., & Pierrehumbert, R. T. (2020). Thermodynamic and Energetic Limits on Continental Silicate Weathering Strongly Impact the Climate and Habitability of Wet, Rocky Worlds. Retrieved from <http://arxiv.org/abs/2004.14058>
- Gutjahr, M., Ridgwell, A., Sexton, P. F., Anagnostou, E., Pearson, P. N., Pälike, H., et al. (2017). Very large release of mostly volcanic carbon during the Palaeocene–Eocene Thermal Maximum. *Nature*, 548(7669), 573–577. <https://doi.org/10.1038/nature23646>
- Held, I., & Soden, B. (2006). Robust responses of the hydrological cycle to global warming. *Journal of Climate*, 19, 5686–5699.
- Hilton, R. G. (2017). Climate regulates the erosional carbon export from the terrestrial biosphere. *Geomorphology*, 277, 118–132. <https://doi.org/10.1016/j.geomorph.2016.03.028>
- Hilton, R. G., & West, A. J. (2020). Mountains, erosion and the carbon cycle. *Nature Reviews Earth and Environment*. Springer US. <https://doi.org/10.1038/s43017-020-0058-6>
- Ibarra, D. E., Caves, J. K., Moon, S., Thomas, D. L., Hartmann, J., Chamberlain, C. P., & Maher, K. (2016). Differential weathering of basaltic and granitic catchments from concentration–discharge relationships. *Geochimica et Cosmochimica Acta*, 190, 265–293. <https://doi.org/10.1016/j.gca.2016.07.006>
- Knutti, R., Rugenstein, M. A. A., & Hegerl, G. C. (2017). Beyond equilibrium climate sensitivity. *Nature Geoscience*, 10, 727–736. <https://doi.org/10.1038/ngeo3017>
- Krissansen-Totton, J., & Catling, D. C. (2017). Constraining climate sensitivity and continental versus seafloor weathering using an inverse geological carbon cycle model. *Nature Communications*, 8, 15423. <https://doi.org/10.1038/ncomms15423>
- Kump, L. R., & Arthur, M. A. (1999). Interpreting carbon-isotope excursions: carbonates and organic matter. *Chemical Geology*, 161(1–3), 181–198. [https://doi.org/10.1016/S0009-2541\(99\)00086-8](https://doi.org/10.1016/S0009-2541(99)00086-8)
- Kump, L. R., Brantley, S. L., & Arthur, M. A. (2000). Chemical weathering, atmospheric CO₂, and climate. *Annual Review of Earth and Planetary Sciences*, 28, 611–667. Retrieved from <http://www.annualreviews.org/doi/abs/10.1146/annurev.earth.28.1.611>
- Larsen, I. J., Montgomery, D. R., & Greenberg, H. M. (2014). The contribution of mountains

to global denudation. *Geology*, 42, 527–530. <https://doi.org/10.1130/G35136.1>

Lau, K. V., Maher, K., Altiner, D., Kelley, B. M., Kump, L. R., Lehrmann, D. J., et al. (2016). Marine anoxia and delayed Earth system recovery after end-Permian extinction. *Proceedings of the National Academy of Sciences*. <https://doi.org/10.1073/pnas.1515080113>

Lemordant, L., Gentine, P., Swann, A. S., Cook, B. I., & Scheff, J. (2018). Critical impact of vegetation physiology on the continental hydrologic cycle in response to increasing CO₂. *Proceedings of the National Academy of Sciences*, 115(16), 201720712. <https://doi.org/10.1073/pnas.1720712115>

Lenton, T. M., & Britton, C. (2006). Enhanced carbonate and silicate weathering accelerates recovery from fossil fuel CO₂ perturbations. *Global Biogeochemical Cycles*, 20(3), 1–12. <https://doi.org/10.1029/2005GB002678>

Li, L., Stewart, B., Zhi, W., Sadayappan, K., Ramesh, S., Kerins, D., et al. (2022). Climate Controls on River Chemistry. *Earth's Future*, 10(6), 1–17. <https://doi.org/10.1029/2021EF002603>

Macdonald, F. A., Swanson-Hysell, N. L., Park, Y., Lisiecki, L., & Jagoutz, O. (2019). Arc-continent collisions in the tropics set Earth's climate state. *Science*, 364, 181–184.

Maher, K., & Chamberlain, C. P. (2014). Hydrologic regulation of chemical weathering and the geologic carbon cycle. *Science*, 343(6178), 1502–1504. <https://doi.org/10.1126/science.1250770>

Mankin, J. S., Seager, R., Smerdon, J. E., Cook, B. I., & Williams, A. P. (2019). Mid-latitude freshwater availability reduced by projected vegetation responses to climate change. *Nature Geoscience*. <https://doi.org/10.1038/s41561-019-0480-x>

Milly, P. C. D., & Dunne, K. A. (2016). Potential evapotranspiration and continental drying. *Nature Climate Change*, 6, 946–949. <https://doi.org/10.1038/NCLIMATE3046>

Milly, P. C. D., & Dunne, K. A. (2020). Colorado River flow dwindles as warming-driven loss of reflective snow energizes evaporation. *Science*, 9187(February), eaay9187. <https://doi.org/10.1126/science.aay9187>

Novick, K. A., Ficklin, D. L., Stoy, P. C., Williams, C. A., Bohrer, G., Oishi, A. C., et al. (2016). The increasing importance of atmospheric demand for ecosystem water and carbon fluxes. *Nature Climate Change*, 6(11), 1023–1027. <https://doi.org/10.1038/nclimate3114>

Otto-Bliesner, B. L. (1995). Continental drift, runoff, and weathering feedbacks: Implications from climate model experiments. *Journal of Geophysical Research*, 100(D6), 11537–

675 11548. <https://doi.org/10.1029/95JD00591>
 676 Overpeck, J. T., & Udall, B. (2020). Climate change and the aridification of North America.
 677 *Proceedings of the National Academy of Sciences*, 0, 202006323.
 678 <https://doi.org/10.1073/pnas.2006323117>
 679 Panchuk, K., Ridgwell, A., & Kump, L. R. (2008). Sedimentary response to Paleocene-
 680 Eocene Thermal Maximum carbon release: A model-data comparison. *Geology*, 36,
 681 315–318. <https://doi.org/10.1130/G24474A.1>
 682 Park, Y., Maffre, P., Godderis, Y., Macdonald, F. A., Anttila, E. S. C. C., Swanson-Hysell,
 683 N. L., et al. (2020). Emergence of the Southeast Asian islands as a driver for Neogene
 684 cooling. *Proceedings of the National Academy of Sciences*, 117(41), 25319–25326.
 685 <https://doi.org/10.1073/pnas.2011033117>
 686 Pendergrass, A. G., & Hartmann, D. L. (2014). The atmospheric energy constraint on global-
 687 mean precipitation change. *Journal of Climate*, 27(2), 757–768.
 688 <https://doi.org/10.1175/JCLI-D-13-00163.1>
 689 Penman, D. E. (2016). Silicate weathering and North Atlantic silica burial during the
 690 Paleocene-Eocene Thermal Maximum. *Geology*, (9). <https://doi.org/10.1130/G37704.1>
 691 Penman, D. E., Turner, K. S., Sexton, P., Norris, R., Dickson, A. J., Boulila, S., et al. (2016).
 692 A carbonate compensation depth overshoot in the aftermath of the Paleocene-Eocene
 693 Thermal Maximum. *Nature Geoscience*. <https://doi.org/10.1038/ngeo2757>
 694 Penman, D. E., Rugenstein, J. K. C., Ibarra, D. E., & Winnick, M. J. (2020). Silicate
 695 weathering as a feedback and forcing in Earth's climate and carbon cycle. *Earth-Science*
 696 *Reviews*, 209, 103298. <https://doi.org/10.1016/j.earscirev.2020.103298>
 697 Piao, S., Friedlingstein, P., Ciais, P., de Noblet-Ducoudre, N., Labat, D., & Zaehle, S. (2007).
 698 Changes in climate and land use have a larger direct impact than rising CO₂ on global
 699 river runoff trends. *Proceedings of the National Academy of Sciences*, 104(39), 15242–
 700 15247. <https://doi.org/10.1073/pnas.0707213104>
 701 Rugenstein, J. K. C., & Winkler, A. J. (2022). CH2O-CHOO v.1.1. Zenodo.
 702 <https://doi.org/10.5281/zenodo.7219627>.
 703 van der Ploeg, R., Selby, D., Cramwinckel, M. J., Li, Y., Bohaty, S. M., Middelburg, J. J., &
 704 Sluijs, A. (2018). Middle Eocene greenhouse warming facilitated by diminished
 705 weathering feedback. *Nature Communications*, 9(1), 2877.
 706 <https://doi.org/10.1038/s41467-018-05104-9>
 707 Roderick, M. L., Greve, P., & Farquhar, G. D. (2015). On the assessment of aridity with
 708 changes in atmospheric CO₂. *Water Resources Research*, 51(7), 5450–5463.

709 <https://doi.org/10.1002/2015WR017031>

710 Sagan, C., & Mullen, G. (1972). Earth and Mars: evolution of atmospheres and surface
711 temperatures. *Science*, 177(4043), 52–56. <https://doi.org/10.1126/science.177.4043.52>

712 Scheff, J., & Frierson, D. M. W. (2014). Scaling potential evapotranspiration with
713 greenhouse warming. *Journal of Climate*, 27(4), 1539–1558.
714 <https://doi.org/10.1175/JCLI-D-13-00233.1>

715 Scheff, J., Seager, R., Liu, H., & Coats, S. (2017). Are glacials dry? Consequences for
716 paleoclimatology and for greenhouse warming. *Journal of Climate*, 30(17), 6593–6609.
717 <https://doi.org/10.1175/JCLI-D-16-0854.1>

718 Schlesinger, W. H., & Jasechko, S. (2014). Transpiration in the global water cycle.
719 *Agricultural and Forest Meteorology*, 189–190, 115–117.
720 <https://doi.org/10.1016/j.agrformet.2014.01.011>

721 Shi, X., & Durran, D. R. (2014). The Response of Orographic Precipitation over Idealized
722 Midlatitude Mountains Due to Global Increases in CO₂. *Journal of Climate*, 27(11),
723 3938–3956. <https://doi.org/10.1175/JCLI-D-13-00460.1>

724 Swann, A. L. S., Hoffman, F. M., Koven, C. D., & Randerson, J. T. (2016). Plant responses
725 to increasing CO₂ reduce estimates of climate impacts on drought severity. *Proceedings*
726 *of the National Academy of Sciences*, 113(36), 10019–10024.
727 <https://doi.org/10.1073/pnas.1604581113>

728 Swanson-Hysell, N. L., & Macdonald, F. A. (2017). Tropical weathering of the Taconic
729 orogeny as a driver for Ordovician cooling. *Geology*, 45(8), 719–722.
730 <https://doi.org/10.1130/G38985.1>

731 Them, T. R., Gill, B. C., Selby, D., Gröcke, D. R., Friedman, R. M., & Owens, J. D. (2017).
732 Evidence for rapid weathering response to climatic warming during the Toarcian
733 Oceanic Anoxic Event. *Scientific Reports*, 7(1), 1–10. [https://doi.org/10.1038/s41598-](https://doi.org/10.1038/s41598-017-05307-y)
734 [017-05307-y](https://doi.org/10.1038/s41598-017-05307-y)

735 Torres, M. A., West, A. J., & Li, G. (2014). Sulphide oxidation and carbonate dissolution as a
736 source of CO₂ over geological timescales. *Nature*, 507, 346–349.
737 <https://doi.org/10.1038/nature13030>

738 Uchikawa, J., & Zeebe, R. E. (2008). Influence of terrestrial weathering on ocean
739 acidification and the next glacial inception. *Geophys. Res. Lett.*, 35(23).
740 <https://doi.org/10.1029/2008GL035963>

741 Ukkola, A. M., Prentice, I. C., Keenan, T. F., van Dijk, A. I. J. M., Viney, N. R., Myneni, R.
742 B., & Bi, J. (2016). Reduced streamflow in water-stressed climates consistent with CO₂

- effects on vegetation. *Nature Climate Change*, 6, 75–78.
<https://doi.org/10.1038/nclimate2831>
- Vervoort, P., Kirtland Turner, S., Rochholz, F., & Ridgwell, A. (2021). Earth System Model Analysis of How Astronomical Forcing Is Imprinted Onto the Marine Geological Record: The Role of the Inorganic (Carbonate) Carbon Cycle and Feedbacks. *Paleoceanography and Paleoclimatology*, 36(10), 1–20.
<https://doi.org/10.1029/2020pa004090>
- Volk, T. (1987). Feedbacks between weathering and atmospheric CO₂ over the last 100 million years. *American Journal of Science*. <https://doi.org/10.2475/ajs.287.8.763>
- Walker, J. C. G., Hays, P. B., & Kasting, J. F. (1981). A negative feedback mechanism for the long-term stabilization of Earth's surface temperature. *Journal of Geophysical Research*, 86(C10), 9776–9782.
- Wang-Erlandsson, L., van der Ent, R. J., Gordon, L. J., & Savenije, H. H. G. (2014). Contrasting roles of interception and transpiration in the hydrological cycle – Part 1: Temporal characteristics over land. *Earth System Dynamics*, 5(2), 441–469.
<https://doi.org/10.5194/esd-5-441-2014>
- Wei, Z., Yoshimura, K., Wang, L., Miralles, D. G., Jasechko, S., & Lee, X. (2017). Revisiting the contribution of transpiration to global terrestrial evapotranspiration. *Geophysical Research Letters*, 44(6), 2792–2801.
<https://doi.org/10.1002/2016GL072235>
- West, A. J. (2012). Thickness of the chemical weathering zone and implications for erosional and climatic drivers of weathering and for carbon-cycle feedbacks. *Geology*, 40(9), 811–814. <https://doi.org/10.1130/G33041.1>
- White, A. F., & Blum, A. E. (1995). Effects of climate on chemical weathering in watersheds. *Geochimica et Cosmochimica Acta*, 59(9), 1729–1747.
[https://doi.org/10.1016/0016-7037\(95\)00078-E](https://doi.org/10.1016/0016-7037(95)00078-E)
- Winkler, A. J., Myneni, R. B., Hannart, A., Sitch, S., Haverd, V., Lombardozzi, D., et al. (2021). Slowdown of the greening trend in natural vegetation with further rise in atmospheric CO₂. *Biogeosciences*, 18(17), 4985–5010. <https://doi.org/10.5194/bg-18-4985-2021>
- Winnick, M., & Maher, K. (2018). Relationships between CO₂, thermodynamic limits on silicate weathering, and the strength of the silicate weathering feedback. *Earth and Planetary Science Letters*, 485, 111–120.
<https://doi.org/https://doi.org/10.1016/j.epsl.2018.01.005>

- Zeebe, R. E., & Caldeira, K. (2008). Close mass balance of long-term carbon fluxes from ice-core CO₂ and ocean chemistry records. *Nature Geoscience*, 1(5), 312–315.
<https://doi.org/10.1038/ngeo185>
- Zeebe, R. E., & Lourens, L. J. (2019). Solar System chaos and the Paleocene–Eocene boundary age constrained by geology and astronomy. *Science*, 365, 926–929.
- Zhang, X., Tang, Q., Zhang, X., & Lettenmaier, D. P. (2014). Runoff sensitivity to global mean temperature change in the CMIP5 Models. *Geophysical Research Letters*, 41, 5492–5498. <https://doi.org/10.1002/2014GL061184>. Received
- Zhu, Z., Piao, S., Myneni, R. B., Huang, M., Zeng, Z., Canadell, J. G., et al. (2016). Greening of the Earth and its drivers. *Nature Climate Change*, 6, 791–795.
<https://doi.org/10.1038/NCLIMATE3004>

Supporting Information References

- Arora, V.K., Boer, G.J., Friedlingstein, P., Eby, M., Jones, C.D., Christian, J.R., Bonan, G., Bopp, L., Brovkin, V., Cadule, P., Hajima, T., Ilyina, T., Lindsay, K., Tjiputra, J.F., Wu, T., 2013. Carbon-concentration and carbon-climate feedbacks in CMIP5 earth system models. *J. Clim.* 26, 5289–5314. <https://doi.org/10.1175/JCLI-D-12-00494.1>
- Arora, V.K., Katavouta, A., Williams, R.G., Jones, C.D., Brovkin, V., Friedlingstein, P., Schwinger, J., Bopp, L., Boucher, O., Cadule, P., Chamberlain, M.A., Christian, J.R., Delire, C., Fisher, R.A., Hajima, T., Ilyina, T., Joetzjer, E., Kawamiya, M., Koven, C.D., Krasting, J.P., Law, R.M., Lawrence, D.M., Lenton, A., Lindsay, K., Pongratz, J., Raddatz, T., Séférian, R., Tachiiri, K., Tjiputra, J.F., Wiltshire, A., Wu, T., Ziehn, T., 2020. Carbon – concentration and carbon – climate feedbacks in CMIP6 models and their comparison to CMIP5 models. *Biogeosciences* 17, 4173–4222.
- Berner, R.A., 2006. GEOCARBSULF: A combined model for Phanerozoic atmospheric O₂ and CO₂. *Geochim. Cosmochim. Acta* 70, 5653–5664.
<https://doi.org/10.1016/j.gca.2005.11.032>
- Brady, P. V., 1991. The effect of silicate weathering on global temperature and atmospheric CO₂. *J. Geophys. Res.* 96. <https://doi.org/10.1029/91jb01898>
- Brook, G., Folkoff, M., Box, E., 1983. A World Model of Soil Carbon Dioxide. *Earth Surf. Process. Landforms* 8, 79–88.
- Caves Rugenstein, J.K., Ibarra, D.E., von Blanckenburg, F., 2019. Neogene cooling driven by land surface reactivity rather than increased weathering fluxes. *Nature* 571, 99–102.
<https://doi.org/10.1038/s41586-019-1332-y>

811 Gaillardet, J., Dupré, B., Louvat, P., Allegre, C., 1999. Global silicate weathering and CO₂
812 consumption rates deduced from the chemistry of large rivers. *Chem. Geol.* 159, 3–30.

813 Graham, R.J., Pierrehumbert, R.T., 2020. Thermodynamic and Energetic Limits on
814 Continental Silicate Weathering Strongly Impact the Climate and Habitability of Wet,
815 Rocky Worlds.

816 Knutti, R., Rugenstein, M.A.A., Hegerl, G.C., 2017. Beyond equilibrium climate sensitivity.
817 *Nat. Geosci.* 10, 727–736. <https://doi.org/10.1038/ngeo3017>

818 Kump, L.R., Brantley, S.L., Arthur, M.A., 2000. Chemical weathering, atmospheric CO₂,
819 and climate. *Annu. Rev. Earth Planet. Sci.* 28, 611–667.

820 Larsen, I.J., Montgomery, D.R., Greenberg, H.M., 2014. The contribution of mountains to
821 global denudation. *Geology* 42, 527–530. <https://doi.org/10.1130/G35136.1>

822 Lenton, T.M., Daines, S.J., Mills, B.J.W., 2018. COPSE reloaded: An improved model of
823 biogeochemical cycling over Phanerozoic time. *Earth-Science Rev.* 178, 1–28.
824 <https://doi.org/10.1016/j.earscirev.2017.12.004>

825 Maher, K., Chamberlain, C.P., 2014. Hydrologic regulation of chemical weathering and the
826 geologic carbon cycle. *Science* (80-.). 343, 1502–1504.
827 <https://doi.org/10.1126/science.1250770>

828 Manabe, S., Wetherald, R.T., Milly, P.C.D., Delworth, T.L., Stouffer, R.J., 2004. Century-
829 scale change in water availability: CO₂-quadrupling experiment. *Clim. Change* 64, 59–
830 76.

831 Milliman, J.D., Droessler, A.W., 1996. Neritic and pelagic carbonate sedimentation in the
832 marine environment: ignorance is not bliss. *Geol. Rundschau* 85, 496–504.

833 Moon, S., Chamberlain, C.P., Hilley, G.E., 2014. New estimates of silicate weathering rates
834 and their uncertainties in global rivers. *Geochim. Cosmochim. Acta* 134, 257–274.
835 <https://doi.org/10.1016/j.gca.2014.02.033>

836 Myhre, G., Highwood, E.J., Shine, K.P., Stordal, F., 1998. New estimates of radiative forcing
837 due to well mixed greenhouse gases. *Geophys. Res. Lett.* 25, 2715–2718.

838 Prentice, I.C., Harrison, S.P., 2009. Ecosystem effects of CO₂ concentration: Evidence from
839 past climates. *Clim. Past* 5, 297–307. <https://doi.org/10.5194/cp-5-297-2009>

840 Scheff, J., Seager, R., Liu, H., Coats, S., 2017. Are glacials dry? Consequences for
841 paleoclimatology and for greenhouse warming. *J. Clim.* 30, 6593–6609.
842 <https://doi.org/10.1175/JCLI-D-16-0854.1>

843 Shields, G.A., Mills, B.J.W., 2017. Tectonic controls on the long-term carbon isotope mass
844 balance. *Proc. Natl. Acad. Sci.* 114, 4318–4323.

<https://doi.org/10.1073/pnas.1614506114>

Volk, T., 1987. Feedbacks between weathering and atmospheric CO₂ over the last 100 million years. *Am. J. Sci.* <https://doi.org/10.2475/ajs.287.8.763>

Wallmann, K., 2001. Controls on the Cretaceous and Cenozoic evolution of seawater composition, atmospheric CO₂ and climate. *Geochim. Cosmochim. Acta* 65, 3005–3025. [https://doi.org/10.1016/S0016-7037\(01\)00638-X](https://doi.org/10.1016/S0016-7037(01)00638-X)

West, A.J., 2012. Thickness of the chemical weathering zone and implications for erosional and climatic drivers of weathering and for carbon-cycle feedbacks. *Geology* 40, 811–814. <https://doi.org/10.1130/G33041.1>

Winnick, M., Maher, K., 2018. Relationships between CO₂, thermodynamic limits on silicate weathering, and the strength of the silicate weathering feedback. *Earth Planet. Sci. Lett.* 485, 111–120. <https://doi.org/https://doi.org/10.1016/j.epsl.2018.01.005>

Zhang, Xuejun, Tang, Q., Zhang, Xuezhen, Lettenmaier, D.P., 2014. Runoff sensitivity to global mean temperature change in the CMIP5 Models. *Geophys. Res. Lett.* 41, 5492–5498. <https://doi.org/10.1002/2014GL061184>.Received