

Paleogene global cooling–induced temperature feedback on chemical weathering, as recorded in the northern Tibetan Plateau

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ABSTRACT

Plate-tectonic processes have long been thought to be the major cause of the Cenozoic global carbon cycle, and global cooling by uplift of the Tibetan Plateau through enhancing silicate weathering and organic carbon burial and/or by weathering of obducted ophiolites during the closure of the Neo-Tethys Ocean. However, the imbalance resulting from accelerated CO₂ consumption and a relatively stable CO₂ input from volcanic degassing during the Cenozoic should have depleted atmospheric CO₂ within a few million years; therefore, a negative feedback mechanism must have stabilized the carbon cycle. Here, we present the first almost-complete Paleogene silicate weathering intensity (SWI) records from continental rocks in the northern Tibetan Plateau showing that silicate weathering in this tectonically inactive area was modulated by global temperature. These findings suggest that Paleogene global cooling was also strongly influenced by a temperature feedback mechanism, which regulated silicate weathering rates and hydrological cycles and maintained a nearly stable carbon cycle. It acted as a negative feedback by decreasing CO₂ consumption resulting from the lower SWI and the kinetic limitations in tectonically inactive areas.

INTRODUCTION

Postulation about the links between silicate weathering and the long-term global carbon cycle has led to an extended debate regarding the role of plate-tectonic processes, including closure of the Tethys Ocean and tectonic uplift of the Tibetan Plateau (TP), in the global carbonate-silicate geochemical cycle via the accelerated weathering of silicate rocks (Raymo and Ruddiman, 1992; France-Lanord and Derry, 1997; Kump et al., 2000; Galy et al., 2015; Jagoutz et al., 2016). Numerous studies have attributed the remarkable increase in Cenozoic seawater Sr, Li, and Os isotopes to the weathering of uplifted continental rocks (Edmond, 1992; Klemm et al., 2005; Misra and Froelich, 2012) or obducted ophiolites (Jagoutz et al., 2016) in the Intertropical Convergence Zone. Therefore, such enhanced weathering of the continent should have induced a rapid depletion of atmospheric CO₂, and requires negative feedbacks to

have stabilized atmospheric CO₂ levels (Berner and Caldeira, 1997). Various negative feedback hypotheses have been proposed; e.g., organic carbon cycle (Raymo and Ruddiman, 1992), metamorphic decarbonation (Bickle, 1996), sulfur cycle (Torres et al., 2014), and temperature (*T*)-dependent weathering (Kump and Arthur, 1997) hypotheses. In the latter case, the negative feedback is thought to occur in areas under “weathering-limited regimes” characterized by silicate weathering rates that are controlled by kinetic (climatic) parameters (Kump et al., 2000; West et al., 2005), such as island basalt (Li and Elderfield, 2013; Li et al., 2016), continental arcs (Lee et al., 2015), and seafloor basalt (Coogan and Dosso, 2015), or by a weatherability-climate linkage (Kump and Arthur, 1997; Caves et al., 2016; Zhang and Planavsky, 2019). These hypotheses of *T*-dependent weathering, and other hypotheses, are difficult to test directly by studying the geological archives at a global

scale (the marine record) because other parts of the world were subjected to weathering that was insensitive to *T*, described as “transport-limited regimes,” where silicate weathering rates scaled directly with erosion (Kump et al., 2000; West et al., 2005).

Instead of building a global record, we used an alternative approach to document the changes in the continental silicate weathering flux. Our hypothesis is that the world beyond the uplifted TP is, on average, globally characterized by stable long-term erosion rates. In that case, the rapid carbon consumption produced by enhanced silicate weathering and faster burial of organic carbon in South and East Asia, where tectonic uplift promotes monsoon development, is nearly balanced by the tight link between a lower silicate weathering intensity (SWI) and global cooling. Thus, the issue of whether the SWI of continental rocks in non-Asian-monsoon regions is closely linked to kinetic parameters, e.g., *p*CO₂ and temperature, will be a crucial test of our working hypothesis.

During the Paleogene, the northern TP was a tectonically inactive area, and the Asian summer monsoon did not reach this region until the beginning of the Neogene (Fig. 1; Guo et al., 2008). The major tectonic uplift of that area occurred in the late Neogene (Tapponnier et al., 2001; Li et al., 2014). Here, we present the first record in this region showing a decrease in the SWI generally coupled with the global temperature during the Paleogene.

MATERIAL AND METHODS

Clay mineral records of SWI were retrieved from the Hongliugou (HLG) section in the

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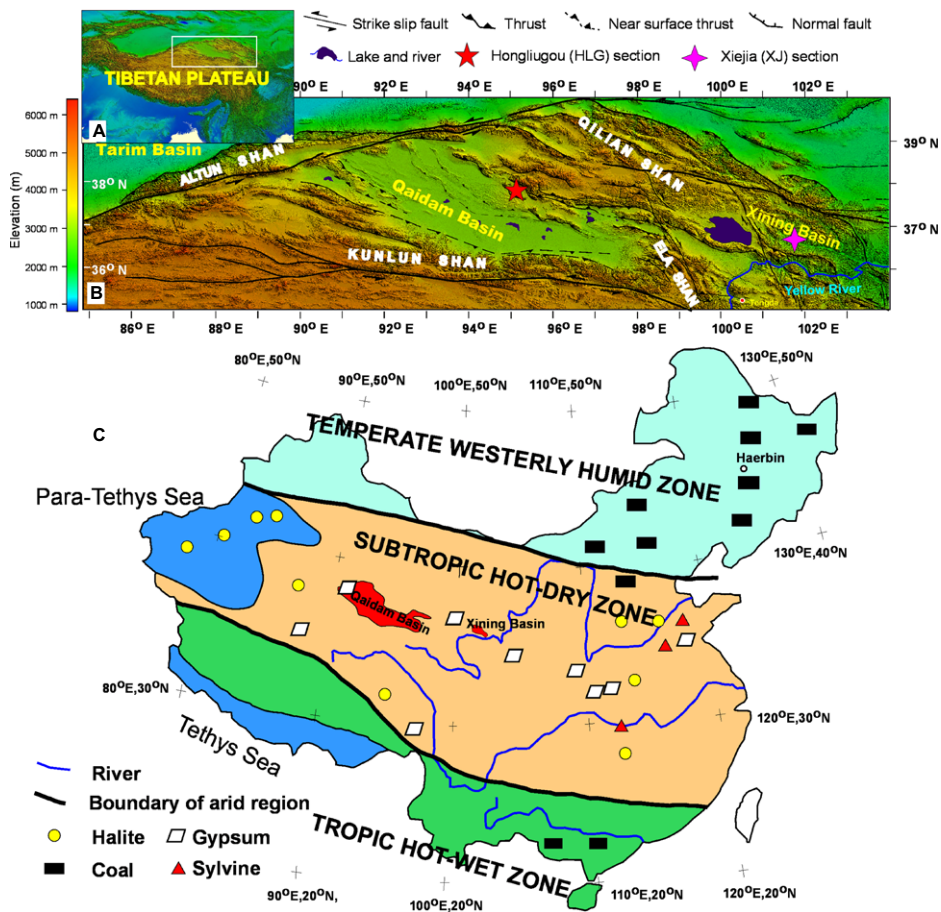


Figure 1. (A,B) Map of the northern Tibetan Plateau showing studied locations: Hongliugou (HLG) section in the Qaidam Basin, and Xiejia (XJ) section in the Xining Basin. (C) Climate zones during the Eocene of China (Guo et al., 2008).

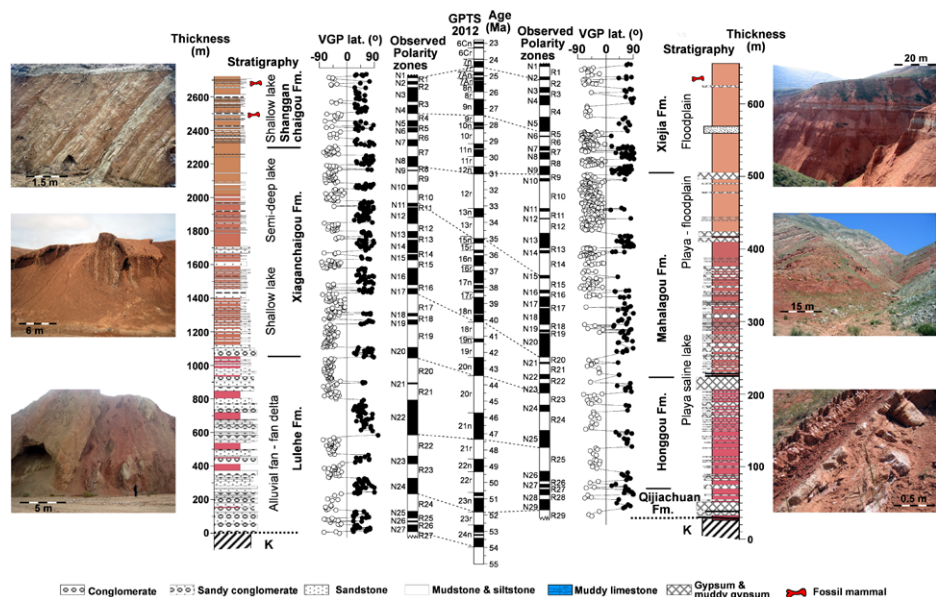


Figure 2. Lithology and magnetostratigraphy of Paleogene sediments in the Hongliugou (HLG) section in the Qaidam Basin, and the Xiejia (XJ) section in the Xining Basin, on the Tibetan Plateau. (A–D) Representative photos, lithofacies, stratigraphy, observed virtual geomagnetic pole (VGP) latitudes (lat.), and polarity zones of the HLG section (Zhang, 2007). Fm—Formation. (E) Reference geomagnetic polarity time scale (GPTS2012; Gradstein and Ogg, 2012). (F–I) Observed polarity zones and VGPs, stratigraphy, and representative photos of lithofacies of the XJ section.

Qaidam Basin and from the Xiejia (XJ) section in the Xining Basin. Both basins are large intermontane basins on the northern TP (Fig. 1). In these basins, detailed magnetostratigraphy and biostratigraphy (Dai et al., 2006; Zhang, 2007) have determined the onset of sedimentation at ca. 54–52 Ma (Fig. 2). The Paleogene strata of the HLG section are dominated in the lower part by red sandstones intercalated with thin mudstone layers representing alluvial-fan deposits, fining-upward to red-brown siltstones-mudstones embedded with green sandstones, which are interpreted as lacustrine deposits (Fig. 2; Zhang, 2007). The Paleogene strata of the XJ section are dominated by homogeneous red siltstones-mudstones intercalated with gypsum layers representing alternations of distal floodplain dry mudflat and saline lake environments (Fig. 2; Dupont-Nivet et al., 2007). The clay fractions were separated following Stokes' law and analyzed by X-ray diffraction. Aliquots of the clay-sized samples were digested with HNO₃/HF, and trace-element concentrations were measured by inductively coupled plasma-mass spectrometry. The hematite contents of bulk samples were identified by the heights of peaks at 565 nm for the first derivatives calculated from diffuse reflectance data (Deaton and Balsam, 1991).

CLAY MINERALS AS SILICATE WEATHERING PROXIES

Clay minerals in both basins are mainly composed of abundant illite/smectite mixed layers (I/S) and illite, with smectite, kaolinite, and chlorite as minor components and the occasional palygorskite (Fig. DR1 in the GSA Data Repository¹). Clay mineral formation is linked to continental crust weathering processes characterized by the disappearance of primary silicate minerals with a loss of base ions and concomitant formation of clay minerals (Nesbitt and Young, 1982). Clay minerals in both basins are assumed to be the residual products of incongruent silicate weathering in drainage areas, and their long-term variations are weakly impacted by postdeposition diagenesis and recycled clays (see Figs. DR2–DR5 and Data Repository text). However, the protolith can have an impact on the mineralogy of clays produced by weathering. The immobile elements La and Th are more abundant in felsic rocks than in mafic rocks, and the opposite is true for Sc. Therefore, the Th/Sc ratio is a useful indicator of the provenance of felsic/mafic rock contributions (Taylor and McLennan, 1985; Cullers et al., 1988), while

¹GSA Data Repository item 2019356, XRD data and details of the clay mineral and trace elements analyses, and test for temperature dependence of chemical weathering rate, is available online at <http://www.geosociety.org/datarepository/2019/>, or on request from editing@geosociety.org.

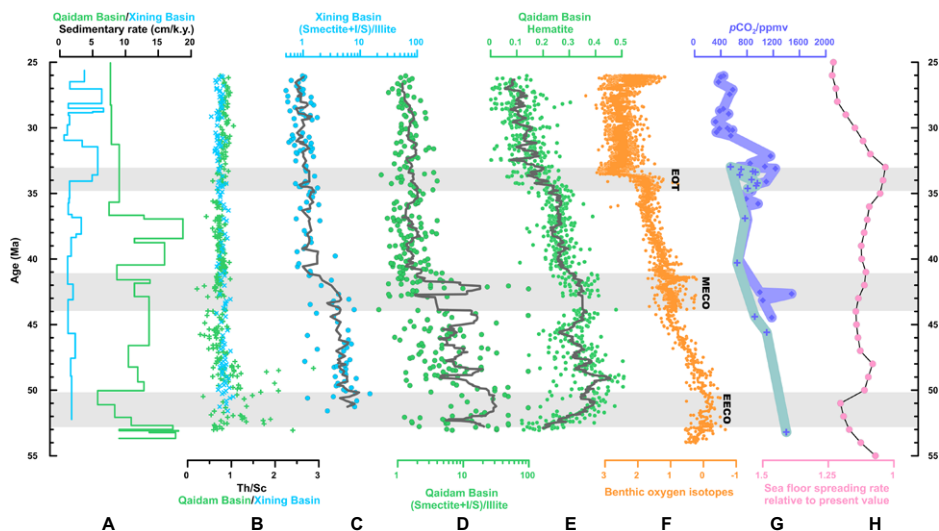


Figure 3. Mineralogy and sedimentation rate of Paleogene sediments in the Hongliugou (HLG) and Xiejia (XJ) sections in the Qaidam and Xining Basins, respectively, on the Tibetan Plateau. (A) Sedimentation rates. (B) Th/Sc ratios. (C–E) (Smectite + I/S)/illite ratios (I/S—illite/smectite mixed layers) and hematite content, with gray curve representing ~1 m.y. running averages (3-point average for C, 11-point average for D and E). Use of two different running point averages accounted for different sampling density. (F) Global marine benthic $\delta^{18}\text{O}$ record (‰, a proxy for global temperature assuming little ice volume in the Eocene; Zachos et al., 2008). EOT—Eocene-Oligocene transition; MECO—middle Eocene climatic optimum; EECO—early Eocene climatic optimum. (G) Atmospheric $p\text{CO}_2$, where blue diamonds indicate marine phytoplanktonic $\delta^{13}\text{C}$ (Pagani et al., 2005), and blue crosses correspond to planktonic foraminiferal $\delta^{11}\text{B}$ (Pearson et al., 2009; Anagnostou et al., 2016). (H) Seafloor spreading rate (Muller et al., 2008).

La/Th should yield a relatively stable value in fine-grained sediments (McLennan et al., 1980). Felsic and mafic rocks are present in the catchments of both basins (Bureau of Geology and Mineral Resources of Qinghai Province, 1989). However, La/Th and Th/Sc in the clay fractions of both sections show very limited variability (Fig. 3; Figs. DR2 and DR3), indicating that the source rock was well mixed by riverine transport prior to deposition, and suggesting a negligible provenance control.

Overall, the clay mineralogy in both sections is likely to represent an invaluable archive to record the past catchment SWI. Chlorite and illite are predominant under cold and/or arid environments, whereas smectite, I/S, and kaolinite reflect a greater SWI under temperate-humid climates (e.g., Chamley, 1989). In both sections, the kaolinite content is generally <10% (Figs. DR2 and DR3), except in a short interval of the early Eocene in the HLG section, where the kaolinite content can reach ~20%. Such a low kaolinite content cannot be used as an index for weathering intensity. I/S is mostly a transitional product (Weaver, 1989), and since smectite, I/S, and illite make up ~80% of the clays, the (smectite + I/S)/illite ratio is the best candidate to trace the SWI at the scale of the drainage basins.

PALEOGENE SILICATE WEATHERING INTENSITY ON THE NORTHERN TIBETAN PLATEAU

The long-term decrease in the (smectite + I/S)/illite ratio in both sections (Fig. 3;

Figs. DR2 and DR3) indicates a long-term weakening of the SWI since the early Eocene, although its absolute values are higher in the Qaidam Basin than in the Xining Basin. The northeastern TP was in the paleo-Asian interior and outside the area influenced by the paleo-Asian monsoon (Guo et al., 2008; Caves et al., 2015). The different absolute values between the two basins might have been caused primarily by a shorter distance from the Qaidam Basin to the westward moisture source, the Para-Tethys Sea (see Fig. 1). The long-term trend (Fig. 3) displays a close temporality between the relative changes in the (smectite + I/S)/illite ratio from both basins and early Cenozoic global cooling (Zachos et al., 2008) and $p\text{CO}_2$ drop (Pagani et al., 2005; Pearson et al., 2009; Anagnostou et al., 2016). This result suggests that global temperature may have been the first-order factor modulating the SWI. The remarkable decreases in the (smectite + I/S)/illite ratio at ca. 42 Ma might be a response to the ephemeral Antarctic glaciation, probably linked to the initial opening of the Drake Passage at ca. 41 Ma (Scher and Martin, 2006), and associated global cooling. The decrease in the SWI in this study can be related to the enhanced drying found in other sparse and low-resolution paleoclimatic records in the Xining Basin (Bosboom et al., 2014) and in the Qaidam Basin (Wang et al., 1999). The Eocene retreat of the Para-Tethys Sea (Sun and Jiang, 2013; Bosboom et al., 2014) may have also exerted some influence on climate by reducing the hydrological cycle in the northern

TP, and thus weakening chemical weathering. However, the retreat of the Para-Tethys Sea occurred over the Eocene, not as an abrupt event at ca. 42 Ma. Thus, its influence would be both long-term and gradual. Local tectonic and differential uplifts are likely to be responsible for the increased sedimentation in the Xining Basin that occurred coeval with the lower sedimentation rate in the Qaidam Basin between 37 and 34 Ma (Fig. 3). The similar trend in the (smectite + I/S)/illite ratios of the two basins during that period further highlights the lack of correlation between the SWI and tectonic forcing in the area. The gradual decrease in the (smectite + I/S)/illite ratio in both sections ca. 35–33 Ma might have been a response to the global cooling associated with the onset of the permanent Antarctic ice sheet at the Eocene-Oligocene transition (EOT). Although the magnitude of the EOT event in our records is smaller than those of the middle Eocene climatic optimum (MECO) and early Eocene climatic optimum (EECO) events (Fig. 3), its magnitude is clear in the Xining Basin in high-resolution records (Zhang and Guo, 2014).

Hematite is a type of iron oxide formed during soil weathering, and its relative content can serve as a SWI proxy (e.g., Clift et al., 2008). The hematite content in the HLG section exhibits a long-term decrease (Fig. 3; Fig. DR2) similar to the variations in the (smectite + I/S)/illite ratio. The long-term decrease in the hematite content also displays large fluctuations and provides a good match to the global temperature record, particularly post-MECO and at the Eocene-Oligocene boundary (Fig. 3). Together with temperature, precipitation is another key climate factor affecting chemical weathering (West et al., 2005; Maher and Chamberlain, 2014). High regional moisture levels likely occurred on the Para-Tethys Sea to the west of the studied regions (Fig. 1) during the Paleogene (Bosboom et al., 2014). Higher temperatures cause greater evaporation of the sea and produce higher moisture levels. Given the results of climate modeling of the area during the Paleogene (Ramstein et al., 1997), this moisture could have been transported to the studied regions by the westerlies, promoting rain and facilitating chemical weathering. Since this mechanism keeps pace with temperature, its effect has been encapsulated in the temperature-driven weathering record.

IMPLICATIONS FOR GLOBAL WEATHERING FEEDBACK

Global temperature seems to be the most likely candidate to explain the decrease in the SWI in the TP two basins. This might also be true in other parts of the world (beyond the impact of the Asian monsoon) where tectonic uplift was also weak during the Cenozoic. Considering an overall stable erosion rate in tectonically

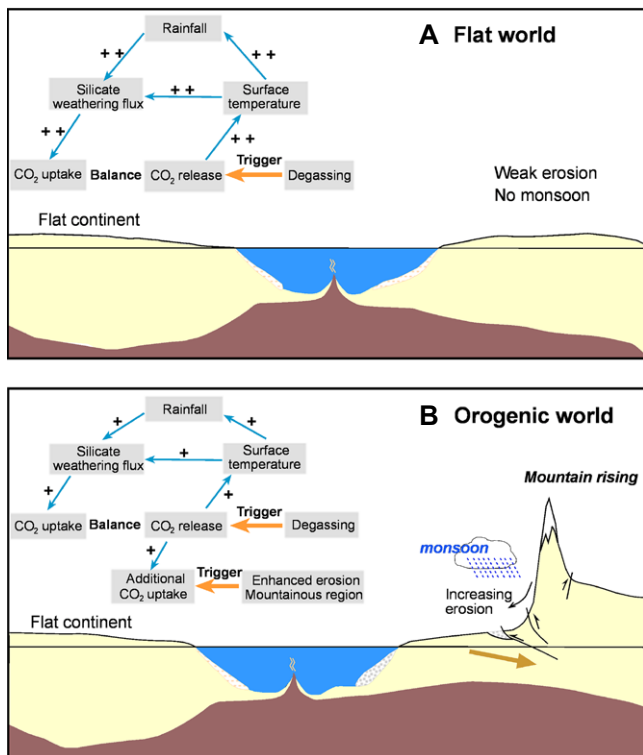


Figure 4. Conceptual models showing negative feedback mechanisms in geological carbon cycles. (A) Flat, tectonically inactive world with weak and stable erosion. (B) Orogenic world with strong erosion in orogenic belts coupled with mountain-induced strong monsoon rainfall and glacier processes. In scenario A, CO₂ degassing from Earth's mantle will be balanced by CO₂ uptake in tectonically inactive areas. Thus, any increase in degassing will lead to an increase in temperature, and ultimately will be balanced by nearly the same amount of increase in CO₂ uptake through increases in silicate weathering intensity (SWI) and thus in silicate weathering flux. In scenario B, CO₂ degassing will be quasi-balanced by combined CO₂ uptake from orogenic belts and in tectonically inactive areas. Given a constant

degassing, any increase in erosion in orogenic belts will result in a decrease in SWI, possibly compensated, or even overwhelmed, by an increase in silicate weathering flux, but clearly an overall increase in CO₂ uptake in orogenic belts primarily related to enhanced organic carbon burial (France-Lanord and Derry, 1997; Galy et al., 2015). Increase in CO₂ uptake in orogenic belts will be quasi-balanced by nearly the same amount of decrease in CO₂ uptake in flat, tectonically inactive regions through decreases in temperature and, thus, in SWI and silicate weathering flux in tectonically inactive areas. Plus signs (+) mark the magnitude of carbon flux involved in various feedback processes. Large carbon flux (++) initially from degassing was involved in temperature feedbacks for scenario A, but part of this carbon flux (+) was directly consumed in orogenic belts, and the remainder (+) was involved in temperature feedbacks for scenario B.

inactive regions, the silicate weathering flux can roughly be regarded as proportional to the SWI. Thus, the correlation of SWI with global temperature (Fig. 3) suggests that global cooling could be a first-order control on the silicate weathering flux in regions beyond the impact of the Indo-Asian collision and associated development of the Asian monsoon.

Our study indeed illustrates two distinct carbon cycle patterns: one in a tectonically inactive world with less-pronounced orogenesis, giving rise to an overall stable and low global erosion flux, and the other in an orogenic world with remarkably high erosion flux derived from a combination of active orogenesis and mountain-induced heavy monsoon precipitation and glacier processes (e.g., the Himalayas; Fig. 4). In the tectonically inactive world, any imbalance in the carbon cycle from CO₂ degassing would ultimately be balanced by changes in the SWI, given an overall stable erosion flux (Fig. 4A). In contrast, in an orogenic world, any increase in erosion from the tectonically active region would lead to an overall increase in carbon sequestration (caused by extremely high erosion flux-induced increases in silicate weathering

flux and organic carbon burial). However, the increase in CO₂ uptake in orogenic belts would be almost completely offset by the decrease in CO₂ uptake flux in tectonically inactive regions through decreases in global temperature and the associated lowering of the hydrological cycle. So, enhanced CO₂ consumption in tectonically active regions would be accommodated by lowered SWI across the rest of the world (Fig. 4B). Such a model is plausible since it is backed up by a first-order calculation of temperature dependence on silicate weathering rate using the Arrhenius law: $\ln(k_{\text{post-uplift}}/k_{\text{pre-uplift}}) = -Ea/R(1/T_{\text{post-uplift}} - 1/T_{\text{pre-uplift}})$, where k is silicate weathering rate; T is reaction temperature; R is a gas constant; and Ea is the activation energy; Figs. DR7 and DR8). When considering that global cooling can also induce a sluggish water cycle, the overall global cooling effect is more pronounced than that caused by the T sensitivity of silicate weathering alone (e.g., Kump et al., 2000). Because the climate system and carbon cycle evolve in a quasi-steady state, the small imbalance that results from the enhanced erosion in the tectonically active areas could drive down atmospheric $p\text{CO}_2$. Recent studies also

have debated that other plate-tectonic processes may act as important drivers of CO₂ drawdown (Van der Meer et al., 2014; Jagoutz et al., 2016; Zhang and Planavsky, 2019). Although all of the proposed mechanisms have substantial uncertainties regarding the evolution of the Paleogene carbon cycle, our study suggests spatially variable negative feedback processes between atmospheric $p\text{CO}_2$ and climate, linking plate tectonics and climate.

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