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1. Introduction

[1] Determining the interplay between tectonic deformation, climate, atmospheric CO2 concentrations and continental weathering and erosion is key to understanding the mechanisms that forced Cenozoic global cooling. In contrast with studies of paleo-climate and pCO2, the history of long-term silicate weathering in the Himalaya and Tibetan Plateau (HTP) during the late Cenozoic remains unclear. We reconstruct 5 m.y. of silicate sedimentary records at Ocean Drilling Program (ODP) Site 1143 in the South China Sea to explore the weathering history of the Mekong River basin that supplied the sediment. Coherent variation of weathering proxies from the South China Sea, Bay of Bengal, Loess Plateau, as well as the Yangtze and Yellow Rivers, indicates weakening chemical weathering intensity since the late Pliocene, as the climate cooled. This cooling, coupled with tectonic activity, shifted the dominant weathering regime from more transport-limited to more weathering-limited, causing less chemical depletion of silicate minerals. While silicate weathering rates became strongly correlated to erosion rates, they were decoupled from chemical weathering intensity. Physical denudation and associated silicate weathering rates in the HTP area increased in the Pliocene, driven by both rock uplift and stronger monsoon precipitation, decreasing atmospheric CO2 concentrations, and so contributing to Northern Hemisphere Glaciation (NHG). Citation: Wan, S., P. D. Clift, A. Li, Z. Yu, T. Li, and D. Hu (2012), Tectonic and climatic controls on long-term silicate weathering in Asia since 5 Ma, Geophys. Res. Lett., 39, L15611, doi:10.1029/2012GL052377.

[2] Continental weathering can affect both the atmosphere through draw-down of CO2 and ocean chemistry through the dissolved load in fluvial discharge. These processes have the ability to affect global climate over time scales of >106 years [Berner and Caldeira, 1997; Raymo and Ruddiman, 1992]. Climate, as represented by temperature and precipitation, has in turn been identified as a critical factor influencing silicate weathering [White and Blum, 1995]. Walker et al. [1981] proposed that the temperature-dependence of silicate weathering rates constitutes a negative feedback on atmospheric CO2 and therefore stabilizes Earth’s surface temperature. In contrast, Raymo and Ruddiman [1992] argued that Cenozoic climatic deterioration was caused by rise of the HTP, and an increase in chemical weathering rates and consumption of atmospheric CO2. However, the absence of both high-resolution silicate weathering records and a mass-balanced carbon cycle made proving an uplift trigger impossible [Berner and Caldeira, 1997]. Carbon modeling suggests that chemical weathering rates should not increase in the absence of enhanced delivery of CO2 to the atmosphere from metamorphic sources, because CO2 would be completely stripped from the atmosphere within a few million years [Berner and Caldeira, 1997]. It is noteworthy that some recent studies suggest that mass balance calculations neglect the metamorphic degassing of CO2 from orogenic zones, including the Himalaya [Gaillardet and Galy, 2008]. However, it remains unclear whether this process is more important than weathering in contributing to the long-term flux of carbon for the whole range. Under these circumstances, a detailed history of long-term silicate weathering in the HTP area through the late Cenozoic is needed to test for linkages between climate, weathering and erosion.

[3] The use of the seawater 87Sr/86Sr record as a monitor of silicate weathering rates is complicated because carbonate weathering and hydrothermal inputs also influence this proxy [Waltham and Grocke, 2006]. Recently, a lithium isotope record was used to trace global Cenozoic silicate weathering [Misra and Froelich, 2012], but this does not isolate the influence of the HTP. An alternative solution is to use silicate sediments in the Asian marginal seas, which preserve material from the large rivers that drain the HTP. Here we focus on sediment records spanning 5 Ma from ODP Site 1143 (Figure 1). The coincidence of progressive plateau uplift [Royden et al., 2008], rapidly declining atmospheric CO2 [Seki et al., 2010], coupled with onset of major NHG in the late Pliocene makes this an ideal period to study the response of weathering to these processes.

2. Materials and Methods

[4] ODP Site 1143 is located in the southern South China Sea, from which a total of 1245 samples were sampled to a sub-seafloor depth of 190.7 m, resulting in a sample resolution of about 4.0 k.y. The section is dominated by hemipelagic, fine-grained terrigenous materials and nanofossil carbonate ooze [Wan et al., 2006]. The age model was built by tuning the benthic δ18O stratigraphy to orbital obliquity and precession [Tian et al., 2002]. We used a combination of mineralogical and geochemical proxies to reconstruct changes in weathering intensity through time. Semi-quantitative clay and bulk mineral analyses were made respectively on the <2 μm size fraction and unoriented bulk sample powder by X-Ray Diffraction (XRD) analysis (see Table DR-1 in...
Major and trace element concentrations were measured on bulk samples using X-Ray Fluorescence (XRF) methods (see Table DR-2 in Text S1 in the auxiliary material). In addition, we synthesized other published weathering records across the HTP region, including the K/Al ratio [Tian et al., 2011] and hematite/goethite [Zhang et al., 2009] at ODP Site 1143, the chemical index of alteration (CIA) at Cores DY in the Yellow River Delta and PD from the Yangtze Delta [Yang et al., 2006], and Baishui loess section from the Chinese Loess Plateau [Xiong et al., 2010], \(^{87}\text{Sr}/^{86}\text{Sr}\) at ODP Site 718 in the Indian Ocean [Derry and France-Lanord, 1996], stacked East Asian summer monsoon (EASM) index from the Lingtai loess section [Sun et al., 2010], terrigenous mass accumulation rate (MAR) at ODP Site 1146 [Wan et al., 2007], stacked sediment flux to the Indian Ocean [Rea, 1992], erosion rates in the Tianshan [Charreau et al., 2011]. The records were normalized and averaged to produce stacked indexes of chemical weathering intensity and physical erosion.

### 3. Chemical Weathering Intensity

Clay minerals and trace element chemistry indicate a stable provenance from the Mekong River for sediment at ODP Site 1143 since 5 Ma [Wan et al., 2006, 2010] suggesting that changes in composition are driven by changes in chemical weathering intensity, which we define as the degree of alteration of the sediments compared to sources rocks as a result of chemical breakdown. We use kaolinite/illite and clay/feldspar ratios, as well as the CIA value to track the intensity of chemical weathering of the sediments. Kaolinite is formed in soils developed in regions with warm, humid climates, whereas illite is the product of physical erosion from bedrock [Chamley, 1989]. An increased proportion of clay minerals relative to feldspar indicates strengthened chemical weathering because certain clay minerals are formed by alteration [Wan et al., 2006]. CIA gives a quantitative measure of chemical weathering intensity by constraining the loss of labile Na, Ca, and K relative to stable Al [Nesbitt and Young, 1982].

The ratios of kaolinite/illite and clay/feldspar, as well as CIA values, indicate that the chemical weathering history of the Mekong can be roughly divided into two periods. There was relatively strong weathering during the Pliocene (5.0–2.7 Ma), followed by a decline after the onset of NHG (<2.7 Ma) (Figure 2b). This observation is consistent with the weathering history indicated by K/Al [Tian et al., 2011], which shows lower values when chemical weathering is more intense because K is more water-mobile than Al.

In addition to chemical weathering and provenance changes, hydraulic sorting and sediment storage may also influence sediment mineralogy and geochemistry. The fall in sea level since the late Pliocene displaced the coastline closer to ODP Site 1143 and thus could have influenced variations in clay/feldspar and CIA. However, this effect must be small in this case because K/Al and kaolinite/illite, which are insensitive to hydrological sorting [Wan et al., 2010], display similar long-term trends to clay/feldspar and CIA (Figure 2c). Further, we also exclude a dominant influence of long-term sediment storage in flood plains because typically such storage is controlled by processes on decades-orbital cycles [Giosan et al., 2012], which are much shorter than >10^6 years timescale in this study. CIA variations in three drill holes (G3, DY and PD) in the Yellow and Yangtze River deltas [Yang et al., 2006], at Baishui loess section on the Loess Plateau [Xiong et al., 2010], as well as \(^{87}\text{Sr}/^{86}\text{Sr}\) in clastic sediments from ODP Site 718 in the Bay of Bengal [Derry and France-Lanord, 1996] (Figures 1 and 2) also show a general reduction in chemical weathering intensity since the onset of NHG across the whole HTP.
chemical weathering intensity [Misra and Froelich, 2012].

[9] Chemical weathering is strongly affected by moisture and temperature [White and Blum, 1995]. In East Asia precipitation is mainly supplied by the EASM, whose Neogene intensity has been reconstructed by magnetic susceptibility and carbonate content in loess sequences [Sun et al., 2010]. These records show that EASM was relatively stable before 3.5 Ma and strengthened gradually from 3.5 to 2.7 Ma. Subsequently, the EASM decreased slightly until 1.5 Ma, after which time it increased again to the present day (Figure 2d). In contrast, temperature, as revealed by benthic δ18O at ODP Site 1143 [Tian et al., 2002] (black line) and Mg/Ca ratio derived sea surface temperature at ODP Site 1143 [Jia et al., 2008] (red line), was relatively high and stable between 5.0 Ma and then gradually decreased after 3.5 Ma (Figure 2a). Weakening chemical weathering after 2.7 Ma does not correlate well with monsoon intensity but does track the temperature proxies, suggesting that this plays a dominant role in controlling the chemical depletion of silicates. The effect of climate on silicate weathering is also demonstrated by global correlations between CIA values of suspended sediments in large modern rivers and local temperature and precipitation (auxiliary material).

[10] Although the intensity of silicate weathering weakened following the onset of NHG, this does not require silicate weathering rates to also have reduced. Chemical weathering intensity is the degree of the chemical depletion of silicates. In contrast, chemical weathering rate is defined as the lost amount of soluble mass per unit area per unit time of the source rock [White and Blum, 1995]. Intensity and rate of chemical weathering are two different concepts and have different controls. Previous work has shown that chemical weathering rates are influenced by physical erosion. Rapid production of fresh surfaces driven by high physical erosion rates is critical to fast chemical weathering rates [West et al., 2005]. This is confirmed by the positive correlation between silicate weathering rate and suspended sediment yield of global large rivers, which is a proxy of erosion rate (see auxiliary material). Both tectonic activity and strong precipitation accelerate physical erosion [West et al., 2005] and thus potentially enhance silicate weathering rates.
[11] On million-year timescales, terrigenous accumulation on continental margins has been used to monitor erosion rates in source regions. The approximately simultaneous increase in terrigenous mass accumulation rate in the South China Sea [Clift, 2006; Wan et al., 2007] and in the Indian Ocean [Rea, 1992] since ~3.5 Ma (Figure 2) is consistent with strengthened physical erosion in Asia after that time. Although this increase might be biased by sea-level fall and the decreasing probability of sediment preservation with age [Sadler, 1999], cosmogenic 10Be concentrations also indicate enhanced erosion in the northern Tian Shan at 3.5–1.7 Ma [Charreau et al., 2011]. Faster physical erosion in Asia may relate to stronger monsoonal precipitation since 3.5 Ma [An et al., 2001] and the disequilibrium states of Quaternary glacial climate [Zhang et al., 2001] coupled with continued strong rock uplift around the periphery of the HTP [An et al., 2001].

[12] Given the generally coherent co-variation of chemical weathering proxies and sediment flux around the HTP, we combine these records by normalizing each proxy and averaging them to produce stacked indexes of chemical weathering intensity and physical erosion (Figure 2f). The gradually weakening chemical weathering intensity correlates with a rapid increase in erosion rate between 4 and 2 Ma. Furthermore, variations in chemical weathering rate can be inferred according to the mass-balance equation:

\[ W = E \times F, \]

where \( W \) is the chemical weathering rate, \( E \) is total denudation rate, and \( F \) is chemical depletion fraction of insoluble silicate sediments [Riebe et al., 2001]. The stacked physical erosion flux and chemical weathering intensity were used to approximately represent \( E \) and \( F \), respectively. The calculated silicate weathering rates rapidly increased between 4.0 and 2.5 Ma and then gradually decreased until 1.5 Ma (Figure 2g) preceding the onset of NHG by ~0.5 m.y., and suggesting that a simple link only to temperature is unjustified.

[13] During the late Cenozoic, rivers draining the HTP have been characterized by high suspended and dissolved discharge, accounting for ~30% of global sediment production from only ~5% of the land area (Raymo and Ruddiman, 1992). The large rivers in these regions are generally enriched in 44Ca and thus rapid rise in marine 44Ca values between 4.0 and 2.7 Ma has been interpreted to reflect enhanced riverine Ca discharge from Asia (Figure 2e) [Fantine and DePaolo, 2005]. This is consistent with strengthening physical erosion and silicate weathering in Asia between 4.0 and 2.5 Ma.

[14] On the million-year timescale, atmospheric CO2 levels are primarily regulated by variations in the rate of volcanic input from the Earth’s interior balanced by the rate of output through silicate weathering and organic carbon burial at the Earth’s surface [Berner and Caldeira, 1997]. At times when atmospheric CO2 levels decreased and global climate cooled (Figure 2g), solid-earth CO2 degassing rates have increased, driven by increased rates of seabed spreading and hotspot magmatism [Cogné and Humler, 2006], and/or metamorphic degassing of CO2 in mountain belts [Gaillardet and Galy, 2008]. The efficient burial of organic carbon in the Bengal Fan has been suggested as an important sink of atmospheric CO2 [Galy et al., 2007]. However, considering the generally reduced organic carbon burial since the Pliocene [Raymo, 1997], the decreased atmospheric CO2 in the late Pliocene [Seki et al., 2010] must have been largely caused by enhanced silicate weathering, especially around the HTP. A 9% increase in 87Sr/86Sr in the Cenozoic, suggesting an increase in global continental weathering and erosion, was accompanied by rapid CO2 drawdown [Misra and Froelich, 2012]. This tight interplay between orogenic erosion and climate is also revealed in our higher-resolution record since 5 Ma, but contrasts with the stable weathering since about 12 Ma indicated by seawater 10Be/9Be [Willenbring and Blanckenburg, 2010]. We suggest that during the Miocene Climatic Optimum, when the tectonic state of the HTP did not change much, CO2-weathering feedback was able to maintain the climate system near a steady state [Wan et al., 2009]. However, during a period of active mountain building, non-steady-state effects (uplift hypothesis) may contribute to long-term trends in CO2 and climate.

5. Conclusions

[15] We envisage incremental plateau uplift as the ultimate driving force changing weathering and erosion. Continued rock uplift of Tibet in the late Pliocene strengthened physical erosion and associated silicate weathering. A larger plateau may have stimulated stronger Asian monsoon precipitation and further enhanced silicate weathering. These weathering processes must have resulted in a pronounced decrease of atmospheric CO2 level between 3.5 and 2.7 Ma, which culminated in NHG. The disequilibrium states of Quaternary glacial climate could in turn further increase the rates of physical erosion on the Earth’s surface. The precise timing of NHG may reflect the HTP passing a threshold value in size and resulting in a stronger monsoon and weathering regime that tipped Earth into an icehouse condition. Our weathering records during the period with active mountain building supports the “uplift-weathering” hypothesis of Raymo and Ruddiman [1992]. Our demonstration of a temperature-dependence to silicate weathering intensity, but not rate, since 5 Ma suggests that the “thermostat” hypothesis [Walker et al., 1981] overestimated the role of negative feedbacks between climate and silicate weathering in regulating Earth’s temperature and atmospheric CO2 at least in the late Cenozoic.

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