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Extreme weathering/erosion during the Miocene Climatic Optimum: Evidence from sediment record in the South China Sea

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[1] Investigating the interplay between continental weathering and erosion, climate, and atmospheric CO_2 concentrations is significant in understanding the mechanisms that force the Cenozoic global cooling and predicting the future climatic and environmental response to increasing temperature and CO_2 levels. The Miocene represents an ideal test case as it encompasses two distinct extreme climate periods, the Miocene Climatic Optimum (MCO) with the warmest time since 35 Ma in Earth's history and the transition to the Late Cenozoic icehouse mode with the establishment of the east Antarctic ice sheet. However the precise role of continental weathering during this period of major climate change is poorly understood. Here we show changes in the rates of Miocene continental chemical weathering and physical erosion, which we tracked using the chemical index of alteration (CIA) and mass accumulation rate (MAR) respectively from Ocean Drilling Program (ODP) Site 1146 and 1148 in the South China Sea. We found significantly increased CIA values and terrigenous MARs during the MCO (ca. 17–15 Ma) compared to earlier and later periods suggests extreme continental weathering and erosion at that time. Similar high rates were revealed in the early-middle Miocene of Asia, the European Alps, and offshore Angola. This suggests that rapid sedimentation during the MCO was a global erosion event triggered by climate rather than regional tectonic activity. The close coherence of our records with high temperature, strong precipitation, increased burial of organic carbon and elevated atmospheric CO_2 concentration during the MCO argues for long-term, close coupling between continental silicate weathering, erosion, climate and atmospheric CO_2 during the Miocene.

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1. Introduction

[2] Continental erosion and weathering can affect both ocean chemistry through fluvial run-off and the atmosphere through draw-down of CO_2 during chemical weathering. These processes have the ability to affect global climate over a range of time scales [e.g., *Berner et al.*, 1983; *Walker et al.*, 1981]. The Miocene is of particular interest for palaeoclimate studies as it is distinguished by an extreme climatic optima alternating with two major glaciations during the Early and the late Middle Miocene [*Zachos et al.*, 2001]. A recent climate model study [*You et al.*, 2009] suggests that the MCO is most likely linked to elevated CO_2 ($\sim$500 ppmv), which is in agreement to the recent CO_2 estimates [*Kürschner et al.*, 2008], but contrast with previous low CO_2 levels reconstruction [*Pearson and Palmer*, 2000; *Pagani et al.*, 2005]. A marked increase in atmospheric CO_2 would increase global temperatures through the greenhouse effect, thereby affecting atmospheric and oceanic circulation, precipitation patterns and intensities, and thus accelerate silicate weathering. An enhanced silicate weathering would in turn limit the increased CO_2 content in the atmosphere through a negative feedback [e.g., *Berner et al.*, 1983; *Walker et al.*, 1981]. A rapid increase in $^{87}Sr/^{86}Sr$ of seawater in the middle Miocene has been related to a major episode of erosion and weathering due to the deformation in the Himalayan orogen at that time [*Raymo*, 1994]. But the use of the sea water $^{87}Sr/^{86}Sr$ record as an ideal monitor of silicate chemical weathering rates has been refuted because this ratio is controlled by non-unique factors [e.g., *Oliver et al.*, 2003; *Quade et al.*, 1997]. Under these circumstances, an independent method to estimate the Miocene continental weathering and erosion will significantly help us to understand the response and feedback of weathering and erosion to changing climate from greenhouse condition to icehouse climate, and thus shed some light on this debate of Miocene CO_2 through consideration of the weathering – climate feedback process in long-term carbon cycle.

2. Materials and Methods

[3] Sedimentary basins in the South China Sea preserve some of the best stratigraphic records available for the continental interior of eastern Asia. Cores from ODP Site 1146 and 1148 in the northern South China Sea represent a nearly complete sequence of essentially unaltered Neogene sediments eroded from the Pearl River system in South China [*Li et al.*, 2003; *Wei et al.*, 2006; *Wan et al.*, 2007]. ODP Site 1146 is located at a water depth of 2092 m, within a small rift basin on the mid-continental slope of the northern South China Sea. Three holes were cored to a sub-seafloor depth of 643 meters composite depth (mcd) [*Wang et al.*, 2000]. For this study, a total of 90 samples were sampled at 1.5 m intervals from 435.62–498.94 mcd and at 3 m intervals from 499.84–642.44 mcd. The lithology of the recovered section is quite homogenous, being

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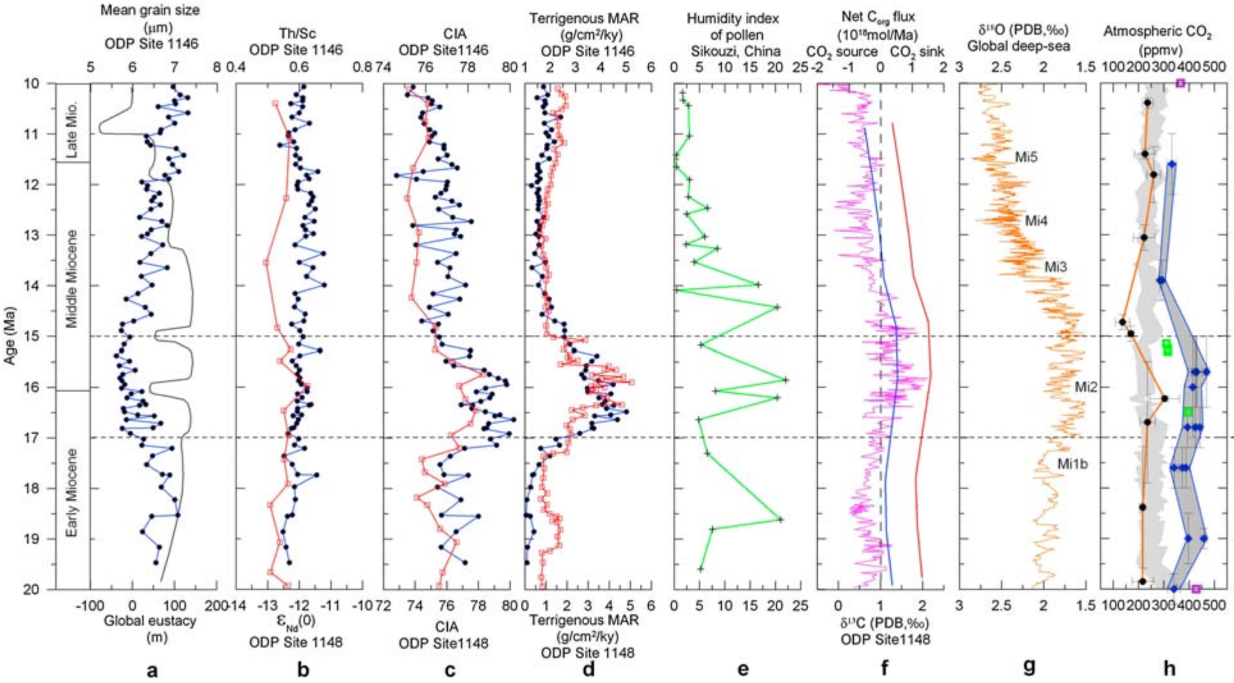

Figure 1. Comparison of silicate chemical weathering, physical erosion, organic carbon burial, climate, atmospheric CO_2 concentration in the Miocene. (a) Mean grain size at ODP Site 1146 and global eustacy curve [*Haq et al.*, 1987]. (b) Trace elements ratio at Site 1146 (filled circle) and Nd isotopes at Site 1148 [*Li et al.*, 2003] (open square). (c) CIA values of silicate sediment at Site 1146 (filled circle) and Site 1148 [*Wei et al.*, 2006] (open square). (d) MARs of silicate sediments at Site 1146 (filled circle) and Site 1148 [*Clift*, 2006] (open square). (e) Humidity index from pollen in North China [*Jiang and Ding*, 2008]. (f) Net organic carbon burial flux (blue line, [*Raymo*, 1997]; red line, [*Derry and France-Lanord*, 1996] and carbon isotope at Site 1148 [*Cheng et al.*, 2004]). (g) Global deep-sea $\delta^{18}O$ [*Zachos et al.*, 2001]. (h) Atmospheric CO_2 concentration based on stomatal records (grey band [*Kürschner et al.*, 2008]; green square [*Royer et al.*, 2001]), Boron isotopes (red line [*Pearson and Palmer*, 2000]), alkenones (grayish band [*Pagani et al.*, 2005]), and GEOCARB III (purple square [*Berner and Kothavala*, 2001]). The two horizontal dotted lines show the interval of extreme weathering and erosion, organic carbon burial consistent with warmer, wetter climate and elevated CO_2 during the MCO. The vertical dashed line indicates zero size of the sedimentary organic C reservoir. When the growth of the sedimentary organic C reservoir is positive, the organic C cycle acts as a sink for atmospheric CO_2 (and visa-versa for negative net growth rates).

dominated by hemipelagic fine-grained terrigenous materials and nannofossil carbonate ooze. Terrigenous materials primarily comprise quartz, feldspar, and clay minerals, accounting for up to 99% of the clastic fraction. Carbonate contents range from 21% to 46% throughout the section [*Wan et al.*, 2007]. The chronostratigraphic framework for ODP Site 1146 was established on the basis of magneto- and biostratigraphy [*Wang et al.*, 2000]. The analyzed sequences span the period ca. 20–10 Ma, with a sample resolution of ca. 100 ky. Geochemical analysis of major and trace elements concentrations was performed on bulk organic-, and carbonate-free sediments. Detailed methods and results of the analysis are provided in Text S1 and Data Set S1 of the auxiliary material.[1]

3. Results and Discussion

[4] Although the topography of the South China Sea and surrounded regions has changed significantly since the Miocene, Nd isotopic and minerals data from sediments collected at ODP sites 1148 and 1146 indicate that their sediment source is very stable through the Neogene [*Li et al.*, 2003; *Wei et al.*, 2006; *Wan et al.*, 2007]. Temporal changes in provenance between 20 and 10 Ma can also be constrained by variations in immobile trace element ratios, such as Th/Sc, Th/Cr, Th/Zr, and La/Cr. The trace elements ratio Th/Sc at ODP Site 1146 and Nd isotopes at ODP Site 1148 display no coherent secular trend during the Miocene (Figure 1), suggesting that there is no major change in sediment source at either site between 20 and 10 Ma. As a result we infer that the rapid increase in terrigenous MAR at 17.2–15 Ma is the result of faster erosion in the source regions rather than provenance change.

[5] The degree of chemical weathering may be estimated by the well established chemical index of alteration (CIA) [*Nesbitt and Young*, 1982; *Wei et al.*, 2006]. CIA is defined as $100 \times Al_2O_3/(Al_2O_3 + CaO^* + Na_2O + K_2O)$ (molecular proportions, with CaO^* being the CaO content in the silicate fraction of the sample). The CIA value provides a quantitative measure of chemical weathering intensity by measuring the loss of labile elements such as Na, Ca, and K relative to stable ones like Al [*Nesbitt and Young*, 1982]. In addition to chemical weathering and provenance changes, hydraulic sorting may influence sedimentary elemental ratios. However, it has been demonstrated that there is no

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[1] Auxiliary material data sets are available at ftp://ftp.agu.org/apend/gl/2009gl040279. Other auxiliary material files are in the HTML.

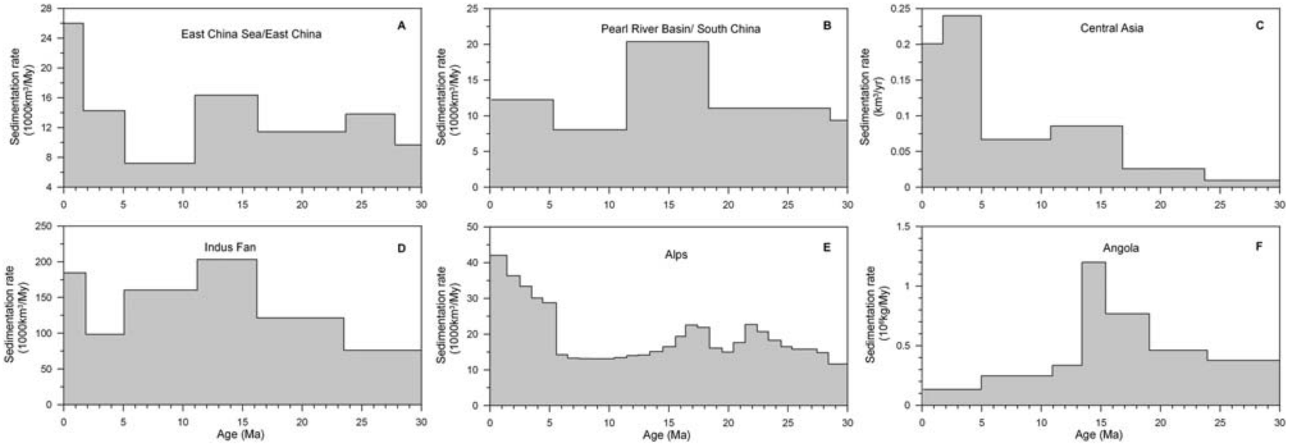

Figure 2. Examples of variations in sedimentation rates, showing an evident increase during the Early-Middle Miocene in different settings including (a) East China Sea, (b) South China Sea [*Clift*, 2006], (c) Central Asia [*Métivier et al.*, 1999], (d) Indus Fan [*Clift*, 2006], (e) Alps [*Kuhlemann et al.*, 2002], and (f) Angola [*Lavier et al.*, 2001].

significant influence of sea-level change on the sedimentation at ODP Site 1146 over the million year time scale, as revealed by the lack of correlation between terrigenous MAR, grain-size, and global sea-level since 20 Ma [*Wan et al.*, 2007]. Therefore, CIA values do not reflect changes in provenance or transport processes, but constrain chemical weathering intensity in southern China which is in agreement with previous studies [*Wei et al.*, 2006].

[6] As shown in Figure 1, the very similar trends of CIA at both ODP Sites 1146 and 1148 clearly indicate an overall slightly decreasing trend in chemical weathering intensity in South China during the Miocene. Intriguingly, there is a very high CIA interval between about 17.2–15 Ma, during which the average CIA value is up to 79, which is very close to the degree of extreme weathering (CIA > 80 [*Fedo et al.*, 1995]). Furthermore, the terrigenous MAR at ODP Site 1146 and 1148 between 17.2–15 Ma averages 3.5 g/cm^2/ky, about 3–4 times higher than those before and after this period in the Miocene (Figure 1). Because the sedimentary provenance did not change at that time, we can conclude on the basis of CIA and terrigenous flux that the climate in South China between 17.2–15 Ma was characterized by extreme chemical weathering together with strong physical erosion.

[7] Our proposed period of extreme erosion in the Early-Middle Miocene is also supported by analysis of a number of regional seismic reflection profiles through the Asian marginal seas [*Clift*, 2006]. This work showed that the Early-Middle Miocene was a time of rapid sedimentation in the major basins of Asia including the Mekong/Nam Con Son Basin, Gulf of Thailand, Pearl River Mouth Basin, Yinggehai-Song Hong Basin, and Indus Fan [*Clift*, 2006] (Figure 2). More importantly, sediment budgets produced for the Central Asia [*Métivier et al.*, 1999], European Alps [*Kuhlemann et al.*, 2002] and offshore Angola [*Lavier et al.*, 2001] also show similar patterns implying fast continental erosion rates worldwide in the Early-Middle Miocene (Figure 2). Together these data suggest that the rapid sedimentation event between ~17–15 Ma must be a global erosion event likely triggered by the global MCO rather than by regional tectonic activity, because there is little unequivocal evidence of global bedrock uplift in the Alps, Africa, the Himalayas, Indochina and throughout Asia at that time [e.g., *Hay et al.*, 2002; *Molnar and England*, 1990]. Moreover, our independent estimate of silicate weathering and erosion rate by CIA and MAR is consistent with the increased ^{87}Sr/^{86}Sr values during the MCO, suggesting high rates of chemical weathering and dissolved riverine fluxes to the oceans during that period [*Raymo*, 1994]. However, the overall stable or slightly decline in CIA through the Miocene is at odds with the secular increase of ^{87}Sr/^{86}Sr values, possibly suggesting the non-uniqueness about the interpretation of seawater strontium isotope ratio.

[8] The period of enhanced erosion and weathering during the Early-Middle Miocene (~17–15 Ma) correlates closely with the MCO, which is the warmest time of the past 35 Ma. At that time the mid-latitude temperatures were as much as 6°C higher than at present [*Zachos et al.*, 2001]. Evidence in support of a middle Miocene temperature and precipitation maximum has come not just from marine isotope records, but also from palaeosols in southeastern China [*Zou et al.*, 2004], and palaeobotanical records in East China [*Jiang and Ding*, 2008] (Figure 1), arctic Canada and Alaska [*White et al.*, 1997], northwest Germany [*Mosbrugger et al.*, 2005], and Serbia [*Utescher et al.*, 2007]. Warm temperatures and an enhanced hydrological cycle have been suggested as the main factors that would accelerate silicate chemical weathering rates, whereas precipitation is the most important factor controlling continental erosion rates after tectonic processes [e.g., *Clift*, 2006]. As a result our findings strongly support the presence of a climatic (i.e., temperature and precipitation) - continental silicate weathering and erosion feedback mechanism through the Miocene.

[9] On the geological timescale, atmospheric CO$_2$ levels are primarily regulated by variations in the rate of volcanic input from the Earth's interior together with the rate of output through silicate weathering and organic carbon burial at the Earth's surface [e.g., *Berner et al.*, 1983]. With regard to the CO$_2$ degassing flux during the MCO, it has been often suggested that the eruption of the huge amount of

Columbia River basalt between 17 and 15 Ma, should have released a significant volume of CO_2 directly to the atmosphere [e.g., *Camp*, 1995]. In addition, a high peak of crustal production of back-arc basins in the period of the MCO may have also contributed to CO_2 degassing [*Kaiho and Saito*, 1994]. However, the Miocene atmospheric CO_2 reconstructions inferred from different proxies are equivocal. There is no clear correlation between Miocene climate evolution, weathering history and the CO_2 proxy record based on alkenone C isotope measurements evident [*Pearson and Palmer*, 2000; *Pagani et al.*, 2005, Figure 1]. However, a recent climate modelling study [*You et al.*, 2009] suggests that MCO is most likely linked to elevated CO_2 (~500 ppmv) which is in agreement with CO_2 estimates based on stomatal frequency data [*Kürschner et al.*, 2008].

[10] The most commonly accepted process for providing rapid negative feedback, which stabilizes the Earth's long-term surface temperature and also exhibits functional dependence on atmospheric CO_2 is the response of silicate weathering to changes in climate [e.g., *Berner et al.*, 1983; *Walker et al.*, 1981]. Moreover, the burial of organic carbon has a significantly larger effect on the carbon cycle than silicate weathering, by a factor of 2–3 and is favored by the major influx of particles and nutrients eroded from mountains by climatic and tectonic factors [*France-Lanord and Derry*, 1997; *Galy et al.*, 2007]. We note that the MCO corresponds to an enhanced silicate chemical weathering and physical erosion in eastern Asia and a synchronous increase in the burial of organic carbon inferred from carbon cycle models [*Raymo*, 1997; *Derry and France-Lanord*, 1996], as well as the carbon isotope record at ODP Site 1148 [*Cheng et al.*, 2004] (Figure 1). Because organic carbon is highly enriched in the light isotope of carbon during photosynthesis, any increase in the fraction of organic carbon removed from the surface carbon reservoir will make the remaining inorganic carbon relatively more enriched in the heavy isotope [e.g., *Raymo*, 1994]. Both increased silicate chemical weathering and organic carbon burial would have acted as an effective sink for atmospheric CO_2. Moreover, decreased CO_2 degassing rate since about 15 Ma resulting from the cessation of Columbia River basalt eruption [e.g., *Camp*, 1995] and the weakened crustal production rate of back-arc basins [*Kaiho and Saito*, 1994] would have resulted into a decline in CO_2 after the MCO and climate cooling. This feedback is in agreement with some CO_2 trend and climate proxy records [*Raymo*, 1994; *Zachos et al.*, 2001; *Kürschner et al.*, 2008]. Recently, the role of vegetation cover has been emphasized as a regulating feedback mechanism weakening rock weathering and therefore stabilizing atmospheric CO_2 during the late Cenozoic [*Pagani et al.*, 2009].

[11] Apart from enhanced weathering rates during the MCO, there is also at around 19 Ma slightly increased terrigenous MAR and CIA, and organic carbon burial (Figure 1) correlate with elevated CO_2 and a warm and more humid period (Figure 1). Earlier in the Cenozoic, enhanced weathering rates along with elevated atmospheric CO_2 level have been suggested for the Early Eocene Climatic Optimum [*Smith et al.*, 2008]. Therefore, our new data gives supporting evidence for the existence of a close, long-term coupling between continental silicate weathering, erosion, organic carbon burial, climate (i.e., temperature and precipitation) and atmospheric CO_2 throughout the Cenozoic.

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