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1 **Cenozoic advance of the East Asian monsoon promoted carbon dioxide**  
2 **consumption by weathering of the Mg-rich southern China upper crust**

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27 **Abstract**

28 The Oligocene-Miocene boundary climatic reorganization linked to the northward  
29 advance of the East Asian monsoon in subtropical China is a potentially important but  
30 poorly constrained atmospheric CO<sub>2</sub> sink. Here, we performed a first-order estimate  
31 of the difference in CO<sub>2</sub> consumption induced by silicate chemical weathering and  
32 organic carbon burial in subtropical China related to this monsoon advance. The  
33 results highlight the significant role of weathering of the Mg-rich upper continental  
34 crust in East China that would contribute significantly to the rise in the Mg content of

35 the ocean during the Neogene. Our results show that an increase in CO<sub>2</sub> consumption  
36 by silicate weathering varies between ~1% and 15% of the current global continental  
37 silicate sink with an ~60% contribution of Mg-silicate weathering since the late  
38 Oligocene but a negligible increase in the global organic carbon burial (<3.5%). The  
39 carbon impact of the northward advance of the East Asian monsoon is therefore  
40 significant but strongly related to the Mg-rich nature of the crust affected by such  
41 climatic change. Further modelling work indicates that the uplift of the northern  
42 Tibetan Plateau plays a key role in this monsoon advance. Our study thus suggests  
43 that the uplift of the Himalayas-Tibetan Plateau can lead to indirect modification of  
44 the carbon cycle and the global climate by changing the regional hydrological cycle in  
45 areas of East Asia that are tectonically inactive in addition to the direct impact of high  
46 erosion and organic carbon burial along the orogenic belt in South Asia.

47 **Keywords:** Tibetan Plateau; Late Oligocene; silicate weathering; carbon cycle;  
48 oceanic Mg cycle

49

## 50 **1. Introduction**

51 Atmospheric CO<sub>2</sub> consumption by silicate weathering and organic carbon burial  
52 are the sinks that control atmospheric CO<sub>2</sub> levels over geological timescales. Complex  
53 feedback mechanisms between climate and tectonics led to a hypothesis that the uplift  
54 of the Himalayas-Tibetan Plateau (TP) was a primary driver of Cenozoic atmospheric  
55 CO<sub>2</sub> decline and global cooling predominantly through accelerating silicate chemical  
56 weathering in the India-Asia collision zone (Raymo and Ruddiman et al., 1992; Kump  
57 et al., 2000), while debated (e.g., Blum et al., 1998), and through effective burial of  
58 organic carbon in the nearby Bengal Fan in South Asia (France-Lanord and Derry,  
59 1997; Galy et al., 2007; Lee et al., 2019). However, orogenic uplift also contributed to  
60 the increase in monsoonal rainfall by the expansion of the East Asian monsoon and, as  
61 such, might have extensive control over the geological carbon cycle by enhancing  
62 silicate chemical weathering and/or burial of organic carbon in East Asia, namely,  
63 outside the Bay of Bengal (Fig. 1).

64 The modern East Asian climatic pattern is likely to have been established at the  
65 Oligocene-Miocene boundary with an arid zone restricted to Northwest China, in  
66 contrast to a relatively broad belt of aridity stretching across China from west to the  
67 east during the Palaeogene (Fig. 1, Guo et al., 2008; Sun and Wang, 2005;). The onset  
68 of the East Asian monsoon was recently proposed to be during the Eocene, but  
69 monsoonal activities were restricted to regions not exceeding the far south of China

70 (e.g., Licht et al., 2014; Spicer et al., 2016; Xie et al., 2020). Monsoon penetration  
71 into modern arid Northwest China may have extensively occurred only from the latest  
72 early Oligocene to early Miocene, as suggested by fossil evidence (Guo et al., 2008;  
73 Miao et al., 2013; Song et al., 2019; Sun and Wang, 2005). This reorganization of the  
74 climate system around the Oligocene-Miocene boundary from arid to modern,  
75 monsoon-like humid in this broad region involves a total area of  $\sim 3.2 \times 10^6 \text{ km}^2$  (Fig.  
76 1). The increase in rainfall due to the monsoon advance over such a broad region is  
77 thus expected to not only enhance silicate weathering but also accelerate terrestrial  
78 organic carbon production. In particular, weathering of Mg-enriched Yangtze and  
79 North China cratons in East China (Gao et al., 1991, 1998), which makes up the  
80 majority of the area affected by climatic reorganization around the  
81 Oligocene-Miocene boundary (Fig. 1b), could yield a more significant atmospheric  
82  $\text{CO}_2$  sink than the weathering of Mg-depleted Himalayan terranes (e.g.,  
83 France-Lanord and Derry, 1997). To test this hypothesis, we performed a first-order  
84 estimate of the changes in  $\text{CO}_2$  consumption flux from silicate weathering and organic  
85 carbon burial through the Palaeogene-Neogene boundary using the chemistry of  
86 sediments in the South China Sea (SCS) and soils in East China regions.

## 87 **2. Material and methods**

### 88 **2.1 Quantifying carbon dioxide consumption by silicate weathering**

89 Quantification of the release of soil base cations in silicate weathering can be  
90 done only by normalizing their abundances to that of an immobile element, and we  
91 used Al since other immobile elements, such as Ti, are more sensitive to changes in  
92 the composition of the regolith (e.g., Ti is relatively enriched in mafic rocks in the  
93 upper crust of eastern China, Gao et al., 1991). Given that Al is an immobile element  
94 during bedrock weathering, the  $\text{CO}_2$  consumption ( $\text{mol}\cdot\text{kg}^{-1}$ ) due to weathering of  
95 silicates in bedrock for a given base cation ( $\Delta X_0$ ) can be estimated by:

$$96 \Delta X_0 = ([X]_{\text{bedrock}} - [Al]_{\text{bedrock}} / [Al]_{\text{soil}} \times [X]_{\text{soil}}) / M_X \times 100 \quad (1),$$

97 where  $[X]$  shows the concentration (%) of element X measured in soil and bedrock  
98 and M is the molar mass for a given element X. The overall  $\text{CO}_2$  consumption on  
99 geological timescales ( $>1 \text{ Myr}$ ) by silicate weathering ( $C_{\text{sil}}$ ) between bedrock and soil  
100 can thus be estimated by the overall difference of soil base cations:

$$101 C_{\text{sil}} = \Delta \text{Ca}_0 + \Delta \text{Mg}_0 + 0.10 \Delta \text{K}_0 + 0.15 \Delta \text{Na}_0 \quad (2),$$

102 assuming that all the alkalinity equivalent to  $\text{Mg}^{2+}$  and  $\text{Ca}^{2+}$  lost from silicate minerals  
103 ultimately precipitates as marine carbonate, that only a fraction of the alkalinity is  
104 associated with alkalis (20% for  $\text{K}^+$  and 30% for  $\text{Na}^+$ ) and that there is no “reverse

105 weathering” as a sink for these base cations in marine settings (France-Lanord and  
106 Derry, 1997).

107 The change in CO<sub>2</sub> consumption by silicate weathering ( $\Delta_{\text{sil}}$ ) can be evaluated by  
108 chemical differences between the soil in the study area after and before climatic  
109 reorganization. Then, the overall difference in average soil base cations between arid  
110 loess and humid red soils ( $\Delta_{\text{sil}}$ ) can be estimated as:

$$\begin{aligned} 111 \Delta_{\text{sil}} &= C_{\text{sil, a}} - C_{\text{sil, b}} \\ 112 &= (\Delta\text{Ca}_{0, \text{a}} - \Delta\text{Ca}_{0, \text{b}}) + (\Delta\text{Mg}_{0, \text{a}} - \Delta\text{Mg}_{0, \text{b}}) + 0.10 (\Delta\text{K}_{0, \text{a}} - \Delta\text{K}_{0, \text{b}}) + 0.15 (\Delta\text{Na}_{0, \text{a}} - \Delta\text{Na}_{0, \text{b}}) \\ 113 &= \Delta\text{Ca} + \Delta\text{Mg} + 0.10\Delta\text{K} + 0.15\Delta\text{Na} \quad (3), \end{aligned}$$

114 where subscripts “b” and “a” denote after and before the monsoon advance,  
115 respectively.

116 Because the tectonic setting in East China involving the assembly of the North  
117 China craton, Qinling Orogen and Yangtze craton formed prior to the Cenozoic  
118 (Zheng et al., 2013), we assumed that bedrock in East China exhibits the same  
119 composition between the Paleogene and the Neogene. Palaeogene arid soil  
120 composition is difficult to estimate due to the lack of data. However, a modern  
121 analogue of soil under semi-arid and arid settings could be the widespread eolian loess  
122 in northern China, which is typical of the well-mixed upper continental crust and is  
123 subject to only weak chemical weathering under arid climates (e.g., Gallet et al.,  
124 1996). Due to loess compositional homogeneity, we used the Xifeng (XF) Pleistocene  
125 loess (Fig. 1, Table S1) from northern China to represent lithosols and cambisols  
126 distributed in the modern arid/semi-arid climate (Fig. 2a). Loess can bear carbonates,  
127 but its composition after acetic acid leaching could be typical of soil silicate  
128 compositions in Palaeogene arid climates. However, loess, lithosols and cambisols  
129 from NW China might not have been formed from the same regolith as the weathered  
130 crust of southern East China. Thus, the estimates of related terms, e.g.,  $C_{\text{sil}}$  and  $\Delta_{\text{sil}}$ ,  
131 could be biased. Such provenance bias is negligible except for  $\Delta\text{Mg}$  due to regional  
132 heterogeneity of Mg-bearing minerals in the upper crust rocks and the peculiar  
133 occurrence of a Mg-rich upper crust in East China (Gao et al., 1991, Gao et al., 1998).

134 Typical soils in humid subtropical China related to late Oligocene climatic  
135 reorganization are red earth and yellow earth, which account for ~80% of red soils in  
136 China (He et al., 2004). Red and yellow earths are similar in composition but different  
137 in colour, as they contain different iron oxides (hematite and goethite, respectively).  
138 Here, we chose the average compositions of four red earth datasets and three yellow  
139 earth datasets that are well spaced and can represent widespread acrisols in  
140 subtropical China (Fig. 2a; Table S1). The red earth datasets included Quaternary red  
141 earth compositions along the Yangtze River (Dongting Lake, DTL, a regional

142 investigation of red soils covering  $1.4 \times 10^4$  km<sup>2</sup>, Zhang et al., 2007; Jiujiang, JJ, a 10.4  
143 m thick red earth profile, and Xuancheng, XC, a 14 m thick red earth profile, Hong et  
144 al., 2013) and a red earth average across the entire South China (77 data of A horizon  
145 of soil profiles; CNEMC, 1990). The yellow earth datasets included Quaternary  
146 typical yellow earth compositions (Nanxiong, NX, a 2.4 m thick yellow earth profile;  
147 Tianyang, TY, a 4.2 m thick yellow earth profile) around the southern boundary of the  
148 subtropical zone (Table S1) and a yellow earth average across the entire South China  
149 (37 data of A horizon of soil profiles across South China, CNEMC, 1990). Gleysols  
150 from the lower reaches of the Yangtze and Yellow Rivers were not considered here  
151 because they are cultivated soils.

152 In our study, the NX and TY yellow earth samples were oven-dried, ground into  
153 powder, and leached by diluted acetic acid. Diluted acetic acid leaching will introduce  
154 few Ca from silicates. The residues were digested by pressurized acid digestion using  
155 a mixture of HNO<sub>3</sub>/HF. Major elements were analysed by inductively coupled plasma  
156 optical emission spectroscopy (ICP-OES, Leeman Labs Prodigy-H) at the Institute of  
157 Tibet Plateau Research, Chinese Academy of Sciences. Concentrations of all elements  
158 were normalized to the weight of the bulk leached residues. Replicate analyses of  
159 samples showed relative standard deviations of <2%. Except NX and TY yellow earth  
160 samples with acetic leaching residues, other data used here are whole rock data. Since  
161 few amounts of carbonates exist in such low-pH environments in south China, the two  
162 types of data are comparable.

163 To impart constraints on changes in soil silicate composition in response to global  
164 cooling and regional climate variability during the Neogene, we also estimated  $\Delta_{\text{sil}}$   
165 variability using the average compositions of diluted acetic acid leached sediments of  
166 Ocean Drilling Program (ODP) site 1146 in the SCS (Fig. 1a). These sediments  
167 represent an average of the eroded silicates from southern East China (e.g., Liu et al.  
168 2003). Considering the variation in silicate weathering intensity, two stages have been  
169 defined at 15-19 Ma (SCS 15-19 Ma) and 0-5 Ma (SCS 0-5 Ma), corresponding to  
170 strong and weak silicate weathering intensity stages, respectively (Wan et al., 2010).  
171 We used these two stages as the Neogene composition and, similar to our soil  
172 approach, compared it to the arid loess in North China.

## 173 **2.2 Quantifying carbon dioxide consumption by organic carbon burial**

174 Changes in the organic carbon burial ( $\Delta_{\text{oc}}$ ) after and before climatic  
175 reorganization can be quantified by measuring the difference in the terrestrial organic  
176 carbon content in East and South Asia clastic sediments. However,  $\Delta_{\text{oc}}$  cannot be

177 directly evaluated using data from marginal sea sediments because it is difficult to  
178 distinguish terrestrial and marine contributions to bulk organic carbon. When  $\Delta_{oc}$  was  
179 directly evaluated using marginal sea sediments, a dominance of marine organic  
180 material was revealed (Shipboard Scientific Party, 2000). However, by analogy with  
181 the findings in the Bay of Bengal (e.g., Galy et al., 2007; Lee et al. 2019), we assumed  
182 that  $\Delta_{oc}$  is principally controlled by terrestrial organic carbon production and burial  
183 efficiency in marine sediments, and  $\Delta_{oc}$  could be simply estimated as:

$$184 \Delta_{oc} = M_a \times f_a - M_b \times f_b \quad (4),$$

185 where  $M$  denotes organic matter content in soils,  $f$  denotes the organic carbon burial  
186 efficiency (percentage of organic carbon preserved in sediments), and the subscripts  
187 “a” and “b” denote after and before the monsoon advance, respectively.

### 188 **2.3. Quantifying monsoon rainfall in response to tectonic uplift**

189 To further constrain the orogenic uplift effect on the precipitation change in  
190 monsoon-influenced region in East China, we used a climate model to test  
191 precipitation differences between the modern topography and Palaeogene topography.  
192 Boundary conditions of the model corresponded to a change in topography only in the  
193 northern part of the TP since 1) uplift of this region exhibited a profound impact on  
194 the East Asian monsoon (Liu et al., 2015) and 2) the major uplift-related topographic  
195 change in this region occurred over the Neogene (e.g., Tapponnier et al., 2001; Lin et  
196 al., 2020). Two experiments were conducted with different topographies. The first  
197 utilized modern conditions with the present topography. In the second experiment, the  
198 northern TP was removed according to the uplift history of the TP (Tapponnier et al.,  
199 2001), which represents the Oligocene condition. Thus, the difference between the  
200 experiments gives the climatic effects of the rise of the northern part of the TP.

201 We employed the Community Earth System Model version 1.2.2 (CESM1.2.2)  
202 developed by the National Center of Atmospheric Research (NCAR). This model  
203 consists of atmosphere, ocean, land, river runoff, sea ice, land ice, and ocean wave  
204 components that interact with each other through a coupler. However, only the first 5  
205 components were active in the model used in this study (see details in Zhang et al.,  
206 2019). Both experiments are run with a  $CO_2$  concentration of 560 ppmv, which is  
207 higher than the present concentration and represents the late Oligocene level based on  
208 various proxies (Beerling and Royer, 2011). The concentrations of  
209 chloro-fluoro-carbon (CFCs) were set to 0, and those of  $CH_4$  and  $N_2O$  were set 760  
210 ppbv and 270 ppbv, respectively. All the other conditions, including  $O_3$  and aerosol  
211 concentrations, the solar constant and orbital parameters, were set to preindustrial

212 conditions. Both experiments were run for 1700 model years, which is long enough  
213 for the atmosphere and ocean surface layer to reach quasi-equilibrium. The last 50  
214 years of data from each experiment were analysed and presented in this study.

### 215 **3 The magnitude of silicate weathering and organic carbon burial change**

#### 216 **3.1 Silicate alteration and Mg contribution**

217 The results of  $C_{\text{sil}}$  show a high contribution of Ca-bearing mineral weathering  
218 ( $\Delta\text{Ca}_0$ ) in all chosen weathered products in humid subtropical China and arid northern  
219 China but a distinct pattern of Mg contribution ( $\Delta\text{Mg}_0$ ) between the two regions (Fig.  
220 3a).  $C_{\text{sil}}^*$ , defined as  $C_{\text{sil}}^* = C_{\text{sil}} - \Delta\text{Mg}_0$ , could be used to account for  $\text{CO}_2$  consumption  
221 by the chemical weathering of Ca-Na-K silicate minerals only (mainly feldspar).  
222 There are significant differences between  $C_{\text{sil}}^*$  and  $C_{\text{sil}}$  for the SCS sediments, the  
223 yellow and red earths, but there are few differences for the XF loess. This contrast  
224 suggests a dominant climate-induced weathering behaviour that is characterized by a  
225 greater depletion of Mg and Ca in humid subtropical China than in arid northern  
226 China. This climate-induced weathering contrast can also be supported by the results  
227 of  $\Delta_{\text{sil}}$  that can be regarded as the composition difference of weathering products  
228 between intense and weak weathering regimes (Fig. 3b). The detail of the contribution  
229 from each cation shows decreasing  $\text{CO}_2$  consumption values with  
230  $\Delta\text{Mg} > \Delta\text{Ca} > \Delta\text{Na} > \Delta\text{K}$  (Fig. 3b, Table S2), in line with the relative mineral stability  
231 during weathering (e.g., Mg-bearing minerals weather the fastest, followed by  
232 Ca-feldspar, Na-feldspar, and finally K-feldspar). Furthermore, the  $\Delta_{\text{sil}}/C_{\text{sil}}$  ratios yield  
233 a mean of  $\sim 50\%$  for SCS sediments and  $\sim 60\%$  subtropical soils, suggesting a  
234 dominant cation yield by intense weathering in humid South China.

235 The  $\Delta_{\text{sil}}$  pattern is suitable for displaying silicate alteration related to late  
236 Oligocene monsoon advance. Similarly,  $\Delta_{\text{sil}}^*$ , defined as  $\Delta_{\text{sil}}^* = \Delta_{\text{sil}} - \Delta\text{Mg}$ , was used to  
237 account for  $\text{CO}_2$  consumption by the chemical weathering of Ca-Na-K silicate  
238 minerals only (mainly feldspar). The difference between  $\Delta_{\text{sil}}$  and  $\Delta_{\text{sil}}^*$  indicates that the  
239 relative contribution of  $\Delta\text{Mg}$  is always  $>60\%$  (Table S3).  $\Delta_{\text{sil}}^*$  ranged from 0.12 to  
240 0.16  $\text{mol}\cdot\text{kg}^{-1}$  for SCS sediments and from 0.18 to 0.22  $\text{mol}\cdot\text{kg}^{-1}$  for modern soils (Fig.  
241 3b). Differences for the SCS sediments (19-15 Ma vs. 5-0 Ma) regarding different  
242 silicate weathering intensities (Table S3) suggest that the variability of Neogene  
243 climate/provenance for  $\Delta_{\text{sil}}^*$  and  $\Delta_{\text{sil}}$  is probably within 20-30%. These  $\Delta_{\text{sil}}^*$  values are  
244 comparable to  $\text{CO}_2$  consumption by silicate weathering in ODP 116 sediments in the  
245 Bengal Fan (Fig. 1), where  $\text{CO}_2$  consumption ( $C_{\text{sil}}$ ) is between 0.17  $\text{mol}\cdot\text{kg}^{-1}$  for  
246 illite-chlorite sediments with no contribution from  $\Delta\text{Mg}$  and 0.23  $\text{mol}\cdot\text{kg}^{-1}$  for



247 smectite-kaolinite sediments with a contribution of only  $\sim 0.03 \text{ mol}\cdot\text{kg}^{-1}$  by  $\Delta\text{Mg}$  (Fig.  
248 3a; France-Lanord and Derry, 1997). This suggests a comparable magnitude of  $\text{CO}_2$   
249 consumption by silicate alteration except for Mg for both regions.

250 Overall, the key difference in monsoon-influenced silicate weathering in  
251 subtropical China is the dominant effect of weathering of Mg-bearing silicates, which  
252 is likely related to the higher Mg concentrations in the Yangtze and Northern China  
253 cratons (e.g., Gao et al., 1991; Gao et al., 1998) than in the average upper continental  
254 crust (UCC, e.g., Rudnick and Gao, 2014) or the Himalayan crust (e.g.,  
255 France-Lanord and Derry, 1997). For example, the relatively low  $\Delta\text{Mg}$  contribution  $<$   
256  $\sim 0.03 \text{ mol}\cdot\text{kg}^{-1}$  in South Asia corresponds to High Himalayan crystalline rocks with  
257 low MgO (1.76%, France-Lanord and Derry, 1997). Further, the erosional products  
258 have higher MgO contents (with a mean of 3.12%,  $n=30$ ) for the fine-grained silicate  
259 sediments in the Yangtze River (He et al., 2015) than those in the XF loess (2.76%)  
260 (Table S1). These observations conform to that East China (carbonate-free  
261 composition), compared with other terranes (e.g., the Canadian shield), contains large  
262 proportions of unmetamorphosed supracrustal units that are considered to have, on  
263 average, higher proportions of mafic volcanics (Gao et al., 1998; Rudnick and Gao,  
264 2004). Generally, the high  $\Delta\text{Mg}$  contribution can result from the facts that 1) Mg-rich  
265 upper crust rocks will be strongly depleted during soil silicate weathering and 2) the  
266 upper crust that experienced the intensification of climate-induced weathering is a  
267 Mg-rich portion. Such low MgO content preserved in red and yellow earths under a  
268 humid climate (Fig. 1) that dominated the Mg-rich upper crust in East China thus  
269 suggests a significantly higher Mg loss by weathering of the East China upper crust  
270 affected by late Oligocene climatic reorganization.

### 271 **3.2 Organic carbon burial**

272 To estimate  $\Delta_{\text{oc}}$ , we first qualified the organic carbon burial efficiency,  $f$ . It is  
273 suggested that a high  $f$  value mainly results from high erosion/rapid deposition  
274 systems (Blair and Aller, 2012), e.g., the Bengal Fan (Galy et al., 2007) and steep  
275 mountain rivers (Hilton et al., 2011). The Neogene monsoon climate in subtropical  
276 China can generate a large delivery of suspended sediments to the ocean in  
277 comparison with that in the Palaeogene (e.g., Métivier et al., 1999; Clift et al., 2004).  
278 Since the Yangtze River is the major carrier of sediments in subtropical China, we  
279 conservatively assumed that both  $f_a$  and  $f_b$  were  $\sim 30\%$ , the  $f$  value for the modern  
280 Yangtze River (Blair and Aller, 2012), which yielded a minimum estimation of  $\Delta_{\text{oc}}$ .

281 The modern soil organic carbon content is 0.8-1.8% in most subtropical China

282 and is 0.4-0.8% in arid-semi-arid China for surface soil horizons (0 to 30 cm) (Fig.  
283 2b). The area-weighted mean values are  $1.3\pm 0.6\%$  (excluding cultivated soils in East  
284 China) and  $0.7\pm 0.3\%$  for humid subtropical China and arid North China, respectively.  
285 Given a mean of  $\sim 0.2\%$  for the fossil carbon content inherited from parent rock in SE  
286 Asia (France-Lanord and Derry, 1997; Hilton et al, 2011) and assuming its burial  
287 efficiency of 100%,  $M_a$  and  $M_b$  were assumed to be 0.89 and  $0.39 \text{ mol}\cdot\text{kg}^{-1}$ ,  
288 respectively, and together yielded a  $\Delta_{oc}$  of  $0.15 \text{ mol C}\cdot\text{kg}^{-1}$ . This  $\Delta_{oc}$  is less than  $\Delta_{sil}$ ,  
289 exhibiting a mean value of  $0.60 \text{ mol}\cdot\text{kg}^{-1}$  in subtropical China, which contrasts with  
290 the Himalayas, where the  $\Delta_{oc}$  is much larger than  $\Delta_{sil}$  (France-Lanord and Derry,  
291 1997).

## 292 4. Discussion

### 293 4.1 Erosion rate related to the late Oligocene monsoon advance

294 Assessing the role of monsoon advance-induced silicate weathering and organic  
295 carbon burial in the context of the global budget requires an historical estimation of  
296 the volume of the crust subjected to erosion. We reconstructed this temporal variation  
297 in erosion flux as described in the following paragraphs.

298 In modern settings, the physical erosion of subtropical China is highly variable  
299 with  $^{10}\text{Be}$ -based erosion rates from  $20\text{-}50 \text{ mm}\cdot\text{kyr}^{-1}$  in low relief areas to  $700\text{-}900$   
300  $\text{mm}\cdot\text{kyr}^{-1}$  in steep, landslide-dominated mountain terrane (Chappel et al., 2006). The  
301 drainage basin of the Yangtze River is mainly located within the study area (Fig. 1a)  
302 and accounts for 59% of the study area. In addition, the Yangtze River drains variable  
303 landscapes and climates and is thus likely to provide an area-averaged erosion rate for  
304 the whole studied area. Therefore, we assumed that the physical erosion of subtropical  
305 China (total area of  $\sim 3.2 \times 10^6 \text{ km}^2$ ) is  $\sim 800 \times 10^6 \text{ t}\cdot\text{yr}^{-1}$  by extrapolation of  $480 \times 10^6$   
306  $\text{t}\cdot\text{yr}^{-1}$  for the Yangtze River (Milliman and Syvitski, 1992).

307 We then assumed that the long-term variations in erosion flux related to  
308 subtropical China could generally scale with the temporal variations in the sediment  
309 budget around the East China marginal basins. The existing Cenozoic sediment  
310 budgets for the major basin systems of East Asia have yielded distinct long-term  
311 variations (Clift, 2004; Metivier et al., 1999). The results of Clift et al. (2004)  
312 including the Pearl River, East China Sea and Bohai Bay display different long-term  
313 variations with peaks of sedimentation in the Mid-Miocene and Quaternary (Clift et  
314 al., 2004; Fig. 4a). Metivier et al. (1999) showed a monotonous increase in the  
315 sedimentation rate in the East China Platform, which includes the East China  
316 Sea/North Taiwan, Okinawa, Yellow Sea and Bohai Bay (Fig. 4a). Clarification of

317 both variations is out of the aim and scope of this study; thus, we used our modern  
318 estimate of erosion flux to multiply the two relative variations of sediment discharges  
319 to construct two records of erosion flux related to subtropical China climatic  
320 reorganization since the late Oligocene (Fig. 4a).

## 321 4.2 Mg flux linked to the monsoon advance

322 The Mg flux variation linked to the monsoon advance from silicate weathering,  
323  $F_{Mg, sil}$ , can be calculated as follows:

$$324 F_{Mg, sil} = E_a \times \Delta Mg_{0, a} - E_b \times \Delta Mg_{0, b} \quad (5),$$

325 where  $E$  is sediment yield ( $kg \cdot yr^{-1}$ ) and it corresponds to our reconstructed erosion  
326 flux (Figs. 4a).  $\Delta Mg_{0, b}$  is  $0.03 \text{ mol} \cdot kg^{-1}$ , identical to XF loess (Fig. 3a).  $0.45 \text{ mol} \cdot kg^{-1}$   
327 for  $\Delta Mg_{0, b}$  is the mean of  $\Delta Mg_0$  for SCS sediments and red soils in subtropical China  
328 (Fig. 3).

329 In addition, the sedimentary cover of East China bears a significant amount of  
330 dolostone lithology (e.g., Gao et al., 1991). The Neogene intensification of the  
331 weathering of that area would also have dissolved a relatively large amount of  
332 Mg-rich carbonate, further delivering Mg to the oceanic cycle. The Mg and Ca  
333 concentrations related to carbonate in the eastern China upper crust can be estimated  
334 by the comparison of the CEC upper crust compositions with and without carbonate  
335 (Gao et al., 1998). Assuming Si resides in only silicates, then the Mg molar  
336 concentration in carbonate ( $[Mg]_{carb}$ ,  $mol \cdot kg^{-1}$ ) can be estimated as follows:

$$337 [Mg]_{carb} = [Mg]_{bulk} - [Mg]_{sil} / [Si]_{bulk} \times [Si]_{sil} \quad (6),$$

338 where the subscripts “bulk” and “sil” denote element molar concentrations the CEC  
339 upper crust compositions with and without carbonates, respectively. The estimated  
340  $[Mg]_{carb}$  is  $0.37 \text{ mol} \cdot kg^{-1}$ , and  $[Ca]_{carb}$  is  $0.80 \text{ mol} \cdot kg^{-1}$ , as calculated by the same  
341 approach. The molar Mg/Ca ratio in carbonate in eastern China is 0.46, which  
342 indicates that approximately 50% of carbonate minerals are dolomite, given a molar  
343 Mg/Ca ratio of 0.08 for calcite (a lower limit of average post-Archean carbonates in  
344 East China, Gao et al., 1991) and a molar Mg/Ca ratio of 1 for dolomite, supporting  
345 the above observation of a dolostone-rich lithology in East China. Since dolomite is  
346 hard to dissolve in arid and semi-arid settings (e.g., in arid NW China), we assumed  
347 that only calcite dissolves in arid settings before the monsoon advance and that both  
348 calcite and dolomite dissolve in humid settings after the monsoon advance. The Mg  
349 flux variation linked to the monsoon advance through carbonate weathering,  $F_{Mg, sil}$ ,  
350 can be calculated as follows:

$$351 F_{Mg, carb} = E_a \times [Mg]_{carb, a} - E_b \times [Mg]_{carb, b} \quad (7)$$

352 Here,  $[Mg]_{carb, a} = 0.37 \text{ mol}\cdot\text{kg}^{-1}$  corresponds to all Mg in carbonates (calcite and  
353 dolomite), and  $[Mg]_{carb, b} = 0.04 \text{ mol}\cdot\text{kg}^{-1}$  corresponds to Mg only in calcite with a  
354 molar Mg/Ca ratio of 0.08 in the East China upper crust.

355 The reconstructed Mg release flux from weathering of silicate ( $F_{Mg, sil}$ ) and  
356 carbonate ( $F_{Mg, carb}$ ) related to the late Oligocene monsoon advance in East Asia shows  
357 that both fluxes account for their respective modern global flux 5-17% based on  
358 erosion flux variation reported by Clift et al. (2004) and  $\sim 1.5$ -17% based on erosion  
359 flux variation reported by Metivier et al. (1999) (Figs. 4b and 4c; the modern Mg flux  
360 from silicate weathering and carbonate weathering is  $2$ - $2.5 \times 10^{12} \text{ mol}\cdot\text{yr}^{-1}$  and  
361  $2$ - $2.2 \times 10^{12} \text{ mol}\cdot\text{yr}^{-1}$ , respectively, according to Higgins and Schrag, 2015). The Mg  
362 oceanic cycle during the Cenozoic is characterized by a rise in the Mg content in the  
363 ocean (e.g., Horita et al., 2002). The rise in the sea water Mg concentration from 38  
364  $\text{mmol}\cdot\text{kg}^{-1}$  at 37 Ma to  $55.1 \text{ mmol}\cdot\text{kg}^{-1}$  at present (Horita et al., 2002) requires an  
365 average rate of increase of  $0.46 \text{ mmol}\cdot\text{Ma}^{-1}$ . The combined Mg flux of silicate and  
366 carbonate weathering related to the late Oligocene monsoon advance in East Asia  
367 accounts for a rate of increase of the seawater Mg concentration is  $0.18$ - $0.45$   
368  $\text{mmol}\cdot\text{Ma}^{-1}$  based on erosion flux variation reported by Clift et al. (2004) and  
369  $0.05$ - $0.46 \text{ mmol}\cdot\text{Ma}^{-1}$  based on erosion flux variation reported by Metivier et al. (1999)  
370 since the late Oligocene. Our results clearly suggest that at least a significant portion  
371 of this rise in seawater Mg content corresponds to the weathering of Mg-silicates and  
372 dolomite-rich carbonates in East China rather than just changes in the amount of Mg  
373 removed from seawater (Higgins and Schrag, 2015).

#### 374 4.3 Carbon consumption flux linked to the monsoon advance

375 Similar to the Mg flux estimation, the flux of long-term  $\text{CO}_2$  consumption by  
376 silicate weathering linked to the monsoon advance,  $F_{sil}$ , can be calculated as follows:

$$377 F_{sil} = E_a \times C_{sil, a} - E_b \times C_{sil, b} \quad (8),$$

378 where  $C_{sil, a}$  is  $1.04 \text{ mol}\cdot\text{kg}^{-1}$ , which is the mean of  $C_{sil}$  values for SCS sediments and  
379 red soils in subtropical China (Fig. 3); and  $C_{sil, b}$  is  $0.43 \text{ mol}\cdot\text{kg}^{-1}$ , identical to XF  
380 loess.

381 The reconstructed fluxes of long-term  $\text{CO}_2$  consumption by silicate weathering  
382 ( $F_{sil}$ ) (Fig. 4e) account for 3-8% of modern global flux of continental rock weathering  
383 ( $8.7 \times 10^{12} \text{ mol}\cdot\text{yr}^{-1}$ , Gaillardet et al. 1999) based on the erosion flux variation by Clift  
384 et al. (2004) and  $\sim 1$ -9% by Metivier et al. (1999). It should be noted that 98% of our  
385 estimated  $F_{sil}$  is attributed to Ca-Mg-silicate weathering since  $(\Delta\text{Mg}_0 + \Delta\text{Ca}_0)$  accounts  
386 for  $\sim 80\%$   $C_{sil}$  of all samples (Fig. 3a). The modern estimate of  $\text{CO}_2$  consumption

387 fluxes by Ca-Mg silicate is  $5 \times 10^{12} \text{ mol} \cdot \text{yr}^{-1}$  (Gaillardet et al. 1999), which is ~60% of  
388 the modern global flux of continental rock weathering. Thus, if only Ca-Mg-silicate  
389 weathering is considered, our estimated flux regarding weathering of Ca-Mg silicate  
390 accounts for modern global flux of continental Ca-Mg-silicate weathering was 5-13%  
391 based on the given erosion flux variation reported by Clift et al. (2004) and ~1-16%  
392 based on the erosion flux variation reported by Métivier et al. (1999).

393 In the same way, the flux of organic carbon burial linked to the monsoon advance,  
394  $F_{oc}$ , could be calculated as follows:

$$395 F_{oc} = E_a \times M_a \times f_a - E_b \times M_b \times f_b \quad (9),$$

396 The reconstructed organic carbon burial flux ( $F_{oc}$ ) shows <3.5% of the modern global  
397 flux of organic carbon for both erosion rate models (Fig. 4f). This organic carbon  
398 burial flux is approximately 25% of the contemporary  $\text{CO}_2$  consumption by silicate  
399 weathering (Fig. 4f), which also contrasts with the Himalayas, where the organic  
400 carbon burial flux is much larger than that of silicate weathering (France-Lanord and  
401 Derry, 1997).

402 The reconstructed long-term  $\text{CO}_2$  consumption fluxes by silicate weathering and  
403 organic burial temporally vary, but the total amount ( $F_{sil} + F_{oc}$ ) is within the same order  
404 as that in the Himalayas ( $1.28 \times 10^{12} \text{ mol} \cdot \text{yr}^{-1}$ ; France-Lanord and Derry, 1997). If the  
405 Cenozoic long-term silicate weathering fluxes are relatively stable (Caves et al., 2016),  
406 our reconstructed long-term  $\text{CO}_2$  consumption fluxes by silicate weathering and  
407 organic burial can induce an imbalance in the geological carbon cycle. For long-term  
408  $\text{CO}_2$  consumption fluxes by silicate weathering, weathering of Mg silicate ( $\Delta_{Mg}$ )  
409 accounts for 64-72% of total  $\text{CO}_2$  consumption ( $\Delta_{sil}$ ), with a mean of ~69% (Table S3).  
410 Considering that the study region occupies ~2% of the total continental area, our  
411 study suggests a significant additional atmospheric  $\text{CO}_2$  sink by silicate weathering  
412 when Mg-bearing minerals are subjected to weathering in response to this climatic  
413 reorganization since the late Oligocene. Such intensification of the weathering of the  
414 Yangtze and North China cratons will also contribute to the required increasing land  
415 surface reactivity during the Neogene (e.g., Kump and Arthur 1997; Caves et al. 2016;  
416 Caves Rugenstein et al., 2019). Additionally, fast and congruent weathering of  
417 dolomite-rich carbonate minerals (~10%) in the upper crust of humid subtropical  
418 China after the monsoon advance may also contribute a long-term carbon sink by  
419 aquatic photosynthesis (e.g., Liu et al., 2018).

## 420 **5 Implications for tectonic-climatic linkage**

421 Our modelling results show a significant increase in annual precipitation, with

422 magnitudes of ~100->400 mm, in response to topographic changes in the northern TP  
423 (Fig. 7). Moreover, most of the annual precipitation increase occurred in summer,  
424 especially over a large part of the Yangtze and North China cratons (Fig. 5), which  
425 indicates that the East Asian monsoon climate system might have been established or  
426 greatly strengthened. In summer, topographically elevated heating leads to  
427 strengthening of the low over Asia, with which the anomalous southeasterly and/or  
428 southwesterly winds mainly occur over East China (Fig. 5d). Thus, the modelling  
429 results indicate that the East Asian summer monsoon circulation has been established  
430 or is greatly stronger than it previously was. This strongly suggests that a  
431 tectonic-climatic linkage induced by the uplift of the northern TP has played a major  
432 role in the establishment of East Asian monsoon cycles in East China (e.g., Tada et al.,  
433 2016). The rapid late Oligocene-early Miocene uplift of the TP and a series of  
434 concomitant tectonic events are evidenced by various studies of tectonic deformation  
435 and volcanic rock emplacement (Lu et al., 2018). The size of the India-Asia collision  
436 thus had a prominent effect on the reorganization of the climatic patterns beyond the  
437 collision zone with the northward advance of the East Asian monsoon in tectonically  
438 inactive subtropical China, which, in turn, induced a globally significant silicate  
439 weathering CO<sub>2</sub> sink.

440 The study of orogenic effects on the global climate during the Cenozoic has  
441 focused almost exclusively on the India-Asia collision zone (e.g., Raymo and  
442 Ruddiman, 1992; France-Lanord and Derry, 1997). Our study suggests that the  
443 significant rise in CO<sub>2</sub> consumption induced by climatic changes and enhanced  
444 erosion in both South and East Asia is an effect of the Indian-Asian collision and  
445 uplift of the TP. This effect is characterized by an approximately equal order of  
446 magnitude between the direct effect of the orogeny via enhanced carbon burial in  
447 South Asia and the indirect effect that enhanced the weathering of Mg-rich silicate of  
448 the Yangtze craton and surrounding terrane. The unusual nature of the exposed crust  
449 in East China suggests that the tectonic impact on Cenozoic cooling via modulation of  
450 the geological carbon cycle is particularly intense, and such forcing might not be fully  
451 extrapolated to older global-scale orogeny. Our study thus illustrates how complex  
452 perturbations of global climate and atmospheric CO<sub>2</sub> levels by orogenic uplift can be  
453 and how important the nature of the crust is not only involved in collisions but also  
454 around collisions. This last point might have generated an imbalance in the global  
455 carbon cycle since it is not involved in any tectonic feedback but is sensitive to  
456 climate feedback, which may have contributed to the sharp atmospheric CO<sub>2</sub> decline  
457 during the late Oligocene and early Miocene transition (e.g., Beerling and Royer,  
458 2011) and land reactivity that even regulates Neogene global cooling (Caves

459 Rugestein et al. et al., 2019). Although there are many unresolvable uncertainties,  
460 e.g., erosion fluctuations over short and long timescales (Métivier and Gaudemer,  
461 1999; Clift et al., 2006), our study has provided a first-order estimate of the global  
462 geochemical cycle regarding carbon and magnesium linked to such complex  
463 tectonic-climate linkages.

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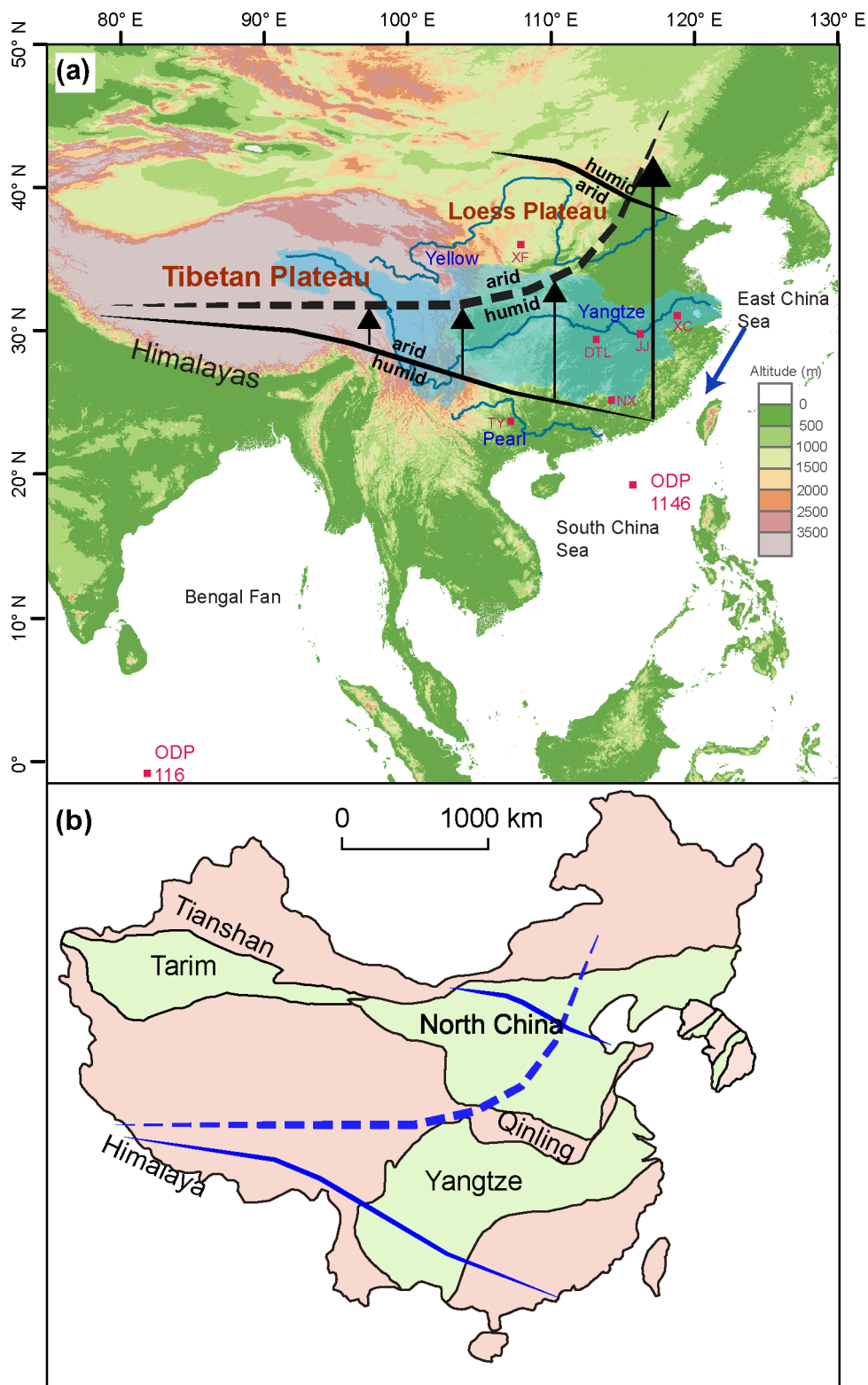
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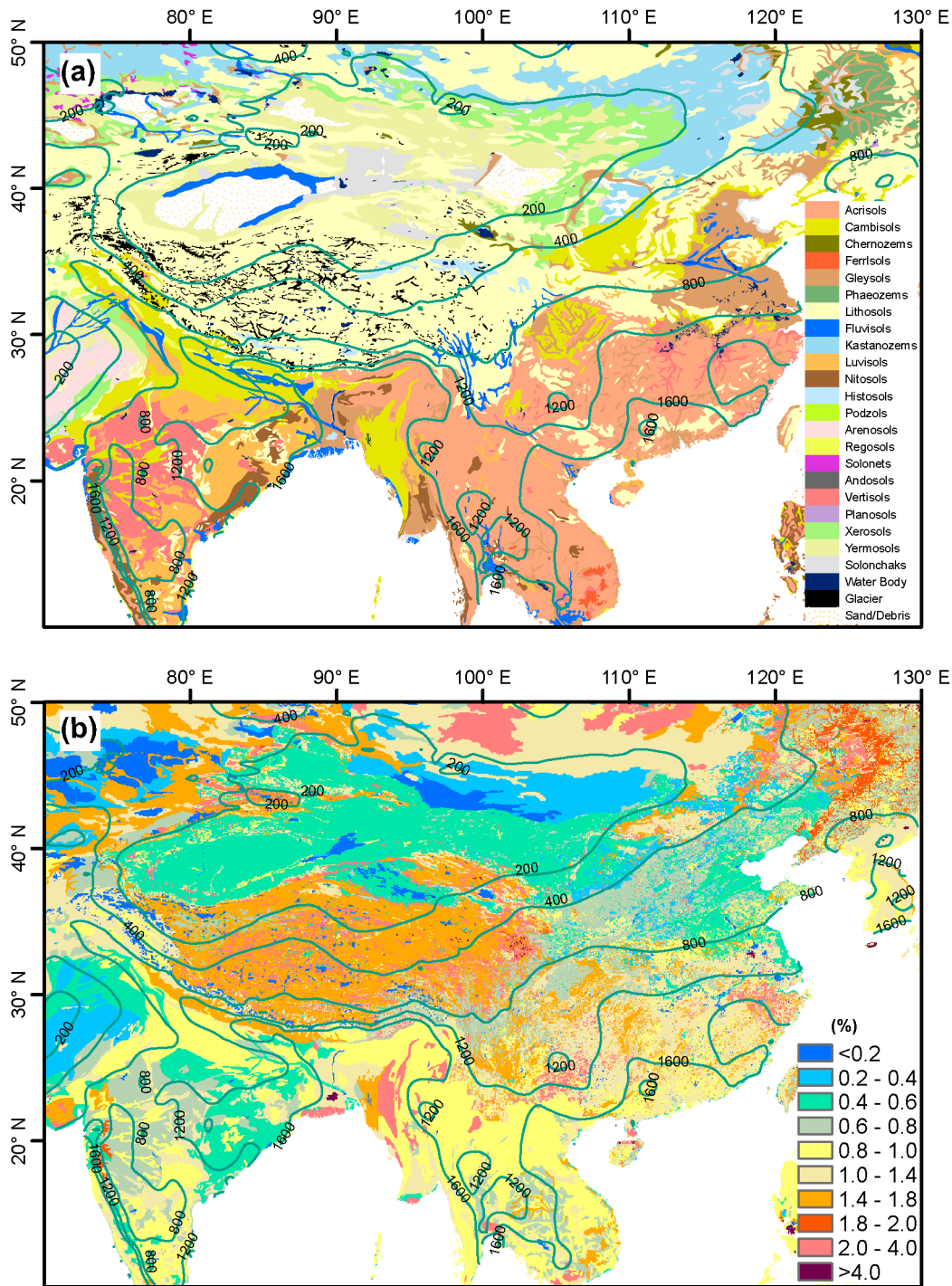
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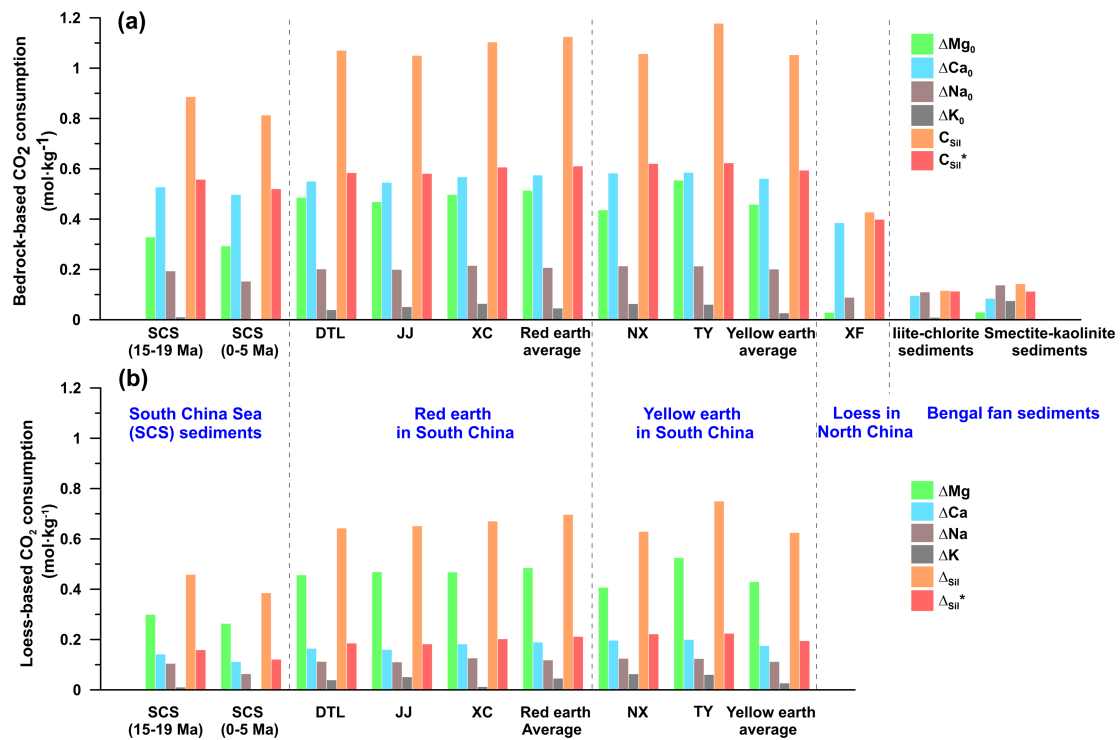
609  
 610 **Figure 1 (a)** Cenozoic humid/arid boundaries in China for the Palaeogene (black bold  
 611 line) and the Neogene to Quaternary (black dashed line). Black arrows show the  
 612 northward migration of the humid zone. Red squares are locations of the studied soils  
 613 (XF, Xifeng loess; DTL, Dongting Lake, JJ, Jiujiang; XC, Xuancheng; NX, Nanxiong;

614 and TY, Tianyang) and sediments (ODP site 1146 in the South China Sea, SCS; ODP  
615 site 116 in the Bengal Fan). The light blue shaded area marks the drainage area of the  
616 Yangtze River. The blue arrow shows the modern transport path of sediments from the  
617 Yangtze River mouth to the SCS (Liu et al., 2003). **(b)** Simplified tectonic map of the  
618 Chinese continent showing the distributions of major cratons (green area) and orogens  
619 (light brown area) (redrawn from Gao et al., 1998 and Zheng et al., 2013). Black lines  
620 mark major faults. The northward migration of the humid zone (the area between the  
621 blue bold and dashed lines) impacted the majority of the Yangtze craton.



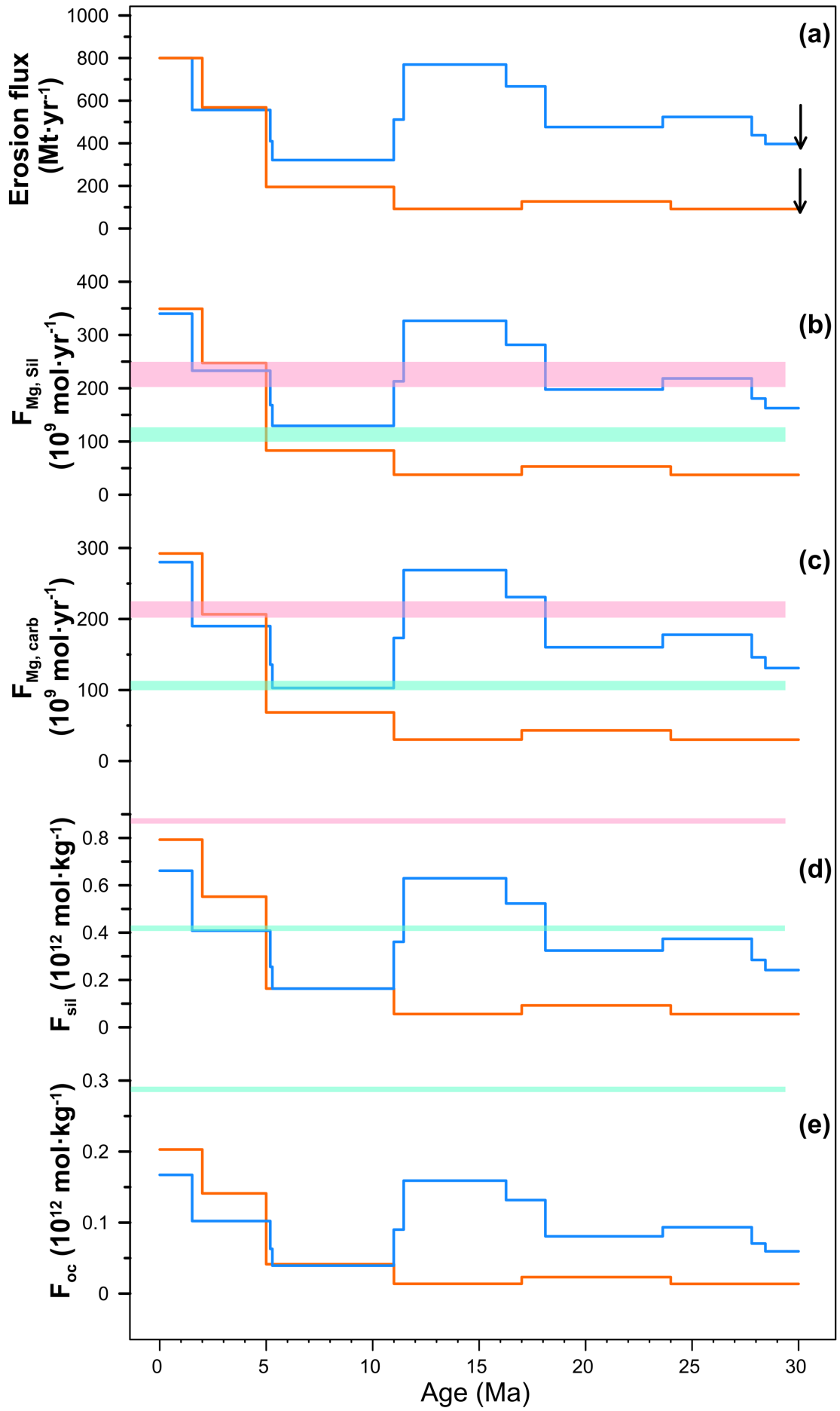
622

623 **Figure 2 (a)** Modern soil type distributions with isohyets (mm) in Asia. Soil type data  
 624 are based on the FAO-UNESCO Soil Map of the World  
 625 (<http://www.fao.org/geonetwork/srv/en/main.home>). **(b)** Soil organic carbon content  
 626 (%) distributions with isohyets (mm) in China. Organic carbon data refer to surface  
 627 soil horizons (0 to 30 cm) from Wieder et al. (2014). Isohyet data are from updated  
 628 high-resolution grids of monthly climatic observations acquired from the CRU TS



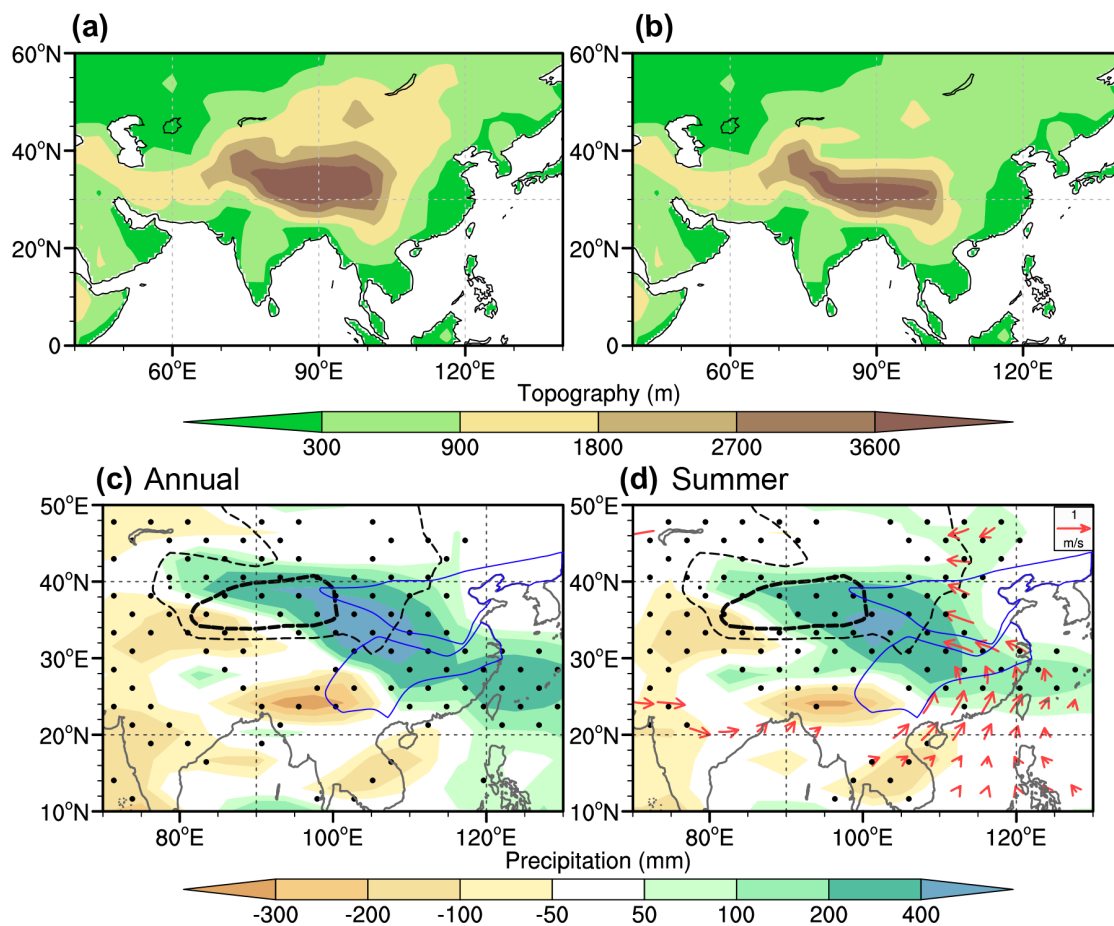
630

631 **Figure 3 (a)** Consumption of atmospheric CO<sub>2</sub> by silicate weathering estimated from  
 632 the difference between the chemistry of various sediments and bedrock. The Bengal  
 633 Fan sediment composite is based on unaltered Himalayan source rocks  
 634 (France-Lanord and Derry, 1997); others are calculated based on the carbonate-free  
 635 upper crust of East China (Gao et al., 1998). **(b)** Difference in atmospheric CO<sub>2</sub>  
 636 consumption by silicate weathering between various sediments used for the post-late  
 637 Oligocene monsoon-influenced region (sediments in the South China Sea  
 638 (SCS)/various soils in South China) and loess from Northwest China.





640 **Figure 4** The estimated erosion flux **(a)**, CO<sub>2</sub> consumption flux **(b)**, and Mg release  
641 fluxes from weathering of silicate ( $F_{Mg, sil}$ ) **(c)** and carbonate ( $F_{Mg, carb}$ ) **(d)** as well as  
642 organic burial flux **(e)**, related to late Oligocene monsoon advance in East Asia. The  
643 orange and blue cruves in **(a-e)** are based on two reconstructed sediment discharge  
644 fluxes based on the East China Platform dataset (Métivier et al., 1999) and integrative  
645 flux of the Pearl River and East China Sea basins (Clift et al., 2004), respectively.  
646 Curves in **(b)** were reconstructed by our estimated modern physical erosion flux of  
647  $\sim 800 \times 10^6 \text{ t}\cdot\text{yr}^{-1}$  for the monsoon-influenced region by multiplying the relative  
648 variations of two sediment discharge models. The black arrows in **(b)** mark the  
649 erosion flux before the late Oligocene climatic reorganization. The pink and green  
650 shaded areas in **(b-e)** mark the 10% and 5% levels of the modern flux for each panel,  
651 respectively.  
652



653  
654 **Figure 5** Modern topography **(a)** and removal of the northern Tibetan Plateau **(b)** over  
655 Asia. Differences in **(c)** annual precipitation (coloured areas) and **(d)** summer  
656 monsoon (May to September) precipitation (coloured areas) and 850 hPa winds (red  
657 arrows) between the climate model experiments with the modern topography. Dotted  
658 areas correspond to places with confidence levels greater than 95% (using Student's

659 *t*-test) in precipitation change. The thick and thin black dashed lines mark the contour  
660 lines of 500 and 1500 m for topography removal, respectively, of the northern Tibetan  
661 Plateau between the two climate models. The blue lines indicate the contours of the  
662 Yangtze and North China cratons.