Dear, Editor Ran Feng,

We have carefully revised and edited the manuscript entitled "Late Miocene-Pliocene climate evolution recorded by the red clay covered on the Xiaoshuizi planation surface, NE Tibetan Plateau" based on your valuable comments and suggestions. This time, we mainly revised the discussion of possible mechanisms for palaeo-EASM expansion during early Pliocene. Many thanks for spending so much time to review our manuscript. Wish you and yours have a happy Spring Festival in advance.

Judging by previous reviews, I lean towards trusting your geochemical and sedimentary analyses. However, I do have concerns about your discussion. I am worried that the discussion of mechanisms accounting for EASM expansion during early Pliocene was built upon misunderstanding of early Pliocene tropical SST records. The records cited in your discussion did not cover late Miocene, and hence should not be used to argue for expansion of monsoon starting from early Pliocene. The long records from western tropical Pacific actually suggest similar and warmer than present-day SSTs for both early Pliocene and late Miocene.

I recommend rewrite and shorten the discussion. Plesae stick with interpretations from sedimentary and geochemical features to monsoonal climate. The discussion of mechanisms reads very weak, and as shown above, erroneous at various places.

Our response: Many thanks for your valuable suggestions. We remove the statement of lines 488-498 and lines 546-570.

See details in the below:

Line 566 to 572, Brierley et al., 2009; Fedorov et al., 2013 did not suggest that equatorial SSTs remain stable or cooled. Instead, Brierley et al. suggests that warm pool was expanded during the early to mid-Pliocene. Zhang et al., (2014, science) actually suggests that western tropical Pacific was significantly warmer during the late Miocene and early Pliocene. The argument from line 566 and 575 is based on misunderstanding of published literatures.

Our response: We would remove this part.

Line 572 to line 575, the cited studies provide no theoretical basis on how the northern high latitude warming would affect ITCZ position. This argument again has no support.

Our response: We modify the statement as "On the other hand, the unusually warm Arctic and the West Antarctic ice-sheet expansion by 6–5 Ma (Zachos et al., 2001, 2008) steepened interhemispheric thermal gradient and could further cause the thermal equator to move northward (Chiang and Friedman, 2012; Broecker and Putnam, 2013). This facilitated the northwestward expansion of the palaeo-EASM, which is also proposed as the driving mechanism for northwestward migration of the monsoon rain belt for the warm Holocene (Yang et al., 2015)."

Line 582 to 584, again, misinterpretation of Brierley et al and Fedorov et al. Notice that the tropical Pacific records discussed in these papers DO NOT cover late Miocene for the most part. You CANNOT cite them to highlight the uniqueness of early Pliocene. In fact, based on

Zhang et al., 2012, the western tropical Pacific during late Miocene is not that different from early Pliocene.

Our response: We remove the statement.

Thanks for taking time to revise the manuscript! I am looking forward to your submission!

Other modifications

#1: In lines 40-42, the statement of "the poleward expansion of the tropical warm pool into subtropical regions, and the freshening of the subtropical Pacific" is removed.

#2: The statement of lines 488-498 is removed.

#3: Lines 533-536, the statement of "The warming of the northern high latitude region led to increases in summer temperature in the mid-latitudes of Eurasia. However, equatorial SSTs remained stable or cooled slightly (Brierley et al., 2009; Fedorov et al., 2013), and thus the land-ocean thermal contrast was intensified. Furthermore," is modified to "Furthermore, a decrease in the input of ice-rafted debris to the sediments of the subarctic northwest Pacific was synchronous with the expansion of the palaeo-EASM during the early Pliocene (Fig. 6). The warming of the Northern Hemisphere and".

#4: In line 537, "also" is removed.

#5: In line 546-548 "and small meridional heat gradient in the Northern Hemisphere pushed the Intertropical Convergence Zone northwards (Chang et al., 2013; Sun et al., 2015), which weakened the westerly circulation and thus facilitated the northwestward expansion of the palaeo-EASM" is modified to "and West Antarctic ice-sheet expansion by 6–5 Ma (Zachos et al., 2001, 2008) steepened interhemispheric thermal gradient and could further cause the thermal equator to move northward (Chiang and Friedman, 2012; Broecker and Putnam, 2013). This facilitated the northwestward expansion of the palaeo-EASM which is also proposed as the driving mechanism for northwestward migration of the monsoon rain belt for the warm Holocene (Yang et al., 2015)."

#6: The statement of lines 549-570 is removed.

#7: In line 571, "In conclusion" is modified to "Therefore".

#8: In line 572-574, "accompanied by the vast poleward expansion of the tropical warm pool into subtropical regions and the freshening of the subtropical Pacific," is modified to "in response to the closure of the Panamanian Seaway".

#9: In line 581, "and SSTs of the low latitude Pacific Ocean" is removed.

#10: In lines 595-597, "the vast poleward expansion of the tropical warm pool into subtropical regions, and the freshening of the subtropical Pacific," is removed.

#11: References of line635, line 698 and line702 are removed and "Broecker, W.S., Putnam A.E.: Hydrologic impacts of past shifts of Earth's thermal equator offer insight into those to be produced by fossil fuel CO2. Proc Natl Acad Sci USA 110(42):16710–16715, 2013. Chiang J. C. H., Friedman A. R.: Tropical cooling, interhemispheric thermal gradients, and tropical climate change. Annu Rev Earth Planet Sci 40(1):383–412, 2012. Yang, S.L., Ding, Z.L., Li, Y.Y., Wang, X., Jiang, W.Y., Huang, X.F.: Warming-induced northwestward migration of the East Asian monsoon rain

belt from the Last Glacial Maximum to the mid-Holocene. Proc. Natl. Acad. Sci. USA 112, 13178–13183, 2015. Zachos, J.C., Dickens, G.R., Zeebe, R.E., 2008. An early Cenozoic perspective on green-house warming and carbon-cycle dynamics. Nature.451, 279–283" are added.

#12: Fig 6 h and i are removed.

Hopefully the revised version is now satisfactory for publication in Climate of the Past.

Best regards,

Jijun Li

Late Miocene-Pliocene climate evolution recorded by the red clay covered on the Xiaoshuizi planation surface, NE Tibetan Plateau

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Abstract

The Pliocene climate and its driving mechanisms have attracted substantial scientific interest because of their potential as an analog for near-future climates. The late Miocene-Pliocene red clay sequence of the main Chinese Loess Plateau (CLP) has been widely used to reconstruct the history of interior Asian aridification and the Asian monsoon. However, red clay sequences deposited on the planation surface of the Tibetan Plateau (TP) are rare. A continuous red clay sequence was recently discovered on the uplifted Xiaoshuizi (XSZ) planation surface in the Maxian Mountains, northeastern (NE) TP. In this study, we analyzed multiple climatic proxies from the XSZ red clay sequence with the aim of reconstructing the late Miocene-early Pliocene climate history of the NE TP and to assess regional climatic differences between the central and western CLP. Our results demonstrate the occurrence of minimal weathering and pedogenesis during the late Miocene, which indicates that the climate was arid. We speculate that precipitation delivered by the palaeo- East Asian Summer Monsoon (EASM) was limited during this period, and that the intensification of the westerlies circulation resulted in arid conditions in the study region. Subsequently, enhanced weathering and pedogenesis occurred intermittently during 4.7-3.9 Ma, which attests to an increase in effective moisture. We ascribe the arid-humid climatic transition near ~4.7 Ma to the expansion of the palaeo-EASM. The warming of the high northern latitudes~~Increasing Arctic temperatures, the poleward expansion of the tropical warm pool into subtropical regions, and the freshening of the subtropical Pacific~~ in response to the closure of the Panamanian Seaway, may have been responsible for the thermodynamical enhancement of the palaeo-EASM system, which permitted more moisture to be transported to the NE TP.

Keywords: Late Miocene-Pliocene; Xiaoshuizi Planation Surface; Red Clay; Palaeo-EASM; Westerly Circulation

1. Introduction

The Pliocene, including the Zanclean (5.33-3.60 Ma) and Piacenzian (3.60-2.58 Ma) stages, is one of the most intensively studied intervals of the pre-Quaternary in climate change research. The Zanclean climate was generally warm and wet and is often used as an analogue for near-future climate conditions in terms of carbon dioxide levels, ranging from 280-415 ppm (Tripati et al., 2009; Pagani et al., 2010), and comparable temperatures in the tropical region (Herbert et al., 2010, 2016). On the other hand, the Zanclean was markedly different from today, although several critical changes in thermohaline and atmospheric circulation towards modern conditions were occurring (Haug et al., 2005; Lawrence et al., 2006; Chaisson and Ravelo, 2000). For example, the early-Pliocene global mean temperature was approximately 4 ℃ warmer (Brierley and Fedorov, 2010), and the sea levels are estimated to have been ~25 m higher, than today (Dowsett et al., 2010). Temperatures at high northern latitudes were considerably higher and therefore continental glaciers were almost absent from the Northern Hemisphere (Ballantyne et al., 2010; Dowsett et al., 2010). The zonal and meridional sea surface temperature gradients in the Northern Hemisphere were weak but gradually became more intensified, changing towards the modern state which has a much more pronounced spatial temperature contrast (Fedorov et al., 2013; Brierley et al., 2009, 2010). The low meridional surface temperature gradient

resulted in weaker meridional circulation during this interval (Fedorov et al., 2013; Brierley et al., 2009), and the minor east-west sea surface temperature contrast in the tropical Pacific during this interval is believed to have given rise to a permanent El Nino Southern Oscillation (Lawrence et al., 2006); however, whether permanent El Nino–like conditions were sustained during the Pliocene is controversial (Wara et al., 2005; Watanabe et al., 2011; Zhang et al., 2014). In addition, the episodic uplift of the TP (Li et al., 2015; Zheng et al., 2000; Fang et al., 2005a, 2005b) and gradual closure of the Panama Seaway (Keigwin et al., 1978; O'Dea et al., 2016) were underway. The former had a substantial climatic impact (An et al., 2001; Ding et al., 2001; Liu et al., 2014) and the latter resulted in the reorganization of the global thermohaline circulation system (Haug et al., 1998, 2001). These features imply a spatial change in the organization of the global climate system from the early Pliocene to the present. In this context, it is important to characterize the response of regional climates to these major global climatic and tectonic changes.

East Asia is one of the key regions for studying the aridification of the Asian interior and the Asian monsoon evolution, which are tightly linked to the uplift of the TP, regional climate change, and the evolution of global temperature and ice volume (An et al., 2001; Ding et al., 2001; Li et al., 2008; Clift et al., 2008; Nie et al., 2014; Ao et al., 2016; Sun et al., 2006a, 2017; Chang et al., 2013; Liu et al., 2014). Previous research has revealed that red clay was widely deposited across the CLP since the late Miocene, indicating that Asian aridification was enhanced (Guo et al., 2001; Song et al., 2007; An et al., 2014; Ao et al., 2016; Li et al., 2017). In the eastern and central CLP, where the climate is dominated by the East Asian Monsoon, palaeontological evidence, mineral magnetic parameters and

geochemical records from the red clay indicate dry climatic conditions during the late Miocene but generally wet climatic conditions during the early Pliocene (Wang et al., 2006; Guo et al., 2001; Wu et al., 2006; Song et al., 2007; Sun et al., 2010; An et al., 2014; Ao et al., 2016). The most controversial climatic change occurred during the interval from 4.8-4.1 Ma, for which climate reconstructions using different proxies indicate conflicting palaeo-environmental trends. For example, field observations and pollen records indicate an intensified summer monsoon intensity, but low magnetic susceptibility values are more consistent with arid rather than wet climatic conditions (Ding et al., 2001; Ma et al., 2005; Song et al., 2007; Sun et al., 2010). It is thought that dissolution of ferrimagnetic minerals and iron reduction resulting from high precipitation significantly affected the climatic significance of magnetic susceptibility records during this period (Ding et al., 2001). In addition to the East Asian Monsoon, the westerlies also had an impact on the climate of East Asia; however, the patterns of climate change in the westerlies-dominated regions were different from the eastern and central CLP during the early Pliocene. Geochemical, stratigraphic and pollen evidence from the Qaidam and Tarim basins has demonstrated that aridification intensified since the early Pliocene (Fang et al., 2008; Sun et al., 2006a, 2017; Chang et al., 2013; Liu et al., 2014). Although the general climatic trends of the main CLP and central Asia during this period are well recorded, palaeoclimatic changes in the NE TP, which is at the junction of the zones of westerlies and monsoonal influences, remain unclear. Therefore, determining the climatic conditions of the NE TP during the early Pliocene not only improves our understanding of the pattern of regional climate change, but it may also provide insights into the responses of the palaeo-EASM and the westerlies to TP uplift and

changes in the global climate system.

A continuous red clay sequence was recently discovered on the uplifted XSZ planation surface in the NE TP and has been dated via high-resolution magnetostratigraphy (Li et al., 2017). Due to its specific geographical location, the XSZ red clay provides the opportunity to reveal the late Miocene-early Pliocene climate history of the NE TP and to determine the climatic differences between the central and western CLP. In this study, we measured multiple climatic proxies from the late Miocene-Pliocene XSZ red clay core. Our aims were to construct a detailed record of precipitation, chemical weathering and pedogenesis during 6.7-3.6 Ma; and to determine the pattern of regional climate evolution and its possible causal mechanisms.

2. Regional background

The XSZ planation surface is located in Yuzhong County in the western Chinese Loess Plateau (Fig. 1). The main XSZ planation surface is at an altitude of 2800 m in the Maxian Mountains where it has truncated Precambrian gneiss. The Maxianshan are rejuvenated mountains which protrude into the broad Longzhong Basin; they are located within a climatically sensitive zone because of the combined influences of the Asian Monsoon and the northern branch of the mid-latitude westerly circulation system. The planation surface is mantled by over 30 m of loess and over 40 m of red clay. Our previous bio-magnetostratigraphic study demonstrates that the red clay sequence covering the XSZ planation surface is dated to ~6.9-3.6 Ma (Li et al., 2017). Here, we use the XSZ drill core to reconstruct and discuss the patterns of regional climate change during the Miocene-Pliocene.

The long, continuous well-dated record of the drill core is superior to that of the Shangyaotan core analyzed in Li et al. (2017). Yuzhong County lies within the semi-arid temperate climate zone at the junction of the eastern monsoon area, the arid area of northwest China, and the cold region of the TP. The mean annual temperature during 1986-2016 was ~7.0 ℃ and the annual precipitation was 260-550 mm; 80% of the precipitation is in summer and autumn (data source: National Meteorological Information Center (http://data.cma.cn/) of the Chinese Meteorological Administration). The spatial distribution of precipitation is uneven, decreasing from south to north in Yuzhong County. Precipitation amount increases with elevation at the rate of 27 mm per 100 m, attaining a maximum of 800 mm at the top of Maxianshan.

3. Material and methods

The XSZ core (35.8115 °N, 103.8623 °E and 2758.1 m above sea level) is composed of 42 m of pure red clay and ~3 m of red clay and there is an increasing content of angular gravel at the base (Fig. 1 b). The red clay interval is composed of brownish red and yellowish clay layers (Fig. 2). The upper 20 m contains numerous horizontal carbonate nodule horizons (Bk), most of which underlie brownish red soil layers (Bw) characterized by loam and moderate medium angular blocky structure. There are also occasional carbonized plant root channels, elliptical worm burrows and fossil snail shell fragments. Fe-Mn stains are more frequent in the brownish layers than in the yellowish layers, which is also the case for the carbonized root channels. The red clay across the XSZ planation surface is similar to that of typical eolian red clay in the CLP; both are characterized by numerous carbonate nodule-rich

154 horizons (Fig. 2 b).

155 Samples for grain-size, carbonate content and magnetic susceptibility measurements
156 were taken at 5-cm intervals, and samples for geochemical analysis were collected at 25-cm
157 intervals. Samples for grain-size measurements were pre-treated with 10% H_2O_2 to remove
158 organic material, with 10% HCl to remove carbonates, and with 0.05 mol/L of $(NaPO_3)_6$ for
159 dispersion. They were then measured with a Malvern Mastersizer 2000 grain-size analyzer
160 with a detection range of 0.02-2000 μm. Magnetic susceptibility was measured using a
161 Bartington Instruments MS2 meter and MS2B dual-frequency sensor at two frequencies (470
162 Hz and 4700 Hz, designated χ_{lf} and χ_{hf}, respectively). Three measurements were made at each
163 frequency and the final results were averaged. The frequency-dependent magnetic
164 susceptibility (χ_{fd}) was calculated as $\chi_{lf}-\chi_{hf}$. Chemical composition was measured via X-ray
165 fluorescence using a Panalytical Magix PW2403 with an error of 0.1%-0.3%. The sample
166 preparation procedure for XRF analysis was as follows: Bulk samples were heated to 35℃
167 for 7 days and then ground with an agate mortar to pass a 75-μm sieve; ~4 g of powdered
168 sample was then pressed into a pellet with a borate coating using a semiautomatic oil-
169 hydraulic laboratory press (model YYJ-40). All the measurements were conducted at the
170 MOE Key Laboratory of Western China's Environmental Systems, Lanzhou University.
171 Silicate-bound CaO (CaO*) can be estimated, in principle, by the equation: CaO*(mol) =
172 CaO(mol) – CO_2(calcite mol) − 0.5 CO_2(dolomite mol) − 10/3 P_2O_5(apatite mol) (Fedo et al.,
173 1995). It is generally calculated based on the assumption that all the P_2O_5 is associated with
174 apatite and all the inorganic carbon is associated with carbonates. Thus, the CaO* of the XSZ
175 red clay was calculated using the following equivalent equation:

176 $$CaO^*(mol) = CaO(mol) - CaCO_3(mol) - \frac{10}{3} * \frac{P_2O_5}{M(P_2O_5)}$$

177 The carbonate content was measured with a calcimeter using the volumetric method of
178 Avery and Bascomb (1974) in the Key Laboratory of Mineral Resources in Western China
179 (Gansu Province), Lanzhou University.

180 We used the coefficient of variation (CV) to measure the variability of the records: the
181 higher the CV, the more variable the record. The CV is defined as:

$$CV = 100 * \frac{Standard\ deviation}{Mean}$$

182 Each sample age was estimated using linear interpolation to derive absolute ages,
183 constrained by our previous magnetostratigraphic study (Fig. 1). The average temporal
184 resolution of the records is 3.8 kyr. Some 80 % of the sequence has a sampling resolution of 4
185 kyr or less. After interpolation to a 3-kyr sampling interval, we performed spectral analysis
186 on detrended records of carbonate content and χ_{pedo} using Redfit, based on the Lomb-Scargle
187 Fourier transform combined with a Welch-Overlapped Segment averaging procedure. We
188 applied Gaussian band-pass filters at frequencies of 0.09090-0.01111, 0.02174-0.02778 and
189 0.04167-0.05556 kyr^{-1} to extract oscillations associated with the 100-kyr, 41-kyr and 21-kyr
190 periodicities, respectively. The significance of the correlations is based on a two-tailed test.

191 **4. Results**

192 Profiles of the various environmental proxies are illustrated in Figure 3. Notably, there is
193 evidence for a relatively wet interval from ~16-5 m (4.7-3.9 Ma) which is reflected in the
194 high-frequency occurrence of Bw horizons with a low carbonate content (< 8 %) and
195 intermittent enhancement of magnetic susceptibility. There is a large contrast in carbonate
196 content between Bw and Bk horizons, which corresponds to variations in elemental contents.

The Bk horizons, with a higher carbonate content, consist of carbonate nodule layers underlying leached zones in the field indicate the substantial translocation of carbonate minerals from Bw horizons to Bk horizons due to greater rainfall (He et al., 2013). In addition, the CV of most of the records is greater during this interval than in other intervals (Table 1). These various forms of evidence suggest that the climate became more humid and variable during 4.7-3.9 Ma. The characteristics of the individual proxy records are described in detail below.

Carbonate content

The carbonate content of the entire core fluctuates from 1.6-39.2% with an average of 15.9 %. From 42-16 m, the average carbonate content is high (17.1%) and the carbonate content decreases upwards. The contrast in the carbonate content between the Bw and Bk horizons is generally low; for the Bw horizons, the carbonate content is ~12% and values <8% are rare. Bk horizons, with a carbonate content of around or above 21%, are frequent (Fig. 3). From 16-5 m, there are fluctuations in carbonate content of large amplitude (1.6-39.1%) but the average value is low (13.3%). Bw-Bk horizons are frequent; the Bw horizons have a carbonate content of <8%, while that of the Bk horizons is >21%. From 5-0 m, the average carbonate content increases to 15.5%; Bw horizons with a carbonate content <8% is absent, and the carbonate content contrast between the Bw and Bk horizons is low.

Element geochemistry

K_2O ranges from 1.9-3.7% with an average of 2.8%; Na_2O ranges from 0.14-1.54% with an average of 1.2%; Rb ranges from 74-134 ppm with an average of 106.2 ppm; and Sr ranges from 141-281 ppm with an average of 212.8 ppm. The variations in CaO exhibit the

same trend as carbonate content with high values in Bk horizons and low values in Bw horizons. The variations in Rb and K_2O are synchronous and roughly inverse to those of CaO. The changes of Sr show some similarity with magnetic susceptibility prior to 4.7 Ma but with CaO after 4.7 Ma. Reference to Table 2 shows that CaO is positively correlated with $CaCO_3$ and Sr, and negatively correlated with the other elements. From 16-5 m, CaO and Sr exhibit low values in Bw horizons and high values in Bk horizons, while the opposite is the case for K_2O and Rb. Finally, from 16-5 m, the amplitudes of the fluctuations in CaO, K_2O, Sr and Rb are greater than in the other intervals.

Magnetic susceptibility

The variations of χ_{hf}, χ_{lf} and χ_{fd} are synchronous. χ_{hf} ranges from 9.6-53.9$\times10^{-8}$ m^3/kg with an average of 21.8$\times10^{-8}$$m^3$/kg; χ_{lf} ranges from 11.4-59.0$\times10^{-8}$ m^3/kg with an average of 23.1$\times10^{-8}$ m^3/kg; and χ_{fd} ranges from 0-4.7$\times10^{-8}$ m^3/kg with an average of 1.2$\times10^{-8}$ m^3/kg. From 42-16 m, the three magnetic parameters are relatively low and uniform. χ_{hf} ranges from 9.6-33.3$\times10^{-8}$ m^3/kg with an average of 19.4$\times10^{-8}$$m^3$/kg; χ_{lf} ranges from 11.4-36.1$\times10^{-8}$ m^3/kg with an average of 20.3$\times10^{-8}$ m^3/kg; and χ_{fd} ranges from 0-2.8$\times10^{-8}$ m^3/kg with an average of 1.0$\times10^{-8}$ m^3/kg. From 16-5 m, the values of the three parameters, together with their amplitudes of variation, are high. χ_{hf} ranges from 13.8-53.9$\times10^{-8}$ m^3/kg with an average of 27.4$\times10^{-8}$ m^3/kg; χ_{lf} ranges from 14.2-59.0$\times10^{-8}$ m^3/kg with an average of 29.0$\times10^{-8}$ m^3/kg; and χ_{fd} ranges from 0-4.7$\times10^{-8}$ m^3/kg with an average of 1.6$\times10^{-8}$ m^3/kg. Within the intervals of 16-15 m, 13-11 m and 7-5 m, the values of the three parameters increase substantially. From 5-0 m, both the values and amplitudes of variation of the three parameters decrease. χ_{hf} ranges from 12.8-32.9$\times10^{-8}$ m^3/kg with an average of 22.0$\times10^{-8}$ m^3/kg; χ_{lf} ranges from 13.6-

241 34.6×10^{-8} m^3/kg with an average of 22.9×10^{-8} m^3/kg; and χ_{fd} ranges from 0-2.5×10^{-8} m^3/kg
242 with an average of 1×10^{-8} m^3/kg. Overall, the fluctuations in magnetic susceptibility are
243 substantially different to those of carbonate content which indicates that the enhancement of
244 magnetic susceptibility was not caused by carbonate leaching.

245 **Grain size**

246 The clay content (<2 μm) ranges from 3.8-13.5% with an average of 8.17%; and the >40
247 um content ranges from 0.7-13.9% with an average of 6%. The fluctuations in clay content
248 are minor, except for maxima at about 15 m, 12 m and 6 m, which correspond to peaks in
249 magnetic susceptibility (Fig. 3). The coarse silt component (>40 μm), mainly carried by the
250 East Asian winter monsoon, exhibits a different trend to that of the clay content. In addition,
251 from 21-5 m the fluctuations in the >40 um fraction are roughly the inverse to those of
252 magnetic susceptibility. From 42-21 m, the variation of the >40 μm fraction is characterized
253 by low values and high-frequency fluctuations, whereas above 21 m it exhibits high values
254 and fluctuations of lower frequency.

255

256 **5. Discussion**

257 **5.1 Palaeoenvironmental interpretation of the proxies**

258 The carbonate content of aeolian sediments can be readily remobilized and deposited in
259 responses to changes in precipitation and evaporation intensity and thus is sensitive to
260 changing climatic conditions. Previous studies demonstrated that the carbonate content of
261 loess-red clay sequences of the CLP varies with precipitation (Fang et al., 1999; Sun et al.,
262 2010). The carbonate is mainly derived from a mixture of airborne dusts (Fang et al., 1999).

Soil micromorphological evidence from the Lanzhou loess demonstrates that the carbonate grains in loess are little altered, whereas those in the palaeosols have undergone a reduction in size as a result of leaching and reprecipitation as secondary carbonate in the lower Bk horizons (Fang et al., 1994, 1999). Furthermore, seasonal alternations between wet and dry conditions are thought to be a key factor driving carbonate dissolution and reprecipitation (Sun et al., 2010). Thus, changes in carbonate content are generally controlled by the effective precipitation. When effective precipitation is high, carbonate leaching increases, and vice versa. Thus, the carbonate content is an effective proxy for characterizing wet-dry oscillations as well as summer monsoon evolution (Fang et al., 1999; Sun et al., 2010).

Chemical weathering intensity is generally evaluated by the ratio of mobile (e.g. K, Ca, Sr and Na) to non-mobile elements (e.g. Al and Rb). In general, Sr shows analogous geochemical behavior to Ca and is readily released into solution and mobilized in the course of weathering; by contrast, Rb is relatively immobile under moderate weathering conditions due to its strong adsorption to clay minerals (Nesbitt et al., 1980; Liu et al., 1993). Thus, the Rb/Sr ratio potentially reflects chemical weathering intensity. However, Sr may substitute for Ca in carbonates which may limit the environmental significance of the Rb/Sr ratio (Chang et al., 2013; Buggle et al., 2011). The correlation between Sr and CaO* (silicate CaO) is significant at the 99% confidence interval, while the correlation between Sr and $CaCO_3$ is not significant. This means that the variations in Sr are determined by weathering intensity, and therefore we speculate that in our samples the Rb/Sr ratio mainly reflects weathering intensity (Fig. 4 c and d). In addition, the K_2O/Na_2O ratio is used to evaluate the secondary clay content in loess and is also a measure of plagioclase weathering, avoiding biases due to

uncertainties in separating carbonate Ca from silicate Ca (Liu et al., 1993; Buggle et al., 2011). Na_2O is mainly produced by plagioclase weathering and is easily lost during leaching as precipitation increases. By contrast, K_2O (mainly produced by the weathering of potash feldspar) is easily leached from primary minerals and is then absorbed by secondary clay minerals with ongoing weathering (Yang et al., 2006; Liang et al., 2013). In the arid and semi-arid regions of Asia, K_2O is enriched in palaeosols compared to loess horizons (Yang et al., 2006). Thus, high K_2O/Na_2O ratios are indicative of intense chemical weathering.

In the red clay-loess sequence of the CLP, magnetic parameters and the clay content are well correlated and thus are regarded as proxies of EASM strength (Liu et al., 2004). Aeolian particles usually have two distinct magnetic components, consisting of detrital and pedogenic material, respectively (Liu et al., 2004). χ_{lf} can reflect the combined susceptibility of both components, but changes in χ_{lf} are mainly affected by changes in the concentration of pedogenic magnetic grains (Liu et al., 2004). The grain-size distribution of pedogenic particles within the superparamagnetic (SP) to single-domain (SD) size range has been shown to be constant (Liu et al., 2004, 2005). Thus, χ_{fd} can be used detect SP minerals produced by pedogenesis and therefore the correlation coefficient between χ_{lf} and χ_{fd} is a measure of the contribution of such grains (<0.03 μm for magnetite) to the bulk susceptibility (Liu et al., 2004; Xia et al., 2014). As shown in Figure 4a, χ_{lf} is positively correlated with χ_{fd}, which means that the magnetic susceptibility of the XSZ red clay mainly reflects pedogenic enhancement of the primary aeolian ferromagnetic content via the in-situ formation of fine-grained ferrimagnetic material. Thus, the magnetic susceptibility primarily reflects pedogenic intensity. Both the original and pedogenic magnetic signals can be separated using a simple

linear regression method (Liu et al., 2004; Xia et al., 2014), which we use to extract the lithogenic (χ_0) and pedogenic magnetite/maghemite (χ_{pedo}) components. We found that pedogenic magnetite/maghemite accounts for 11% of the susceptibility ($\chi_{pedo} = \chi_{fd} / 0.11$).

Pedogenesis results in enhanced secondary clay formation (Sun and Huang, 2006b); however, not all of the clay particles are derived from in situ pedogenesis, but rather are inherited from aeolian transport and deposition. Clay particles can adhere to coarser silt and sand particles (Sun and Huang, 2006b). In the western CLP, the coarse silt (>40 μm) content is regarded as a rough proxy of winter monsoon strength (Wang et al., 2002). Therefore, to eliminate this signal from the primary clay particles, the <2 μm/>40 μm ratio is proposed to evaluate pedogenic intensity. Furthermore, the similarity of the variations of <2 μm/>40 μm ratio and χ_{pedo} confirms that in this case <2 μm/>40 μm ratio has the potential to evaluate the pedogenic intensity (Fig. 6).

5.2 Time- and frequency- domain analysis of carbonate content and χ_{pedo}

The power spectral analyses of carbonate content and χ_{pedo} show different dominant cycles (Fig. 5 a-b). In detail, χ_{pedo} is concentrated in the eccentricity (100 kyr), obliquity (41 kyr) and precession (21 kyr) bands and other periodicities (71 kyr and 27 kyr) are also evident. By contrast, the carbonate signal is concentrated in the precession (21 kyr) and obliquity (41 kyr) bands, but it also exhibits even more prominent periodicities of 56 kyr and 30 kyr. Furthermore, the fluctuations in $CaCO_3$, weathering and pedogenesis indices agree well with orbital eccentricity variations during 4.7-3.9 Ma (Fig. 5 d). Three orbital periodicities were also detected at other sites in CLP, in the interval from the late Miocene to the early Pliocene, confirming that changes in orbital parameters had a substantial impact of

the climate of the CLP (Han et al., 2011).

King (1996) proposed that non-orbital cycles may originate from harmonic effects or interactions of the orbital cycles, while Lu (2004) ascribed them to unstable dust depositional processes followed by varying degrees of pedogenesis in palaeosol units. In the XSZ section, the deposition rate is low and uneven, which potentially resulted in the incomplete preservation of the paleoclimatic signal, especially for the relatively short precession cycles. In addition, pedogenesis and post-depositional compaction would also weaken the orbital signals and produce spurious cycles. Moreover, the carbonate content at various depths is affected by leaching which means that the record integrates soil polygenetic processes, thus obscuring orbital forcing trends related to precipitation amount. Therefore, we speculate that uneven and low deposition rates, combined with compaction and leaching processes, may have weakened the orbital signals and may be responsible for the presence of non-orbital cycles in the XSZ section.

To investigate the post-6.7 Ma frequency domain evolution of the climate signals in the XSZ section, we filtered the carbonate content and χ_{pedo} time series at the periods of 100, 41, and 21 kyr, using Gaussian band filters centered at frequencies of 0.01, 0.02439, and 0.04762, respectively. We then compared the results with the equivalent filtered components of the stacked deep-sea benthic oxygen isotope record. The results show that the fluctuations of the three filtered components (especially the 41-kyr component) of both proxies change from a low amplitude during 6.7-4.7 Ma to a relatively high amplitude during 4.7-3.9 Ma (Fig. 5 c). The enhanced orbital-scale variability of the two proxies from 4.7-3.9 Ma implies increased seasonality and wet-dry contrasts. This shift is not observed in the Earth orbital parameters

but is observed in the filtered 41-kyr component of the stacked deep-sea benthic oxygen isotope record ($\delta^{18}O$). This may mean that the enhancement of wet-dry contrasts at the XSZ site was not driven directly by changes in solar radiation intensity but rather was linked with changes in ice volume or global temperature.

5.3 Late Miocene-Pliocene climate history revealed by the Xiaoshuizi red clay

5.3.1 Multi-proxy evidence for a dry climate during the late Miocene

We used the proxies of pedogenesis and chemical weathering to reconstruct the late Miocene and early Pliocene climatic history of the Xiaoshuizi planation surface. During the late Miocene, the relatively high carbonate values with minor fluctuations indicate that the climate was dry, and low Rb/Sr and K_2O/Na_2O ratios also support the occurrence of weak chemical weathering. Notably, both the Rb/Sr and K_2O/Na_2O ratios show opposite trends to that of carbonate content, meaning that low effective precipitation resulted in weak chemical weathering. Furthermore, the pedogenic proxies (χ_{pedo} and χ_{lf}), characterised by low values with minor fluctuations, generally support the occurrence of weak pedogenesis under an arid climate. Thus, the climate at the XSZ site was relatively arid during this interval, resulting in weak chemical weathering and pedogenic intensity. However, there are several subtle differences between the carbonate and pedogenic indexes. It is evident that the carbonate content decreases with an increased amplitude of variation after 5.5 Ma, which is consistent with the cycles of carbonate nodules within paleososol horizons observed in the field (Li et al., 2017). It is possible that increased precipitation since 5.5 Ma induced eluviation and the redeposition of carbonate. However, the pedogenic indexes indicate that the generally arid climate was interrupted by two episodes of enhanced pedogenesis, at 5.85-5.7 Ma and 5.5-

5.35 Ma. The subtle differences may result from differences in the sensitivity of magnetic susceptibility and carbonate content to precipitation variability when precipitation is low (Sun et al., 2010). In addition, a coeval mollusk record from the western Liupanshan showed that cold-aridiphilous species dominated, which also indicates that cold and dry climatic conditions occurred in the western CLP during the late Miocene (Fig. 7 g).

Coeval pollen, mollusk and magnetic records from the central and eastern CLP also indicate generally dry and cold climatic conditions (Wang et al., 2006; Wu et al., 2006; Nie et al., 2014). However, the principal difference is that at the XSZ site, the arid climate was relatively stable, while the climate of the central and eastern CLP was interrupted by several humid stages. For example, two humid stages (6.2-5.8 Ma and 5.4-4.9 Ma) are recorded by the magnetic susceptibility of the red clay in the central and eastern CLP but are absent in the magnetic susceptibility record at the XSZ site (Fig. 7). Notably, the 41-kyr filtered component of thermo-humidiphilous species from Dongwan was damped in the late Miocene (Li et al., 2008). Similarly, the amplitude of the orbital periodicities, filtered from the XSZ carbonate content and χ_{pedo} records, was obviously damped during 6.7-4.7 Ma. However, the three periodicities in the Summer Monsoon index from the central CLP show no obvious difference between the late Miocene and Pliocene, but only a slight reduction in variability after 4.2 Ma (Sun et al., 2010). Therefore, we agree that a dry climate prevailed on the CLP during the late Miocene; however, the difference was that the climate in the central and eastern CLP fluctuated more substantially than was the case in the vicinity of the XSZ red clay section.

The especially damped response of the wet-dry climatic oscillations in the western CLP

395 to obliquity forcing may indicate that the influence of the palaeo-EASM in the western CLP
396 was negligible. It is widely known that the summer monsoon intensity decreases from
397 southeast to northwest across the CLP. A regional climate model experiment demonstrated
398 that the modern East Asian Summer Monsoon was not fully established in the late Miocene
399 and had only a small impact on northern China (Tang et al., 2011). A weak palaeo-EASM
400 intensity from 7.0-4.8 Ma was revealed by hematite/goethite and smectite/kaolinite ratios at
401 ODP Site 1148 in the South China Sea (SCS) (Fig. 7 i and j). Therefore, we infer that the
402 palaeo-EASM was weak and had only a minor impact on the climate in the study region. In
403 addition, previous studies indicated that the red clay may have been transported by both low-
404 level northerly winds and the upper-level westerlies (Sun et al., 2004; Vandenberghe et al.,
405 2004) and thus the impact of the westerly circulation on the study region cannot be ignored.
406 Notably, the variation of the pedogenic proxies roughly parallels that of the stacked deep-sea
407 benthic foraminiferal oxygen isotope curve (Fig. 6), and that that χ_{pedo} has a positive
408 relationship with $\delta^{18}O$ (Fig. 4 e). This indicates that when the global temperature was low,
409 pedogenic intensity in the study area increased. It is unreasonable to conclude that
410 precipitation in the study area was dominated by the palaeo-EASM and thus we speculate that,
411 during the late Miocene, precipitation transported by the palaeo-EASM was limited and that
412 the westerly circulation probably dominated the regional climate.

413 The simultaneous reduction in amplitude of the 41-kyr filtered components from the
414 western CLP and the deep sea $\delta^{18}O$ record during the late Miocene likely indicates that the
415 dry climate was related to changes in global temperature and ice volume. A sustained cooling
416 occurred in both hemispheres during the late Miocene which culminated between 7 and 5.4

417 Ma (Herbert et al., 2016). $\delta^{18}O$ records from DSDP and ODP sites show an increase of ~1.0‰
418 during the late Miocene which resulted from the increased ice volume and the associated
419 decrease in global temperature (Zachos et al., 2001). In the Northern Hemisphere, transient
420 glaciations occurred when the cooling culminated (Herbert et al., 2016). Records from high-
421 latitude regions of the Northern Hemisphere indicate continuously decreasing temperatures
422 and increasing ice volume during the late Miocene (Jansen and Sjøholm, 1991; Mudie and
423 Helgason, 1983; Haug et al., 2005). During the Quaternary, a dry climate prevailed during
424 glacial periods when the global average temperature (especially in summer) was low. Cool
425 summers could result in a small land-sea thermal contrast which in turn weakened the palaeo-
426 EASM. Furthermore, the increased ice volume in the Northern Hemisphere resulted in an
427 increased meridional temperature gradient (Herbert et al., 2016), thus strengthening the
428 westerlies and driving them southwards. This would have prevented the northwestward
429 penetration of the Asian Summer Monsoon, which is also proposed as the driving mechanism
430 for a weak EASM in northern China during glacial periods (Sun et al., 2015). Thus, the
431 southward shift of the westerlies had a significant impact on the XSZ region. However,
432 moisture sources for the westerly air flow are distant from the CLP (Nie et al., 2014), and
433 only a relatively small amount of moisture was carried to the CLP, resulting in a dry and
434 stable climate in the XSZ region. In conclusion, global cooling and increasing ice volume in
435 the Northern Hemisphere contributed to the dry climatic conditions in the study region.

436 **5.3.2 Intermittently humid climate during the early Pliocene**

437 During the early Pliocene, the proxy evidence indicates that the previously arid climate
438 of the XSZ area became humid from ~4.7 Ma. The carbonate content was low on average but

with large fluctuations, indicating that the climate was generally humid with increased dry-wet oscillations, especially during 4.7-3.9 Ma. Several eluvial-illuvial cycles are evident during 4.7-3.9 Ma; the carbonate content in the eluvial horizons was less than 8%, whereas in illuvial horizons it was >21% (Fig. 6). Research on the migration process of carbonate indicate that a seasonally wet/dry climate is a key factor in driving carbonate dissolution and reprecipitation, and strong seasonally-biased precipitation enhances the leaching process and produces thick leached horizons (Rossinsky and Swart, 1993; Zhao, 1995, 1998). The occurrence of high-frequency cycles of carbonate eluviation-redeposition indicates that seasonal precipitation increased during this interval. Furthermore, the variations of Rb/Sr and K_2O/Na_2O ratios are very similar to those of carbonate content, which suggests that weathering intensity was related to precipitation amount. Generally, high values of the <2 μm / >40 μm ratio, χ_{pedo} and χ_{lf} correspond to large contrasts in carbonate content between eluvial and illuvial horizons; thus, increased precipitation had a significant influence on pedogenic intensity. Seasonal precipitation was intermittently enhanced from 4.7-3.9 Ma, and so was weathering and pedogenic intensity. Pedogenesis and weathering intensity reached a maximum during 4.60-4.25 Ma, as did precipitation intensity, manifested by the enhanced eluviation and carbonate accumulation. Notably, during this interval of peak precipitation (4.6-4.25 Ma), the enhancement of the <2 μm />40 μm ratio is not as strong as that of χ_{pedo}, which may indicate that the former is of limited value when pedogenic intensity is strong. During 3.9-3.6 Ma, precipitation decreased, and weathering and pedogenic intensity also weakened. Consistent with the records of the XSZ section, mollusk records from Dongwan also indicate the occurrence of warm and humid conditions in the western CLP during the

461 early Pliocene (Fig. 7 h).

462 Palynological and terrestrial mollusk records from the central CLP also indicate
463 relatively humid conditions during the early Pliocene (Wang et al., 2006; Wu et al., 2006).
464 Magnetic susceptibility records from the central and eastern CLP are similar to that from the
465 XSZ section in that both the magnitude and the variability are high during the early Pliocene.
466 From 4.1-3.9 Ma, the increased magnetic susceptibility indicates that humid climatic
467 conditions prevailed across the entire CLP (Fig. 7). Evidently, when precipitation amount
468 peaked in the vicinity of the XSZ section during 4.60-4.25 Ma, the magnetic susceptibility
469 values at Xifeng, Lingtai and Chaona were low. However, a record of Fe_2O_3 ratio from
470 Lingtai reveals extremely high values, corresponding to the presence of abundant clay
471 coatings, during 4.8-4.1 Ma and this interval was interpreted as experiencing the strongest
472 EASM intensity in the CLP since 7.0 Ma (Ding et al., 2001). In addition, the relative intensity
473 of pedogenic alteration of the grain-size distribution was the strongest during the interval
474 from 4.8-4.2 Ma in the Lingtai section (Sun et al., 2006c). Pollen assemblages at Chaona
475 indicate a substantially warmer and more humid climate from 4.61-4.07 Ma (Ma et al., 2005).
476 These various lines of evidence indicate that during 4.60-4.25 Ma the climate was warm and
477 humid in the central CLP. Gleying has been implicated in reducing the value of magnetic
478 susceptibility as a record of precipitation during this period (Ding et al., 2001). When soil
479 moisture regularly exceeds the critical value, dissolution of ferrimagnetic minerals occurs and
480 the susceptibility signal is negatively correlated with pedogenesis (Liu et al., 2003). This
481 alone indicates that precipitation was likely to have been very high during this interval.

482 In summary, a wet climate prevailed across the CLP in the early Pliocene. At the same

time, the hematite/goethite ratio in the sediments of the South China Sea also indicates enhanced precipitation amount and the smectite/kaolinite ratio indicates increased seasonality at ~4.7 Ma (Fig. 7 i and j) and thus the enhancement of the palaeo-EASM (Clift et al., 2006, 2014). Therefore, we regard the climatic change evident in XSZ section to reflect the expansion of the palaeo-EASM.

~~The remarkably increased amplitude of the 41-kyr filtered components from the XSZ section and the deep sea $\delta^{18}O$ record at about 4.7 Ma indicates that the expansion of the palaeo-EASM may have been related to changes in global temperature and ice volume. Furthermore, a decrease in the input of ice-rafted debris to the sediments of the subarctic northwest Pacific was synchronous with the expansion of the palaeo-EASM during the early Pliocene (Fig. 6). In addition, from 4.8-4.7 Ma and 4.6-4.25 Ma, the high values of the three pedogenic indices at the XSZ section indicate that strong pedogenic intensity corresponded with high SSTs in the Eastern Equatorial Pacific (EEP). This coherence between the record of the XSZ section and marine records implies that phases of enhanced precipitation were correlative with changes in SST and ice volume (or temperature) at northern high latitudes.~~

~~5.4 Possible driver of palaeo-EASM expansion during the early Pliocene~~

Ding (2001) proposed that the uplift of the TP to a critical elevation resulted in an enhanced summer monsoon system during 4.8-4.1 Ma. TP uplift was shown to have had profound effects on the EASM in terms of its initiation and strength, as well as in changing the distribution of the band of high precipitation in East Asia (Li et al., 1991, 2014; An et al., 2001). A detailed modeling study demonstrated that the uplift of the northern TP mainly resulted in an intensified summer monsoon and increased precipitation in northeast Asia

505 (Zhang et al., 2012). From 8.26-4.96 Ma, massive deltaic conglomerates were widely
506 deposited and the sediment deposition rate increased, indicating the uplift of the Qilian
507 Mountains (Song et al., 2001). At the same time, the Laji Mountains underwent pronounced
508 uplift by thrusting at ~8 Ma, which resulted in the current basin-range pattern (Li et al., 1991;
509 Fang et al., 2005a; Zheng et al., 2000). However, geological and palaeontological records
510 indicate that the uplift of the eastern and northern margins of the TP was very minor from the
511 late Miocene to the middle Pliocene (Li et al., 1991, 2015; Zheng et al., 2000; Fang et al.,
512 2005a, 2005b). Therefore, we speculate that uplift of the TP was not the major cause of the
513 expansion of the palaeo-EASM at ~4.7 Ma.
514
515 The occurrence of a humid climate across the CLP was synchronous with the gradual
516 closure of the Panama Seaway (Keigwin, 1978; O'Dea et al., 2016). Nie (2014) proposed that
517 the freshening of Eastern Equatorial and North Pacific surface water, resulting from the
518 closure of the Panama Seaway since 4.8 Ma (Haug et al., 2001), led to sea ice formation in
519 the North Pacific Ocean, which enhanced the high-pressure cell over the Pacific and
520 increased the strength of southerly and southeasterly winds. However, there was a warming
521 trend in the Northern Hemisphere from 4.6 Ma (Haug et al., 2005; Lawrence et al., 2006).
522 The gradual closure of the Panama Seaway resulted in the reorganization of surface currents
523 in the Atlantic Ocean. Notably, the Gulf Stream was enhanced and began to transport warm
524 surface waters to high northern latitudes, thus strengthening the Atlantic meridional
525 overturning circulation and warming the Arctic (Haug and Tiedemann, 1998; Haug et al.,
526 2005). Three independent proxies from an early Pliocene peat deposit in the Canadian High

Arctic indicate that Arctic temperatures were 19 ℃ warmer during the early Pliocene than today (Ballantyne et al., 2010). This warmth is also confirmed by other records from high northern latitude regions: diatom abundances and assemblages, pollen data, magnetic susceptibility and sedimentological evidence from Siberia all indicate that the climate was warm and wet in the early Pliocene (Baikal Drilling Project Memb, 1997, 1999). Furthermore, a decrease in the input of ice-rafted debris to the sediments of the subarctic northwest Pacific was synchronous with the expansion of the palaeo-EASM during the early Pliocene (Fig. 6). ~~The warming of the northernhigh latitude region led to increases in summer temperature in the mid-latitudes of Eurasia.However, equatorial SSTs remained stable or cooled slightly (Brierley et al., 2009; Fedorov et al., 2013), and thus the land-ocean thermal contrast was intensified. Furthermore~~T, he warming of the Northern Hemisphere and external heating derived from a reduced ice albedo at high northern latitudes ~~also~~ enhanced the thermal contrast between the Pacific and Eurasian regions (Dowsett et al., 2010). This large land-ocean thermal contrast was essential for enhancing the palaeo-EASM. On the other hand, the unusually warm high northern latitudes and the West Antarctic ice-sheet expansion by 6–5 Ma (Zachos et al., 2001, 2008) steepened interhemispheric thermal gradient and further caused the thermal equator to move northward (Chiang and Friedman, 2012; Broecker and Putnam, 2013). This facilitated the northwestward expansion of the palaeo-EASM, which is also proposed as the driving mechanism for northwestward migration of the monsoon rain belt for the warm Holocene (Yang et al., 2015).

~~- small meridional heat gradient in the Northern Hemisphere pushed the Intertropical Convergence Zone northwards (Chang et al., 2013; Sun et al., 2015), which weakened the~~

~~westerly circulation and thus facilitated the northwestward expansion of the palaeo-EASM.~~

~~Figure 6 shows that high values of pedogenic indices in the XSZ section correspond with high SSTs in the EEP. This appears to be contradictory to the case of the modern ENSO (when the EEP temperature is high, the precipitation amount in the western CLP is low). The discrepancy may indicate that the nature of sea-air interactions during the early Pliocene was different from today. During 4.8-4.0 Ma, the thermohaline circulation was reorganizing and creating a precondition for the development of the modern equatorial Pacific cold tongue (Chaisson and Ravelo, 2000). Several crucial changes linked with the summer monsoon occurred: There was a vast expansion of the western Pacific warm pool into subtropical regions in the early Pliocene (Brierley et al., 2009; Fedorov et al., 2013), and temperatures at the edge of the warm pool showed a warming trend of ~2 ℃ from the latest Miocene to the early Pliocene (Karas et al., 2011). This enhanced thermal state of the WEP warm pool significantly enhanced the summer monsoon and its northward extension. A modelling experiment indicates that the precipitation of the CLP would increase when the tropical warm pool expended into subtropical region (Brierley et al., 2009). Today, when the northern part of the western pacific warm pool is warm, convection over and around the Philippines is enhanced; in addition, the northern extent of the western Pacific subtropical high shifts northwards from the Yangtze River valley to the Yellow River valley and moisture is introduced across the entire CLP (Huang et al., 2003). Further research is needed to determine if this was also the case during the early Pliocene. However, the warming and freshening of the subtropical Pacific would have promoted increased evaporation which would have provided enhanced moisture for the palaeo-EASM, resulting in increased rainfall**

~~across the CLP.~~

~~In conclusion,~~ Therefore, we infer that the warming of high northern latitudes in response to the closure of the Panamanian Seaway ~~, accompanied by the vast poleward expansion of the tropical warm pool into subtropical regions and the freshening of the subtropical Pacific,~~ may have facilitated the expansion of the palaeo-EASM during the early Pliocene. However, there are several uncertainties associated with such an explanation. For example, the timing of the closure of the Panama Seaway is still debated (Bacon et al., 2015; O'Dea et al., 2016), and it is unclear how strongly these changes influenced the palaeo-EASM. Addressing these questions requires more geological evidence and precise model simulations of the early Pliocene climate. The value of our study lies in proposing the potential linkage of the evolution of palaeo-EASM and changes in temperatures of high northern latitudes during the early Pliocene ~~and SSTs of the low latitude Pacific Ocean in the early Pliocene~~.

6. Conclusions

The continuous late Miocene-Pliocene red clay sequence preserved on the planation surface in the NE Tibetan Plateau provides the opportunity to elucidate the history of the Asian monsoon in the western CLP. Multi-proxy records from the XSZ section, together with other paleoclimatic records from the CLP, reveal the major patterns of climatic change from 6.7-3.6 Ma. During the late Miocene, both the amount and variability of precipitation over the XSZ section were small; however, they were much greater in the central and eastern CLP; thus, the palaeo-EASM had little influence on the climate of the western CLP at this time. During the early Pliocene, the records from the XSZ section indicate that both the amount

and variability of precipitation increased from 4.7-3.9 Ma. The climate was characterized by abrupt increases in the seasonality of precipitation, which attests to a major northwestward extension and enhancement of the summer monsoon. Multiple paleoclimatic proxies clearly show that the strongest summer monsoon occurred during 4.60-4.25 Ma. The expansion of the palaeo-EASM may have been caused by warming of the ~~Arctic region~~high northern latitudes~~, the vast poleward expansion of the tropical warm pool into subtropical regions, and the freshening of the subtropical Pacific,~~ in response to the closure of the Panamanian Seaway during the early Pliocene.

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References

An, Z. S., Kutzbach, J. E., Prell, W. L., Porter, S. C.: Evolution of Asian monsoons and phased uplift of the Himalayan Tibetan plateau since Late Miocene times. Nature, 411, 62–66, 2001.

An, Z. S.: Late Cenozoic Climate Change in Asia. Springer Netherlands, 2014.

615 Ao, H., Roberts, A. P., Dekkers, M. J., Liu, X., Rohling, E. J., Shi, Z., An, Z. S., and Zhao, X.: Late
616 Miocene–Pliocene Asian monsoon intensification linked to Antarctic ice-sheet growth. Earth &
617 Planetary Science Letters,444, 75-87, 2016.
618 Avery, B. W., and Bascomb, C. L.: Soil survey laboratory methods /, 1974.
619 Bacon, C. D., Silvestro, D., Jaramillo, C., Smith, B. T., Chakrabarty, P., and Antonelli, A.: Biological
620 evidence supports an early and complex emergence of the Isthmus of Panama. Proceedings of the
621 National Academy of Sciences,112(19), 6110-6115, 2015.
622 Ballantyne, A. P., Greenwood, D. R., SinninghеDamste, J. S., Csank, A. Z., Eberle, J. J., and Rybczynski,
623 N.: Significantly warmer arctic surface temperatures during the pliocene indicated by multiple
624 independent proxies. Geology, 38(7), 603-606, 2010.
625 Brierley, C. M., Fedorov, A. V., Liu, Z., Herbert, T. D., Lawrence, K. T., and Lariviere, J. P.: Greatly
626 expanded tropical warm pool and weakened Hadley circulation in the early Pliocene. Science,
627 323(5922), 1714-8, 2009.
628 Brierley, C. M., and Fedorov, A. V.: Relative importance of meridional and zonal sea surface temperature
629 gradients for the onset of the ice ages and pliocene-pleistocene climate evolution. Paleoceanography,
630 25(4), 2010.
631 Broecker, W.S., Putnam A.E.: Hydrologic impacts of past shifts of Earth's thermal equator offer insight
632 into those to be produced by fossil fuel CO_2. Proc Natl Acad Sci USA 110(42):16710–16715, 2013.
633 Buggle, B., Glaser, B., Hambach, U., Gerasimenko, N., and Markovi, S.: An evaluation of geochemical
634 weathering indices in loess–paleosol studies. Quaternary International, 240(1–2):12-21, 2011.
635 Chiang J. C. H., Friedman A. R.: Tropical cooling, interhemispheric thermal gradients, and tropical climate
636 change. Annu Rev Earth Planet Sci 40(1):383–412, 2012.

~~Chaisson, W. P., and Ravelo, A. C.: Pliocene development of the east-west hydrographic gradient in the equatorial Pacific. Paleoceanography, 15(5), 497-505, 2000.~~

Chang, H., An, Z. S., Wu, F. L., Jin, Z., Liu, W. G., and Song, Y. G.: A Rb/Sr record of the weathering response to environmental changes in westerly winds across the tarim basin in the late Miocene to the early Pleistocene. Palaeogeography Palaeoclimatology Palaeoecology, 386(6), 364-373, 2013.

Clift, P. D.: Controls on the erosion of cenozoicasia and the flux of clastic sediment to the ocean. Earth & Planetary Science Letters, 241(3–4), 571-580, 2006.

Clift, P. D., Hodges, K. V., Heslop, D., Hannigan, R., Long, H. V., and Calves, G.: Correlation of Himalayan exhumation rates and Asian monsoon intensity. Nature Geoscience, 1(12), doi:10.1038/ngeo351, 2008.

Clift, P. D., Wan, S. M., and Blusztajn, J.: Reconstructing chemical weathering, physical erosion and monsoon intensity since 25 Ma in the northern South China Sea: a review of competing proxies. Earth-Science Reviews, 130(3), 86-102, 2014.

Compo, G., Whitaker, J., Sardeshmukh, P., and Mccoll, C.: The quality control system of the 20th century reanalysis dataset. Egu General Assembly, 15, 2013.

Ding, Z. L., Yang, S. L., Sun, J. M., and Liu, T. S.: Iron geochemistry of loess and red clay deposits in the Chinese loess plateau and implications for long-term Asian monsoon evolution in the last 7.0 ma. Earth & Planetary Science Letters, 185(1), 99-109, 2001.

Dowsett, H. J., Robinson, M., Haywood, A., Salzmann, U., Hill, D., Sohl, L. E., Chandler, M., Williams, M., Foley, K., and Stoll, D. K.: The PRISM3D paleoenvironmental reconstruction. Stratigraphy, 7, 123-139, 2010.

Fedorov, A. V., Brierley, C. M., Lawrence, K. T., Liu, Z., Dekens, P. S., and Ravelo, A. C.: Patterns and

659 mechanisms of early Pliocene warmth. Nature, 496 (7443), 43, 2013.
660 Fang, X. M, Li, J. J, Derbyshire, E., Fitzpatrick, E. A., and Kemp, R. A.: Micromorphology of the Beiyuan
661 loess-paleosol sequence in Gansu province, China: geomorphological and paleoenvironmental
662 significance. Palaeogeography Palaeoclimatology Palaeoecology, 111(3–4), 289-303, 1994.
663 Fang, X. M., Ono, Y., Fukusawa, H., Pan, B. T., Li, J. J., Guan, D. H., Oi. K., Tsukamotoc, S., Torii, M.,
664 and Mishima, T.: Asian summer monsoon instability during the past 60,000 years: magnetic
665 susceptibility and pedogenic evidence from the western Chinese Lloess plateau. Earth & Planetary
666 Science Letters, 168(3–4), 219-232, 1999.
667 Fang, X. M., Yan, M. D., Voo, R. V. D., Rea, D. K., Song, C. H., Parés, J. M., Gao J. P., Nie J. S., and Dai
668 S: Late Cenozoic deformation and uplift of the NE Tibetan plateau: evidence from high-resolution
669 magnetostratigraphy of the Guide basin, Qinghai province, China. Geological Society of America
670 Bulletin,117(9), 1208-1225, 2005a.
671 Fang, X., Zhao, Z. J., Li J. J., Yan, M. D, Pan, B. T., Song, C. H., and Dai, S.: Magnetostratigraphy of the
672 late Cenozoic Laojunmiao anticline in the northern Qilian mountains and its implications for the
673 Northern Tibetan plateau uplift. Science in China, 48(7), 1040-1051, 2005b.
674 Fang, X. M, Wu, F. L., Hai, W. X., Wang, Y. D., Zhang, X. Z., and Zhang, W. L.: Plio-Pleistocene drying
675 process of Asian inland-sporopollen and salinity records from Yahu section in the central Qaidam
676 basin(in Chinese). Quaternary Sciences 28(5): 874-882, 2008.
677 Fedo, C. M., Nesbitt, H. W., and Young, G. M.: Unraveling the effects of potassium metasomatism in
678 sedimentary rocks and paleosols, with implications for paleoweathering conditions and
679 provenance. Geology, 23(10), 921-924, 1995.
680 Guo, Z. T., Peng, S. Z., Hao, Q. Z., Biscaye, P. E., and Liu, T. S.: Origin of the Miocene–Pliocene red-

earth formation at Xifeng in northern China and implications for paleoenvironments. Palaeogeography Palaeoclimatiology Palaeoecology, 170(1-2), 11-26, 2001.

Han, W., Fang, X., Berger, A., and Yin, Q.: An astronomically tuned 8.1 ma eolian record from the Chinese Loess plateau and its implication on the evolution of Asian monsoon. Journal of Geophysical Research Atmospheres, 116(D24114), 2011.

Haug, G. H., and Tiedemann, R.: Effect of the formation of the isthmus of Panama on Atlantic ocean thermohaline circulation. Nature, 393(3), 673-676, 1998.

Haug, G. H., Tiedemann, R., Zahn, R., and Ravelo, A. C.: Role of Panama uplift on oceanic freshwater balance. Geology, 29(3), 207-210, 2001.

Haug, G. H., Ganopolski, A., Sigman, D. M., Rosell-Mele, A., Swann, G. E., Tiedemann, R., Jaccard, S. L., Bollmann, B. J., Maslin, M. A., Leng, M. J., and Eglinton, G.: North pacific seasonality and the glaciation of north America 2.7 million years ago. Nature, 433(7028), 821-825, 2005.

He, T., Chen, Y., Balsam, W., Qiang. X.K., Liu, L.W., Chen, J., Li, F.J.: Carbonate leaching processes in the Red Clay Formation, Chinese Loess Plateau: Fingerprinting East Asian summer monsoon variability during the late Miocene and Pliocene. Geophysical Research Letters, 40(1):194-198, 2013.Herbert, T. D., Peterson, and Liu, Z.: Tropical ocean temperatures over the past 3.5 million years. Science, 328(5985), 1530-4, 2010.

Herbert, T. D., Lawrence, K. T., Tzanova, A., Peterson, L. C., Caballerogill, R., and Kelly, C. S.: Late Miocene global cooling and the rise of modern ecosystems. Nature Geoscience, 9(11), 2016.

~~Huang, R. H., Zhou L.T., and Chen, W.: The progresses of recent studies on the variabilities of the East Asian monsoon and their causes, Adv. Atmos. Sci., 20, 55-69, 2003.~~

Jansen, E., and Sjøholm, J.: Reconstruction of glaciation over the past 6 Myr from ice-borne deposits in the

703 Norwegian sea. Nature,349(6310), 600-603, 1991.

704 ~~Karas, C., Nürnberg, D., Tiedemann, R., and Garbe-Schönberg, D. : Pliocene climate change of the~~
705 ~~southwest Pacific and the impact of ocean gateways. Earth & Planetary Science Letters, 301(1–2),~~
706 ~~117–124, 2011.~~

707 Keigwin, L. D.: Pliocene closing of the isthmus of Panama, based on biostratigraphic evidence from
708 nearby Pacific ocean and Caribbean sea cores. Geology, 6(10), 630, 1978.

709 King, T.: Quantifying non-linearity and geometry in time series of climate. Quaternary Science Reviews
710 15, 247-266, 1996.

711 Laskar, J.: Long-term solution for the insolation quantities of the earth. Proceedings of the International
712 Astronomical Union, 2(14), 101-106, 2004.

713 Lawrence, K. T., Liu, Z., and Herbert, T. D.: Evolution of the eastern tropical pacific through Plio-
714 Pleistocene glaciation. Science, 312(5770), 79-83, 2006.

715 Liang, L., Sun, Y., Beets, C. J., Prins, M. A., Wu, F., and Vandenberghe, J.: Impacts of grain size sorting
716 and chemical weathering on the geochemistry of Jingyuan loess in the northwestern Chinese Loess
717 plateau. Journal of Asian Earth Sciences, 69(12), 177-184, 2013.

718 Li, F. J., Rousseau, D. D., Wu, N., Hao, Q., and Pei, Y.: Late Neogene evolution of the East Asian
719 monsoon revealed by terrestrial mollusk record in western Chinese Loess plateau: from winter to
720 summer dominated sub-regime. Earth & Planetary Science Letters, 274(3–4), 439-447, 2008.

721 Li, J. J.: The environmental effects of the uplift of the Qinghai-Xizang plateau. Quaternary Science
722 Reviews, 10(6), 479-483, 1991.

723 Li, J. J., Fang, X., Song, C., Pan, B., Ma, Y., and Yan, M., Late Miocene–quaternary rapid stepwise uplift
724 of the ne Tibetan plateau and its effects on climatic and environmental changes. Quaternary Research,

725 81(3), 400-423, 2014.
726 Li, J. J., Zhou, S. Z., Zhao, Z. J., and Zhang, J.: The Qingzang movement: the major uplift of the Qinghai-
727 Tibetan plateau. Science China Earth Sciences, 58(11), 2113-2122, 2015.
728 Li, J. J., Ma, Z. H, Li, X. M., Peng, T. J., Guo, B. H., Zhang, J., Song, C. H., Liu, J. Hui, Z. C., Yu, H.,
729 Ye, X. Y., Liu, S. P., Wang, X. X.: Late Miocene-Pliocene geomorphological evolution of the
730 Xiaoshuizi peneplain in the Maxian Mountains and its tectonic significance for the northeastern
731 Tibetan Plateau. Geomorphology. 295 393-405, 2017.
732 Liu, X. M., Rolph, T., An, Z., and Hesse, P.: Paleoclimatic significance of magnetic properties on the red
733 clay underlying the loess and paleosols in China. Palaeogeography Palaeoclimatology Palaeoecology,
734 199(1), 153-166, 2003.
735 Liu, C. Q., Masuda, A., Okada, A., Yabuki, S., Zhang, J., and Fan, Z. L.: A geochemical study of loess and
736 desert sand in northern China: implications for continental crust weathering and composition.
737 Chemical Geology, 106 (3-4), 359-374, 1993.
738 Liu, Q., Jackson, M. J., Yu, Y., Chen, F., Deng, C., and Zhu, R.: Grain size distribution of pedogenic
739 magnetic particles in Chinese loess/paleosols. Geophysical Research Letters, 312(22), 359-393, 2004.
740 Liu, Q., Torrent, J., Maher, B. A., Yu, Y., Deng, C. L., Zhu, R., and Zhao X. X.: Quantifying grain size
741 distribution of pedogenic magnetic particles in Chinese loess and its significance for pedogenesis.
742 Journal of Geophysical Research Atmosphere, 110(B11), 2005.
743 Liu, W., Liu, Z., An, Z., Sun, J., Chang, H., Wang, N., andDong, J. B.: Late Miocene episodic lakes in the
744 arid Tarim basin, western China. Proceedings of the National Academy of Sciences, 111(46), 16292-6,
745 2014.
746 Lu, H., Zhang, F., Liu, X., and Duce, R. A: Periodicities of palaeoclimatic variations recorded by loess-

747 paleosol sequences in China. Quaternary Science Reviews, 23(18–19), 1891–1900, 2004.
748 Ma, Y. Z., Wu, F. L., Fang, X. M., Li, J. J., An, Z. S., and Wei, W.: Pollen record from red clay sequence
749 in the central Loess plateau between 8.10 and 2.60 Ma. Chinese Science Bulletin, 50(19), 2234-2243,
750 2005.Baikal Drilling Project Memb.: Preliminary results of the first scientific drilling on lake Baikal,
751 Buguldeika site, southeastern Siberia.Quaternary International, 37(2), 3-17, 1997.
752 Baikal Drilling Project Memb.: Continuous paleoclimate record recovered for last 5 million years. Eos
753 Transactions American Geophysical Union, 78(51), 597-601, 1999.
754 Mudie, P. J., and Helgason, J.: Palynological evidence for Miocene climatic cooling in eastern iceland
755 about 9.8 myr ago. Nature, 303(5919), 689-692, 1983.
756 Nesbitt, H. W., Markovics, G., and Price, R. C.: Chemical processes affecting alkalis and alkaline earths
757 during continental weathering. Geochimica Et Cosmochimica Acta, 44(11), 1659-1666, 1980.
758 Nie, J. S., Stevens, T., Song, Y., King, J. W., Zhang, R., Ji, S. C, Gong L. S., and Cares, D.: Pacific
759 freshening drives Pliocene cooling and Asian monsoon intensification. Scientific Reports, 4, 5474,
760 2014.
761 O'Dea, A., Lessios H. A., Coates, A. G., Eytan, R. I., Restrepo-Moreno, S. A., Cione, A. L., Collins, L. S.,
762 Queiroz, A. D., Farris, D. W., Norris, R. D., Stallard, R. F., Woodburne, M. O., Aguilera, O., Aubry,
763 M. P., Berggren, W. A., Budd, A. F., Cozzuol, M. A., Coppard, S. E., Duque-Caro, H., Finnegan, S.,
764 Gasparini, G. M., Grossman, E. L., Johnson, K. G., Keigwin, L. D., Knowlton, N., Leigh, E. G.,
765 Leonard-Pingel, J. S., Marko, P. B., Pyenson, N. D., Rachello-Dolmen, P. G., Soibelzon, E.,
766 Soibelzon, L., Todd, J. A., Vermeij, G. J., and Jackson, J. B. C.: Formation of the Isthmus of Panama.
767 Science Advances, 2(8), 2016.
768 Pagani, M., Liu, Z., Lariviere, J., and Ravelo, A. C.: High earth-system climate sensitivity determined from

769 Pliocene carbon dioxide concentrations. Nature Geoscience, 3(1), 27-30, 2010.
770 Rossinsky V. J., and Swart, P. K.,: Influence of climate on the formation and isotopic composition of
771 calcretes. In: Swart, P.K., Lohmann, K.C., McKenzie, J., Savin, S. (Eds.), Climate Change in
772 Continental Isotopic Records, American Geophysical Union: Geophysical Monography, 78, pp. 67-75,
773 1993.
774 Song, C. H., Fang, X. M., Li, J. J., Gao, J., Zhao, Z. J., and Fan, M. J.,: Tectonic uplift and sedimentary
775 evolution of the Jiuxi basin in the northern margin of the Tibetan plateau since 13 Ma BP. Science
776 China Earth Sciences,44(1), 192-202, 2001.
777 Song, Y. G., Fang, X. M., Torii, M., Ishikawa, N., Li, J. J., and An, Z. S.: Late Neogene rock magnetic
778 record of climatic variation from Chinese eolian sediments related to uplift of the Tibetan
779 plateau. Journal of Asian Earth Sciences, 30(2), 324-332, 2007.
780 Sun, D. H., Bloemendal, J., Rea, D. K., An, Z. S., Vandenberghe, J.,and Lu, H., Su, R. X., and Liu, T.: .
781 Bimodal grain-size distribution of Chinese loess, and its palaeoclimatic implications. Catena, 55(3),
782 325-340, 2004.
783 Sun, J. M., and Liu, T. S. : The age of the Taklimakan desert. Science, 312 (5780), 1612-1621, 2006a.
784 Sun, J. M., and Huang, X.: Half-precessional cycles recorded in Chinese loess: response to low-latitude
785 insolation forcing during the last interglaciation. Quaternary Science Reviews, 25(9–10), 1065-1072,
786 2006b.
787 Sun, J. M., Liu, W. G., Liu, Z., Deng, T., Windley, B. F., and Fu, B.: Extreme aridification since the
788 beginning of the Pliocene in the Tarim basin, western China. Palaeogeography Palaeoclimatology
789 Palaeoecology, 2017.
790 Sun, Y. B., Lu, H. Y., and An, Z S.: Grain size of loess, palaeosol and red clay deposits on the Chinese
791 Loess plateau: significance for understanding pedogenic alteration and palaeomonsoon

792 evolution. Palaeogeography Palaeoclimatology Palaeoecology, 241(1), 129-138, 2006c.
793 Sun, Y. B., An, Z. S., Clemens, S. C., Bloemendal, J., andVandenberghe, J.: Seven million years of wind
794 and precipitation variability on the Chinese Loess plateau. Earth & Planetary Science Letters, 297(3–
795 4), 525-535, 2010.
796 Sun, Y. B., Kutzbach, J., An, Z., Clemens, S., Liu, Z., Liu, W., Liu, X. D., Shi, Z. G., Zheng, W. P., Liang,
797 L., Yan, Y., and Li, Y.: Astronomical and glacial forcing of East Asian summer monsoon
798 variability. Quaternary Science Reviews, 115, 132-142, 2015.
799 Tripati, A. K., Roberts, C. D., and Eagle, R. A.: Coupling of CO_2 and ice sheet stability over major climate
800 transitions of the last 20 million years. Science, 326(5958), 1394-1397, 2009.
801 Tang, H., Micheels, A., Eronen, J., and Fortelius, M.: Regional climate model experiments to investigate
802 the Asian monsoon in the late Miocene. Climate of the Past, 7(3), 847-868, 2011.
803 Vandenberghe, J., H. Lu, D. Sun, J. Huissteden, V., and Konert, M.: The late Miocene and Pliocene
804 climate in East Asia as recorded by grain size and magnetic susceptibility of the Red Clay deposits
805 (Chinese Loess Plateau), Palaeogeography Palaeoclimatology Palaeoecology, 204, 239–255,
806 doi:10.1016/S0031-0182(03)00729-6, 2004.
807 Wan, S. M., Tian, J., Steinke, S., Li, A., and Li, T.: Evolution and variability of the East Asian summer
808 monsoon during the Pliocene: evidence from clay mineral records of the South China
809 Sea. Palaeogeography Palaeoclimatology Palaeoecology, 293(1–2), 237-247, 2010.
810 Wang, H. B., Chen, F. H., and Zhang, J. W.: Environmental significance of grain size of loess-paleosol
811 sequence in western part of Chinese Loess plateau. Journal of Desert Research, 22(1), 21-26, 2002.
812 Wang, L., Lu, H. Y., Wu, N. Q., Li, J., Pei, Y. P., Tong, G. B., and Peng, S. Z.:. Palynological evidence for
813 Late Miocene-Pliocene vegetation evolution recorded in the red clay sequence of the central Chinese

814 Loess Plateau and implication for palaeo-environmental change. Palaeogeography Palaeoclimatology
815 Palaeoecology. 241, 118–128, 2006.
816 Wara, M. W., Ravelo, A. C., and Delaney, M. L.: Permanent EI niño-like conditions during the Pliocene
817 warm period. Science, 309(5735), 758-61, 2005.
818 Watanabe, T., Suzuki, A., Minobe, S., Kawashima, T., Kameo, K., &Minoshima, K., Aguilar, Y. M., Wan,
819 R., Kawahata, H., Sowa, K., Nagai, T., and Kase, T.: Permanent EI niño during the Pliocene warm
820 period not supported by coral evidence. Nature, 471 (7337), 209-211, 2011.
821 Wu, N., Pei, Y., Lu, H., Guo, Z., Li, F., and Liu, T.: Marked ecological shifts during 6.2–2.4 ma revealed
822 by a terrestrial molluscan record from the chinese red clay formation and implication for
823 palaeoclimatic evolution. Palaeogeography Palaeoclimatology Palaeoecology, 233(3-4), 287-299,
824 2006.
825 Xia, D. S., Jia, J., Li, G., Zhao, S., Wei, H. T., and Chen, F. H.: Out-of-phase evolution between summer
826 and winter East Asian monsoons during the Holocene as recorded by Chinese Loess deposits.
827 Quaternary Research, 81(3), 500-507, 2014.
828 Yang, S. L., Ding, F., and Ding, Z. L.: Pleistocene chemical weathering history of Asian arid and semi-arid
829 regions recorded in loess deposits of China and Tajikistan. Geochimica Et Cosmochimica Acta, 70(7),
830 1695-1709, 2006.
831 Yang, S. L., Ding, Z.L., Li, Y.Y., Wang, X., Jiang, W.Y., Huang, X.F.: Warming-induced northwestward
832 migration of the East Asian monsoon rain belt from the Last Glacial Maximum to the mid-Holocene.
833 Proc. Natl. Acad. Sci. USA 112, 13178–13183, 2015.
834 Zachos, J., Pagani, M., Sloan, L., Thomas, E., and Billups, K.: Trends, rhythms, and aberrations in global
835 climate 65 Ma to present. Science, 292(5517), 686-93, 2001.

836 Zachos, J.C., Dickens, G.R., Zeebe, R.E.: An early Cenozoic perspective on green-house warming and
837 carbon-cycle dynamics. Nature.451, 279–283, 2008.
838 Zhang, R., Jiang, D. B., Liu, X. D., and Tian, Z. P.: Modeling the climate effects of different subregional
839 uplifts within the Himalaya-Tibetan plateau on Asian summer monsoon evolution. Science
840 Bulletin, 57(35), 4617-4626, 2012.
841 Zhang, Y. G., Pagani, M., and Liu, Z.: A 12-million-year temperature history of the tropical Pacific
842 ocean. Science, 344(6179), 84, 2014.
843 Zhao, J. B.: A study of the $CaCO_3$ illuvial horizons of paleosols and permeated pattern far rain water, J
844 Geogr Sci, 15(4), 344-350, 1995.
845 Zhao, J. B.: Illuvial $CaCO_3$ layers of paleosol in loess and its environmental significance, Journal of Xi'an
846 Engineering University, 20(3), 46-49, 1998.
847 Zheng, H. B, Mcaulay Powell, C., An, Z. S., Zhou, J., and Dong, G. R.: Pliocene uplift of the northern
848 Tibetan plateau. Geology, 28(8), 715, 2000.
849

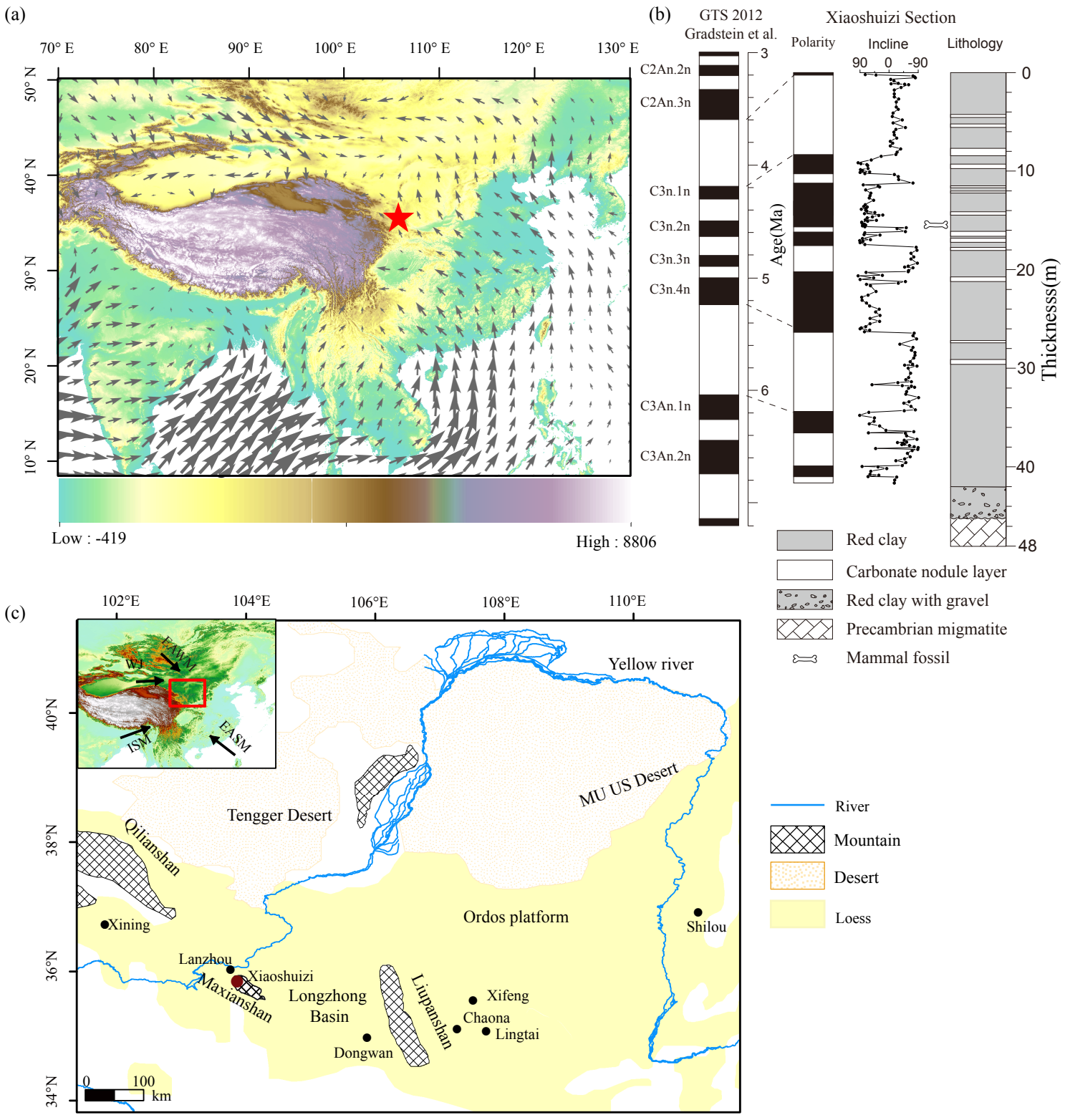

Fig. 1. Location of the study area and atmospheric circulation patterns. (a) 850 hPa vector wind averaged from June to August for 1982-2012 based on NOAA Earth System Research Laboratory reanalysis data (Compo et al., 2013). (b) Lithology and magnetostratigraphy of the XSZ drill core. (c) The Chinese Loess Plateau with locations of the studied Xiaoshuizi site and other sections mentioned in the text.

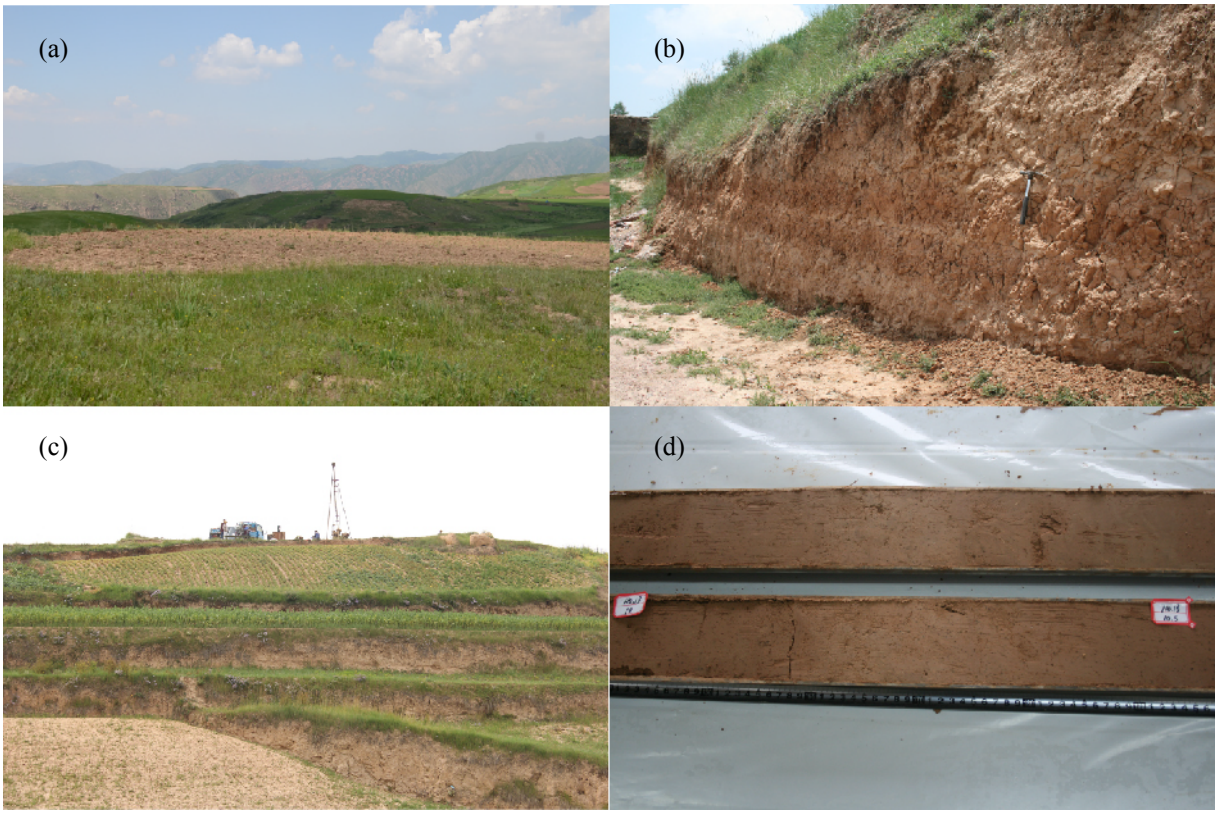

Fig. 2. Photos of the XSZ planation surface and the red clay. (a) XSZ planation surface. (b) Red clay outcrop, XSZ. (c) Position of the XSZ drill hole. (d) The XSZ drill core.

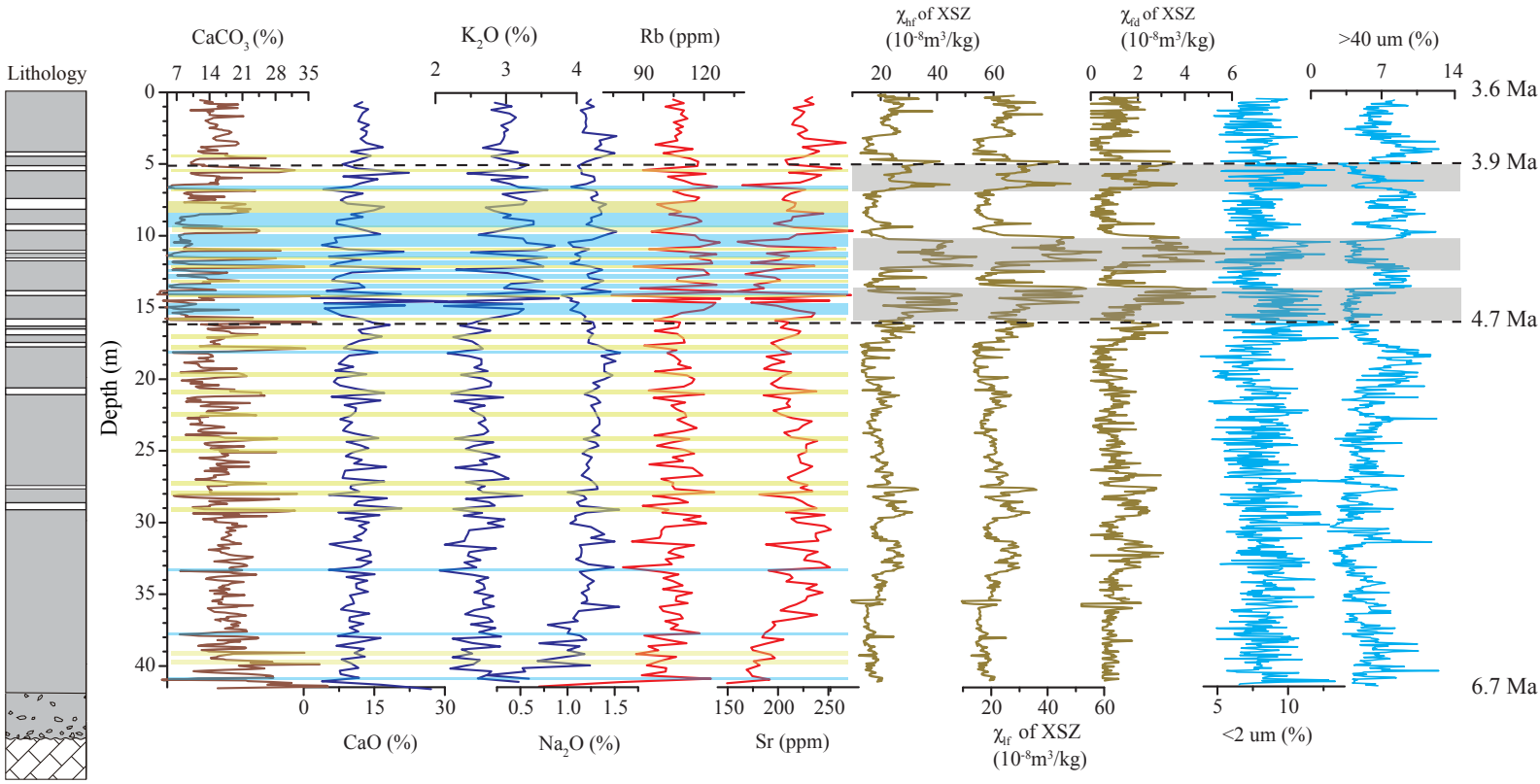

Fig. 3. Variations in carbonate content, major element concentration, minor element concentration, magnetic susceptibility and grain size for the XSZ red clay section (6.7-3.6 Ma). Yellow shading indicates Bk horizons with carbonate content >21%; blue shading indicates Bw horizons with carbonate content <8%; gray shading indicates intervals with high magnetic susceptibility. Dashed lines are upper and lower boundaries of the relatively wet interval.

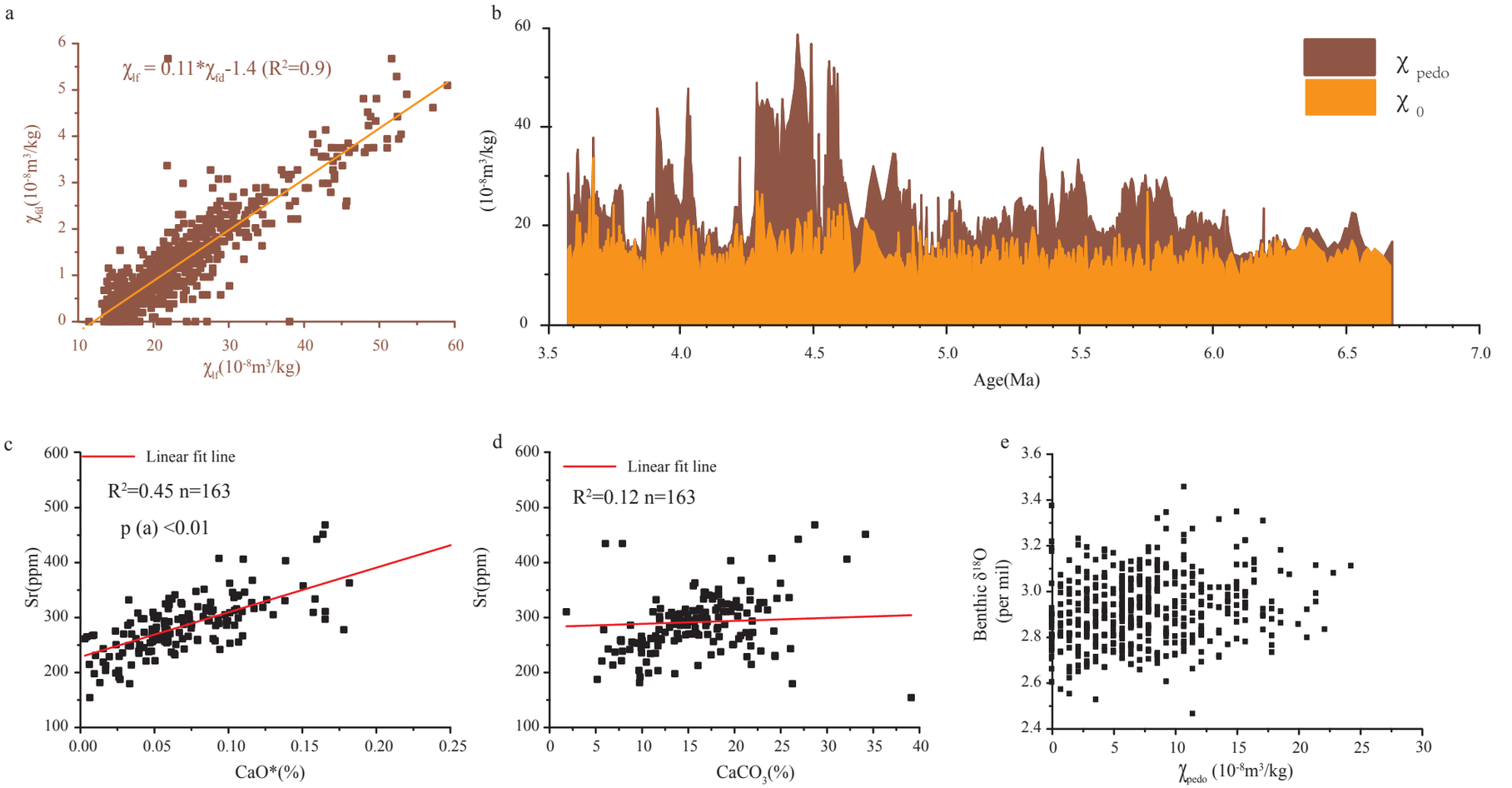

Fig. 4. (a) Scatter plot of χ_{lf} versus χ_{fd}. (b) Separation of χ_{pedo} and χ_0. (c) Scatter plot of Sr versus CaO*. (d) Scatter plot of Sr versus $CaCO_3$. (e) Scatter plot of benthic $\delta^{18}O$ versus χ_{pedo} during 6.7-5.2 Ma. χ_{pedo} is the magnetic susceptibility of pedogenic origin and χ_0 is the magnetic susceptibility of the detrital material.

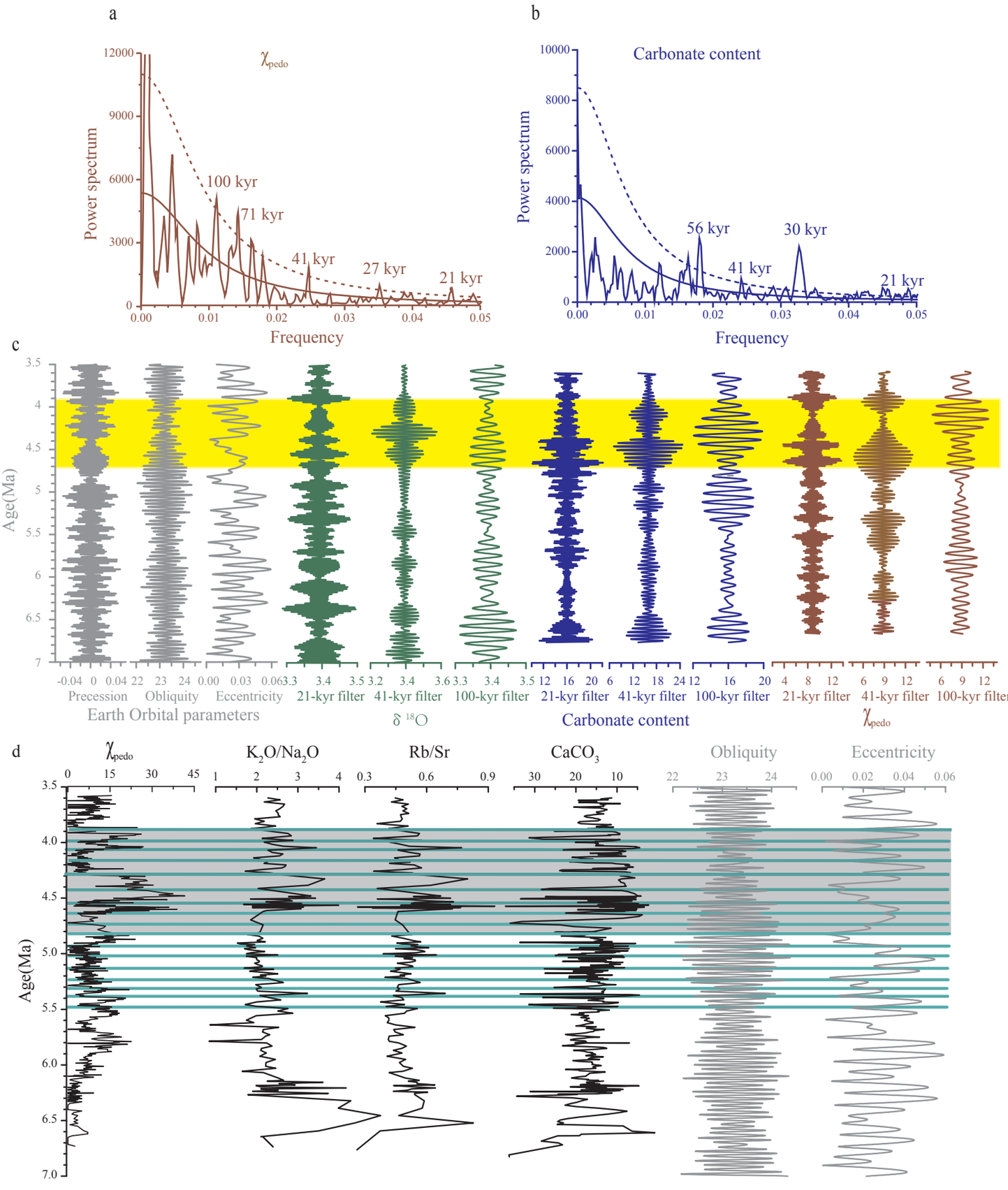

Fig. 5. Spectrum analysis results of the XSZ red clay section. (a) χpedo and (b) carbonate content (blue). (c) Comparison of orbital parameters (eccentricity, obliquity and precession - Laskar et al., 2004) with filtered components of the carbonate content, χpedo and δ18O records (Zachos et al., 2001) in the 18-24 kyr, 36-46 kyr, and 90-110 kyr bands. Yellow shading denotes increased amplitude of the filtered components of carbonate and χpedo within the three orbital bands. (d) Carbonate, weathering and pedogenic indicators linked to eccentricity and obliquity orbital variations during 4.7–3.9 Ma.

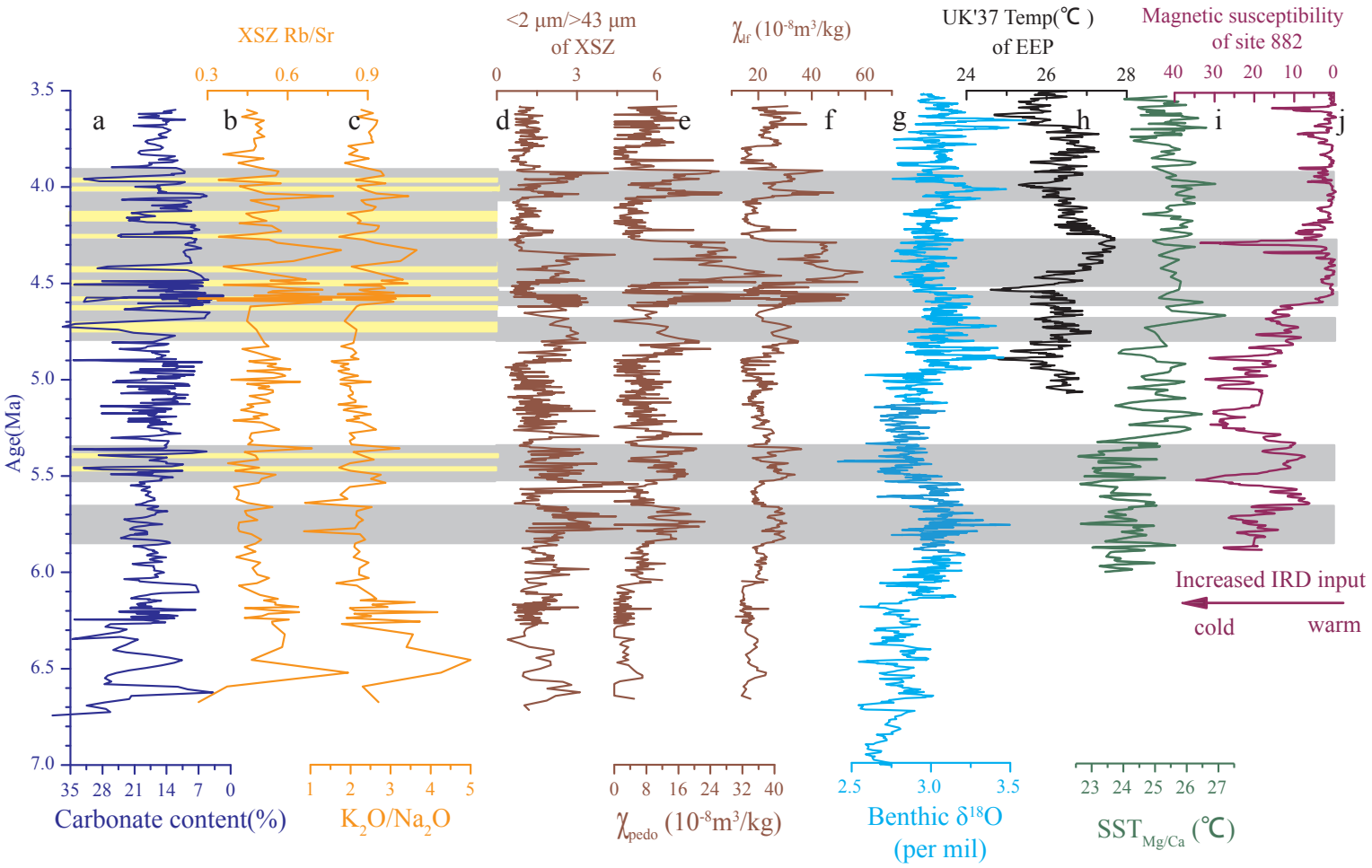

Fig. 6. Comparison of the paleoclimatic record of the XSZ red clay section with climate records from elsewhere. (a) Effective precipitation record for the XSZ section; (b-c) chemical weathering records for the XSZ section; (d-f) pedogenic intensity records for the XSZ section; (g) stacked deep-sea benthic foraminiferal oxygen isotope curve compiled from data from DSDP and ODP sites (Zachos et al., 2001); (h) ~~reconstructed sea surface temperature in the eastern equatorial Pacific (EEP) from ODP Site 846 (Lawrence et al., 2006); (i) reconstructed temperature at the edge of the warm pool in the southwest Pacific Ocean, from ODP Site 590B (Karas et al., 2011); (j)~~ magnetic susceptibility record from ODP Site 882 (Haug et al., 2005). The gray shading indicates relatively wet periods and the light-yellow shading shows intervals of carbonate accumulation.

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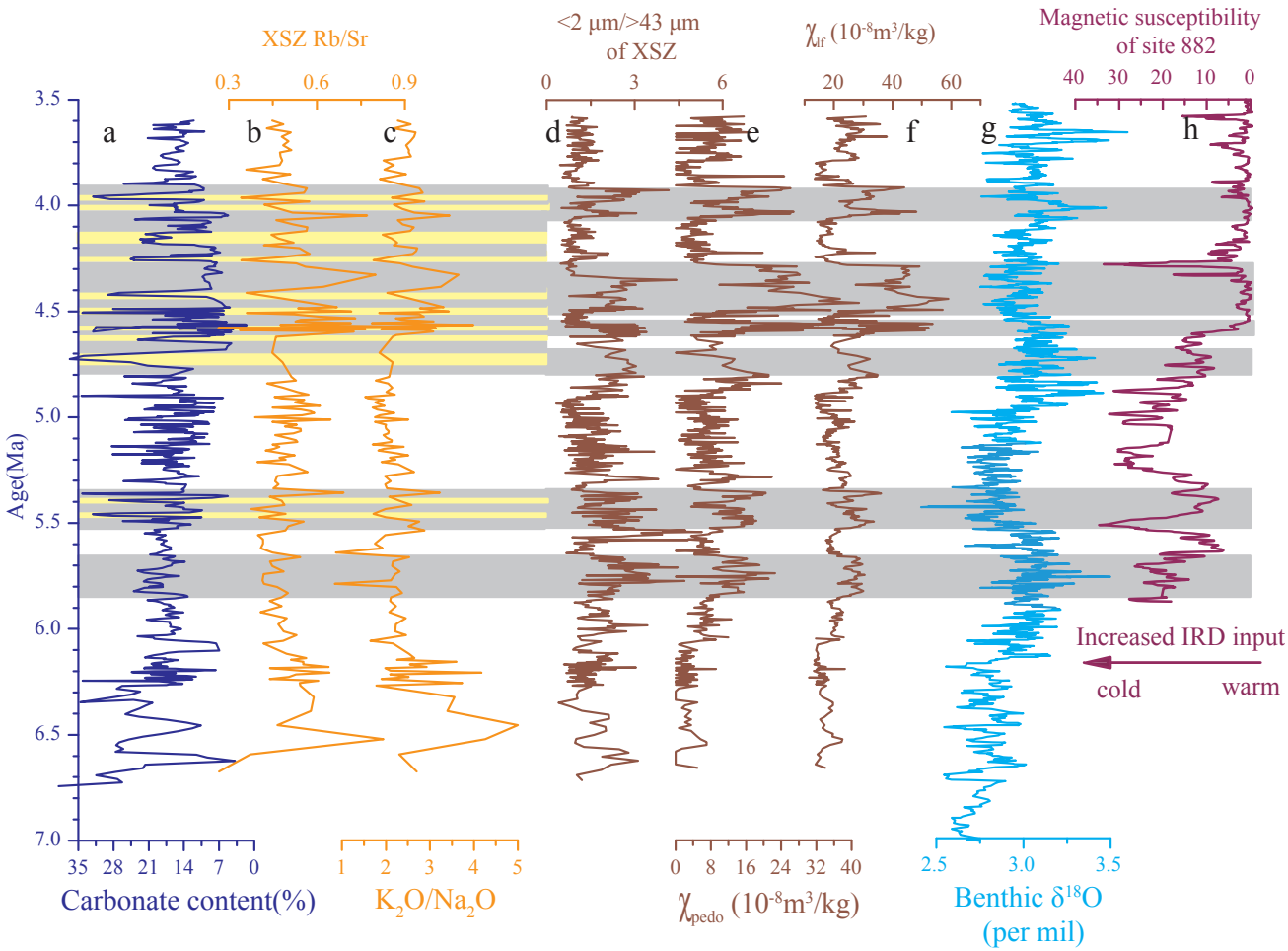

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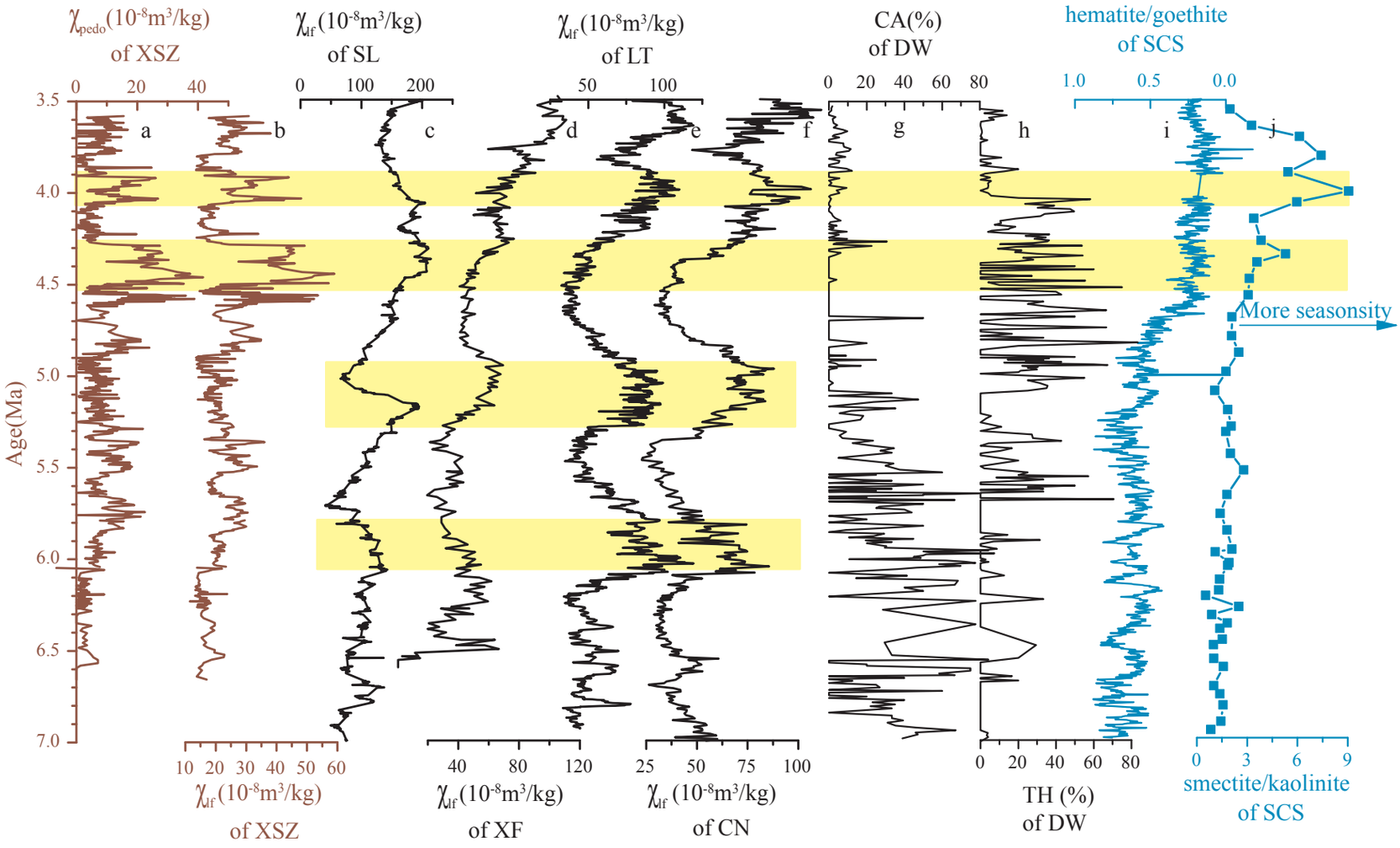

Fig. 7. Comparison of late Miocene-Pliocene paleoclimatic records from Asia. (a-b) χ_{pedo} and χ_{lf} from the XSZ section. (c-f) χ_{lf} record from Shilou (Ao et al., 2016), Xifeng (Guo et al., 2001), Lingtai (Sun et al., 2010) and Chaona (Song et al., 2007). (g-h) Percentages of cold-aridiphilous (CA) mollusks and thermo-humidiphilous (TH) mollusks from Dongwan (Li et al., 2008), (i) hematite/goethite ratio from sediments of the South China Sea (Clift, 2006), (j) smectite/kaolinite ratio from the South China Sea (Wan et al., 2010; Clift et al., 2014).

Table. 1. Average values and coefficients of variation of the geophysical and geochemical data for the XSZ section

		$CaCO_3$(%)	CaO(%)	K_2O(%)	Na_2O(%)	Sr(ppm)	Rb(ppm)
3.9-3.6 Ma	Average	15.5	12.8	3.0	1.25	228.1	106.6
	CV	16.0	12.5	10.8	9.3	5.8	4.6
4.7-3.9 Ma	Average	13.3	11.2	3.1	1.21	210.4	111.0
	CV	53.6	45.3	13.8	10.5	14.0	12.1
6.7-4.7 Ma	Average	17.1	11.2	2.6	1.22	211.7	103.9
	CV	28.2	31.7	10.3	20.9	9.9	10.8
		χ_{hf}	χ_{lf}	χ_{fd}	χ_{pedo}	Rb/Sr	K_2O/Na_2O
3.9-3.6 Ma	Average	21.9	22.9	0.95	8.7	0.47	2.36
	CV	20.6	21.3	67.0	67.0	8.7	11.1
4.7-3.9 Ma	Average	27.4	29	1.6	14.5	0.55	2.58
	CV	36.2	37.8	78.9	78.9	25.0	20.9
6.7-4.7 Ma	Average	19.4	20.3	1.0	9.1	0.49	2.21
	CV	21.0	22.4	72.8	72.8	15.8	25.1

Table. 2. Correlation coefficients for geochemical data for the XSZ section

Pearson correlation	CaO	$CaCO_3$	K_2O
CaO	1	0.51	-0.67
Na_2O	-0.06	-0.10	-0.38
K_2O	-0.67	-0.47	1
Rb	-0.20	-0.36	0.12
Sr	0.24	0.34	-0.29
$CaCO_3$	0.51	1	-0.47

Table. 3. Results of a significance test for the correlations between CaO*, $CaCO_3$ and Sr

Pearson correlation	CaO*	$CaCO_3$
Sr	0.67**	0.34
sig.(2-tailed)	0.000	0.063
n	163	163

**Correlation is significant at the 0.01 level