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 the valuable comments and suggestions from four anonymous reviewers. We have canceled the subdivision of climate stages and modified the boundary of humid period from 4.8 Ma to 4.7 Ma. Below please find the detailed responses.

Response to Report #1

This is my second round of review of this manuscript. Compared with the last version, the revised manuscript is significantly improved. I have no major concerns but some minor questions and suggestions as listed below.

Our response: Many thanks for reviewing our manuscript again. We specially thank you for the valuable questions and suggestions. They have really helped improve our paper. Here are responses.

P82-84: Red clay indicates aridification? And why this indicates uplift of TP? The logics here are not clear. In the next sentence, the authors state that red clay could reflect either arid or humid climate.

Our response: Yes, the wide deposition of red clay during the late Miocene indicates an increased dust source area (related to the Asian aridification) and enhancement of the East Asian Winter Monsoon. It does not mean a limited local aridification but rather a generalized Asian aridification (Sun et al., 1998; Li et al., 2017). Asian aridification is not evidence for the uplift of the TP but it is related to the uplift. This expression is ambiguous. We remove "related to the uplift of the TP".

P103: what are the climate trends in the northern TP? There is no description. Maybe "central Asia" or "NW China" is more accurate?

Our response: Thanks for pointing it out. We would modify the "northern TP" to "central Asia".

P112: "climatic characteristics"? what does this mean?

Our response: The statement is ambiguous. We would modify the "The distinctive geomorphological and climatic characteristics of the XSZ red clay sequence differentiates it from the main CLP red clay, and " to " Due to its specific geographical location, the XSZ red clay ".

P124: better to show "Maxianshan" and "Longzhong Basin" in Fig. 1c.

Our response: We indicate the "Maxianshan" and "Longzhong Basin" in Fig.1c.

P147: From Fig. 1b, carbonate nodule horizons are more abundant in the upper part, e.g., < 4.8 Ma. So does appearance of carbonate horizons also reflect climate changes?

Our response: Yes, the Bk horizons with larger carbonate content consist of carbonate nodule horizons underlying leached zones in the field indicate a significant translocation of carbonate minerals from Bw horizons to deeper Bk horizon due to greater rainfall (He et al., 2013).

P192: I can understand that larger CV indicates more variable climate, but how could you infer more humid from CV?

Our response: Larger CV is not evidence for a more humid climate. A humid period is reflected in high

frequency occurrence of leaching (Bw) horizons with low carbonate content (< 8 %), a larger carbonate content contrast between Bw horizons and accumulation (Bk) horizons and intermittent enhancement of magnetic susceptibility. The Bk horizons with larger carbonate content consisting of carbonate nodule horizons underlying leached zones in the field indicate the significant translocation of carbonate minerals from Bw horizons to deeper Bk horizon due to greater rainfall.

P193: The depositional rate is not a good evidence here. As there are also other abrupt drops or increase. Why do you pick the 4.8 Ma out? I suggest to delete this evidence.

Our response: Thanks for your suggestion. We will take it into consideration.

L331: which part of Fig.5? There are many sub-parts in Fig.5, better to denote each clearly.

Our response: Many thanks. We modify "Fig.5" as "Fig.5 c".

L345-346: From Table 3, comparing the two intervals, the Rb/Sr difference is only 0.02 and the K₂O/Na₂O difference is 0.14, no more than 3.5 % of the exhibited value range (e.g., 0.3-0.9 for Rb/Sr; and 1-5 for K₂O/Na₂O). Does this small difference truly reflect climatic difference? What are the analytical errors of these parameters? Could this small difference be interpreted as analytical errors, or the small difference actually indicate no difference between interval I and interval II.

Our response: Firstly, the measurement error ranged from 0.1%-0.3%. Secondly, changes in chemical weathering indices show some similarity with the carbonate content that weathering intensity increased in Bw horizons and remarkably decreased in Bk horizons, which means that the differences between the two intervals are not analytical errors. Thirdly, the unusually low weathering indices values of interval II resulting from translocation of mobile elements from Bw horizons to deeper Bk horizons make the difference between the two intervals small.

L348-350: Here, the authors indicate that the <2um/>40um ratio is characterized by low values. However, this contradicts with values presented in Table 3. This is apparently a false claim.

Our response: Thanks for pointing it out. We would consider the limitations of the <2um/>40um ratio as a pedogenesis index. We add the statement: "Noticeably, during this interval of peak precipitation (4.6-4.25 Ma), the enhancement of $<2 \mu m />40 \mu m$ 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" after "carbonate accumulation" in line 441.

L435: apparently, from Table 3, the <2um/>40um ratio is low during interval II

Our response: We consider the limitation of the <2um/>40um ratio as a pedogenesis index. We add the statement "Noticeably, during this interval of peak precipitation (4.6-4.25 Ma), the enhancement of $<2 \mu m />40 \mu m$ 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" after "carbonate accumulation" in line 441.

Response to Report #2

The authors investigate a new red-clay serious with special regard to palaeoclimate reconstruction in a critical region of the Tibetan Plateau. The authors find an abrupt change in precipitation character (to wetter and seasonally more varied conditions) after 4.8Ma, which they interpret as an increased

westward reach of the East Asian Summer Monsoon (EASM). This work is important as it helps constrain this westward extension of EASM on the Tibetan Plateau during the late Miocene and Pliocene. Our response: Many thanks for reviewing our manuscript and giving approval to our work. Here, we provide some quick replies to your questions.

General Comments:

Overall, the manuscript is well structured and well written. However, important methodical details are omitted in the presentation of results. The language is good, although minor mistakes and occasional poor phrasing can be found throughout the manuscript. I recommend proofreading of a proficient English speaker. My major concern pertains to the lack of use or description of statistics and signal processing. The main example of this is the seemingly arbitrary subdivision of the record into Interval I and Interval II. I suggest either finding a more robust way for change point detection, or refraining from this subdivision altogether. The broad implications of this study remain the same even without the somewhat unnecessary subdivision. Furthermore, the term significant is used several times, but it is unclear how significance was determined. Furthermore, it is unclear how the statistical dependence between several variables was established. I urge the authors to take advantage of the existing tools in statistics to substantiate such claims. There needs to be a section (in "3. Materials and Methods") describing these methods.

Our response: Many thanks for the valuable suggestions which we take into consideration in the revised version. We have canceled the subdivision of climate stages. We add the statement: "After interpolation to a 3-kyr sampling interval we performed spectral analysis on detrended records of carbonate content and χ_{pedo} using Redfit bases on a Lomb-Scargle Fourier transform combined with a Welch-Overlapped Segment Averaging procedure. We applied Gaussian band-pass filters at frequency of 0.09090-0.01111, 0.02174-0.02778 and 0.04167-0.05556 kyr-1 to extract oscillations associated with the 100-kyr, 41-kyr and 21-kyr periodicities. The significance of correlation is based on two-tailed test" after "4kyr or less" in line183.

Lastly, while the inclusion of discussion of possible climate change drivers is important, I urge the authors to highlight the uncertainties of these more, since only limited evidence is presented here.

Our response: Thanks for your valuable suggestions. We add statement: "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 and SSTs of the low latitude Pacific Ocean in the early Pliocene" at the end of line 546.

Note that I cannot comment on details about sample preparation and methods pertaining to proxy reconstructions as those lie outside my fields of expertise. I strongly recommend that other reviewers with

complementary expertise comments on these to compensate.

While I do believe this work to be valuable to several geoscientific communities, I can recommend publication only after the points above have been adequately addressed and a reviewer with a stronger geochemical background has commented on the manuscript.

Additional Specific Comments:

In the introduction, the authors correctly point out many problems of using the Pliocene as an analogue for future climate. An additional factor compromising the Pliocene as an analogue are the differences in palaeotopography.

L 50 "in" instead of "on" Our response: Many thanks. We modify "on" to "in".

L63 It should say "contrast" (not plural) to be consistent with the rest of the sentence, and maybe using "gradient" would be less confusing. What zonal gradients are you referring to? Do you simply mean zonal differences (differences along the same latitudes)? In this case, the term gradient may be a tad misleading, as it is used to describe the slope or one directional change as you have in case of meridional gradients.

Our response: Thanks for your suggestions. We modify the "contrasts" in line 64 to "contrast". Zonal gradient refers to the sea surface temperature contrast between the eastern and western equatorial Pacific. We modify "low east-west sea surface temperature gradient" in line 66 to "minor east-west surface temperature contrast".

L74 "latter" instead of "later"

Our response: Many thanks. We modify "later" to "latter".

L75-76 By "structural changes", I assume you are referring to spatial structure and mean that the regional expressions of global climate change were highly varied? Our response: Yes. We add "spatial" before "structural" in line 75.

L76 change to "... the regional climate is like ..."

Our response: We add "was" before "like".

L84 "was enhanced" instead of "enhanced"

Our response: We add "was" before "enhance".

L96 needs some rephrasing

Our response: We modify "waterlogging" to "dissolution of ferrimagnetic minerals" in line 95.

L112 What are these distinct geomorphological and climatic characteristics? Do you mean the above

described geographical and climatic setting?

Our response: Yes. The statement is ambiguous. We modify the "The distinctive geomorphological and climatic characteristics of the XSZ red clay sequence differentiates it from the main CLP red clay, and " to "Due to its specific geographical location, the XSZ red clay ".

L187 Does the coefficient of variation change significantly for ALL of the records? How was this established? By looking at it purely qualitatively, I would subdivide the section above the division up again into a higher variability lower part and lower variability upper part (that is similar to what is below the currently drawn line). In other words, the subdivision into interval I and interval II seems rather arbitrary. It looks to me like there is only a brief period of higher variability from 15-10m, interrupting the period of relatively low variability.

Our response: Many thanks for your questions. In fact, other reviewers have mentioned this problem before. We have been looking for evidence to show that two periods are different but we seem to be failed. The prime reason why we subdivide the records into two intervals is for the sake of discussion. We now remove the subdivision and highlight the humid and more variable climate period from ~16-5 m (4.7-3.9 Ma). We modify the statement of lines 186-195 to "Profiles of the various environmental proxies are illustrated in Figure 3. Notably, there is evidence for a relatively wet interval from ~16-5 m (4.7-3.9 Ma) which is reflected in the high-frequency occurrence of Bw horizons with a low carbonate content (< 8 %) and intermittent enhancement of magnetic susceptibility. There is a large contrast in carbonate content between Bw horizons and accumulation (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)."

L264 What correlation analysis is this based on and how was significance determined?

Our response: It is based on a two-tailed test. We now give the significance test table for correlation between CaO^* , $CaCO_3$ and Sr (Table 3).

L342 How was the significance of this change determined (see above)?

Our response: The "significant" used here is qualitative and subjective. We would remove the subdivision of climate stages and remove the sentence of lines 342-344.

L393 Again, how was this relationship and significance determined?

Our response: It is based on a t-test. The statement may be inappropriate. We remove "a significant" and "at 80% confidence interval".

Fig. 1c: Please use standard units like km and hPa, not miles and mb

Our response: We modify it.

References

- 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.
- 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., Ye, X.Y., Liu, S. P., Wang, X. X.: Late Miocene-Pliocene geomorphological evolution of the Xiaoshuizi peneplain in the Maxian Mountains and its tectonic significance for the northeastern Tibetan

Plateau. Geomorphology. 295,393-405, 2017.

Sun, D.H., An, Z.S., Shaw, J., Bloemendal, J., Sun, Y.B.: Magnetostratigraphy and palaeoclimatic significance of Late Tertiary aeolian sequences in the Chinese Loess Plateau. Geophysical Journal International, 134(1):207-212, 1998.

Other modifications

1. Modifications in abstract and introduction

#1: In line 34, "6.7-4.8 Ma" is modified to "late Miocene".

#2: In line 38, "occurred during 4.8-3.6 Ma" is modified to "occurred intermittently during 4.7-3.9 Ma".

#3: In line 39, "4.8" is modified to "4.7".

#4: In line 50, "on" is modified to "in".

#5: In line 64, "contrasts" is modified to "contrast".

#6: In line 67, "low" is modified to "minor" and "gradient" is modified to "contrast".

#7: In line 74, "later" is modified to "latter".

#8: In line 76, "spatial" is added before "change".

#9: In line 86, "related to the uplift of the TP" is modified to "was".

#10: In line 98, "waterlogging" is modified to "dissolution of ferrimagnetic minerals".

#11: In line 106, "northern TP" is modified to "central Asia".

#12: In line 114, "The distinctive geomorphological and climatic characteristics of the XSZ red clay sequence differentiates it from the main CLP red clay, and " is modified to " Due to its specific geographical location, the XSZ red clay ".

#13: In line 158, "was" is modified to "were pre-treated with 10% H_2O_2 to remove organic material, with 10% HCl to remove carbonates, and with 0.05 mol/L of (NaPO₃)₆ for dispersion. They were then".

2. Modifications in material and methods

#1: In lines 150-151, "red layers" is modified to "red soil layers (Bw) characterized by loam and moderate medium angular blocky structure."

#2: In line 166, "with an error of 0.1%-0.3%" is added after "PW2403".

#3: In line 188, "After interpolation to a 3-kyr sampling interval, we performed spectral analysis on detrended records of carbonate content and χ_{pedo} using Redfit, based on the Lomb-Scargle Fourier transform combined with a Welch-Overlapped Segment averaging procedure. We applied Gaussian band-pass filters at frequencies of 0.09090-0.01111, 0.02174-0.02778 and 0.04167-0.05556 kyr⁻¹ to extract oscillations associated with the 100-kyr, 41-kyr and 21-kyr periodicities, respectively. The significance of the correlations is based on a two-tailed test" was added after "4 kyr or less".

3. Modifications in results

#1: In lines 196-206, "and there is an obvious difference in the character of the fluctuations above and below the depth of 16.5 m (~ 4.8 Ma). Above 16.5 m, the carbonate content fluctuates at a lower level but with greater amplitude accompanied by the noted increase in nodule horizons underlaying leached zones in the field, and the magnetic susceptibility also fluctuates at greater amplitude" is modified to "Notably, there is evidence for a relatively wet interval from ~16-5 m (4.7-3.9 Ma) which is reflected in the high-frequency occurrence of leaching (Bw) horizons with a low carbonate content (< 8 %) and intermittent enhancement of magnetic susceptibility. There is a large contrast in carbonate content between Bw horizons and accumulation (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)." #2: In line 207, "above the boundary than below" is modified to "during this interval than in other intervals" #3: In line 208, "It suggests" is modified to "These various forms of evidence suggest".

#4: In line 209, "after 4.8 Ma" is modified to "during 4.7-3.9 Ma" and sentence of lines 209-211 was removed. #5: The statement of lines 215-228 is modified to "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%). Leaching-accumulation horizons (Bw-Bk) 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."

#6: The statement of lines 230-240 is modified to " K_2O ranges from 1.9-3.7% with an average of 2.8%; Na₂O 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 changes of Sr show some similarity with magnetic susceptibility prior to 4.7 Ma but with CaO after 4.7 Ma."

#7: In line 241, "Accordingly, table 2" is modified to "Reference to Table 2".

#8: In lines 242-243, "From 16-5 m, CaO and Sr exhibit low values in Bk horizons and high values in Bw horizons, while the opposite is the case for K_2O and Rb" was added.

#9: The statement of lines 244-247 is modified to "Finally, during 4.7-3.9 Ma, the amplitudes of the fluctuations in CaO, K_2O , Sr and Rb are greater than in the other intervals."

#10: The statement of lines 249-269 is modified to "Variations of χ_{hf} , χ_{lf} and χ_{fd} are synchronous. χ_{hf} ranges from 9.6-53.9×10⁻⁸ m³/kg with an average of 21.8×10⁻⁸ m³/kg; χ_{lf} ranges from 11.4-59.0×10⁻⁸ m³/kg with an average of 23.1×10⁻⁸ m³/kg; and χ_{fd} ranges from 0-4.7×10⁻⁸ m³/kg with an average of 1.2×10⁻⁸ m³/kg. From 42-16 m, three magnetic parameters show relatively flat and low values: χ_{hf} ranges from 9.6-33.3×10⁻⁸ m³/kg; and χ_{fd} ranges from 11.4-36.1×10⁻⁸ m³/kg with an average of 20.3×10⁻⁸ m³/kg; and χ_{fd} ranges from 0-2.8×10⁻⁸ m³/kg with an average of 1.0×10⁻⁸ m³/kg. From 16-5 m, the values and amplitudes of three parameters are high: χ_{hf} ranges from 13.8-53.9×10⁻⁸ m³/kg; and χ_{fd} ranges from 0-4.7×10⁻⁸ m³/kg. From 16-5 m, the values and amplitudes of three parameters are high: χ_{hf} ranges from 13.8-53.9×10⁻⁸ m³/kg; and χ_{fd} ranges from 0-4.7×10⁻⁸ m³/kg with an average of 1.6×10⁻⁸ m³/kg. From 16-15 m, 13-11 m and 7-5 m, the values of the three parameters obviously increase. From 5-0 m, both the value and amplitudes of three parameters decrease: χ_{hf} ranges from 12.8-32.9×10⁻⁸ m³/kg; with an average of 22.0×10⁻⁸ m³/kg; χ_{lf} ranges from 13.6-34.6×10⁻⁸ m³/kg. The fluctuation of magnetic susceptibility is substantially different from that of carbonate content which indicates enhancement of magnetic susceptibility did not caused by leaching of the carbonate."

#11: The sentence of lines 271-273 is modified to "The clay content ($\leq 2 \mu m$) ranges from 3.8-13.5% with an average of 8.1%; and the >40 um content ranges from 0.7-13.5% with an average of 6%."

#12: In line 274, "which correspond to peaks in magnetic susceptibility" is added after "6 m".

#13: In line 276, "as well as to other proxies described above" is removed and "In addition, from 21-5 m the fluctuations in the >40 um fraction are roughly the inverse to those of magnetic susceptibility" is added.

#14: In line 278, "From 6.7-4.8 Ma" is modified to "From 42-21m".

#15: In line 279, "4.8 Ma" is modified to "21m".

4. Modifications in the discussion

#1: In line 347, "Fig. 5" is modified to "Fig. 5 a-b".

#2: In line 352, "4.8-3.9 Ma (Fig. 5 d)" is modified to "4.7-3.9 Ma (Fig. 5 d)".

#3: In lines 374-375, all of the "4.8 Ma" are modified to "4.7 Ma" and "Fig. 5" is modified to "Fig. 5d".

#4: In line 383, "6.7-4.8Ma" is modified to "the late Miocene".

#5: In line 385-388, "As shown in Figure 6 and Table 3, there is a significant change in most of the proxies

(carbonate, Rb/Sr, K2O/Na2O and xpedo) near 4.8-4.7 Ma, and therefore the climatic record can be divided into

two intervals. During interval I (6.7-4.8 Ma)," is modified to "During the late Miocene".

#6: In line 393, "<2 µm/>40 µm ratio" is removed.

#7: In line 421, "interval of 6.7-4.8 Ma" is modified to "the late Miocene".

#8: In line 347, "a significant" is removed.

#9: In line 348, "at 80 % confidence interval (Fig. 4 f)" is modified to "(Fig. 4 e)".

#10: In line 441, "from 6.7-4.8 Ma" is modified to "during the late Miocene".

#11: In line 445, "from 6.7-4.8 Ma" is modified to "during the late Miocene".

#12: In line 469, "Humid climate with pronounced fluctuations during 4.8-3.6 Ma" is modified to

"Intermittently humid climate during the early Pliocene".

#13: In line 470, "interval II (4.8-3.6 Ma)" is modified to "the early Pliocene".

#14: In line 471, "from ~4.7 Ma" is added after "humid".

#15: In lines 473-474, all of "4.8 Ma" are modified to "4.7 Ma".

#16: In lines 475, "7%" and "20%" are modified to "8%" and "21%" respectively.

#17: The sentence of the lines 485-486 is modified to "Seasonal precipitation was intermittently enhanced from

4.7-3.9 Ma, and so was weathering and pedogenic intensity".

#18: In lines 489-491, "Notably, during this interval of peak precipitation (4.6-4.25 Ma), the enhancement of the $<2 \mu m />40 \mu m$ 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" is added.

#19: In line 498, "4.8-3.6 Ma" is modified to "the early Pliocene".

#20: In line 523, "4.8 Ma" is modified to "4.7 Ma".

#21: In line 523, "the" is added before "early".

#22: In line 547, "4.8 Ma" is modified to "4.7 Ma".

#23: In line 549, "et al" is removed.

#24: In lines 586-588, "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)" is added after "extention". #25: In line 599, " may have" is added before "facilitated".

#26: In line 600, the statement of "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 and SSTs of the low latitude Pacific Ocean in the early Pliocene" is added.

5. Modifications in coclusion

#1: In line 613, "two interval of" is modified to "the".

#2: In line 614, "the first interval (6.7-4.8 Ma)," is modified to "the late Miocene".

#3: In line 617, "the second interval (4.8-3.6 Ma)," is modified to "the early Pliocene".

#4: In line 619, "was large" is modified to "increased from 4.7-3.9 Ma".

6. Modifications in references

References of "Bacon, C. D., Silvestro, D., Jaramillo, C., Smith, B. T., Chakrabarty, P., and Antonelli, A.: Biological evidence supports an early and complex emergence of the Isthmus of Panama. Proceedings of the National Academy of Sciences,112(19), 6110-6115, 2015" and "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" are added.

7. Modifications in Figures and tables

#1: In Fig. 1 c, "Longzhong Basin" and "Maxianshan" are added.

#2: Fig. 3 and Fig.4 are replaced by new Fig.3.and Fig.4 respectively.

#3: In Fig. 5c, the dash line is removed.

#4: In Fig. 6, the dash line is removed and labels are used in each graph.

#5: In Fig. 7, the boundary line is removed.

#6: Table1 Table2 and Table3 are all replaced with new Tables.

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

Best regards,

Jijun Li

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

The Pliocene climate and its driving mechanisms have attracted substantial scientific 24 25 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 26 reconstruct the history of interior Asian aridification and the Asian monsoon. However, red 27 clay sequences deposited on the planation surface of the Tibetan Plateau (TP) are rare. A 28 continuous red clay sequence was recently discovered on the uplifted Xiaoshuizi (XSZ) 29 planation surface in the Maxian Mountains, northeastern (NE) TP. In this study, we analyzed 30 31 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 32 differences between the central and western CLP. Our results demonstrate the occurrence of 33 minimal weathering and pedogenesis during -6.7-4.8 Mathe late Miocene, which indicates 34 that the climate was arid. We speculate that precipitation delivered by the palaeo- East Asian 35 Summer Monsoon (EASM) was limited during this period, and that the intensification of the 36 37 westerlies circulation resulted in arid conditions in the study region. Subsequently, enhanced weathering and pedogenesis occurred intermittently during 4.87-3.6-9 Ma, which attests to an 38 39 increase in effective moisture. We ascribe the arid-humid climatic transition near ~ 4.87 Ma to the expansion of the palaeo-EASM. Increasing Arctic temperatures, the poleward 40 expansion of the tropical warm pool into subtropical regions, and the freshening of the 41 subtropical Pacific in response to the closure of the Panamanian Seaway, may have been 42 responsible for the thermodynamical enhancement of the palaeo-EASM system, which 43 permitted more moisture to be transported to the NE TP. 44

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

47

48 **1. Introduction**

The Pliocene, including the Zanclean (5.33-3.60 Ma) and Piacenzian (3.60-2.58 Ma) 49 stages, is one of the most intensively studied intervals of the pre-Quaternary on-in climate 50 change research. The Zanclean climate was generally warm and wet and is often used as an 51 52 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 53 tropical region (Herbert et al., 2010, 2016). On the other hand, the Zanclean was markedly 54 different from today, although several critical changes in thermohaline and atmospheric 55 circulation towards modern conditions were occurring (Haug et al., 2005; Lawrence et al., 56 2006; Chaisson and Ravelo, 2000). For example, the early-Pliocene global mean 57 temperature was approximately 4 °C warmer (Brierley and Fedorov, 2010), and the sea 58 levels are estimated to have been ~ 25 m higher, than today (Dowsett et al., 2010). 59 60 Temperatures at high northern latitudes were considerably higher and therefore continental 61 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 62 Hemisphere were weak but gradually became more intensified, changing towards the 63 modern state which has a much more pronounced spatial temperature contrasts (Fedorov et 64 al., 2013; Brierley et al., 2009, 2010). The low meridional surface temperature gradient 65

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

80 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 81 climate change, and the evolution of global temperature and ice volume (An et al., 2001; 82 Ding et al., 2001; Li et al., 2008; Clift et al., 2008; Nie et al., 2014; Ao et al., 2016; Sun et 83 al., 2006a, 2017; Chang et al., 2013; Liu et al., 2014). Previous research has revealed that 84 red clay was widely deposited across the CLP since the late Miocene, indicating that Asian 85 aridification related to the uplift of the TPwas enhanced (Guo et al., 2001; Song et al., 2007; 86 An et al., 2014; Ao et al., 2016; Li et al., 2017). In the eastern and central CLP, where the 87

88	climate is dominated by the East Asian Monsoon, palaeontological evidence, mineral
89	magnetic parameters and geochemical records from the red clay indicate dry climatic
90	conditions during the late Miocene but generally wet climatic conditions during the early
91	Pliocene (Wang et al., 2006; Guo et al., 2001; Wu et al., 2006; Song et al., 2007; Sun et al.,
92	2010; An et al., 2014; Ao et al., 2016). The most controversial climatic change occurred
93	during the interval from 4.8-4.1 Ma, for which climate reconstructions using different
94	proxies indicate conflicting palaeo-environmental trends. For example, field observations
95	and pollen records indicate an intensified summer monsoon intensity, but low magnetic
96	susceptibility values are more consistent with arid rather than wet climatic conditions (Ding
97	et al., 2001; Ma et al., 2005; Song et al., 2007; Sun et al., 2010). It is thought that dissolution
98	of ferrimagnetic minerals waterlogging and iron reduction resulting from high precipitation
99	significantly affected the climatic significance of magnetic susceptibility records during this
100	period (Ding et al., 2001). In addition to the East Asian Monsoon, the westerlies also had an
101	impact on the climate of East Asia; however, the patterns of climate change in the
102	westerlies-dominated regions were different from the eastern and central CLP during the
103	early Pliocene. Geochemical, stratigraphic and pollen evidence from the Qaidam and Tarim
104	basins has demonstrated that aridification intensified since the early Pliocene (Fang et al.,
105	2008; Sun et al., 2006a, 2017; Chang et al., 2013; Liu et al., 2014). Although the general
106	climatic trends of the main CLP and northern TPcentral Asia during this period are well
107	recorded, palaeoclimatic changes in the NE TP, which is at the junction of the zones of
108	westerlies and monsoonal influences, remain unclear. Therefore, determining the climatic
109	conditions of the NE TP during the early Pliocene not only improves our understanding of

110 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. 111 A continuous red clay sequence was recently discovered on the uplifted XSZ planation 112 surface in the NE TP and has been dated via high-resolution magnetostratigraphy (Li et al., 113 2017). Due to its specific geographical location, the XSZ red clay The distinctive 114 geomorphological and climatic characteristics of the XSZ red clay sequence differentiates it 115 from the main CLP red clay, and provides the opportunity to reveal the late Miocene-early 116 Pliocene climate history of the NE TP and to determine the climatic differences between the 117 central and western CLP. In this study, we measured multiple climatic proxies from the late 118 Miocene-Pliocene XSZ red clay core. Our aims were to construct a detailed record of 119 precipitation, chemical weathering and pedogenesis during 6.7-3.6 Ma; and to determine the 120 pattern of regional climate evolution and its possible causal mechanisms. 121 122 2. Regional background 123 The XSZ planation surface is located in Yuzhong County in the western Chinese Loess 124 Plateau (Fig. 1). The main XSZ planation surface is at an altitude of 2800 m in the Maxian 125 Mountains where it has truncated Precambrian gneiss. The Maxianshan are rejuvenated 126

mountains where it has indicated freedinorian gleiss. The Maxianshal 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 biomagnetostratigraphic 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 132 reconstruct and discuss the patterns of regional climate change during the Miocene-Pliocene. 133 134 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 135 zone at the junction of the eastern monsoon area, the arid area of northwest China, and the 136 cold region of the TP. The mean annual temperature during 1986-2016 was ~7.0 °C and the 137 annual precipitation was 260-550 mm; 80% of the precipitation is in summer and autumn 138 (data source: National Meteorological Information Center (http://data.cma.cn/) of the Chinese 139 Meteorological Administration). The spatial distribution of precipitation is uneven, 140 decreasing from south to north in Yuzhong County. Precipitation amount increases with 141 elevation at the rate of 27 mm per 100 m, attaining a maximum of 800 mm at the top of 142 143 Maxianshan.

144

145 **3. Material and methods**

The XSZ core (35.8115 N, 103.8623 °E and 2758.1 m above sea level) is composed of 146 42 m of pure red clay and ~3 m of red clay and there is an increasing content of angular 147 gravel at the base (Fig. 1 b). The red clay interval is composed of brownish red and yellowish 148 clay layers (Fig. 2). The upper 20 m contains numerous horizontal carbonate nodule horizons 149 (Bk), most of which underlie brownish red soil layers (Bw) characterized by loam and 150 moderate medium angular blocky structure. There are also occasional carbonized plant root 151 channels, elliptical worm burrows and fossil snail shell fragments. Fe-Mn stains are more 152 frequent in the brownish layers than in the yellowish layers, which is also the case for the 153

154 carbonized root channels. The red clay across the XSZ planation surface is similar to that of
155 typical eolian red clay in the CLP; both are characterized by numerous carbonate nodule-rich
156 horizons (Fig. 2 b).

Samples for grain-size, carbonate content and magnetic susceptibility measurements 157 were taken at 5-cm intervals, and samples for geochemical analysis were collected at 25-cm 158 intervals. Samples for grain-size measurements waswere pre-treated with 10% H₂O₂ to 159 remove organic material, with 10% HCl to remove carbonates, and with 0.05 mol/L of 160 (NaPO₃)₆ for dispersion. They were then measured with a Malvern Mastersizer 2000 grain-161 162 size analyzer with a detection range of 0.02-2000 µm. Magnetic susceptibility was measured using a Bartington Instruments MS2 meter and MS2B dual-frequency sensor at two 163 frequencies (470 Hz and 4700 Hz, designated χ_{lf} and χ_{hf} , respectively). Three measurements 164 165 were made at each frequency and the final results were averaged. The frequency-dependent magnetic susceptibility (χ_{fd}) was calculated as $\chi_{lf}-\chi_{hf}$. Chemical composition was measured 166 via X-ray fluorescence using a Panalytical Magix PW2403 with an error of 0.1%-0.3%. The 167 sample preparation procedure for XRF analysis was as follows: Bulk samples were heated to 168 35° C for 7 days and then ground with an agate mortar to pass a 75-µm sieve; ~4 g of 169 powdered sample was then pressed into a pellet with a borate coating using a semiautomatic 170 oil-hydraulic laboratory press (model YYJ-40). All the measurements were conducted at the 171 MOE Key Laboratory of Western China's Environmental Systems, Lanzhou University. 172

Silicate-bound CaO (CaO*) can be estimated, in principle, by the equation: CaO*(mol) $= CaO(mol) - CO_2(calcite mol) - 0.5 CO_2(dolomite mol) - 10/3 P_2O_5(apatite mol) (Fedo et al., 1995).$ It is generally calculated based on the assumption that all the P_2O_5 is associated with apatite and all the inorganic carbon is associated with carbonates. Thus, the CaO* of the XSZ red clay was calculated using the following equivalent equation:

178

$$CaO^{*}(mol) = CaO(mol) - CaCO_{3}(mol) - \frac{10}{3} * \frac{P_{2}O_{5}}{M(P_{2}O_{5})}$$

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

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

184

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

Each sample age was estimated using linear interpolation to derive absolute ages, 185 186 constrained by our previous magnetostratigraphic study (Fig. 1). The average temporal resolution of the records is 3.8 kyr. Some 80 % of the sequence has a sampling resolution of 4 187 kyr or less. After interpolation to a 3-kyr sampling interval, we performed spectral analysis 188 on detrended records of carbonate content and χ_{pedo} using Redfit, based on the Lomb-Scargle 189 Fourier transform combined with a Welch-Overlapped Segment averaging procedure. We 190 applied Gaussian band-pass filters at frequencies of 0.09090-0.01111, 0.02174-0.02778 and 191 0.04167-0.05556 kyr⁻¹ to extract oscillations associated with the 100-kyr, 41-kyr and 21-kyr 192 periodicities, respectively. The significance of the correlations is based on a two-tailed test. 193 194 4. Results 195 Profiles of the various environmental proxies are illustrated in Figure 3.- and Notably, 196

197	there is an obvious evidence for a relatively wet interval difference in the character of the
198	fluctuations from above and below the depth of ~16.5 m-5 m (~4.87-3.9 Ma) - Above 16.5 m,
199	which is reflected in the high-frequency occurrence of Bw horizons with a low carbonate
200	content (< 8 %) and intermittent enhancement of magnetic susceptibility. There is a large
201	contrast in carbonate content between Bw and Bk horizons, which corresponds to variations
202	in elemental contents. The Bk horizons, with a higher carbonate content, the carbonate
203	content fluctuates at a lower level but with greater amplitude accompanied by consist of
204	carbonate the noted increase in nodule layers underlying leached zones in the field indicate
205	the substantial translocation of carbonate minerals from Bw horizons to Bk horizons due to
206	greater rainfall (He et al., 2013)., and the magnetic susceptibility also fluctuates at greater
207	amplitude. In addition, the CV of most of the records is greater during this interval above the
208	boundary than belowthan in other intervals (Table 1). It-These various forms of evidence
209	suggests that the climate became more humid and variable after 4.8 Maduring 4.7-3.9 Ma.
210	Meanwhile, a noticeable drop in deposition rate around 4.8 Ma occurred (Li et al., 2017).
211	Thus, the red clay sequence was divided into two intervals: Interval I (6.7-4.8 Ma) and
212	Interval II (4.8-3.6 Ma). The characteristics of the individual proxy records are described in
213	detail below.
214	
215	Carbonate content
216	During Interval I, Tthe carbonate content of the entire core fluctuates from 3.81.6-39.2%
217	with - and has an high-average (17.4%) of 15.9 %. From 42-16 m, the average carbonate

218 <u>content is high (17.1%)</u> and the carbonate content decreases upwards.; <u>t</u> The contrast in the

219	carbonate content between the leach layers <u>Bw</u> and accumulation layers <u>Bk horizons</u> is
220	generally low; for the Bw horizons, the <u>carbonate content is ~12% and</u> values <8% are rare.
221	Bk horizons, with a carbonate content of around or above 21%, are frequent and the
222	carbonate content decreases upwards (Fig. 3). From 29-16.5 m, the fluctuations are of greater
223	amplitude than during 42-29 m. From During Interval <u>H</u> 16-5 m, there are fluctuations in
224	carbonate content of a-large amplitude (1.6-39.1%) but the average value is low
225	(13.83%)From 16.5-4.5 m there are several <u>Bw-Bk horizons are frequent;</u> the Bw horizons
226	have a carbonate content of $<78\%$, while that of the <u>leached layersBk_horizons</u> is $>2021\%$.
227	From the accumulation layers 5-0 m, the average carbonate content increases to 15.5%; Bw
228	horizons with a carbonate content <8% is absent, and the carbonate content contrast between
229	the Bw and Bk horizons is low.
230	Element geochemistry
231	The XSZ red clay consists mainly of SiO ₂ , Al ₂ O ₃ , CaO and Fe ₂ O ₃ with low
232	concentrations (<5%) of MgO, K ₂ O, Na ₂ O, Sr, Rb and Ba (Table 1). During Interval [], K ₂ O
233	ranges from 1.9-3. $\frac{37}{9}$ % with an average of 2. $\frac{68}{9}$ %; Na ₂ O ranges from 0.14-1. $\frac{5254}{9}$ % with an
233 234	ranges from 1.9-3.37% with an average of 2.68%; Na ₂ O ranges from 0.14-1.5254% with an average of 1.2%; Rb ranges from $\frac{8074}{125}$ ppm with an average of 103.96.2 ppm; and
233 234 235	ranges from 1.9-3.37% with an average of 2.68%; Na ₂ O ranges from 0.14-1.5254% with an average of 1.2%; Rb ranges from $\frac{8074}{125}$ ppm with an average of $103.96.2$ ppm; and Sr ranges from $\frac{150}{252141}$ ppm with an average of $\frac{211.7212.8}{212.8}$ ppm. During Interval II,
233 234 235 236	ranges from 1.9-3. <u>37</u> % with an average of 2. <u>68</u> %; Na ₂ O ranges from 0.14-1. <u>5254</u> % with an average of 1.2%; Rb ranges from <u>8074</u> - <u>125-134</u> ppm with an average of 10 <u>3.96.2</u> ppm; and Sr ranges from <u>150-252141-281</u> ppm with an average of <u>211.7212.8</u> ppm. <u>During <i>Interval II</i></u> , <u>K₂O ranges from 2-3.7% with an average content of 3%. Na₂O ranges from 0.94-1.54 % with</u>
 233 234 235 236 237 	ranges from 1.9-3. <u>37</u> % with an average of 2. <u>68</u> %; Na ₂ O ranges from 0.14-1. <u>5254</u> % with an average of 1.2%; Rb ranges from <u>8074-125-134</u> ppm with an average of 10 <u>3.96.2</u> ppm; and Sr ranges from <u>150-252141-281</u> ppm with an average of <u>211.7212.8</u> ppm. <u>During <i>Interval II</i></u> , K ₂ O ranges from 2 3.7% with an average content of 3%. Na ₂ O ranges from 0.94-1.54 % with an average content of 1.23%. Rb ranges from 74-134 ppm with an average content of 109.9
 233 234 235 236 237 238 	ranges from 1.9-3. <u>37</u> % with an average of 2. <u>68</u> %; Na ₂ O ranges from 0.14-1. <u>5254</u> % with an average of 1.2%; Rb ranges from <u>8074-125-134</u> ppm with an average of 10 <u>3.96.2</u> ppm; and Sr ranges from <u>150-252141-281</u> ppm with an average of <u>211.7212.8</u> ppm. <u>During <i>Interval II</i></u> , K ₂ O ranges from 2 3.7% with an average content of 3%. Na ₂ O ranges from 0.94-1.54 % with an average content of 1.23%. Rb ranges from 74-134 ppm with an average content of 109.9 ppm. Sr ranges from 141-281 ppm with an average content of 214.6 ppm. The variations in
 233 234 235 236 237 238 239 	ranges from 1.9-3.37% with an average of 2.68%; Na ₂ O ranges from 0.14-1.5254% with an average of 1.2%; Rb ranges from 8074-125-134 ppm with an average of 103.96.2 ppm; and Sr ranges from 150-252141-281 ppm with an average of 211.7212.8 ppm. During <i>Interval II</i> , K ₂ O ranges from 2 3.7% with an average content of 3%. Na ₂ O ranges from 0.94-1.54% with an average content of 1.23%. Rb ranges from 74-134 ppm with an average content of 109.9 ppm. Sr ranges from 141-281 ppm with an average content of 214.6 ppm. The variations in CaO exhibit the same trend as carbonate content_with high values in Bk horizons and low
 233 234 235 236 237 238 239 240 	ranges from 1.9-3.37% with an average of 2.68%; Na ₂ O ranges from 0.14-1.5254% with an average of 1.2%; Rb ranges from 8074-125-134 ppm with an average of 103.96.2 ppm; and Sr ranges from 150-252141-281 ppm with an average of 211.7212.8 ppm. During Interval II, K ₂ O ranges from 2-3.7% with an average content of 3%. Na ₂ O ranges from 0.94-1.54% with an average content of 1.23%. Rb ranges from 74-134 ppm with an average content of 109.9 ppm. Sr ranges from 141-281 ppm with an average content of 214.6 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 changes of Sr show some similarity with magnetic susceptibility

241	before 4.8 The variations in Rb and K ₂ O are synchronous and roughly inverse to those of CaO
242	The changes of Sr show some similarity with magnetic susceptibility prior to 4.7 Ma but with
243	CaO after 4.7 Ma. Accordingly, Reference to table 2 Table 2 shows that CaO is positively
244	correlated with $CaCO_3$ and Sr, and negatively correlated with the other elements. From 16-5
245	m, CaO and Sr exhibit low values in Bw horizons and high values in Bk horizons, while the
246	opposite is the case for K_2O and Rb. Finally, from 4.8 3.6 Ma16-5 m, the amplitudes of the
247	fluctuations in CaO, K_2O , Sr and Rb are greater than in the other intervals. those during 6.7-
248	4.8 Ma, which is also indicated by the CV of these elements (Table 1)
249	Magnetic susceptibility
250	The Magnetic susceptibility also shows pronounced differences between the two
251	intervals (Fig. 3). During Interval I , variations of $\chi_{hf_*}\chi_{lf}$ and χ_{fd} are synchronous. χ_{hf} ranges
252	from 9.6-53.9×10 ⁻⁸ m ³ /kg with an average of 21.8×10 ⁻⁸ m ³ /kg; χ_{lf} ranges from 11.4-59.0×10 ⁻⁸
253	m^{3}/kg with an average of 23.1×10 ⁻⁸ m ³ /kg; and χ_{fd} ranges from 0-4.7×10 ⁻⁸ m ³ /kg with an
254	average of 1.2×10^{-8} m ³ /kg. From 42-16 m, the three magnetic parameters are relatively low
255	and uniform. χ_{hf} ranges from 9.6-33.3×10 ⁻⁸ m ³ /kg with an average of 19.4×10 ⁻⁸ m ³ /kg; χ_{lf}
256	ranges from 11.4-36.1×10 ⁻⁸ m ³ /kg with an average of 20.3×10^{-8} m ³ /kg; and χ_{fd} ranges from 0-
257	2.8×10^{-8} m ³ /kg with an average of 1.0×10^{-8} m ³ /kg. From During Interval II, <u>16-5 m</u> , the
258	<u>values</u> of the <u>three parameters</u> , together with their amplitudes of variation, are <u>high</u> . χ_{hf} ranges
259	from $12.83.8$ -53.9×10 ⁻⁸ m ³ /kg with an average of $25.47.4$ ×10 ⁻⁸ m ³ /kg; χ_{lf} ranges from $13.64.2$ -
260	59.0×10^{-8} m ³ /kg with an average of $\frac{26.929.0}{29.0} \times 10^{-8}$ m ³ /kg; and χ_{fd} ranges from 0-4.7×10 ⁻⁸
261	m ³ /kg with an average of 1.26×10^{-8} m ³ /kg. Within the intervals of Clearly, the average values
262	of the three parameters are higher during Interval II than during Interval I. The amplitudes

263	of the fluctuations in the three parameters during Interval II are also larger than those during
264	Interval I. 16-15 m, 13-11 m and 7-5 m, the values of the three parametersare high increase
265	substantially. From 5-0 m, both the values and amplitudes of variation of the three parameters
266	decrease. χ_{hf} ranges from 12.8-32.9×10 ⁻⁸ m ³ /kg with an average of 22.0×10 ⁻⁸ m ³ /kg; χ_{lf} ranges
267	from 13.6-34.6×10 ⁻⁸ m ³ /kg with an average of 22.9×10 ⁻⁸ m ³ /kg; and χ_{fd} ranges from 0-2.5×10 ⁻¹
268	$\frac{8}{10}$ m ³ /kg with an average of 1×10^{-8} m ³ /kg. Overall, the fluctuations in magnetic susceptibility
269	are substantially different to those of carbonate content which indicates that the enhancement
270	of magnetic susceptibility was not caused by carbonate leaching.
271	Grain size
272	The average-clay content (<2 μ m) ranges from 3.8-13.5% with an average of is 8.217%;
273	and the ≥ 40 um content ranges from 0.7-13.9% with an average of 6%. during Interval I
274	and 8.0% during Interval-II. The fluctuations in clay content are minor, except for maxima at
275	about 15 m, 12 m and 6 m, which correspond to peaks in magnetic susceptibility (Fig. 3). The
276	coarse silt component (>40 µm), mainly carried by the East Asian winter monsoon, exhibits a
277	different trend to that of the clay content, as well as to other proxies described above. In
278	addition, from 21-5 m the fluctuations in the >40 um fraction are roughly the inverse to those
279	of magnetic susceptibility. From $\frac{6.742}{4.8}$ Ma 21 m, the variation of the >40 µm fraction is
280	characterized by low values and high-frequency fluctuations, whereas above 4.8 Ma21 m it
281	exhibits high values and fluctuations of lower frequency.
282	

5. Discussion 283

5.1 Palaeoenvironmental interpretation of the proxies 284

285 The carbonate content of aeolian sediments can be readily remobilized and deposited in responses to changes in precipitation and evaporation intensity and thus is sensitive to 286 changing climatic conditions. Previous studies demonstrated that the carbonate content of 287 loess-red clay sequences of the CLP varies with precipitation (Fang et al., 1999; Sun et al., 288 <u>2010</u>). The carbonate is mainly derived from a mixture of airborne dusts (Fang et al., 1999). 289 Soil micromorphological evidence from the Lanzhou loess demonstrates that the carbonate 290 grains in loess are little altered, whereas those in the palaeosols have undergone a reduction 291 in size as a result of leaching and reprecipitation as secondary carbonate in the lower Bk 292 horizons (Fang et al., 1994, 1999). Furthermore, seasonal alternations between wet and dry 293 conditions are thought to be a key factor driving carbonate dissolution and reprecipitation 294 (Sun et al., 2010). Thus, changes in carbonate content are generally controlled by the 295 296 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 297 oscillations as well as summer monsoon evolution (Fang et al., 1999; Sun et al., 2010). 298

Chemical weathering intensity is generally evaluated by the ratio of mobile (e.g. K, Ca, 299 Sr and Na) to non-mobile elements (e.g. Al and Rb). In general, Sr shows analogous 300 geochemical behavior to Ca and is readily released into solution and mobilized in the course 301 of weathering; by contrast, Rb is relatively immobile under moderate weathering conditions 302 due to its strong adsorption to clay minerals (Nesbitt et al., 1980; Liu et al., 1993). Thus, the 303 Rb/Sr ratio potentially reflects chemical weathering intensity. However, Sr may substitute for 304 305 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 306

307	significant at the 99% confidence interval, while the correlation between Sr and CaCO ₃ is not
308	significant. This means that the variations in Sr are determined by weathering intensity, and
309	therefore we speculate that in our samples the Rb/Sr ratio mainly reflects weathering intensity
310	(Fig. 4 d-c and ed). In addition, the K_2O/Na_2O ratio is used to evaluate the secondary clay
311	content in loess and is also a measure of plagioclase weathering, avoiding biases due to
312	uncertainties in separating carbonate Ca from silicate Ca (Liu et al., 1993; Buggle et al.,
313	<u>2011</u>). Na ₂ O is mainly produced by plagioclase weathering and is easily lost during leaching
314	as precipitation increases. By contrast, K_2O (mainly produced by the weathering of potash
315	feldspar) is easily leached from primary minerals and is then absorbed by secondary clay
316	minerals with ongoing weathering (Yang et al., 2006; Liang et al., 2013). In the arid and
317	semi-arid regions of Asia, K ₂ O is enriched in palaeosols compared to loess horizons (Yang et
318	<u>al., 2006</u>). Thus, high K_2O/Na_2O ratios are indicative of intense chemical weathering.
319	In the red clay-loess sequence of the CLP, magnetic parameters and the clay content are
320	well correlated and thus are regarded as proxies of EASM strength (Liu et al., 2004). Aeolian
321	particles usually have two distinct magnetic components, consisting of detrital and pedogenic
322	material, respectively (Liu et al., 2004). χ_{1f} can reflect the combined susceptibility of both
323	components, but changes in χ_{1f} are mainly affected by changes in the concentration of
324	pedogenic magnetic grains (Liu et al., 2004). The grain-size distribution of pedogenic
325	particles within the superparamagnetic (SP) to single-domain (SD) size range has been shown
326	to be constant (Liu et al., 2004, 2005). Thus, χ_{fd} can be used detect SP minerals produced by
327	pedogenesis and therefore the correlation coefficient between χ_{lf} and χ_{fd} is a measure of the
328	contribution of such grains (<0.03 μ m for magnetite) to the bulk susceptibility (<u>Liu et al.</u> ,

329	<u>2004; Xia et al., 2014</u>). As shown in Figure 4a, χ_{lf} is positively correlated with χ_{fd} , which
330	means that the magnetic susceptibility of the XSZ red clay mainly reflects pedogenic
331	enhancement of the primary aeolian ferromagnetic content via the in-situ formation of fine-
332	grained ferrimagnetic material. Thus, the magnetic susceptibility primarily reflects pedogenic
333	intensity. Both the original and pedogenic magnetic signals can be separated using a simple
334	linear regression method (Liu et al., 2004; Xia et al., 2014), which we use to extract the
335	lithogenic (χ_0) and pedogenic magnetite/maghemite (χ_{pedo}) components. We found that
336	pedogenic magnetite/maghemite accounts for 11% of the susceptibility ($\chi_{pedo} = \chi_{fd} / 0.11$).
337	Pedogenesis results in enhanced secondary clay formation (Sun and Huang, 2006b);
338	however, not all of the clay particles are derived from in situ pedogenesis, but rather are
339	inherited from aeolian transport and deposition. Clay particles can adhere to coarser silt and
340	sand particles (Sun and Huang, 2006b). In the western CLP, the coarse silt (>40 μ m) content
341	is regarded as a rough proxy of winter monsoon strength (Wang et al., 2002). Therefore, to
342	eliminate this signal from the primary clay particles, the <2 μ m/>40 μ m ratio can be used is
343	<u>proposed</u> to evaluate pedogenic intensity. Furthermore, the similarity of the variations of <2
344	μ m/>40 μ m ratio and χ_{pedo} confirms that in this case both proxies are sensitive to <2 μ m/>40
345	<u>um ratio has the potential to evaluate the pedogenic intensity (Fig. 6).</u>
346	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 347 cycles (Fig. 5<u>a-b</u>). In detail, χ_{pedo} is concentrated in the eccentricity (100 kyr), obliquity (41 348 kyr) and precession (21 kyr) bands and other periodicities (71 kyr and 27 kyr) are also 349 evident. By contrast, the carbonate signal is concentrated in the precession (21 kyr) and 350

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obliquity (41 kyr) bands, but it also exhibits even more prominent periodicities of 56 kyr and 30 kyr. Furthermore, the fluctuations in CaCO₃, weathering and pedogenesis indices agree well with orbital eccentricity variations during 4.87-3.9 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 373 three filtered components (especially the 41-kyr component) of both proxies change from a 374 low amplitude during 6.7-4.8-7 Ma to a relatively high amplitude during 4.87-3.9 Ma (Fig. 5 375 c). The enhanced orbital-scale variability of the two proxies from 4.87-3.9 Ma implies 376 increased seasonality and wet-dry contrasts. This shift is not observed in the Earth orbital 377 parameters but is observed in the filtered 41-kyr component of the stacked deep-sea benthic 378 oxygen isotope record (δ^{18} O). This may mean that the enhancement of wet-dry contrasts at 379 the XSZ site was not driven directly by changes in solar radiation intensity but rather was 380 381 linked with changes in ice volume or global temperature.

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

384 5.3.1 Multi-proxy evidence for a dry climate during 6.7-4.8 Mathe late Miocene

We used the proxies of pedogenesis and chemical weathering to reconstruct the late 385 Miocene and early Pliocene climatic history of the Xiaoshuizi planation surface. As shown in 386 Figure 6 and Table 3, there is a significant change in most of the proxies (carbonate, Rb/Sr, 387 K_2O/Na_2O and χ_{pedo}) near 4.8-4.7 Ma, and therefore the climatic record can be divided into 388 two intervals. During interval I (6.7-4.8 Ma) the late Miocene, the relatively high carbonate 389 values with minor fluctuations indicate that the climate was dry, and low Rb/Sr and 390 K₂O/Na₂O ratios also support the occurrence of weak chemical weathering. Notably, both the 391 Rb/Sr and K₂O/Na₂O ratios show opposite trends to that of carbonate content, meaning that 392 low effective precipitation resulted in weak chemical weathering. Furthermore, the pedogenic 393 proxies ($<2 \mu m/>40 \mu m$ ratio, χ_{pedo} and χ_{lf}), characterised by low values with minor 394

395 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 396 chemical weathering and pedogenic intensity. However, there are several subtle differences 397 between the carbonate and pedogenic indexes. It is evident that the carbonate content 398 decreases with an increased amplitude of variation after 5.5 Ma, which is consistent with the 399 cycles of carbonate nodules within paleososol horizons observed in the field (Li et al., 2017). 400 It is possible that increased precipitation since 5.5 Ma induced eluviation and the redeposition 401 of carbonate. However, the pedogenic indexes indicate that the generally arid climate was 402 403 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 404 carbonate content to precipitation variability when precipitation is low (Sun et al., 2010). In 405 addition, a coeval mollusk record from the western Liupanshan showed that cold-aridiphilous 406 species dominated, which also indicates that cold and dry climatic conditions occurred in the 407 western CLP during the late Miocene (Fig. 7 g). 408

409 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 410 al., 2014). However, the principal difference is that at the XSZ site, the arid climate was 411 relatively stable, while the climate of the central and eastern CLP was interrupted by several 412 humid stages. For example, two humid stages (6.2-5.8 Ma and 5.4-4.9 Ma) are recorded by 413 the magnetic susceptibility of the red clay in the central and eastern CLP but are absent in the 414 magnetic susceptibility record at the XSZ site (Fig. 7). Notably, the 41-kyr filtered 415 component of thermo-humidiphilous species from Dongwan was damped in the late Miocene 416

417 (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 6.7-4.8-7 Ma. However, 418 the three periodicities in the Summer Monsoon index from the central CLP show no obvious 419 difference between the late Miocene and Pliocene, but only a slight reduction in variability 420 after 4.2 Ma (Sun et al., 2010). Therefore, we agree that a dry climate prevailed on the CLP 421 during the interval of 6.7-4.8 Malate Miocene; however, the difference was that the climate in 422 the central and eastern CLP fluctuated more substantially than was the case in the vicinity of 423 the XSZ red clay section. 424

425 The especially damped response of the wet-dry climatic oscillations in the western CLP to obliquity forcing may indicate that the influence of the palaeo-EASM in the western CLP 426 was negligible. It is widely known that the summer monsoon intensity decreases from 427 428 southeast to northwest across the CLP. A regional climate model experiment demonstrated that the modern East Asian Summer Monsoon was not fully established in the late Miocene 429 and had only a small impact on northern China (Tang et al., 2011). A weak palaeo-EASM 430 431 intensity from 7.0-4.8 Ma was revealed by hematite/goethite and smectite/kaolinite ratios at ODP Site 1148 in the South China Sea (SCS) (Fig. 7 i and j). Therefore, we infer that the 432 palaeo-EASM was weak and had only a minor impact on the climate in the study region. In 433 addition, previous studies indicated that the red clay may have been transported by both low-434 level northerly winds and the upper-level westerlies (Sun et al., 2004; Vandenberghe et al., 435 2004) and thus the impact of the westerly circulation on the study region cannot be ignored. Notably, the variation of the pedogenic proxies roughly parallels that of the stacked deep-sea benthic for aminiferal oxygen isotope curve (Fig. 6), and that that χ_{pedo} has a a significant 438

439 positive relationship with δ^{18} O at 80 % confidence interval (Fig. 4 fc). This indicates that 440 when the global temperature was low, pedogenic intensity in the study area increased. It is 441 unreasonable to conclude that precipitation in the study area was dominated by the palaeo-442 EASM and thus we speculate that, from 6.7-4.8 Maduring the late Miocene, precipitation 443 transported by the palaeo-EASM was limited and that the westerly circulation probably 444 dominated the regional climate.

The simultaneous reduction in amplitude of the 41-kyr filtered components from the western CLP and the deep sea δ^{18} O record from 6.7-4.8 Ma during the late Miocene likely indicates that the dry climate was related to changes in global temperature and ice volume. A sustained cooling occurred in both hemispheres during the late Miocene which culminated between 7 and 5.4 Ma (Herbert et al., 2016). δ^{18} O records from DSDP and ODP sites show an increase of ~1.0‰ during the late Miocene which resulted from the increased ice volume and the associated decrease in global temperature (Zachos et al., 2001). In the Northern Hemisphere, transient glaciations occurred when the cooling culminated (Herbert et al., 2016). Records from high-latitude regions of the Northern Hemisphere indicate continuously decreasing temperatures and increasing ice volume during the late Miocene (Jansen and Sigholm, 1991; Mudie and Helgason, 1983; Haug et al., 2005). During the Quaternary, a dry climate prevailed during glacial periods when the global average temperature (especially in summer) was low. Cool summers could result in a small land-sea thermal contrast which in turn weakened the palaeo-EASM. Furthermore, the increased ice volume in the Northern Hemisphere resulted in an increased meridional temperature gradient (Herbert et al., 2016), thus strengthening the westerlies and driving them southwards. This would have prevented

the northwestward penetration of the Asian Summer Monsoon, which is also proposed as the driving mechanism for a weak EASM in northern China during glacial periods (<u>Sun et al.</u>, <u>2015</u>). Thus, the southward shift of the westerlies had a significant impact on the XSZ region. However, moisture sources for the westerly air flow are distant from the CLP (<u>Nie et al.</u>, <u>2014</u>), and only a relatively small amount of moisture was carried to the CLP, resulting in a dry and stable climate in the XSZ region. In conclusion, global cooling and increasing ice volume in the Northern Hemisphere contributed to the dry climatic conditions in the study region.

5.3.2 Intermittently hHumid climate during the early Pliocene 4.8-3.6 Ma

During interval II (4.8-3.6 Ma) the early Pliocene, the proxy evidence indicates that the previously arid climate 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 4.87-3.9 Ma. Several eluvial-illuvial cycles are evident during 4.87-3.9 Ma; the carbonate content in the eluvial horizons was less than $\frac{78}{8}$, whereas in illuvial horizons it was $>\frac{2021}{6}$ (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 K₂O/Na₂O ratios are very similar to those of carbonate content,

483	which suggests that weathering intensity was related to precipitation amount. Generally, high
484	values of the <2 μm / >40 μm ratio, χ_{pedo} and χ_{lf} correspond to large contrasts in carbonate
485	content between eluvial and illuvial horizons; thus, increased precipitation had a significant
486	influence on pedogenic intensity. High pSeasonal precipitation was persisted intermittently
487	enhanced from 4.87-3.9 Ma, and so was weathering and pedogenic intensity were strong.
488	Pedogenesis and weathering intensity reached a maximum during 4.60-4.25 Ma, as did
489	precipitation intensity, manifested by the enhanced eluviation and carbonate accumulation.
490	Notably, during this interval of peak precipitation (4.6-4.25 Ma), the enhancement of the <2
491	μ m />40 μ m ratio is not as strong as that of χ_{pedo} , which may indicate that the former is of
492	limited value when pedogenic intensity is strong. During 3.9-3.6 Ma, precipitation decreased,
493	and weathering and pedogenic intensity also weakened. Consistent with the records of the
494	XSZ section, mollusk records from Dongwan also indicate the occurrence of warm and
495	humid conditions in the western CLP during the early Pliocene (Fig. 7 h).
496	Palynological and terrestrial mollusk records from the central CLP also indicate
497	relatively humid conditions during the early Pliocene (Wang et al., 2006; Wu et al., 2006).
498	Magnetic susceptibility records from the central and eastern CLP are similar to that from the
499	XSZ section in that both the magnitude and the variability are high during 4.8-3.6 Mathe
500	early Pliocene. From 4.1-3.9 Ma, the increased magnetic susceptibility indicates that humid
501	climatic conditions prevailed across the entire CLP (Fig. 7). Evidently, when precipitation
502	amount peaked in the vicinity of the XSZ section during 4.60-4.25 Ma, the magnetic
503	susceptibility values at Xifeng, Lingtai and Chaona were low. However, a record of Fe ₂ O ₃
504	ratio from Lingtai reveals extremely high values, corresponding to the presence of abundant

clay coatings, during 4.8-4.1 Ma and this interval was interpreted as experiencing the 505 strongest EASM intensity in the CLP since 7.0 Ma (Ding et al., 2001). In addition, the 506 relative intensity of pedogenic alteration of the grain-size distribution was the strongest 507 during the interval from 4.8-4.2 Ma in the Lingtai section (Sun et al., 2006c). Pollen 508 assemblages at Chaona indicate a substantially warmer and more humid climate from 4.61-509 4.07 Ma (Ma et al., 2005). These various lines of evidence indicate that during 4.60-4.25 Ma 510 the climate was warm and humid in the central CLP. Gleying has been implicated in reducing 511 the value of magnetic susceptibility as a record of precipitation during this period (Ding et al., 512 2001). When soil moisture regularly exceeds the critical value, dissolution of ferrimagnetic 513 minerals occurs and the susceptibility signal is negatively correlated with pedogenesis (Liu et 514 al., 2003). This alone indicates that precipitation was likely to have been very high during this 515 516 interval.

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 ~4.8-7_Ma (Fig. 7 i and j) and thus the enhancement of the palaeo-EASM (<u>Clift et al., 2006,</u> <u>2014</u>). 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 δ^{18} O record at about 4.8–7 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 527 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 528 pedogenic indices at the XSZ section indicate that strong pedogenic intensity corresponded 529 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 (Zhang et al., 2012). From 8.26-4.96 Ma, massive deltaic conglomerates were widely deposited and the sediment deposition rate increased, indicating the uplift of the Qilian Mountains (Song et al., 2001). At the same time, the Laji Mountains underwent pronounced uplift by thrusting at ~8 Ma, which resulted in the current basin-range pattern (Li et al., 1991; Fang et al., 2005a; Zheng et al., 2000). However, geological and palaeontological records indicate that the uplift of the eastern and northern margins of the TP was very minor from the late Miocene to the middle Pliocene (Li et al., 1991, 2015; Zheng et al., 2000; Fang et al., 2005a, 2005b). Therefore, we speculate that uplift of the TP was not the major cause of the expansion of the palaeo-EASM at $\sim 4.8-7$ Ma. 548

549 The occurrence of a humid climate across the CLP was synchronous with the gradual closure of the Panama Seaway (Keigwin-et al., 1978; O'Dea et al., 2016). Nie (2014) 550 proposed that the freshening of Eastern Equatorial and North Pacific surface water, resulting 551 from the closure of the Panama Seaway since 4.8 Ma (Haug et al., 2001), led to sea ice 552 formation in the North Pacific Ocean, which enhanced the high-pressure cell over the Pacific 553 and increased the strength of southerly and southeasterly winds. However, there was a 554 warming trend in the Northern Hemisphere from 4.6 Ma (Haug et al., 2005; Lawrence et al., 555 2006). The gradual closure of the Panama Seaway resulted in the reorganization of surface 556 557 currents in the Atlantic Ocean. Notably, the Gulf Stream was enhanced and began to transport warm surface waters to high northern latitudes, thus strengthening the Atlantic meridional 558 overturning circulation and warming the Arctic (Haug and Tiedemann, 1998; Haug et al., 559 560 2005). Three independent proxies from an early Pliocene peat deposit in the Canadian High Arctic indicate that Arctic temperatures were 19 °C warmer during the early Pliocene than 561 today (Ballantyne et al., 2010). This warmth is also confirmed by other records from high 562 northern latitude regions: diatom abundances and assemblages, pollen data, magnetic 563 susceptibility and sedimentological evidence from Siberia all indicate that the climate was 564 warm and wet in the early Pliocene (Baikal Drilling Project Memb, Memb B. D. P., 1997, 565 1999). The warming of the northern high latitude region led to increases in summer 566 temperature in the mid-latitudes of Eurasia. However, equatorial SSTs remained stable or 567 cooled slightly (Brierley et al., 2009; Fedorov et al., 2013), and thus the land-ocean thermal 568 contrast was intensified. Furthermore, external heating derived from a reduced ice albedo at 569 high northern latitudes also enhanced the thermal contrast between the Pacific and Eurasian 570

571	regions (Dowsett et al., 2010). This large land-ocean thermal contrast was essential for
572	enhancing the palaeo-EASM. On the other hand, the unusually warm Arctic and small
573	meridional heat gradient in the Northern Hemisphere pushed the Intertropical Convergence
574	Zone northwards (Chang et al., 2013; Sun et al., 2015), which weakened the westerly
575	circulation and thus facilitated the northwestward expansion of the palaeo-EASM.
576	Figure 6 shows that high values of pedogenic indices in the XSZ section correspond
577	with high SSTs in the EEP. This appears to be contradictory to the case of the modern ENSO
578	(when the EEP temperature is high, the precipitation amount in the western CLP is low). The
579	discrepancy may indicate that the nature of sea-air interactions during the early Pliocene was
580	different from today. During 4.8-4.0 Ma, the thermohaline circulation was reorganizing and
581	creating a precondition for the development of the modern equatorial Pacific cold tongue
582	(Chaisson and Ravelo, 2000). Several crucial changes linked with the summer monsoon
583	occurred: There was a vast expansion of the western Pacific warm pool into subtropical
584	regions in the early Pliocene (Brierley et al., 2009; Fedorov et al., 2013), and temperatures at
585	the edge of the warm pool showed a warming trend of ~2 $^{\circ}$ C from the latest Miocene to the
586	early Pliocene (Karas et al., 2011). This enhanced thermal state of the WEP warm pool
587	significantly enhanced the summer monsoon and its northward extension. A modelling
588	experiment indicates that the precipitation of the CLP would increase when the tropical warm
589	pool expended into subtropical region (Brierley et al., 2009). Today, when the northern part
590	of the western pacific warm pool is warm, convection over and around the Philippines is
591	enhanced; in addition, the northern extent of the western Pacific subtropical-high shifts
592	northwards from the Yangtze River valley to the Yellow River valley and moisture is

593	introduced across the entire CLP (Huang et al., 2003). Further research is needed to
594	determine if this was also the case during the early Pliocene. However, the warming and
595	freshening of the subtropical Pacific would have promoted increased evaporation which
596	would have provided enhanced moisture for the palaeo-EASM, resulting in increased rainfall
597	across the CLP.
598	
599	In conclusion, we infer that the warming of high northern latitudes, accompanied by the
600	vast poleward expansion of the tropical warm pool into subtropical regions and the
601	freshening of the subtropical Pacific, may have facilitated the expansion of the palaeo-EASM
602	during the early Pliocene. However, there are several uncertainties associated with such an
603	explanation. For example, the timing of the closure of the Panama Seaway is still debated
604	(Bacon et al., 2015; O'Dea et al., 2016), and it is unclear how strongly these changes
605	influenced the palaeo-EASM. Addressing these questions requires more geological evidence
606	and precise model simulations of the early Pliocene climate. The value of our study lies in
607	proposing the potential linkage of the evolution of palaeo-EASM and changes in
608	temperatures of high northern latitudes and SSTs of the low latitude Pacific Ocean in the
609	early Pliocene.
610	
611	6. Conclusions
612	The continuous late Miocene-Pliocene red clay sequence preserved on the planation
613	surface in the NE Tibetan Plateau provides the opportunity to elucidate the history of the
614	Asian monsoon in the western CLP. Multi-proxy records from the XSZ section, together with
615	other paleoclimatic records from the CLP, reveal -two intervals of the major patterns of

616 climatic change from 6.7-3.6 Ma. During the first interval (6.7-4.8 Ma) the late Miocene, both the amount and variability of precipitation over the XSZ section were small; however, they 617 618 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 second interval (4.8-3.6 Ma)early 619 Pliocene, the records from the XSZ section indicate that both the amount and variability of 620 precipitation were large increased from 4.7-3.9 Ma. The climate was characterized by abrupt 621 increases in the seasonality of precipitation, which attests to a major northwestward extension 622 and enhancement of the summer monsoon. Multiple paleoclimatic proxies clearly show that 623 624 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, the vast poleward expansion 625 of the tropical warm pool into subtropical regions, and the freshening of the subtropical 626 Pacific, in response to the closure of the Panamanian Seaway during the early Pliocene. 627

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Figures and tables



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 $\chi_{\rm tf}$ versus $\chi_{\rm tfd}$. (b) Scatter plot of $\chi_{\rm tf}$ versus $\chi_{\rm pede}$ during 4.8-3.6 Ma. – (c) Scatter plot of $\chi_{\rm tf}$ versus $\chi_{\rm pede}$ during 6.7-4.8 Ma. (d) Scatter plot of Sr versus CaCO₃. (e) Scatter plot of Sr versus CaO*. (f) Scatter plot of benthic δ^{18} O versus $\chi_{\rm pede}$ during 6.7-4.8 Ma. (g) Separation of $\chi_{\rm pede}$ and χ_{0} . Solid squares and triangles are the average values during 4.8-3.6 Ma and 6.7-4.8 Ma, respectively. $\chi_{\rm pede}$ is the magnetic susceptibility of pedogenic origin and χ_{0} is the magnetic susceptibility of the detrital material.



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 CaCO₃. (e) Scatter plot of benthic δ^{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) χ pedo 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, χ pedo and δ 18O 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 χ pedo 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.



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).