New chronology of the Chinese loess-paleosol sequence by leaf wax δD records during the past 800 k.y.

Zheng Wang1,2, Weiguo Liu1,2,3,*, Hong Wang4,*, Yunning Cao1,2, Jing Hu1,2, Jibao Dong1,2, Hongxuan Lu1,2, Huangye Wang1,2, Meng Xing1,2 and Hu Liu1,2
1State Key Laboratory of Loess and Quaternary Geology, Institute of Earth Environment, Chinese Academy of Sciences (CAS), Xi’an, 710061, China
2CAS Center for Excellence in Quaternary Science and Global Change, Xi’an, 710061, China
3University of Chinese Academy of Sciences, Beijing 100049, China
4Interdisciplinary Research Center of Earth Science Frontier, Beijing Normal University, Beijing, 100875, China

ABSTRACT

The Chinese loess-paleosol sequences provide important archives for studying paleoenvironmental changes. However, the lack of independent and accurate time scales hinders the study between loess and other records. Asian stalagmite δ18O records indicate synchronous patterns of paleoprecipitation δ18O over large geographic regions. The record of hydrogen isotopic composition of plant wax (δDwax) in Chinese loess is also controlled by rainfall δD. Both share a common origin. The linear relationship between rainfall δ18O and δD variance provides the basis to tie together chronologies of the same climate event in different records. Here, we show a new loess chronology by correlating chronologies of marker boundaries of prominent climate chronozones in stalagmite δ18O and summer insolation to the equivalent climate stratigraphy in the loess δDwax sequence. We first developed and tested this novel methodology with data since the last interglacial on a millennial scale, and then applied this approach to the loess δDwax sequence for the past 800 k.y. to improve the traditional chronology based on magnetic susceptibility and grain size. The new δDwax time series provides not only an improved chronology for studying paleoclimate changes during interglacial intervals, but also represents a unique database with which to better understand the links between the Asian monsoon changes in the Chinese loess and other global climate events, especially for the periods prior to 640 ka, for which stalagmite records are not available.

INTRODUCTION

Loess-paleosol sequences are widely distributed in Europe, North America, and Asia, of which the Chinese Loess Plateau (CLP) is the most extensive loess deposition region (Liu et al., 1991; An, 2014). Loess deposits, as the longest continuous continental records, along with marine sediment, stalagmite, and ice records, play key roles for better understanding global paleoenvironmental changes from millennial to orbital time scales. Based on the correspondence between loess-paleosol stratigraphic alternations and interglacial-glacial cycles, it is known that loess formed during glacial periods and paleosol formed during interglacial periods, and so orbital parameter chronologies are commonly used to tune time series of loess-paleosol sequences (Kukla et al., 1988; Ding et al., 1994; Lu et al., 1998; Zhou et al., 2014). For example, magnetic susceptibility and grain-size records, which are regarded as indexes of East Asian summer and winter monsoon intensity, have provided reasonable time controls for loess-based climate and environmental studies (Yang and Ding, 2003; Guo et al., 2009; Sun et al., 2012). However, dissolution of magnetic minerals and translocation of loess particle sizes, especially during interglacial intervals, could seriously alter the original depths of these proxies, which challenges the integrity of these climate time series (Liu et al., 2014; Torrent, 2005). Consequently, the lack of independent and accurate time scales hinders the comparison between loess and other global records in better understanding the details of paleoclimate and environmental changes and associated mechanisms.

Asian stalagmite δ18O values record rainfall δ18O variations over the past 640 k.y. with highly precise, independent, and radiometric chronology (Wang et al., 2008; Cheng et al., 2012; Cai et al., 2015), which demonstrates a tight correlation with the summer insolation over large regions of the Northern Hemisphere (Cheng et al., 2016a). Moreover, stalagmite δ18O demonstrates that the rainfall δ18O varied consistently in different regions throughout glacial-interglacial cycles, defined as a “supra-regional mode” (Cheng et al., 2016b). The loose soil structure inhibits pervasive forest cover over a large area of the CLP, where steppe and grasslands are common in the “Yuan” area (Li et al., 2003; Jiang et al., 2013; Sun et al., 2017). Loess hydrogen isotopic composition of plant wax (δDwax) also inherits the δD signal of rainfall (Sessions et al., 1999; Sachse et al., 2012). Although δDwax shows different fractionation between grasses and woody plants (Liu and Yang, 2008), the grassland-dominated plants of the CLP ensure that loess δDwax is not significantly affected by vegetation changes. Thus, the loess δDwax values mainly record the changes in rainfall isotope values, assuming that vegetation changes are not significant in order to change the fractionation in the δDwax. Asian stalagmite δ18O records as an indicator of rainfall δ18O, and loess δDwax records as an indicator of rainfall δD, should have a shared common origin of paleoprecipitation isotopes over monsoonal climate–affected regions, and over the CLP as well. Both δ18O variance and δD variance of rainwater events have a linear relationship when rain belts migrate with fractionalation (Craig, 1961), which provides the basis for correlating chronologies of stalagmite δ18O and summer insolation to the loess-paleosol δDwax sequence. Meanwhile, leaf wax is widely preserved in the loess sequences, which ensures that loess δDwax records can be easily obtained.

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Therefore, in theory, tuning loess $\delta D_{\text{wax}}$ time series by stalagmite $\delta^{18}O$ and summer insolation chronologies can provide a more reliable and accurate method of determining loess chronologies than magnetic susceptibility and grain-size proxies, for establishing a highly accurate loess-paleosol time-climate sequence.

In this study, we report a $\delta D_{\text{wax}}$ sequence spanning eight glacial-interglacial cycles to test the feasibility of establishing a new $\delta D_{\text{wax}}$ chronology in order to provide a more reliable and accurate time-climate series for the Chinese loess-paleosol sequence for the past 800 k.y. We also aim to detect discrepancies between the newly established loess $\delta D_{\text{wax}}$ record and the traditional magnetic susceptibility and grain-size time series, with a focus on sub-glacial-interglacial climate cycles.

RESULTS AND DISCUSSION

$\delta D_{\text{wax}}$ Record from Layer L8 Upward in the Central CLP

We collected samples from the S1 layer, which represents the last interglacial in the Xifeng loess section of the central CLP, at 5 cm increments, and samples from the L1 to S0 layers at 20 cm increments. The magnetic susceptibility record is consistent with previous reports (Fig. 1). According to the classical age-depth framework of the loess-paleosol sequence, stratigraphic units S1–S0 are linked to Marine Isotope Stages 5–1 (MIS 5–1). The average resolution of $\delta D_{\text{wax}}$ of n-alkane $C_{30}$ in the Xifeng section was $0.9 \pm 0.6$ k.y. per increment, yielding a millennial-scale climate change record.

The millennial-scale $\delta D_{\text{wax}}$ index in the Xifeng loess-paleosol sequence shows a series of periodic fluctuations with amplitudes obviously larger than those of the magnetic susceptibility and the grain-size variances. The fluctuations of the loess $\delta D_{\text{wax}}$ since the last interglacial represented by the S1 layer are comparable with the clear marker peaks in the stalagmite $\delta^{18}O$ curve (Wang et al., 2008; Cheng et al., 2012, 2016a; Cai et al., 2015), and the summer insolation since MIS 5e (Laskar et al., 2004). However, under the traditionally tuned time scale on the magnetic susceptibility and grain-size records, these marker peaks in the $\delta D_{\text{wax}}$ time series at the same depth from the CLP clearly lag behind the marker peaks of the stalagmite $\delta^{18}O$ index and summer insolation index (Fig. 1).

Following previous work (Liu and Huang, 2005; Z. Wang et al., 2018), we report new $\delta D_{\text{wax}}$ data from loess layers L8 to L3 (in 25 cm increments) in the central CLP and compile a complete $\delta D_{\text{wax}}$ data record from layer L8 upward in the central CLP. The $\delta D_{\text{wax}}$ values oscillate uniformly around $-185%$, with an amplitude of $\pm 15%$, indicating strong periodic fluctuations of paleoprecipitation $\delta D$ values for the past 800 k.y. (Fig. 2).

Synchrinicity of Precipitation Isotopes in the CLP are Essential for the New $\delta D_{\text{wax}}$ Chronology since the Last Interglacial

Loess $\delta D_{\text{wax}}$ values reflect the $\delta D$ of precipitation. Because vegetation ecosystem changes through time are minimal in the central CLP, and plant-affected isotopic fractionation would be minimal, loess $\delta D_{\text{wax}}$ records could directly represent the variation of signals of paleo precipitation isotopes.

Stalagmite $\delta^{18}O$ also reflects changes in precipitation isotopes. Note that stalagmite $\delta^{18}O$ values from southwest China (Cai et al., 2015), Central Asia (Cheng et al., 2012), and east China (Wang et al., 2008; Cheng et al., 2016a) all show variations of paleoprecipitation $\delta^{18}O$ in phase with the summer insolation (Fig. 1).

The loess $\delta D_{\text{wax}}$ fluctuations indicate at least six periodicities in Xifeng in the central CLP since the last interglacial (Fig. 1), which are synchronous with the loess $\delta D_{\text{wax}}$ variance in Weinan in the southern CLP (Thomas et al., 2016). Assuming loess $\delta D_{\text{wax}}$ varies synchronously within the CLP and with the stalagmite $\delta^{18}O$ and summer insolation, we tuned the age chronology of the Xifeng loess $\delta D_{\text{wax}}$ curve to match the stalagmite $\delta^{18}O$ and summer insolation parameter chronology for S1–S0. The comparison shows that the average lag time for the new chronology on the loess $\delta D_{\text{wax}}$ curve is $5.4 \pm 3.6$ k.y. behind the traditional magnetic susceptibility chronology, with a lag time range between $-1.8$ and $10.0$ k.y. (Fig. 1). Because the resolution of $\delta D_{\text{wax}}$ in the S1 layer is 0.9 k.y., there is a difference of 5 k.y. between the two chronologies of S1–S0 stratigraphy. For glacial-interglacial cycles, the age uncertainty of 5 k.y. is only 5% over a 100 k.y. period, which indicates that both the traditional magnetic susceptibility–based loess time scale and the loess $\delta D_{\text{wax}}$ time series are consistent. However, on the sub–glacial-interglacial scale, e.g., on precession periods, the 5 k.y. age uncertainty is large enough.
to impact research results. We recommend use of the $\delta D_{\text{wax}}$ chronology to replace the traditional magnetic susceptibility chronology for loess-paleosol sequences, especially for studying climate changes during the last interglacial interval.

**Loess $\delta D_{\text{wax}}$ Chronology in the CLP During the Past 800 k.y.**

We compared loess $\delta D_{\text{wax}}$ records of layers L2–S2 in Xifeng (Z. Wang et al., 2018) and Weinan (Thomas et al., 2016), which show in-phase fluctuations (Fig. S2 in the Supplemental Material1). Under the traditional chronology tuned by magnetic susceptibility values, the $\delta D_{\text{wax}}$ records display a lag phase compared to the chronologies of stalagmite $\delta^{18}O$ and summer insolation (Fig. S2). This means the chronological uncertainties of the traditional loess-paleosol sequence remain measurable not only in the younger, but also in the older, segments of the sequence.

To apply the method of tuning to older loess $\delta D_{\text{wax}}$ chronology, we correlated the chronologies of stalagmite and summer insolation to the $\delta D_{\text{wax}}$ record in Xifeng up to the L8 loess layer (Fig. 2). Based on the loess-paleosol stratigraphy corresponding to glacial-interglacial cycles, the loess $\delta D_{\text{wax}}$ and summer insolation records are synchronous (Fig. 2). Although the average 3.4 k.y. resolution of 25 cm increments is too low to identify climate events (indicated by low value peaks of the indexes in the lower paleosol stratigraphies), the possibility of synchronous climate events between the summer insolation and $\delta D_{\text{wax}}$ records up to layer L8 is still measurably clear.

Because stalagmite $\delta^{18}O$ records only date back to 640 ka, corresponding to loess-paleosol stratigraphy back to the bottom of layer S5, the summer insolation chronology was used to establish the chronology of the loess $\delta D_{\text{wax}}$ proxy below the S5 unit. We found that the loess $\delta D_{\text{wax}}$ variation, particularly, better matches the stalagmite $\delta^{18}O$ and summer insolation during interglacial periods than during glacial periods. Therefore, the loess $\delta D_{\text{wax}}$ time series has higher reliability in paleosol than in loess layers. Under the traditional chronology, an average 5 k.y. time lag occurs between low value peaks of $\delta D_{\text{wax}}$ and the corresponding maximum values of summer insolation, which is consistent with the 5 k.y. lag time above layer S1 (Fig. 2). Thus, the new chronology has significance for studying longer loess-paleosol sequences in the CLP. When the vegetation changes between grass and trees through time are taken into account, we recommend using loess $\delta D_{\text{wax}}$ time series as a more accurate age-controlled monsoon precipitation proxy to study loess-paleosol deposits and associated climate and environment changes.

**GEOLOGICAL IMPLICATION**

The analysis of loess records based on orbital cycles, such as eccentricity or precession, is a common method to demonstrate the driving factors of climatic or environmental changes in the CLP. The wavelet power spectrum analysis indicates that $\delta D_{\text{wax}}$ variation in Xifeng has clear periods of 23 k.y. and 41 k.y. in the newly tuned time series (Fig. S3). This means the chronological correspondences between the loess $\delta D_{\text{wax}}$ and marine or stalagmite records in other regions (Fig. 2). The phase differences between the loess records in the CLP and marine or stalagmite records in other regions are synchronous (indicated by low value peaks of the indexes in the lower paleosol stratigraphies) with the 5 k.y. lag time above layer S1 (Fig. 2). Thus, the new chronology has significance for studying longer loess-paleosol sequences in the CLP. When the vegetation changes between grass and trees through time are taken into account, we recommend using loess $\delta D_{\text{wax}}$ time series as a more accurate age-controlled monsoon precipitation proxy to study loess-paleosol deposits and associated climate and environment changes.

1Supplemental Material. Materials, methods, and interpretations, Figure S1 (site description and loess records from layer S1), Figure S2 (comparing $\delta D_{\text{wax}}$ of layer S2 in central and south CLP), Figure S3 (spectrum analysis of loess $\delta D_{\text{wax}}$), and Figure S4 (the differences between two age-models in paleosol and loess layers). Please visit https://doi.org/10.1130/GEOL.S.14233565 to access the supplemental material, and contact editing@geosociety.org with any questions.
et al., 2016). However, the new δD\text{wax} chronology shows that the traditional chronology of the loess-paleosol alternation has an ~5 k.y. time lag (Fig. 2), which leads to uncertainty of the phase difference by 80° on precession cycles. In some loess stratigraphy, the phase uncertainty due to the error of the traditional chronology (developed from either magnetic susceptibility or grain-size proxies) becomes as high as 180°. On the sub–glacial-interglacial scale, the inherent differences between the loess record and other records may have been partially or completely covered by the uncertainty of the traditional loess chronology. The error of the traditional chronology itself has actually upset the interpretation of existing climate-driven mechanisms.

The strong hydrophobicity of leaf wax ensures that loess δD\text{wax} will not be affected by precipitation leaching, which is an advantage over the traditional proxies. We believe that the high sensitivity of δD\text{wax} variations to monsoon rainfall strength over the CLP makes it the best proxy for tuning the Chinese loess-paleosol chronology, which will eliminate the uncertainty for comparison studies that seek the true inherent climate characteristics of the CLP. It also implies that magnetic susceptibility or grain-size chronologies are no longer the best option for loess and climate change research on sub–glacial-interglacial scales, and need to be replaced by the new δD\text{wax} chronology.

At present, a growing body of evidence shows that the δ18O and δD values on the Tibetan Plateau (Chen et al., 2020), adjacent to the CLP, also vary in a similar pattern as the stalagmite δ18O, though the local temperature and humidity obviously differ from region to region (Rao et al., 2016; Chen et al., 2020). This indirectly indicates that loess-paleosol alternation has an influence of the Indian monsoon precipitation over the past 250 ka: Journal of Asian Earth Sciences, v. 159, p. 292–305, https://doi.org/10.1016/j.jseaes.2017.11.008.

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