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Key Points:

- Backfield remanence data of Chinese loess and red clay consist of two components
- Loess and red clay have similar pedogenic but different detrital components
- The Loess Plateau experienced a cooling trend from the late Miocene to the Quaternary

Supporting Information:

- Readme
- Figures S1–S7
- Text S1

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Late Miocene–early Pleistocene paleoclimate history of the Chinese Loess Plateau revealed by remanence unmixing

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Abstract Previous studies show that the Quaternary loess sequence and the late Miocene–Pliocene red clay sequence on the Chinese Loess Plateau (CLP) share similar magnetic properties despite their being deposited under different climate conditions. To solve this paradox, we apply a novel remanence unmixing technique and demonstrate that loess and red clay on the central CLP contain a similar low-coercivity pedogenic component, but their high-coercivity components differ dramatically, reflecting different oxidation and temperature conditions. We infer that temperatures on the Chinese Loess Plateau cooled from the late Miocene to the Quaternary, in a manner similar to sea surface temperature records for the same time interval. This coherency between marine and terrestrial records argues for a CO₂ forcing on long-term paleoclimatic variations.

1. Introduction

Environmental magnetism was established in the 1980s and has played an important role in solving many geological and environmental problems [Evans and Heller, 2001; King and Channell, 1991; Liu et al., 2012; Thompson and Oldfield, 1986; Verosub and Roberts, 1995]. Magnetic proxies are appealing because most magnetic measurements are nondestructive, fast, and can be applied to large sample sets. However, compared with other paleoclimate proxies, magnetic methods are less quantitative, and often the processes that link past environmental signals to observed magnetic parameters in sediments or soils are poorly constrained. This is mainly because analyses have been limited to bulk magnetic parameters, which represent an integrated response of all the magnetic mineral components within a sample. However, this problem can be resolved if different magnetic mineral components can be separated and individually linked to environmental processes [Egli, 2004; Heslop and Dillon, 2007; Just et al., 2012; Necula et al., 2013; Spassov et al., 2003].

Chinese loess and red clay sequences are important archives of paleoclimate and paleoenvironmental information [An et al., 2001; Deng et al., 2005; Hao et al., 2008; Zhu et al., 2007]. However, most previous rock magnetism studies of these sediments focused on bulk magnetic parameters [Florindo et al., 1999; Liu et al., 2004; Nie et al., 2007, 2010]. Here we numerically unmix magnetic backfield isothermal remanent magnetization (IRM) data from Chaona (Figure S1 in the supporting information) on the central Chinese Loess Plateau (CLP) [Nie et al., 2007], based on the methods developed by Heslop and Dillon [2007] (Text S1 in the supporting information). We then discuss the paleoclimatic significance of the individual unmixed magnetic components.

2. Results

A two end-member (EM) mixing model provides a good approximation of the loess–paleosol remanence data (Figure 1a), while addition of a third EM provides only a minor improvement in terms of the model fit. The remanence gradients of two EMs (Figure 1a) peak at ~72 mT (EM1) and 21 mT (EM2), respectively. Variations in the abundance of EM2 have an inverse relationship with EM1 abundance and the mass-specific magnetic susceptibility (χ , Figure 2 and Figure S2 in the supporting information).

Figure 1. Unmixing of backfield IRM curves of Chaona (a) loess-paleosol and (b) red clay samples.

In a similar manner to the loess-paleosol sequence, the red clay remanence data can be represented by two EMs, with a minimal improvement in the model upon the addition of a third component. The remanence gradients of two red clay EMs peak at ~ 32 mT (EM1) and ~ 21 mT (EM2, Figure 1b). As with the loess-paleosol samples, the variation of EM2 abundance in the red clay samples mimics the χ record (Figure 3 and Figure S3 in the supporting information). However, unlike the loess-paleosol samples, the negative relationship between EM2 and EM1 abundances in the red clay samples is less apparent, especially for red clay deposited prior to 4.5 Ma (Figure S3a in the supporting information). Instead, variations in EM1 abundance are related to the mean bulk grain size data on

a submillion year timescale [Lü *et al.*, 2001] (Figures 3c and 3d).

One typical paleosol ($\chi = 17.4 \times 10^{-7} \text{ m}^3 \text{ kg}^{-1}$), one typical loess sample ($\chi = 2.4 \times 10^{-7} \text{ m}^3 \text{ kg}^{-1}$), and three red clay samples corresponding to the upper ($\chi = 14.0 \times 10^{-7} \text{ m}^3 \text{ kg}^{-1}$), middle ($\chi = 3.0 \times 10^{-7} \text{ m}^3 \text{ kg}^{-1}$), and lower ($\chi = 2.5 \times 10^{-7} \text{ m}^3 \text{ kg}^{-1}$) parts of the red clay sequence were subjected to first-order reversal curve (FORC) measurements (Figures 2 and 3, Figure S4 and Text S1 in the supporting information).

Both the paleosol and the upper red clay samples show a dominant peak highlighted by small open contours located near the origin of the FORC diagram, indicating a significant contribution from superparamagnetic (SP) particles [Roberts *et al.*, 2000] (Figure S4 in the supporting information). The small divergence and the triangular shape of outer contours also indicate a small contribution from the pseudo-single domain (PSD) grains [Pike *et al.*, 2001; Roberts *et al.*, 2000]. The small, central ridge concentrated along the Bc axis (at $B_u = 0$) represents the signature of noninteracting SD particles [Egli *et al.*, 2010; Roberts *et al.*, 2000].

The loess as well as the middle and lower part of the red clay section show more divergent behavior of the outermost

Figure 2. Variations of (a) magnetic susceptibility (χ), (b) EM2 component, (c) EM1 component, and (d) mean grain size of the Chaona loess-paleosol sequence during ~ 2.8 – 1.2 Ma. The stratigraphic column is shown on the left (ages are from Sun *et al.* [2006]). “Sx” represents the xth paleosol layer counting from the surface. Gray and white bars correspond to paleosol and loess layers, respectively. Half-shaded bars indicate subpaleosol layers within loess layers. Filled and open squares correspond to loess and paleosols, respectively. The two samples marked by circles in Figure 2a were selected for FORC analysis.

Figure 3. Variations of (a) magnetic susceptibility (χ), (b) EM2 component, (c) EM1 component, and (d) mean grain size of the Chaona red clay sequence. The gray line corresponds to the ~4.5 Ma climate transition on the CLP. The three samples marked by circles in Figure 3a were selected for FORC analysis.

ferrimagnets formed during pedogenesis (exceptions do occur in the southern part of the Chinese Loess Plateau, where precipitation may have crossed a threshold, inhibiting the ability of χ to reflect paleoprecipitation) [Guo *et al.*, 2001; Liu *et al.*, 2003; Zhou *et al.*, 1990]. Therefore, covariations in χ and EM2 abundance for both the loess-paleosol and red clay sequences indicate that the EM2 component represents a pedogenic contribution. It is noted that the EM2 component for these two sequences has a consistent peak remanence coercivity of 21 mT, indicating that the Quaternary loess-paleosol and late Miocene-Pliocene red clay had a similar magnetic enhancement mechanism controlled by the degree of soil formation. In turn the degree of soil formation depends on precipitation, as shown by Nie *et al.* [2008b, 2013a, 2013b] who associated susceptibility variations with paleoprecipitation.

Unlike the EM2 component, EM1 abundances for loess and paleosols are negatively correlated with χ and have a coercivity peak of 73 mT, which is close to the coercivity value of the detrital component found in the nearby Lingtai section [Spassov *et al.*, 2003]. This component has previously been interpreted to be partially oxidized detrital magnetite [Liu *et al.*, 2003; van Velzen and Zijdeveld, 1995]. Based on detailed magnetic, X-ray diffraction and scanning electron microscope experiments, Liu *et al.* [2003] found that loess has a higher coercivity of remanence than paleosols and attributed this difference to a higher degree of oxidation in paleosols, which tends to decrease the coercivity contrast between a particle's oxidized maghemite rim and its magnetite core. Therefore, we attribute the lower EM1 contribution in paleosol layers to a higher level of oxidation.

A coercivity of remanence of 32 mT corresponds to smaller PSD detrital ferrimagnetic grains. For such PSD particles, coercivity decreases with increasing grain size [Hu *et al.*, 2013; Maher, 2007]. We attribute the lower coercivity of the red clay compared to the loess-paleosol sequences to a higher degree of oxidation in the red

contours, with a pronounced triangular shape at low coercivities. Both the loess and the middle and lower red clay samples have open contours close to the ordinate axis indicating a contribution from SP particles. The second maximum along the Bc axis (indicated by closed contours) demonstrates that the loess and the middle and lower red clay samples also contain noninteracting SD ferrimagnetic particles.

3. Discussion and Conclusions

3.1. Origin of Different Magnetic Components

The Pliocene was the last warm interval before Earth entered a period with ice sheets periodically forming at Northern Hemisphere high latitudes [Zachos *et al.*, 2001]. Thus, the late Miocene-Pliocene red clay sequence was formed in a different climate regime to the overlying Quaternary loess-paleosol sequence. Nevertheless, the loess-paleosol and red clay sequences are both dominated by detrital and pedogenic components [Liu *et al.*, 2003; Nie *et al.*, 2007], which can be further attested by the consistent two end-member models (EM1 and EM2) for the whole profile. It is well known that χ enhancement of Chinese loess and paleosols is controlled by fine-grained

Figure 4. Variations of (a) magnetic susceptibility (χ), (b) EM1 component, (c) EM2 component, (d) EM3 component, and (e) mean grain size of the Chaona loess-paleosol and red clay sequence. The gray lines in Figures 4b and 4c divide the two curves into three time intervals, and the mean contribution from EM1 and EM2 in these intervals are shown (units are as for the x axis).

contrast at orbital timescales before the onset of Northern Hemisphere glaciation [Nie, 2011], it is reasonable to attribute coercivity changes at submillion year timescales to variations in wind strength rather than degree of particle oxidation. More coarse (large PSD) grains were transported to the CLP, and the sediments have a correspondingly larger EM1 contributions when the wind was stronger [Maher, 2007]. Thus, EM1 and EM2 for the red clay, especially those sediments deposited during the late Miocene–early Pliocene, have a weaker negative correlation compared to the Quaternary loess-paleosols (Figures 2 and 3 and Figures S2 and S3 in the supporting information), instead, variations of EM1 in the red clay are more similar to inverted mean grain size at the submillion year timescale (Figure 3). The EM1 contribution to the remanence has a decreasing trend from ~4.5 to 2.7 Ma, accompanied by an increasing contribution from the EM2 component (Figures 3b and 3c).

Previous studies report that the CLP has a wetting/cooling trend from 4.5 to 2.7 Ma [Nie et al., 2013b; Sun et al., 2010]. Paleoceanographic data demonstrate that closure of the Panama Seaway reached a critical point around 4.5 Ma, which significantly changed oceanic thermohaline circulation and salinity distribution patterns [Haug and Tiedemann, 1998]. Such adjustments would be likely to cause changes in precipitation and air temperature on the CLP [Nie et al., 2008a, 2013b]. Sediment cores from Qinghai Lake reveal an eolian to lacustrine transition at ~4.5 Ma [Fu et al., 2013], supporting a wetter climate in China after 4.5 Ma.

Variation in EM2 abundance is consistent with such a wetting trend, and EM1 abundance is consistent with a cooling trend that would result in slower oxidation rates. If this is the case, samples from 4.5 to 2.7 Ma should have a higher contribution to the remanence from the 72 mT component compared to the underlying red clay. To test this hypothesis, we unmixed loess and red clay backfield IRM data together (Figure S7 in the supporting information and Figure 4). The new unmixing results confirm this inference. The ~32 mT component makes a minor contribution to the Quaternary loess-paleosol, whereas the ~72 mT makes a minor contribution to the red clay deposited before 4.5 Ma (Figures 4b and 4c). In contrast, the 72 mT component makes a slightly increased contribution to the remanence of the red clay deposited between 4.5 and 2.7 Ma when compared to older red clay (Figures 4b and 4c).

clay resulting from the warmer climate of the late Miocene–Pliocene, which would decrease the coercivity contrast between maghemite and magnetite [Liu et al., 2003].

The red clay EM1 and EM2 components both have a smaller peak at ~300 mT (Figure 1b), possibly corresponding to the coercivity range of pedogenic hematite [Hu et al., 2013]. If goethite was formed rapidly in the red clay, it may have subsequently undergone long-term dehydration to hematite. Thus, to highlight the possible hematite component, we fitted a three end-member mixing model. Results (Figures S5 and S6 in the supporting information) reveal that the 21 mT (EM2) and the 330 mT (EM3) components covary, which indicates that an increase in pedogenic hematite was accompanied by an increase in pedogenic SP ferrimagnetic materials, which is an expected result following the magnetic enhancement pathway of ferrihydrite to maghemite to hematite [Torrent et al., 2006].

3.2. Paleoclimate History on the CLP

In conditions with decreased climate

Therefore, the unmixing results are consistent with previous bulk parameter studies that reported intensified precipitation on the CLP from 4.5 to 2.7 Ma [Nie *et al.*, 2013b; Sun *et al.*, 2010]. However, the unmixing data reveal, for the first time, that the Quaternary loess-paleosol and late Miocene-Pliocene red clay sediments have different magnetic properties related to differing degrees of oxidation, suggesting that temperature on the CLP gradually decreased from the late Miocene to early Pliocene into the late Pliocene and Quaternary. This inference is consistent with sea surface temperature records from multiple midlatitude and high-latitude locations [Herbert *et al.*, 2013], supporting decreases in atmospheric CO₂ concentration as a common forcing mechanism [Herbert *et al.*, 2010].

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