New High-Temperature Dependence of Magnetic Susceptibility-Based Climofunction for Quantifying Paleoprecipitation From Chinese Loess

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Abstract The ferrimagnetic properties of soils are used to quantitatively reconstruct paleomonsoon precipitation from Chinese loess. Numerous magneto-climofunctions have been established based on the magnetic proxies that are selectively sensitive to neoformation of fine-grained superparamagnetic (SP) or single-domain (SD) ferrimagnetic particles. Accumulating evidence has indicated that maghemite is the final product of the ferrimagnetic phases during pedogenesis in loessic soils. Quantitative estimates of abundance of maghemite of both SP and SD grains are therefore still required in developing magneto-climofunctions. Here, we present detailed measurements on a suite of modern soil samples from the Chinese Loess Plateau to determine pedogenic ferrimagnetic mineralogy and to develop a new magneto-climofunction based on a new parameter derived from the high-temperature-dependent magnetic susceptibility. Particle size fractionation processes combined with magnetic measurements indicate that fine-grained SP and SD maghemite is the dominant pedogenic ferrimagnetic phases. High-temperature-dependent susceptibility measurements show that the thermally induced susceptibility drops between ~230 and ~400 °C during heating mainly result from the conversion of maghemite to hematite. We proposed a new parameter quantifying changes in the temperature dependence of magnetic susceptibility between 230 and 400 °C, \( \chi_{td} \), that captures the concentration of pedogenically formed maghemite. Results show that \( \chi_{td} \) has a strong correlation with known quantities of maghemite in synthetic standard samples and that \( \chi_{td} \) of modern soils correlates with modern mean annual precipitation quite well (\( R^2 = 0.82, n = 24 \)). The established \( \chi_{td} \)-mean annual precipitation climofunction provides a new approach to reconstructing paleorainfall during past warm interglacials from paleosols in Chinese loess.

1. Introduction

The alternating loess-soil sequences in the Chinese Loess Plateau (CLP) in north central China provide a long-term and near-complete terrestrial archive of the East Asian monsoon climate over the past 22 Ma (An et al., 1990; Guo et al., 2002; Hao & Guo, 2004; Kukla & An, 1989; Liu, 1985). The soil-forming intervals correspond to periods with enhanced monsoon precipitation and higher ambient temperature. Therefore, the hundreds of soil layers preserved in the loess sequences allow for their use in reconstructing variations in paleomonsoon precipitation during the past warm periods, which, in turn, may provide invaluable insights into the impact of modern global warming on the monsoon climate.

Empirical transfer functions between mean annual precipitation (MAP) and soil properties, including magnetic properties, are one of the most important approaches for reconstructing paleorainfall in terrestrial systems (Geiss et al., 2008; Maher & Possolo, 2013; Maxbauer et al., 2016, and references therein). Pioneering works of Lü et al. (1994) and Maher et al. (1994) first reported the positive correlations between the low-field magnetic susceptibility (\( \chi_{lf} \)) of modern soils and MAP on the CLP, and Maher et al. (1994) initially...
established the climofunction between the susceptibility of the pedogenic component (the calculated difference in susceptibility of the B and C horizons) and MAP. Subsequently, additional publications have further extended the positive correlations between MAP and magnetic properties of modern soils to the whole CLP (Gao et al., 2018; Han et al., 1996; Liu et al., 1995; Nie et al., 2014; Song et al., 2014; Xia et al., 2012), Europe (Maher & Thompson, 1995), the Russia steppe (Maher et al., 2002), the midwestern U.S. (Geiss et al., 2008), and North Africa (Balsam et al., 2011; Lyons et al., 2010).

Separating pedogenic signals from the magnetic properties of bulk samples are one of the primary challenges in establishing magneto-climofunctions, as the magnetic properties of loess and paleosol samples represent a mixture of pedogenic and detrital signals (Chen et al., 1995; Hao et al., 2008, 2012; Zheng et al., 1991). Generally, three approaches have been used to derive the pedogenic signals. The first approach, adopted by Maher et al. (1994), is to calculate the differences between the susceptibility of the soil layers and the parent loess layers. Many subsequent studies use the magnetic properties of the bulk samples following the same approach. The second approach is to use magnetic properties that are selectively sensitive to the pedogenic fine-grained superparamagnetic (SP) or single-domain (SD) ferrimagnetic particles, which mainly account for the magnetic enhancement of soils (Maher & Thompson, 1991; Verosub et al., 1993; Zhou et al., 1990). Low-temperature magnetic properties (Liu et al., 1995), $X_{\text{TA}}$ % (Xia et al., 2012) and $X_{\text{TA}}$ (Song et al., 2014), have been suggested to characterize the abundance of SP particles; $X_{\text{ARM}}$/IRM (Geiss et al., 2008) and $X_{\text{ARM}}$ (Song et al., 2014) have been proposed to characterize the abundance of SD particles. The third approach is to use the magnetic properties of clay-size fractions separated by the time-consuming pipette method (Chen et al., 1995; Hao et al., 2008, 2012; Oldfield et al., 2009; Sartori et al., 2005; Zheng et al., 1991). These properties can then be used to separately identify the pedogenic components (Gao et al., 2018). Accumulating evidence has indicated that fine-grained (SP and SD) maghemite is the final product of the ferrimagnetic phases during pedogenesis in paleosols (Deng et al., 2000; Eyre & Dickson, 1995; Hao et al., 2012; Spassov et al., 2003; Verosub et al., 1993). As the routine ferrimagnetic proxies are either magnetic phase dependent or magnetic domain state dependent (Maher, 1988; Thompson & Oldfield, 1986), a new proxy, which exclusively targets the abundance of the pedogenic maghemite, is therefore still required in developing any magneto-climofunction.

Several earlier thermomagnetic analyses on loess/paleosol samples from the CLP opened up a new avenue for estimating maghemite concentrations (Deng et al., 2000, 2001; Zhu et al., 2001). Previous work on the metastable nature of maghemite during heating documents that maghemite begins to transform to hematite at temperatures exceeding 200 °C (Deng et al., 2001; Florindo et al., 1999; Liu, Deng, et al., 2005; Oches & Banerjee, 1996; Sun et al., 1995). Importantly, the degree of the thermally induced transformation from maghemite to hematite shows a gradually increasing trend from the northwestern CLP to the southeastern CLP, consistent with parallel variations in the modern MAP (Deng et al., 2000, 2001; Zhu et al., 2001). Therefore, thermomagnetic properties have been suggested as useful indicators of past climatic change (Deng et al., 2000, 2001; Zhu et al., 2001). Ferrimagnetic mineral assemblages are dominantly controlled by MAP on the CLP (Gao et al., 2018; Maher et al., 1994, 2002); however, there has been no quantitative assessment of the correlation between MAP and thermomagnetic properties.

In this study, we present results of routine magnetic susceptibility and remanent magnetization analysis, high- and low-temperature-dependent magnetic measurements, and detailed hysteresis investigations for a suite of representative bulk and particle-fractioned modern soil samples. These samples mainly lie on a NW-SE transect on the CLP (Figure 1). The main goals of the present study are to (1) detail and unambiguously identify the nature and origin of the magnetic minerals and grain sizes, responsible for the signature upon which the climofunction is based, for the CLP soils along the study transect, through investigation of thermomagnetic and other magnetic properties of clay-sized, silt-sized, and bulk samples for the modern surface soils, and (2) investigate the quantitative relationship between pedogenic maghemite determined by the thermomagnetic properties, and MAP.

2. Samples and Methods
2.1. Modern Soil Samples
Twenty-four modern soil samples on the CLP were selected from a set of 180 soils previously used to clarify the relationships between the formation of ferrimagnetic, antiferromagnetic minerals and climatic variables...
by Gao et al. (2018). In the field, after removing leaves, roots, and small crushed stones, samples at 2–5 cm beneath the surface in A-horizon (topsoil) were collected. Sixteen of 24 representative samples are located on three northwest to southeast (NW-SE) transect (red) and 8 samples collected from two west to east (W-E) transects (green). On the W-E transects, four samples collected along the precipitation isoline of 400 mm and four samples along the precipitation isoline of 500 mm. Eight samples from the NW-SE transect of MS-77–MS-107 have been subject to detailed magnetic investigations with the aim to identify the nature and origin of the magnetic minerals and grain sizes. These samples cover a modern MAP gradient from 278.6 mm in the north to 601.1 mm in the south, which represent soils with increasing degree of pedogenesis.

2.2. Particle Separation

For each sample, three fractions, clay (<4 μm), silt (4–61 μm), and sand (>61 μm), were separated using the pipette and wet-sieve methods. Detailed procedures were outlined in Hao et al. (2009) and Text S1 in the supporting information. Buffered acetic acid was used in removing carbonate to prevent significant loss of soft and hard magnetic signals (Hao et al., 2008, 2009). To avoid any possible oxidation of iron minerals, the extracted subsamples were rinsed with deionized water at least three times, centrifuged, and then freeze-dried. Material in the size range larger than 61 μm, separated by the wet-sieved method, mainly comprised sand and plant debris. Therefore, this fraction was not analyzed.
2.3. Magnetic Measurements

Low-frequency magnetic susceptibility ($\chi_{ld}$) at 470 Hz and high-frequency magnetic susceptibility ($\chi_{hd}$) at 4700 Hz were measured using a Bartington MS3 meter. Frequency-dependent magnetic susceptibility ($\chi_{fd}$), which is sensitive to the abundance of fine-grained magnetic particles around the SP/SD boundary, was defined as the difference between the measured susceptibility values at the low and high frequency (Evans & Heller, 2003; Maher, 1988; Thompson & Oldfield, 1986).

Anhysteretic remanent magnetization (ARM) was imparted in a peak alternating field of 100 mT with a bias field of 100 µT using a 2G Enterprises Model 760R Cryogenic magnetometer. ARM susceptibility ($\chi_{ARM}$), which is selectively sensitive to the abundance of ferrimagnets within the stable single-domain (SSD) range (Dunlop & Özdemir, 1997; King et al., 1982; Maher, 1988), was calculated from ARM normalized by the bias field.

Saturation Isothermal Remanent Magnetization (SIRM) was produced in an applied field of 1 T, followed by reverse fields of 40 mT, using a 2G Enterprises Pulse Magnetizer. All remanence measurements were made using a 2G Enterprises Model 760-R cryogenic magnetometer situated in a magnetically shielded room. $\text{Soft}_{40\mu T}$, which is often used as an indicator of the abundance of ferrimagnetic minerals, was defined after Thompson and Oldfield (1986): $\text{Soft}_{40\mu T} = 0.5 \times (\text{SIRM-IRM}_{40\mu T})$.

High-temperature-dependent susceptibility ($\chi_T$) measurements were performed using a KLY-3 Kappabridge with a CS-4 high-temperature furnace. For bulk measurement, the KLY-3 modular system has a sensitivity of $3 \times 10^{-8}$ SI to susceptibility changes, and the CS-4 temperature control unit has a temperature accuracy of ±2 °C. About 0.3 g of each powdered sample was heated from room temperature up to 700 °C with a heating rate of ~13.7 °C/min and then cooled back to room temperature. The variations in susceptibility were measured with an operating frequency of 976 Hz and a field intensity of 200 Am$^{-1}$.

To prevent any possible oxidation during heating and cooling cycles, all the procedures were carried out in an argon atmosphere with a gas flow rate around 100 ml/min.

A new parameter $\chi_{td}$ temperature-dependent magnetic susceptibility, to quantitatively estimate the degree of the thermally induced transformation, is introduced. $\chi_{td}$ is defined as the difference between the measured susceptibility values around 230 and 400 °C, $\chi_{230^\circ C}-\chi_{400^\circ C}$, on the heating curves of high-temperature-dependent susceptibility. Here, the subscript 230 and 400 °C are not specific temperatures. The mean value of $\chi_T$ data at three adjacent temperature points around the peak value between 230 and 250 °C, and the trough value between 400 and 460 °C has been used. For some weakly magnetic samples, first-order derivative of high-temperature magnetic susceptibility can help to identify the peaks and troughs.

To verify if the thermomagnetic properties accurately capture the abundance of maghemite (Deng et al., 2000, 2001; Zhu et al., 2001), a series of synthetic maghemite samples with known concentrations were admixed to the modern soil matrix, which was subjected to thermomagnetic experiments. Detailed procedures were designed as follows: (1) We chose eight samples on the NW-SE transect of MS-77–MS-107 on the CLP (Figure 1). (2) Each sample was treated with the CBD (citrate-bicarbonate-dithionite) method (Mehra & Jackson, 1960), which has been proved effective in reducing the pedogenic fine-grained secondary iron oxides, including maghemite, magnetite, hematite, and goethite in soils (Verosub et al., 1993; Hunt et al., 1995; Deng et al., 2000). During this treatment, coarse-grained eolian magnetic minerals survived. A total of 4.5 g of sodium dithionite was added to 1.5 g of sample in three times at 70–75 °C, and then the residues were rinsed three times, centrifuged and air-dried. (3) Known quantities of synthetic maghemite were added to 0.3 g of each powdered sample matrix. In the present study, nanosized (mean 20 nm) magnetite (produced by Beijing Dk Nano technology Co., LTD), which was close to the magnetic grain size of the pedogenic ferrimagnetic minerals in loess/paleosol deposits (Liu, Torrent, et al., 2005), was used. Finally, eight samples were produced with maghemite contents varying from 0.2 to 1.6 mg in increments of 0.2 mg (0.2, 0.4, … , 1.4, and 1.6 mg) and then subjected to high-temperature-dependent susceptibility ($\chi_T$) measurements. For comparison, the residues after CBD treatment were also measured.

Low-temperature thermal demagnetization of SIRM measurements was conducted using a Quantum Designs Magnetic Properties Measurement System. After cooling to 15 K in zero field, SIRM was imparted in a field of 2.5 T. Then the samples were warmed from 15 K up to 300 K at a constant rate of 3 K/min, and remanence was measured recurrently in field-free space.
Hysteresis loops, Isothermal remanent magnetization (IRM) acquisition curves and backfield demagnetization of SIRM curves, were obtained using a MicroMag 3900 vibrating sampling magnetometer (VSM 3900) with an applied maximum field of 1.0 T (Figure 5). From hysteresis loops, saturation magnetization ($M_s$), saturation remanence ($M_{rm}$), and coercivity ($B_c$) were calculated after correction for the paramagnetic contribution (Thompson & Oldfield, 1986). Paramagnetic susceptibility ($\chi_{para}$) was calculated from the slope of the hysteresis loop between 0.7 and 1.0 T. The coercivity of remanence ($B_{cr}$) was obtained from the backfield demagnetization of SIRM curves.

First-Order Reversal Curve (FORC) measurements were also performed on VSM 3900. For each sample, 180 FORCs were measured with a field rate of ~0.8 mT and maximum field of 1.0 T. FORC diagrams were produced using the FORCinel software of Harrison and Feinberg (2008).

For the present work, magnetic measurements were made at the Paleomagnetism and Geochronology Laboratory, Institute of Geology and Geophysics, Chinese Academy of Sciences; particle separation and magnetic susceptibility measurements were made at the Laboratory of Soil Geology and Environment, Institute of Geology and Geophysics, Chinese Academy of Sciences.

2.4. Meteorological Data
Modern climate data for a 40-year interval (1951–1990) were sourced from the China Meteorological Administration. Climatic factors for each sampling site were obtained from inverse-distance weighting spatial interpolation. The accuracy of the climatic variables derived from this spatial interpolation has been confirmed by Song et al. (2014).

3. Results and Discussion

3.1. Magnetic Properties of the Bulk Samples, Pedogenic, and Detrital Fractions
The present study systematically investigates the magnetic susceptibility and remanent magnetization, temperature-dependent, and hysteresis magnetic properties of a suite of representative bulk samples and the clay and silt subsamples separated from these bulk samples. These properties are widely used in evaluating magnetic domain state and detecting magnetic phases (Dunlop & Özdemir, 1997). Particle size fractionation processes combined with magnetic measurements may provide considerable insight into the magnetic properties of the bulk samples (Oldfield & Yu, 1994).

3.1.1. Magnetic Susceptibility and Remanent Magnetization
Figure 2 shows the results of a range of room temperature magnetic measurements on the bulk samples and the clay and silt subsamples for the selected eight samples on the NW-SE transect of MS-77–MS-107 (original data can be found in supporting information Table S1).

For the absolute values of various magnetic properties, the bulk samples have intermediate $\chi_{lf}$, $\chi_{fd}$, $\chi_{ARM}$, and Soft40mT values. The clay subsamples have generally high and more variable values. In contrast, the silt subsamples have overall low and less variable values. For the spatial variation changes, a gradual southeastward increasing trend can be found for both of the bulk samples and clay subsamples, whereas the silt subsamples show almost uniform values on the NW-SE transect.

By now, the pedogenic origin for the magnetic enhancement of paleosols on the CLP has been widely demonstrated and accepted (Maher & Thompson, 1991; Verosub et al., 1993; Zhou et al., 1990). Our fractionation process-based measurements show that the clay subsamples have a strong signal in magnetic properties with significant southeastward increasing gradient, while the silt subsamples have overall uniform values. These results further support the view that the magnetic properties of the bulk modern soil samples, and its spatial variation are primarily controlled by pedogenic components in the clay fraction.

3.1.2. High-Temperature-Dependent Magnetic Susceptibility
The selected eight bulk and particle-sized modern soil samples on the NW-SE transect of MS-77–MS-107 were subjected to measurements of the high-temperature dependence of magnetic susceptibility. Figure 3 shows the heating curves for representative bulk samples, and the clay and silt subsamples. Generally, the heating curves of all the bulk samples, clay and silt subsamples, exhibit a similar pattern, a steady rise of susceptibility from room temperature to 230 °C, a loss of susceptibility between 230 and 400 °C, a sharp hump around 510 °C, and a dramatic reduction of susceptibility at 585 °C.
Figure 2. Routine magnetic properties of the bulk samples, clay, and silt subsamples for eight representative modern soils collected along the NW-SE transect of MS-77–MS-107. The left-to-right direction of the graph represents samples from northwest to southeast. MAP values for samples from the NW-SE transect of MS-77–MS-107 are 278.63 mm for MS-77, 349.78 mm for MS-70, 458.10 mm for MS-65, 471.13 mm for MS-58, 563.31 mm for MS-50, 560.90 mm for MS-42, 568.65 mm for MS-38, and 601.10 mm for MS-107, respectively.

Figure 3. Temperature dependence of magnetic susceptibility of the bulk samples (a), clay (b), and silt (c) subsamples for eight representative modern soils collected along the NW-SE transect of MS-77–MS-107.
The steady rise in susceptibility with increasing temperature up to ~230 °C may relate to the gradual unblocking of the fine-grained SD particles, which exhibit SP-like behavior with increasing temperature (Deng et al., 2000; Liu, Deng, et al., 2005).

From 230 to 400 °C, the loss of susceptibility has been shown to primarily originate from the conversion of metastable maghemite to a more stable phase, hematite (Deng et al., 2001; Florindo et al., 1999; Liu, Deng, et al., 2005; Oches & Banerjee, 1996; Sun et al., 1995). For the magnitude of the decrease in susceptibility during this interval, both of the bulk samples and clay subsamples show a rapid and significant decrease (Figures 3a and 3b). In contrast, the silt subsamples display a relatively small amplitude decrease (Figure 3c). These results indicate that maghemite in the bulk samples has its origin predominantly in the pedogenic clay fraction.

The noteworthy susceptibility peak around 510 °C is commonly attributed to the neoformation of high-susceptibility magnetite transformed from iron-bearing silicates during heating (Deng et al., 2001; Hoffmann et al., 1999; Hunt et al., 1995; Liu, Deng, et al., 2005). The sharp decrease in magnetic susceptibility near 585 °C, the Curie temperature of magnetite, indicates the presence of magnetite. Considering the neoformation of strong magnetic minerals when heating above 400 °C, we cannot determine whether the Curie temperature around 585 °C reflects the original magnetite presenting in the natural samples or the neoformed magnetite during heating.

The most interesting phenomenon is the way in which the magnitude of the loss in the magnetic susceptibility between 230 and 400 °C gradually increases from the northern CLP to the southern CLP for the bulk samples and clay subsamples (Figure 3), in accordance with the observed trend in MAP (Figure 1). This indicates that the abundance of pedogenic maghemite gradually increased from north to south and its spatial changes are probably dependent on climatic conditions (mainly MAP). While the silt subsamples show a less clear spatial pattern with a much smaller loss in magnetic susceptibility, it is clear that spatial changes in the magnitude of loss in the magnetic susceptibility of the bulk samples are dominantly controlled by the magnetic properties of the clay fraction.

3.1.3. Low-Temperature-Dependent Remnant Magnetization

Four of the 8 bulk samples, as well as the clay and silt subsamples on the NW-SE transect of MS-77–MS-107, were subject to thermal demagnetization of low-temperature saturation isothermal remnant magnetization measurement (LT-SIRM). Figure 4 shows the LT-SIRM warming curves for the representative bulk samples and the clay and silt subsamples. Visually, the LT-SIRM curves of the bulk samples resemble those of the silt subsamples (Figures 4a and 4c). Both curves display an abrupt drop in the intensity of remanence around 120 K, corresponding to the Verwey transition for pseudo-single domain (PSD), multidomain (MD), or some certain SD magnetite particles (Özdemir et al., 1993; Rochette et al., 1990). The magnitude of this drop progressively decreases from north to south. Previous studies have demonstrated that oxidation of magnetite...
broadens and suppresses the Verwey transitions (Banerjee et al., 1993). Our observations may be indicative of the enhanced effects of oxidation from the northern Loess Plateau to the southern Loess Plateau.

The shape of LT-SIRM curves of the clay subsamples contrasts with that of the bulk samples and silt subsamples. The clay subsamples (Figure 4b) show a gradual decay of remanence with increasing temperature, which indicates the dominant contribution of maghemite or the fine viscous SP magnetite (Banerjee et al., 1993). A much less marked Verwey transition around 103 K was observed for MS-77 in the northernmost of the four samples, possibly indicating the presence of trace amount of coarse-grained magnetic particles.

3.1.4. Hysteresis Loops, Day Plot, and FORC Diagrams and Magnetic Domain State

Detailed hysteresis measurements were conducted on the eight bulk samples and the clay and silt subsamples on the NW-SE transect of MS-77–MS-107. Figure 5 shows the hysteresis loops. Table 1 gives the main parameters derived from the hysteresis measurements. Figure 6 shows the corresponding Day plot. Figure 7 shows the FORC diagrams.

The various shapes of hysteresis loops and the derived hysteresis parameters are frequently used to estimate changes in the magnetic domain state of geological materials (Tauxe et al., 1996). In Figure 5, all samples reach saturation above 200–300 mT, indicative of dominance by the magnetic properties of the low-coercivity (ferrimagnetic) component. Except for the slightly wider loops for a few clay subsamples from the northern CLP, no apparent spatial variations can be recognized either for the bulk samples or for particle-sized subsamples.

In recent years, the usefulness of Day plot has been frequently challenged and questioned (Hao et al., 2012; Roberts et al., 2018; Tauxe et al., 2010); however, there are some recent publications still using the Day plot to estimate magnetic domain state. Particle-sized samples in this study provide an opportunity to further check the validity of Day plot in characterizing the magnetic domain state for the loess and paleosol samples. The present results show that all the bulk samples, the clay, and silt subsamples fall in the PSD region on the Day plot (Day et al., 1977; Dunlop, 2002a, 2002b; Figure 6). The clay subsamples have relatively higher $M_{cr}/M_s$ values and lower $B_{cr}/B_s$ values with less variable ranges (Table 1 and Figure 6), corresponding to a tightly clustered region on the Day plot. In contrast, the bulk samples and silt subsamples have relatively lower $M_{cr}/M_s$ values and higher $B_{cr}/B_s$ values with much variable ranges, corresponding to a slightly spread region along the $M_{cr}/M_s$ axis on the Day plot. The bulk samples and silt subsamples did not show a significant difference in $M_{cr}/M_s$ and $B_{cr}/B_s$ values (Table 1 and Figure 6), which suggests that the Day plot fails to detect the presence of the pedogenic fine-grained ferrimagnetic minerals.

To further characterize the magnetic domain state, we employed FORC measurements. Generally, the bulk samples exhibit tightly closed inner contours (Figure 7), which is particularly evident in the southern CLP, indicating the presence of noninteracting SD particles (Pike et al., 2001; Roberts et al., 2000). Divergent outer contours for the bulk samples indicate the coexistence of PSD or a mixture of SD and MD particles (Muxworthy & Dunlop, 2002; Roberts et al., 2014; Smirnov, 2006), which is more evident in the northern CLP. In addition, secondary peaks near the origin of the FORC diagrams are observed for most of the bulk samples (Figure 7), as would be expected for SP particles (Carvallo et al., 2004; Egli et al., 2010). For the clay subsamples (Figure 7), the tightly closed contours with negligible vertical spread indicate the presence of magnetically noninteracting SD particles; moreover, the observed secondary peaks at the origin of the FORC diagrams indicate the presence of SP particles. In contrast, the silt subsamples (Figure 7) exhibit closed inner contours and divergent outer contours, indicating the coarse-grained PSD or a mixture of SD + MD particles.

It is evident that in contrast to the Day plots, FORC diagrams show differences in the magnetic grain size among the clay subsamples, silt subsamples, and bulk samples (Figure 7), which conform with the conclusions derived from routine and thermomagnetic measurements of particle size fractions and bulk samples (Figures 2 and 3). Therefore, our present results further demonstrate that Day plots may often provide misleading information on the magnetic grain size of geological materials (Hao et al., 2012; Roberts et al., 2018; Tauxe et al., 2010), even in characterizing the magnetic properties of bulk, prefractonated samples. FORC diagrams show great advantages in evaluating the magnetic domain state for the loess and paleosols.
3.1.5. Ferrimagnetic Mineralogy of the Clay Fractions

Our rock magnetic investigations indicate that the dominant ferrimagnet in the clay fraction is fine-grained maghemite. The absence of a Verwey transition near 120 K on the LT-SIRM curves (Figure 4b) indicates the presence of maghemite or fine-grained SP magnetite particles. FORC diagrams (Figure 7) indicate that the magnetic assemblage of the clay subsamples consists of noninteracting finer SP and SD particles. In

Figure 5. Hysteresis loops after paramagnetic correction for the bulk samples, clay, and silt subsamples of four of the eight representative modern soils collected along the NW-SE transect of MS-77–MS-107. Figure of all the samples is given in Figure S3. The top-to-bottom direction of the graph represents samples from northwest to southeast.
addition, higher values of ferrimagnetic properties ($\chi_{lf}$, $\chi_{fd}$, $\chi_{ARM}$, and Soft40mT; Figure 2) indicate that both SP grains, which contribute significantly to the magnetic susceptibility, and SD grains, which act as the efficient remanence carriers, dominate the magnetic assemblages of the clay fraction. The direct evidence for the presence of maghemite comes from the high-temperature-dependent susceptibility measurements. On the heating curves, the clear susceptibility drop between ~230 and ~400 °C (Figure 3b) indicates the presence of maghemite (Deng et al., 2001; Florindo et al., 1999; Liu, Deng, et al., 2005; Oches & Banerjee, 1996; Sun et al., 1995).

Under natural environments, the process of production and transformation of iron oxide minerals has been investigated over 50 years (Chukhrov et al., 1973; Feitknecht & Michaelis, 1962; Schwertmann, 1966). However, whether the dominant ferrimagnet under natural pedogenesis is maghemite or magnetite has remained controversial (Ahmed & Maher, 2018; Maher, 1998; Torrent et al., 2006). Maher (1998) proposed that magnetite is the dominant ferrimagnet during pedogenesis. However, a dry season with oxidizing conditions will result in the rapid oxidation of the fine-grained magnetite to maghemite and then to hematite. As the fine-grained magnetite would be oxidized to maghemite rapidly under the natural pedogenic environment (Dunlop & Özdemir, 1997; Özdemir et al., 1993; Van Velzen & Dekkers, 1999), then, irrespective of the possibility that the initial pedogenic ferrimagnet may be magnetite, maghemite may be regarded as the dominant ferrimagnetic minerals after a certain period of pedogenesis. Inversely, based on in vitro experiments, Barrn and Torrent (2002) stated that maghemite was the dominant ferrimagnetic mineral in the transformation of ferrihydrite to hematite. Later, a conceptual model for this transformation sequence has been further proposed in Torrent et al. (2006), as ferrihydrite $\rightarrow$ superparamagnetic maghemite $\rightarrow$ stable...

### Table 1

| Sample    | $M_{rs}$ (10$^{-3}$ Am$^2$/kg) | $M_s$ (mT) | $B_{cr}$ (mT) | $M_{rs}/M_s$ | $B_{cr}/B_c$ | $\chi_{para}$ (10$^{-8}$ m$^3$/kg) | $\chi_{para}/\chi_{lf}$ (%) |
|-----------|-------------------------------|------------|---------------|--------------|-------------|-----------------------------------|-----------------------------|
| **Bulk samples** |
| MS-77     | 3.55                          | 26.86      | 40.30         | 11.96        | 0.13        | 3.37                              | 16.64                       |
| MS-70     | 5.26                          | 47.57      | 40.48         | 10.86        | 0.11        | 3.73                              | 12.68                       |
| MS-65     | 6.24                          | 42.82      | 40.46         | 11.75        | 0.15        | 3.44                              | 14.46                       |
| MS-58     | 6.52                          | 39.80      | 28.60         | 9.77         | 0.16        | 2.93                              | 10.67                       |
| MS-50     | 8.57                          | 109.33     | 30.42         | 9.43         | 0.08        | 3.23                              | 8.91                        |
| MS-42     | 10.96                         | 72.75      | 29.63         | 9.69         | 0.15        | 3.06                              | 8.04                        |
| MS-38     | 12.50                         | 75.46      | 27.73         | 9.75         | 0.17        | 2.84                              | 5.26                        |
| MS-107    | 14.12                         | 106.23     | 25.08         | 8.23         | 0.13        | 3.05                              | 5.11                        |
| **Clay subsamples** |
| MS-77     | 10.28                         | 48.56      | 35.81         | 13.31        | 0.21        | 2.69                              | 20.81                       |
| MS-70     | 9.36                          | 52.07      | 31.87         | 11.13        | 0.18        | 2.86                              | 16.00                       |
| MS-65     | 8.88                          | 41.44      | 27.21         | 11.10        | 0.21        | 2.45                              | 17.16                       |
| MS-58     | 11.56                         | 53.10      | 22.77         | 9.51         | 0.22        | 2.39                              | 10.85                       |
| MS-50     | 15.46                         | 70.25      | 23.58         | 9.13         | 0.22        | 2.58                              | 9.49                        |
| MS-42     | 20.08                         | 98.90      | 23.40         | 9.17         | 0.20        | 2.55                              | 8.24                        |
| MS-38     | 21.84                         | 110.51     | 21.63         | 8.74         | 0.20        | 2.47                              | 5.99                        |
| MS-107    | 24.59                         | 118.98     | 20.64         | 8.81         | 0.21        | 2.34                              | 5.26                        |
| **Silt subsamples** |
| MS-77     | 6.58                          | 64.31      | 40.46         | 10.78        | 0.10        | 3.75                              | 10.99                       |
| MS-70     | 7.00                          | 70.30      | 43.11         | 10.80        | 0.10        | 3.99                              | 11.07                       |
| MS-65     | 6.88                          | 53.06      | 44.83         | 13.00        | 0.13        | 3.45                              | 14.93                       |
| MS-58     | 6.05                          | 38.39      | 41.28         | 13.09        | 0.16        | 3.15                              | 14.52                       |
| MS-50     | 8.10                          | 57.36      | 40.53         | 12.48        | 0.14        | 3.25                              | 11.83                       |
| MS-42     | 10.61                         | 79.05      | 38.21         | 11.51        | 0.13        | 3.32                              | 10.14                       |
| MS-38     | 13.80                         | 85.78      | 35.72         | 12.34        | 0.16        | 2.90                              | 7.09                        |
| MS-107    | 11.04                         | 82.83      | 36.23         | 11.42        | 0.13        | 3.17                              | 7.62                        |

Note. $M_{rs}$: saturation remanence; $M_s$: saturation magnetization; $B_{cr}$: coercivity of remanence; $B_c$: coercivity; $\chi_{para}$: Paramagnetic susceptibility.
In our present work, no direct evidence indicates the existence of pedogenic magnetite. Consequently, we conclude that maghemite is the dominant ferrimagnetic minerals present in the clay fraction, consistent with previous studies on loess-paleosol samples (Deng et al., 2000; Eyre & Dickson, 1995; Hao et al., 2012; Spassov et al., 2003). Furthermore, magnetic grain size indicators of interparametric ratios (published in Gao et al., 2018) and present FORC diagrams (Figure 7) do not show obvious spatial variation on the main part of the CLP, which indicates that there is no apparent spatial variation in the pedogenic magnetic grain size. Therefore, the spatial changes of the magnetic properties of the clay fraction are mainly controlled by variations in the abundance of maghemite.

3.1.6. Ferrimagnetic Mineralogy of the Silt Fractions

In contrast to the clay fraction, the magnetic assemblages of the silt fraction mainly consist of coarse-grained magnetite. The direct evidence comes from the LT-SIRM analysis and FORC diagrams. The clear Verwey transition around 120 K (Figure 4b) and the divergent outer contours of the FORC diagrams (Figure 7) indicate the existence of the coarse-grained magnetite, at least in part likely to be PSD or the mixture of SD and MD particles. This is consistent with previous conclusions that the detrital ferrimagnetic component is coarse-grained magnetite (Banerjee et al., 1993; Deng et al., 2006; Hao et al., 2012; Liu et al., 2003; Oldfield et al., 2009).

However, our evidence shows that maghemite is also present in the detrital silt fraction. The reductions in susceptibility between ~270 and ~400 °C in the heating curves of the silt subsamples (Figure 3c) indicate the presence of thermally unstable maghemite. FORC diagrams (Figure 7) show that there is a trend with the southern silt subsamples containing an increased contribution of SD particles inferred from the closed

Figure 6. Day plot (Day et al., 1977; Dunlop, 2002a, 2002b) of the hysteresis data for the bulk samples (blue diamond), clay (orange dots), and silt (green dots) subsamples from the NW-SE transect of MS-77–MS-107. The mixing curves are after Dunlop (2002a, 2002b); related numbers indicate volume abundance of SP or MD and SD particles, and the grain size for SP particles on the mixing curves with SP-SD mixtures.
inner contours and of fine viscous SP particles inferred from the observed peaks at the origin of the FORC diagrams for southern subsamples. This may result from contamination of finer grades during particle separation and low-temperature oxidation of detrital magnetite. During particle separation, some clay particles may stick on the surface of the large particles and go into the silt fraction. Maghemitization has been often observed at the surface of magnetite particle (Dunlop & Özdemir, 1997), and this kind low-temperature oxidation of magnetite is a common process for a range of sediments (Van Velzen & Zijderveld, 1995, Van Velzen & Dekkers, 1999). The reduced amplitude of decrease at ~120 K in SIRM in LT-SIRM curves for the southern silt subsamples indicates an increase in the low-temperature oxidation.

Figure 7. FORC diagrams for four of the eight representative bulk samples, clay, and silt subsamples collected along the NW-SE transect of MS-77–MS-107. For comparison, the same scales of Hu-axis and Hc-axis are used for all diagrams. The results of four of the eight clay and silt subsamples (MS-77, MS-65, MS-50, and MS-38) have been published in Gao et al. (2018). Here, we further present results of eight bulk samples and another four of the 8 clay and silt subsamples collected along the NW-SE transect of MS-77–MS-107 (Figure S4). The top-to-bottom direction of the graph represents samples from northwest to southeast.
3.2. A New Paleoprecipitation Indicator Based On High-Temperature Dependence of Magnetic Susceptibility

Our rock magnetic results indicate that the spatial changes of the magnetic properties measured here are mainly dependent on variations in the abundance of pedogenic maghemite. For this reason, the most reliable magnetic proxies for paleorainfall reconstruction should be the ones that most effectively capture changes in the relative abundance of fine-grained maghemite.

As the room temperature ferrimagnetic proxies are strongly influenced by the variation magnetic phases and are selectively sensitive to any specific magnetic domain state, they may have some inherent limitations in quantifying the abundance of pedogenic maghemite. For climatic significance, $\chi_{T}$ works under the assumption that the contribution of paramagnetic and antiferromagnetic particles is negligible. However, our hysteresis analysis shows that the contributions of paramagnetic minerals to the total susceptibility of the clay subsamples are as high as 20.81% in the northern CLP (Table 1) and exhibit wide variations, ranging on the NW-SE transect, ranging from 20.81% to 5.26%. Furthermore, the canted antiferromagnetic minerals as reflected by HIRM also show large spatial variation across the loess plateau (Gao et al., 2018). Routine $\chi_{T}$ just detects the response of a very specific, narrow proportion of the SP particles (Maher, 1988; Worm, 1998; Worm & Jackson, 1999). $\chi_{ARM}$ is selectively sensitive to SD particles (Thompson & Oldfield, 1986). Additionally, coarse magnetic particles that are detrital in origin can also make a major contribution to the remanence proxies (Gao et al., 2018; Hao et al., 2009).

Thermomagnetic analysis possibly provides a practical way to quantify the abundance of the fine-grained, pedogenic maghemite. In the heating curves of high-temperature-dependent susceptibility measurements (Figures 3a and 3b), both bulk samples and clay subsamples show a proportional loss in susceptibility between ~230 and ~400 °C, and the amplitude of the susceptibility loss becomes more prominent from north to south. As the conversion of metastable maghemite to hematite mainly accounts for this susceptibility drop (Florindo et al., 1999; Liu, Deng, et al., 2005; Oches & Banerjee, 1996; Sun et al., 1995), the degree of the thermally induced alteration should be proportional to the concentration of the pedogenic maghemite and our newly proposed parameter $\chi_{T}$, which is defined as the difference between the measured susceptibility values around 230 and 400 °C on the heating curves of high-temperature-dependent susceptibility (Figure 8), might be a potentially powerful paleorainfall indicator (detailed procedures on calculating $\chi_{T}$ are given in section 2).

A series of quantitatively determined synthetic maghemite mixed samples have been used to test if $\chi_{T}$ captures the variation of abundance of maghemite. Before measurement of the mixed samples, CBD-treated samples were subject to $\chi-T$ experiment to understand the nature of the sample matrix. The heating curves of samples after CBD treatment show a steady increasing susceptibility under ~300 °C and a flat pattern up to ~410 °C or 450 °C (Figure 9a). Then, all samples show a significant increase of susceptibility, and an obvious peak at 520 °C or 550 °C, which mainly result from the neoformation of high-susceptibility magnetite transformed from iron-bearing silicates during heating (Deng et al., 2001; Hoffmann et al., 1999; Hunt et al., 1995; Liu, Deng, et al., 2005). In addition, susceptibility curves of all CBD-treated samples show a sharp drop at ~585 °C, marking the Curie point of magnetite. It is clear that by contrast with the natural modern soil samples (Figure 3a), the susceptibility drop between ~230 and 400 °C on heating is absent in the post-
Figure 9. Temperature dependence of magnetic susceptibility of representative modern soils collected along the NW-SE transect of MS-77–MS-107 after CDB treatment (a) and after adding known quantities synthetic maghemite (b), and the correlation between calculated $\chi_{td}$ and concentration of synthetic maghemite for the mixed samples (c).

Figure 10. Relationships between MAP and $\chi_{td}$ of the bulk samples (a, c) and clay subsamples (b, d) for eight samples from the MS-77–MS-107 transect (a, b) and 24 samples (c, d), with 16 samples from three NW-SE transect (solid circles) and 8 samples from two W-E transects along the precipitation isoline of 400 and 500 mm (open circles). Please note that the sample with MAP lower than 200 mm has extremely low $\chi_{td}$ values. Ignoring this sample in the calculation, the $R^2$ will be 0.90 for the bulk samples and 0.85 for the clay subsamples.
Figure 11. Relationships between MAP and routine ferrimagnetic proxies of the bulk samples (a–d) and clay subsamples (e–h) for 24 samples, with 16 samples from three NW–SE transect (solid circles) and 8 samples from two W–E transects along the precipitation isoline of 400 and 500 mm (open circles). Please note that the sample with MAP lower than 200 mm has extremely low $\chi_f$, $\chi_{fd}$, $\chi_{ARM}$, and Soft$^{40mT}$ values. By ignoring this sample in the calculation, the $R^2$ increases from 0.87 to 0.90 for the bulk samples and from 0.73 to 0.79 for the clay subsamples.
CBD samples (Figure 9a), confirming that the fined-grained pedogenic maghemite grains have been completely removed by CBD treatment. After adding known quantities of nanosized synthetic maghemite to the residues after CBD treatment, all samples show a significant susceptibility loss between ~250 and 400 °C (Figure 9b). The natural samples show susceptibility loss between ~230 and ~430 °C, slightly different from the synthetic samples, which may mainly result from the different grain size distribution between them. The high coefficient of determination ($R^2 = 0.99$) between $\chi_{\text{td}}$ and maghemite concentration for the synthetic samples (Figure 9c) robustly proves that the proposed new parameter $\chi_{\text{td}}$ enables semiquantitative estimation of the abundance of pedogenic maghemite.

In light of these results, the relationship between $\chi_{\text{td}}$ in modern soils and MAP was investigated in order to explore the potential for relating pedogenic maghemite concentrations to variations in MAP. Figures 10a and 10b show the correlations between $\chi_{\text{td}}$ values and MAP for the eight representative bulk samples and clay subsamples along the MS-77–MS-107 transect, which is nearly perpendicular to the MAP isolines on the CLP. Both of the $\chi_{\text{td}}$ values of the bulk samples and clay subsamples show strong exponential correlations with MAP, which indicates that the proposed new parameter $\chi_{\text{td}}$ is an attractive paleorainfall indicator. The strong correlations are well supported by the results of another 16 bulk samples and clay subsamples (eight samples along the precipitation isoline of 400 and 500 mm Figure S2 and eight samples from two N-S transects Figure S5). Figures 10c and 10d show the correlation between $\chi_{\text{td}}$ values and MAP for a total of the 24 bulk samples and clay subsamples (original data can be found in Table S1 in supporting information). The strong correlations convincingly demonstrate the effectiveness of this new parameter in quantifying MAP.

With the aim to clearly show the pros and cons of the present magneto-climofunction, we make a comparison with the relationships between routine magnetic susceptibility, remnant magnetization, and MAP. Figure 11 shows the correlations between the routine ferrimagnetic proxies and MAP for the same 24 bulk samples and clay subsamples over the CLP. Overall, routine ferrimagnetic proxies of the bulk samples and clay subsamples all exhibit a strong and exponential correlation with MAP (Figure 11). The relatively higher coefficient of determination ($R^2 = 0.82$ for the bulk samples and clay subsamples) and the lower standard error (SE = ±45.45 for the bulk samples and SE = ±45.80 for the clay subsamples) for the $\chi_{\text{td}}$-MAP climofunctions (Figures 10c and 10d) than those of climofunctions based on the routine ferrimagnetic proxies (Figure 11) suggest the great potential of $\chi_{\text{td}}$ in quantifying paleorainfall. However, it should be noted that high-temperature-dependent susceptibility experiment is a time-consuming and labor-intensive work. It will cost at least 1 hr to finish the heating and cooling measurements (even just heating to 500 °C) for one sample. Some routine parameters also work well in quantifying paleorainfall. For example, our present results suggest that $\chi_{\text{ARM}}$ is a superior proxy (Figure 11).

It is interesting to note that the magneto-climofunctions based on the $\chi_{\text{td}}$ of the bulk samples and clay sub-samples (Figures 10c and 10d) have similar slopes (99.70 for bulk samples and 120.24 for clay sub-samples) and same coefficient of determination ($R^2 = 0.82$). In addition, for the eight samples from the MS-77–MS-107 transect, the coefficient of determination of the bulk samples is even relative higher than that of the clay subsamples. The possible explanation is that the pedogenic fine-grained maghemite occurs in two forms. One form is pedogenic maghemite in clay fractions. Another form occurs in the silt fraction, mainly originating from the low-temperature oxidation of coarse-grained magnetite. The degree of the oxidation shows an increasing trend from northern CLP to southern CLP, which indicates that maghemite concentrations in the silt fraction are dependent on the degree of pedogenesis, pointing to pedogenic origin for maghemite in the silt fraction. Therefore, $\chi_{\text{td}}$ of the bulk samples captures all the pedogenic signals and, for quantifying paleorainfall, may have the advantage over measurements of the clay fraction alone.

4. Conclusions

We have presented here a systematic multiparameter magnetic investigation of the magnetic properties of a suite of the bulk and particle-fractioned modern soil samples mainly along a NW-SE transect on the CLP to develop a new magneto-climofunction for paleoprecipitation reconstruction. The main conclusions are the following:
1. Magnetic measurements on the bulk soil samples and the clay and silt fractions derived from them improve our understanding of the results of some frequently used magnetic measurements on the loess and paleosols. Our results show that low-temperature saturation isothermal remnant magnetization measurements of the bulk soil samples mainly reflect the properties of the silt fraction. Day plot parameters of the hysteresis loops may provide misleading and even erroneous information on the magnetic domain state of samples. FORC diagrams can provide detailed information about the magnetic grains in a sample.

2. Rock magnetic studies on particle-sized subsamples indicate that the final product of pedogenic ferrimagnetic minerals is a mixture of finer SP and SD maghemite. Spatial variations in pedogenic magnetic properties are predominantly dependent on changes in the abundance of fine-grained maghemite.

3. We establish a new magneto-climofunction for paleoprecipitation reconstruction from Chinese loess. A new parameter $\chi_{ab}$, which quantifies the degree of the thermally induced alteration between $-230$ and $-400 \degree C$ on the heating curves of high-temperature-dependent susceptibility measurements, exclusively reflects the abundance of pedogenic maghemite. The new $\chi_{ab}$-MAP climofunction shows a high coefficient of determination and lower standard error than those climofunction based on routine ferrimagnetic proxies. It therefore provides an improved approach to quantitatively reconstruct the history of past variations in paleomonsoon precipitation using the record from Chinese loess-paleosol sequences.

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