Grain size distribution of pedogenic magnetic particles in Chinese loess/paleosols

Qingsong Liu,[1,2,3] Michael J. Jackson,[1] Yongjae Yu,[4] Fahu Chen,[5] Chenglong Deng,[6] and Rixiang Zhu[6]

Received 23 July 2004; revised 11 October 2004; accepted 18 October 2004; published 18 November 2004.

[1] Variations in magnetic parameters of the long Chinese loess/paleosol sequences reflect paleoclimatic fluctuations. However, there remain ambiguities whether the magnetic proxies are controlled by the concentration or by the distribution of magnetic grain sizes. We systematically investigated the frequency-dependence of low-field magnetic susceptibility (χ_{fd}%) and the decay-rate (DR) dependence of anhysteretic remanence magnetization (ARM) susceptibility ($\chi_{ARM,DR}$%) for a set of representative loess/paleosol samples from the Yuanbao section, western Chinese loess plateau. After removing the effect of aeolian coarse-grained magnetites, both χ_{fd}% and $\chi_{ARM,DR}$% are nearly independent of the degree of pedogenesis, indicating a presence of fine-grained magnetic particles ranging from superparamagnetic to single domain with an almost constant grain size distribution. Therefore, the magnetic enhancement of magnetic susceptibility or ARM is caused dominantly by changes in the concentration of these fine-grained pedogenic grains. *INDEX TERMS:* 1512 Geomagnetism and Paleomagnetism: Environmental magnetism; 1519 Geomagnetism and Paleomagnetism: Magnetic mineralogy and petrology; 1540 Geomagnetism and Paleomagnetism: Rock and mineral magnetism. **Citation:** Liu, Q., M. J. Jackson, Y. Yu, F. Chen, C. Deng, and R. Zhu (2004), Grain size distribution of pedogenic magnetic particles in Chinese loess/paleosols, *Geophys. Res. Lett.*, *31*, L22603, doi:10.1029/2004GL021090.

1. Introduction

[2] The Chinese loess is an excellent terrestrial material for recording the long-term (~2.5 Myr) paleoclimatic variations in eastern Asia. For a characteristic loess profile, paleosols always have higher susceptibility than the interbedded (less pedogenically altered) loess units, reflecting dominant monsoon patterns in terms of large-scale atmospheric circulation. The loess/paleosol rhythms have been successfully correlated to the marine oxygen isotope records [*Heller and Liu*, 1984; *Kukla et al.*, 1988; *Hovan et al.*, 1989; *Bloemendal et al.*, 1995].

[3] Pedogenic magnetic particles (maghemite, γ-Fe_2O_3) have long been recognized as the dominant carriers for the susceptibility enhancements of the Chinese loess/paleosols [*Zhou et al.*, 1990; *Maher and Thompson*, 1991; *Hus and Han*, 1992; *Verosub et al.*, 1993; *Heller and Evans*, 1995; *Deng et al.*, 2000; *Liu et al.*, 2004a]. *Zhou et al.* [1990] showed that the enhanced susceptibility of paleosols is strongly linked to the ultrafine superparamagnetic (SP, <20–25 nm) particles produced through pedogenesis. A recent study revealed that single-domain (SD) magnetic particles also contribute more than half of the magnetic susceptibility of paleosol samples [*Liu et al.*, 2004b].

[4] The magnetic properties of these fine-grained (SP + SD) particles are determined both by the grain size distribution and concentrations. It has been generally regarded that variations in the concentrations of these pedogenic particles are more important than changes in grain sizes [*Florindo et al.*, 1999; *Deng et al.*, 2004], but no sound evidence was provided. To provide a rigorous interpretation of the pedogenic signals in terms of grain size, concentration, or a combination of two, we investigated the frequency-dependence of magnetic susceptibility (sensitive to SP particles) [*Maher*, 1988] and the decay-rate dependence of anhysteretic remanent magnetization (ARM, sensitive to SD particles) [*Yu and Dunlop*, 2003].

2. Sampling and Experiments

[5] The paleosol unit S1, corresponding to the Marine Oxygen Isotope Stage 5 (MIS 5), of the Yuanbao (YB) section (35°38′N/103°10′E) consists of three well-developed sub-paleosol units (S1S1/MIS5a, S1S2/MIS5c, and S1S3/MIS5e) and two interbedded sub-loess layers (S1L1/MIS5b and S1L2/MIS5d) [*Chen et al.*, 1999]. This study focuses on the S1S3 section that represents the interglacial maximum (Figure 1).

[6] The low- and high-frequency magnetic susceptibility (χ, mass-specific) was initially measured with a dual-frequency Bartington Susceptometer at 470 and 4700 Hz, respectively. The parameters χ_{fd} and χ_{fd}% were defined as $\chi_{470Hz} - \chi_{4700Hz}$ and $100\%*(\chi_{470Hz} - \chi_{4700Hz})/\chi_{470Hz}$, respectively. Hereafter, χ alone also refers to χ_{470Hz}.

[7] ARM was imparted in a peak alternating field (AF) of 200 mT with a bias field of 100 μT using a Dtech D2000 instrument. χ_{ARM} is calculated from ARM normalized by the bias field. We used five different decay rates (DR), 2, 5, 10, 15, and 20 μT/cycle. DR-dependence of χ_{ARM} ($\chi_{ARM,DR}$) is defined as $\chi_{ARM,2} - \chi_{ARM,20}$, where $\chi_{ARM,2}$ and $\chi_{ARM,20}$ represent χ_{ARM} measured with DRs of 2 and

[1]Institute for Rock Magnetism, Department of Geology and Geophysics, University of Minnesota, Minneapolis, Minnesota, USA.
[2]Formerly at Paleomagnetism Laboratory, Institute of Geology and Geophysics, Chinese Academy of Sciences, Beijing, China.
[3]Now at Department of Earth Sciences, University of California, Santa Cruz, California, USA.
[4]Geosciences Research Division, Scripps Institution of Oceanography, La Jolla, California, USA.
[5]Chinese Academy of Engineering Physics, College of Earth and Environment Science, University of Lanzhou, Lanzhou, China.
[6]Paleomagnetism Laboratory, Institute of Geology and Geophysics, Chinese Academy of Sciences, Beijing, China.

0094-8276/04/2004GL021090$05.00

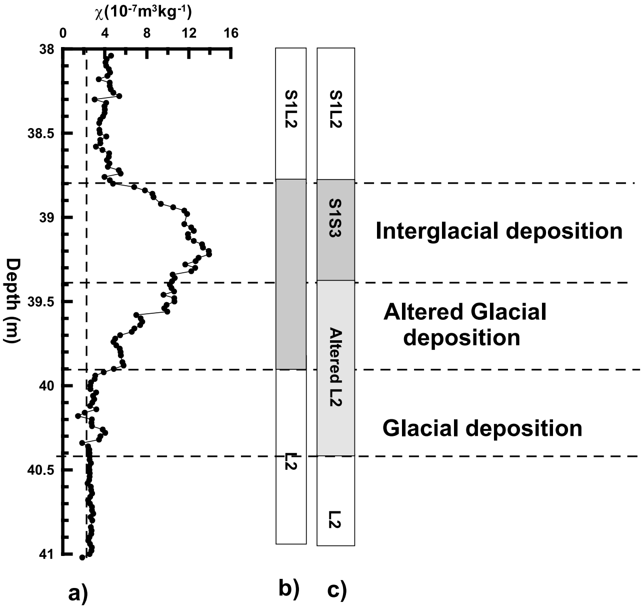

Figure 1. Plots of susceptibility (a), and stratigraphy of the Yuanbao profile interpreted by *Chen et al.* [1999] (b), and by *Liu et al.* [2004a] (c).

20 μT/cycle, respectively. In addition, $\chi_{ARM,DR}$% is defined as a percentage difference, $\chi_{ARM,2}/\chi_{ARM,20} - 1$.

3. Results

[8] χ_{fd} (sensitive to the absolute concentration of SP particles) and χ (contributed from both aeolian and pedogenic particles) show a perfect linear correlation (Figure 2a). The x-axis intercept (hereafter denoted χ_0) is $\sim$2.0 $\times$ 10^{-7} m^3 kg^{-1}, which represents the initial aeolian inputs without pedogenic overprints [*Liu et al.*, 2004b]. χ_{fd}% (sensitive to the grain size distribution of SP particles) is positively but non-linearly correlated with χ (Figure 2b), and reaches a plateau ($\sim$12%) when $\chi > \sim 10 \times 10^{-7}$ m^3 kg^{-1}. To compensate for the effect of the initial aeolian signals, the corrected χ_{fd}% is calculated from $(\chi_{470Hz} - \chi_{4700Hz})/(\chi_{470Hz} - \chi_0)$, which ($\sim$14%) is almost independent of the bulk susceptibility (Figure 2b). Note that correction of χ_o is not needed for the numerator $(\chi_{470Hz} - \chi_{4700Hz})$ because χ_0 is frequency-independent and has been canceled out by subtraction of χ measured at two frequencies. Assuming that χ_{fd}% remains constant ($\sim$14%) in our particular dataset, then χ_{fd} of the pedogenic magnetic particles is 0.14*$(\chi - \chi_0)$. The simulated χ_{fd}% (14%*$(\chi - \chi_0)/\chi$) (gray curve in Figure 2b) fits the observed χ_{fd}% well.

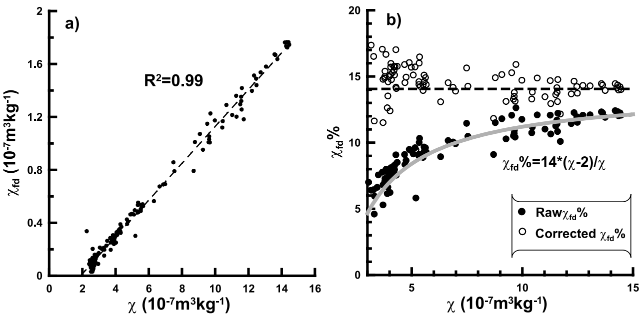

Figure 2. Correlation between χ_{fd} and χ (a), χ_{fd}% and χ (b). Open and solid circles in represent the corrected and the raw χ_{fd}%, respectively. The horizontal dashed line shows that the corrected χ_{fd}% is almost independent of χ. The gray curve is the simulated χ_{fd}% based on the equation $\chi_{fd}\% = 14\%*(\chi_{fd} - \chi_0)/\chi$, where $\chi_0 = 2*10^{-7}$ m^3 kg^{-1}.

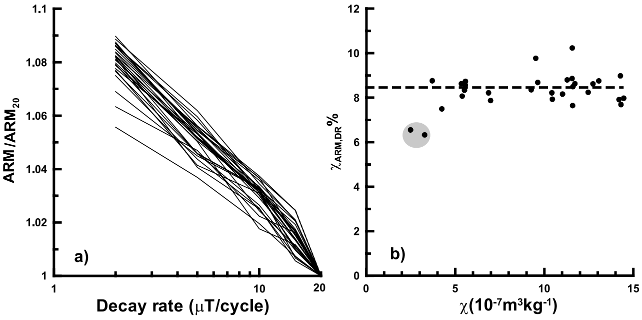

Figure 3. (a) Plots of normalized ARM versus decay rate. ARM_{20} represents ARM measured with DR of 20 μT/cycle. (b) DR-dependence of ARM ($\chi_{ARM,DR}$%) versus the bulk susceptibility (measured at 470 Hz). The horizontal dashed line shows that $\chi_{ARM,DR}$% is almost independent of χ when χ is slightly enhanced above $\sim 4 \times 10^{-7}$m^{3}kg^{-1}. The shaded circle in (b) marks the least-altered loess samples with lower $\chi_{ARM,DR}$% values.

[9] Decay-rate dependence of ARM is shown in Figure 3a. With an order of magnitude increase in DR, ARM intensities gradually decrease by $\sim$8%. $\chi_{ARM,DR}$% is almost independent of the bulk susceptibility except for samples with $\chi < 4 \times 10^{-7}$ m^3 kg^{-1} (Figure 3b). Both $\chi_{ARM,DR}$ and χ_{fd} exhibit a strongly positive correlation, whose extrapolated line passes the origin, indicating that the concentration of SP and SD magnetic particles linearly co-vary with the degree of pedogenesis (Figure 4).

4. Discussion

4.1. SP (Superparamagnetic) Indicator

[10] χ_{fd}% is a sensitive parameter for detecting the presence of SP grains in soils. However, this parameter is

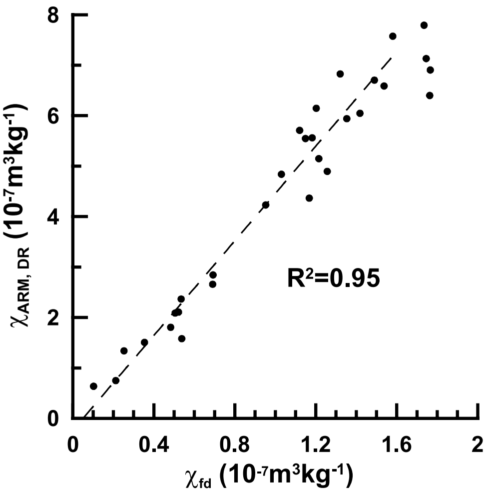

Figure 4. Correlation between $\chi_{ARM,DR}$ and χ_{fd}. The dashed line is the linear trend with R^2 = 0.95.

preferentially sensitive to "viscous SP" (VSP) particles (just smaller than the blocking volume, $\sim$20 nm for magnetite and maghemite) with a very narrow grain size distribution [*Maher*, 1988]. Above this VSP range, particles become blocked and are in SD states. Below the VSP size, particles are in ideal SP states (independent of frequency because their relaxation times are much smaller than the time constants for the susceptibility measurements). Theoretically, $\chi_{fd}\%$ could be $\sim$100% for only viscous SP particles. On the contrary, most observations on soils show a "saturation" of $\chi_{fd}\%$ at $\sim$15% [e.g., *Stephenson*, 1971; *Mullins*, 1977; *Oldfield et al.*, 1985; *Forster et al.*, 1994; *Chen et al.*, 1999]. On the other hand, for samples with narrower size distributions of SP + SD particles, higher $\chi_{fd}\%$ values were documented [*Worm*, 1998; *Worm and Jackson*, 1999]. Both *Worm* [1998] and *Worm and Jackson* [1999] found that $\chi_{fd}\%$ inversely relates to the width of grain size distribution.

[11] After removing the effect of the initial aeolian signal, the corrected $\chi_{fd}\%$ ($\sim$14%), corresponding to pure pedogenic particles (SP + SD), is independent of the degree of pedogenesis (Figure 2b). This observation strongly indicates that the pedogenic particles have a wide grain size distribution. Meanwhile χ_{fd} is linearly correlated to the bulk χ. Therefore, it is highly unlikely that the grain size distribution of the pedogenic particle changes with the degree of pedogenesis. Otherwise, very special adjustment of the grain size distribution is needed to keep both the linear relationship between χ_{fd} and χ and the constant $\chi_{fd}\%$.

4.2. SD (Single-Domain) Indicator

[12] ARM is influenced by several factors: grain size [*Dunlop and Argyle*, 1997; *Egli and Lowrie*, 2002], concentration [*Yamazaki and Ioka*, 1997], the peak AF intensity, the instrument [*Sagnotti et al.*, 2003], and DR [*Sagnotti et al.*, 2003; *Yu and Dunlop*, 2003]. For natural samples, magnetic particles are generally well dispersed, thus the effect of volume concentration would be negligible. Despite the limitation of the instrument factor [*Sagnotti et al.*, 2003], the relative changes in $\chi_{ARM,DR}\%$ for a fixed instrument surely reflect changes in grain sizes (e.g., Figures 3 and 4). As a result, a direct comparison of $\chi_{ARM,DR}\%$ should be limited only for one-type of instrument, unless the information of the inter-laboratory calibration is established [e.g., *Sagnotti et al.*, 2003].

[13] Our loess/paleosol samples show $\sim$8–9% variation of $\chi_{ARM,DR}\%$ (Figure 3), agreeing well with the reported DR-dependence of ARM for SD grains [*Yu and Dunlop*, 2003]. A prevailing dominance of SD particle loess/paleosol samples was also inferred from a detailed rock magnetic analysis [*Liu et al.*, 2004b]. The pedogenesis-independence of $\chi_{ARM,DR}\%$ indicates that the grain size distribution of these ARM carriers changes very little, whereas their concentrations change significantly with pedogenesis (Figure 3b). The lower $\chi_{ARM,DR}\%$ for the least-altered loess samples (Figure 3b) is due to the significant effects of aeolian coarse-grained magnetic particles. A linear correlation between $\chi_{ARM,DR}$ and χ_{fd} (Figure 4) shows that the concentration of pedogenic SD and viscous SP co-varies.

5. Conclusions

[14] In summary, pedogenic processes produce a wide grain size range of magnetic particles, but with an overall relatively fixed grain size distribution. Therefore, fluctuations in susceptibility and ARM of the Chinese loess/paleosols are caused dominantly by changes in concentrations of these fine-grained pedogenic particles.

[15] **Acknowledgments.** We thank B. Carter-Stiglitz for collecting samples, and two anonymous reviewers for insightful comments. This study is supported by NSFC grants 40325011 and 40221402, NSF Grants No. EAR 0003421, and EAR/IF 9818704. The fieldwork and sampling is partly supported by NSFC Grant 40125001. C. Deng acknowledges further support from the Royal Society in the form of a BP Amoco Fellowship. All rock magnetic measurements were made at the Institute for Rock Magnetism (IRM), which is supported by the W.M. Keck Foundation, the Earth Science Division of the US National Science Foundation, and the University of Minnesota. This is IRM publication #0411.

References

Bloemendal, J., X. M. Liu, and T. C. Rolph (1995), Correlation of the magnetic susceptibility stratigraphy of Chinese loess and the marine oxygen isotope record: Chronological and palaeoclimatic implications, *Earth Planet. Sci. Lett.*, *131*, 371–380.

Chen, F. H., J. Bloemendal, Z. D. Feng, J. M. Wang, E. Parker, and Z. T. Guo (1999), East Asian monsoon variations during Oxygen Isotope Stage 5: Evidence from the northwestern margin of the Chinese loess plateau, *Quat. Sci. Rev.*, *18*, 1127–1135.

Deng, C. L., R. X. Zhu, K. L. Verosub, M. J. Singer, and B. Y. Yuan (2000), Paleoclimatic significance of the temperature-dependent susceptibility of Holocene loess along a NW-SE transect in the Chinese loess plateau, *Geophys. Res. Lett.*, *27*, 3715–3718.

Deng, C., R. Zhu, K. L. Verosub, M. J. Singer, and N. J. Vidic (2004), Mineral magnetic properties of loess/paleosol couplets of the central loess plateau of China over the last 1.2 Myr, *J. Geophys. Res.*, *109*, B01103, doi:10.1029/2003JB002532.

Dunlop, D. J., and K. S. Argyle (1997), Thermoremanence, anhysteretic remanence and susceptibility of submicron magnetites: Nonlinear field dependence and variation with grain size, *J. Geophys. Res.*, *102*, 20,199–20,210.

Egli, R., and W. Lowrie (2002), Anhysteretic remanent magnetization of fine magnetic particles, *J. Geophys. Res.*, *107*(B10), 2209, doi:10.1029/2001JB000671.

Florindo, F., R. Zhu, B. Guo, L. Yue, Y. Pan, and F. Speranza (1999), Magnetic proxy climate results from the Duanjiapo loess section, southernmost extremity of the Chinese loess plateau, *J. Geophys. Res.*, *104*, 645–659.

Forster, T., M. E. Evans, and F. Heller (1994), The frequency dependence of low field susceptibility in loess sediments, *Geophys. J. Int.*, *118*, 636–642.

Heller, F., and M. E. Evans (1995), Loess magnetism, *Rev. Geophys.*, *33*, 211–240.

Heller, F., and T. S. Liu (1984), Magnetism of Chinese loess deposits, *Geophys. J. R. Astron. Soc.*, *77*, 125–141.

Hovan, S. A., D. K. Rea, N. G. Pisias, and N. J. Shackleton (1989), A direct link between the China loess and marine $\delta^{18}O$ records: Aeolian flux to the North Pacific, *Nature*, *340*, 296–298.

Hus, J. J., and J. M. Han (1992), The contribution of loess magnetism in China to the retrieval of past global changes: Some problems, *Phys. Earth Planet. Inter.*, *7*, 154–168.

Kukla, G. J., F. Heller, X. M. Liu, T. C. Xu, T. S. Liu, and Z. S. An (1988), Pleistocene climates in China dated by magnetic susceptibility, *Geology*, *16*, 811–814.

Liu, Q. S., S. K. Banerjee, M. J. Jackson, F. H. Chen, Y. X. Pan, and R. X. Zhu (2004a), Determining the climatic boundary between the Chinese loess and palaeosol: Evidence from aeolian coarse-grained magnetite, *Geophys. J. Int.*, *156*, 267–274.

Liu, Q. S., S. K. Banerjee, M. J. Jackson, B. A. Maher, Y. X. Pan, R. X. Zhu, C. L. Deng, and F. H. Chen (2004b), Grainsizes of susceptibility and anhysteretic remanent magnetization carriers in Chinese loess/paleosol sequences, *J. Geophys. Res.*, *109*, B03101, doi:10.1029/2003JB002747.

Maher, B. A. (1988), Magnetic properties of some synthetic sub-micron magnetites, *Geophys. J.*, *94*, 83–96.

Maher, B. A., and R. Thompson (1991), Mineral magnetic record of the Chinese loess and paleosol, *Geology*, *19*, 3–6.

Mullins, C. E. (1977), Magnetic susceptibility of the soil and its significance in soil science: A review, *J. Soil Sci.*, *28*, 223–246.

Oldfield, F., B. A. Maher, J. Donoghue, and J. Pierce (1985), Particle-size related, mineral magnetic source-sediment linkages in the Rhode River catchment, Maryland, USA, *J. Geol. Soc. London*, *142*, 1035–1046.

Sagnotti, L., P. Rochette, M. Jackson, F. Vadeboin, J. Dinares-Turell, A. Winkler, and MAG-NET Science Team (2003), Inter-laboratory

calibration of low-field magnetic and anhysteretic susceptibility measurements, *Phys. Earth Planet. Inter.*, *138*, 25–38.
Stephenson, A. (1971), Single domain grain distributions I. A method for the determination of single domain grain distributions, *Phys. Earth Planet. Inter.*, *4*, 353–360.
Verosub, K. L., P. Fine, M. J. Singer, and J. TenPas (1993), Pedogenesis and paleoclimate: Interpretation of the magnetic susceptibility record of Chinese loess-paleosol sequences, *Geology*, *21*, 1011–1014.
Worm, H.-U. (1998), On the superparamagnetic-stable single domain transition for magnetite, and frequency dependency of susceptibility, *Geophys. J. Int.*, *133*, 201–206.
Worm, H.-U., and M. Jackson (1999), The superparamagnetism of Yucca Mountain Tuff, *J. Geopys. Res.*, *104*, 25,415–25,425.
Yamazaki, T., and N. Ioka (1997), Cautionary note on magnetic grain-size estimation using the ratio of ARM to magnetic susceptibility, *Geophys. Res. Lett.*, *24*, 751–754.
Yu, Y. J., and D. J. Dunlop (2003), Decay-rate dependence of anhysteretic remanence: Fundamental origin and paleomagnetic applications, *J. Geophys. Res.*, *108*(B12), 2550, doi:10.1029/2003JB002589.
Zhou, L. P., F. Oldfield, A. G. Wintle, S. G. Robinson, and T. J. Wang (1990), Partly pedogenic origin of magnetic variations in Chinese loess, *Nature*, *346*, 737–739.

F. Chen, CAEP, College of Earth and Environment Science, University of Lanzhou, Lanzhou, China. (fhchen@lzu.edu.cn)

C. Deng and R. Zhu, Paleomagnetism Laboratory, Institute of Geology and Geophysics, Chinese Academy of Sciences, Beijing 1000029, China. (cldeng@mail.igcas.ac.cn; rxzhu@mail.igcas.ac.cn)

M. J. Jackson, Institute for Rock Magnetism, Department of Geology and Geophysics, University of Minnesota, 310 Pillsbury Drive S.E., Minneapolis, MN 55455, USA. (irm@umn.edu)

Q. Liu, Department of Earth Sciences, University of California, Santa Cruz, 1156 High Street, Santa Cruz, CA 95064, USA. (qliu@es.ucsc.edu)

Y. Yu, Geosciences Research Division, Scripps Institution of Oceanography, 9500 Gilman Drive, La Jolla, CA 92093-0220, USA. (yjyu@ucsd.edu)