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RESEARCH NOTE

Frequency dependence of magnetic susceptibility for populations of single-domain grains

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SUMMARY

A recent paper (Dearing *et al.* 1996) on the use of frequency dependence of magnetic susceptibility (X_{fd}) in magnetic grain-size investigations of environmental materials has proposed a fundamentally new model for the magnetic susceptibility of single-domain (SD) grains measured at different frequencies, and is divergent from the now well-established theory of SD behaviour. This new model appears to be founded on a confusion about the behaviour of SD grains, as predicted by theory, and magnetic measurements of real materials, which naturally have a distribution of magnetic grain sizes. Here I try to clear up this confusion by showing how different log-normal distributions of magnetic grain sizes affect the frequency dependence of magnetic susceptibility measurement. This analysis highlights that the limiting maximum X_{fd} value of ~15 per cent obtained for environmental materials can be explained simply as a constraint imposed by the magnetic grain-size distribution having a finite minimum width (~1.0 on a natural log scale). There is no reason to suppose that if the grain-size dispersion was narrower higher X_{fd} values could not be observed. Furthermore, the X_{fd} value is non-unique and so cannot be used quantitatively to determine the amount or grain-size distribution of SD grains in a sample

INTRODUCTION

Magnetic susceptibility measurements are now routinely made in magnetic investigations of environmental materials. This is largely because the measurement is quick and simple to carry out and the equipment required is relatively cheap. Magnetic susceptibility values on their own do not reveal much information about the magnetic mineral assemblage of a material because magnetic susceptibility values are non-uniquely affected by magnetic concentration, mineralogy and grain size. In most situations further magnetic measurements are required to characterize a magnetic assemblage in greater detail in order to deduce possible environmental influences. Often, as is the case with the widely employed Bartington dual-frequency susceptibility probe, magnetic susceptibility can be obtained at two different frequencies. This allows for the determination of a further magnetic parameter, X_{fd}, which is the difference in magnetic susceptibility obtained at the two frequencies (470 and 4700 Hz for the Bartington). This has led to considerable interest in what additional information this parameter provides. It is considered loosely to be an indication of the amount of superparamagnetic material present in a sample, but is it possible to use the parameter more quantitatively to determine grain sizes?

THEORY

Origin of the frequency dependence of magnetic susceptibility

Below a critical size, thermal energy becomes large enough to spontaneously switch the magnetic moments of small, single-domain grains in a relatively short time (Néel 1949). For example, if a population of such grains is magnetized and then the field removed, a measurement of remanence after a time $(t)=t$ will reveal that the value has decreased to 1/e of its initial value. This is known as superparamagnetic relaxation and t is known as the relaxation time. The importance of time in magnetic measurements of small grains is exploited in the measurement of the frequency dependence of magnetic susceptibility. The difference between magnetic susceptibility measurements obtained at the two different frequencies is a function of the concentration of grains that have relaxation frequencies (i.e. $1/t$) that lie between the two measuring frequencies. The relationship between the critical blocking volume (v_b) of a SD grain and the relaxation frequency (≈ 2 times the measurement frequency f_m) is governed by the equation

$$\log\left(\frac{2f_m}{f_0}\right) = -\left(\frac{Kv_b}{kT}\right), \tag{1}$$

where K is the effective anisotropy constant, k is Boltzmann's constant, T is temperature and f_0 is a constant $= 10^{12}$ Hz (Dickson *et al.* 1993). Taking magnetite as an example, and assuming that the stable single-domain (SSD) to superparamagnetic (SP) threshold lies at ~ 30 nm for a laboratory measurement time of 100 s (Dunlop 1973), the two measurement frequencies of 470 and 4700 Hz correspond to critical grain volumes of 1.24×10^{-23} and 1.10×10^{-23} m^3 (or grain diameters of 28.7 and 27.6 nm), respectively. The exponential relationship between relaxation time and grain volume means that the threshold between grains that behave superparamagnetically and those that behave stably is very narrow. At room temperature the magnetic susceptibility of grains that are just below the SSD/SP threshold is of the order of 14 times that of grains that lie above (Stephenson 1971). X_{fd} (per cent) is taken to be the difference in magnetic susceptibility measured at the two frequencies as a percentage of the magnetic susceptibility measured at the low frequency [i.e. X_{fd} (per cent)$=$ $(X_{\mathrm{lf}} - X_{\mathrm{hf}}) \times 100/(X_{\mathrm{lf}})$]. It follows that if all grains in a sample 'block in' between the two measuring frequencies (that is, they change from being superparamagnetic to being stable), X_{fd} will be roughly 90 per cent. The variation of magnetic susceptibility of magnetite grains (at room temperature) with grain volume is illustrated in Fig. 1 for the two different measuring frequencies. As can be seen, the only difference between the two measurements lies in the grain-volume range between 1.24×10^{-23} and 1.10×10^{-23} m^3. The magnetic susceptibilities are identical for the rest of the SD grains outside this range. This behaviour and the following analysis can be generalized to other SD magnetic minerals, although the critical blocking volumes and the magnetic susceptibilities will be different.

Frequency dependence for distributions of grain sizes

Naturally occurring populations of grains will have a distribution of grain sizes and hence their magnetic susceptibility will be a convolution of the susceptibility function $X\{v\}$, shown in Fig. 1, and a grain-size distribution. For purposes of modelling, a log-normal distribution of SD magnetic grains is assumed (Nunes & Yu 1989).

For the number of grains with volume v, the following function can be written:

$$f\{v,v_m,\sigma\} \propto \exp\left[\frac{-(\log[v]-\log[v_m])^2}{2\sigma^2}\right], \tag{2}$$

where v_m is the mode of the distribution and σ is the width on a log scale. Multiplying this function by v gives the volume of grains at each volume. The total volume of grains is normalized in all cases to unity. This is then convolved with the susceptibility function $X\{v\}$ to give a magnetic susceptibility value for the whole grain population (X_{pop}) for different modal positions of the distribution:

$$X_{\mathrm{pop}}\{v_m,\sigma\} = \sum_v f\{v,v_m,\sigma\} v X\{v\}. \tag{3}$$

X_{pop} was modelled for both measurement frequencies and the frequency dependence derived in the same manner as described previously. Fig. 2 shows the variation of X_{fd} for different modal positions (v_m) of the grain-volume distribution. Seven curves have been calculated corresponding to seven grain-volume distributions with widths (σ) from 0.4 to 1.0. As is clear from intuition, the highest values of X_{fd} are associated with narrow grain distributions that are centred around the SP/SSD boundary. The X_{fd} values fall off rapidly for v_m values above the SP/SSD boundary; this is a reflection of the log-normal distribution, which has a steeper gradient on the lower side. For wider distributions, below the SP/SSD boundary, the variation of X_{fd} with v_m becomes less pronounced.

DISCUSSION

As highlighted by Dearing *et al.* (1996), studies of environmental materials have failed to find X_{fd} values higher than 15 per cent. The same is true for studies on geological and synthetic materials that contain magnetic grains (Heller & Evans 1995; Maher 1988). The question is whether this is some failing of SD theory or whether there is some other explanation. As I have shown here, a simple explanation involves distributions

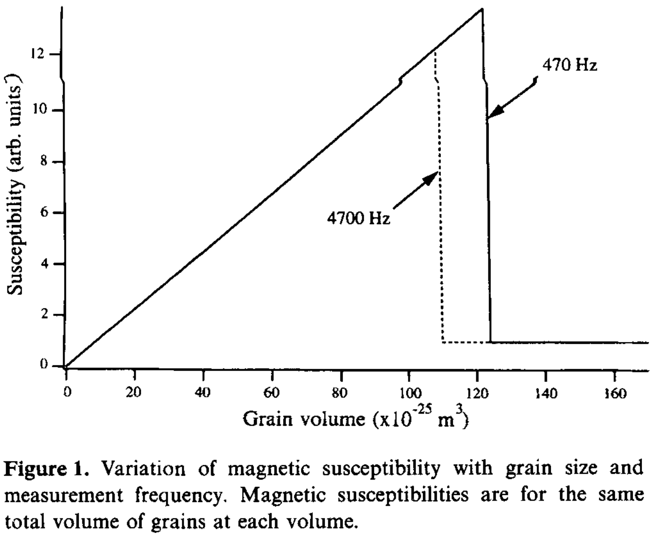

Figure 1. Variation of magnetic susceptibility with grain size and measurement frequency. Magnetic susceptibilities are for the same total volume of grains at each volume.

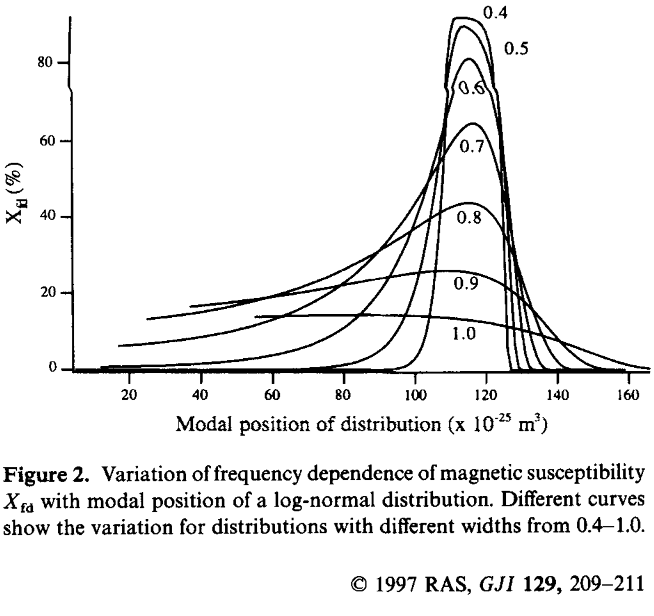

Figure 2. Variation of frequency dependence of magnetic susceptibility X_{fd} with modal position of a log-normal distribution. Different curves show the variation for distributions with different widths from 0.4–1.0.

of grain sizes, which logically have a finite minimum width. This minimum width is too broad to allow a large proportion of the grains to 'block in' between the two frequencies and so X_{fd} values fall far short of the maximum predicted by theory. A minimum width for a distribution of magnetite grains of ~ 1.0 can be deduced from Fig. 2, although this may need to be revised given the present uncertainties about the position of the SP/SSD threshold. Fig. 2 also demonstrates that, even for samples containing a single magnetic component, X_{fd} is not a particularly useful parameter for determining grain sizes. For example, for the distribution with $\sigma = 1.0$, X_{fd} is quite constant for v_{m} below the SP/SSD threshold. Further information is required for a more detailed determination of the SD grain-size distribution. For example, this could be magnetic susceptibility obtained at a third frequency or magnetic susceptibility measured at low temperature.

On the general theme of fine-grained magnetic material in environmental materials, it is perhaps significant to note that the magnetic characteristics of the 'enhancing fraction' in palaeosols developed on Chinese and Tadjikistan loess, and in soils developed on silt and sand deposits on the Californian coast (Evans & Heller 1994; Eyre & Shaw 1994; Forster, Evans & Heller 1994; Singer *et al.* 1992) are so similar as to be practically indistinguishable. The implication of this is that the magnetic fraction responsible for magnetic susceptibility enhancement in soils (and possibly in a wider range of environments) may prove to be from a common origin and be composed of magnetic grains with a distinct, non-variable grain-size distribution. If this is the case, then the parameter $X_{\mathrm{lf}} - X_{\mathrm{hf}}$ could in many situations be used as a direct measure of the relative concentration of this component.

REFERENCES

Dearing, J.A., Dann, R.J.L., Hay, K., Lees, J.A., Loveland, P.J., Maher, B.A. & O'Grady, K., 1996. Frequency-dependent susceptibility measurements of environmental materials, *Geophys. J. Int.*, **124,** 228–240.

Dickson, D.P.E., Reid, N.M.K., Hunt, C., Williams, H.D., El-Hilo, M. & O'Grady, K., 1993. Determination of f_0 for fine magnetic particles, *J. Magn. Magnetic Mat.*, **125,** 345–350.

Dunlop, D.J., 1973. Superparamagnetic and single domain threshold sizes in magnetite, *J. geophys. Res.*, **78,** 1780–1793.

Evans, M.E. & Heller, F., 1994. Magnetic enhancement and palaeoclimate: Study of a loess/palaeosol couplet across the Loess Plateau of China, *Geophys. J. Int.*, **117,** 257–264.

Eyre, J.K. & Shaw, J., 1994. Magnetic enhancement of the Chinese loess – the role of γFe_2O_3, *Geophys J. Int.*, **117,** 265–271.

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

Heller, F. & Evans, M.E., 1995. Loess magnetism, *Rev. Geophys.*, **33** (2), 211–240.

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

Néel, L., 1949. Théorie du traînage magnétique des ferromagnétiques en grains fins avec application aux terres cuites, *Ann. Geophys.*, **5,** 99–136.

Nunes, A.C. & Yu, Z.-C., 1989. Size and quality dispersion of a single preparation of colloidal magnetic particles, *J. Magn. Magnetic Mat.*, **78,** 241–246.

Singer, M.J., Fine, P., Verosub, K.L. & Chadwick, O.A., 1992. Time dependence of magnetic susceptibility of soil chronosequences on the California coast, *Quaternary Res.*, **37,** 323–332.

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.