

Models of frequency-dependent susceptibility of rocks and soils revisited and broadened

František Hrouda^{1,2}

¹AGICO Ltd., Brno, Czech Republic. E-mail: fhrouda@agico.cz

²Institute of Petrology and Structural Geology, Charles University, Prague, Czech Republic

Accepted 2011 September 7. Received 2011 August 30; in original form 2010 November 10

SUMMARY

Mathematical models of the frequency-dependent susceptibility in rocks, soils and environmental materials have been adapted to measurements performed with multiple operating frequencies (465, 976, 3904, 4650, 15 616, 100 000 and 250 000 Hz) on the basis of log-normal volume distribution of magnetic particles. The X_{FD} parameter depends, in addition to the amount of SP particles, also on the operating frequencies, whose values should be therefore also presented. The model curves of the X_{FD} parameter versus arithmetical mean (μ) of the logarithms of grain volume are roughly bell-like shaped. The width and peak position of these curves is controlled by mean and standard deviation of the logarithmic volume distribution. Magnetic susceptibility contributions from paramagnetic minerals, and from ferrimagnetic particles not belonging to a unimodal SP/SD volume distribution, tend to decrease the X_{FD} parameter. Therefore, low X_{FD} values do not therefore necessarily indicate low amount of SP particles, but can also be indicative of the presence of the paramagnetic fraction. A new parameter X_R is introduced based on susceptibility measurements at three operating frequencies; it is insensitive to dia- and paramagnetic fractions and helps us to differentiate between wide and narrow size distributions of ferromagnetic particles. A new X_{FB} parameter is introduced that originates through normalizing the X_{FD} parameter by the difference of natural logarithms of operating frequencies and related to the decade difference between the frequencies. It is convenient for comparison of the Bartington MS-2 Susceptibility Meter data with the MFK1-FA Kappabridge data.

Key words: Environmental magnetism; Magnetic and electrical properties; Magnetic mineralogy and petrology.

INTRODUCTION

In environmental sciences and palaeoclimatology, the frequency-dependent magnetic susceptibility of rocks, soils and environmental materials is traditionally interpreted as resulting from the interplay between superparamagnetic (SP) and stable single domain (SSD) or even multidomain (MD) magnetic particles even though some other phenomena, such as eddy currents, may also play a role mainly at high operating frequencies. This approach was pioneered by Dearing *et al.* (1996), who introduced a parameter quantitatively characterizing the frequency dependence and developed a model for predicting the frequency-dependent susceptibility in environmental materials. Eyre (1997) extended this model considering populations of grains with variable grain sizes following the log-normal distribution and Worm (1998) considered also distribution of grain coercivities. All the above models were elaborated for two operating frequencies, viz. those possessed by the Bartington MS-2 Susceptibility Meter (465 and 4650 Hz).

The recently developed MFK1-FA Multi-Function Kappabridge (Pokorný *et al.* 2006) measures the magnetic susceptibility at three operating frequencies, viz. 976, 3904 and 15 616 Hz, in variable fields ranging from 2 to 700 A m⁻¹ at 976 Hz, from 2 to 350 A m⁻¹ at 3904 Hz and from 2 to 200 A m⁻¹ at 15 616 Hz. The sensitivity in measuring bulk susceptibility is in the order of 10⁻⁸ (SI), the sensitivity in measuring mass susceptibility being in the order of 10⁻¹¹ m³ kg⁻¹.

Assessment of the volume distribution of SP particles is also possible from temperature variation of susceptibility (for summary, see Shcherbakov & Fabian 2005; Egli 2009) or from magnetic hysteresis loops (for summary see Tauxe *et al.* 1996). As both these methods are very time consuming, the rapid frequency-dependent susceptibility is worth of being further developed.

The purpose of this paper is to adapt the above models for the frequencies of the MFK1-FA Kappabridge, those of the Bartington instrument, and the frequencies of 100 and 250 kHz. This provides us with a theoretical basis for comparing the data by the MFK1-FA

Kappabridge and the Bartington MS-2 instrument and, moreover, it enables us to investigate whether multiple frequencies have at least theoretical advantages compared to two frequencies approach used till now.

THEORETICAL BACKGROUND

The frequency-dependent susceptibility can be characterized by the following commonly accepted parameter introduced by Dearing *et al.* (1996)

$$X_{FD} = 100(X_{LF} - X_{HF})/X_{LF} (\%), \quad (1)$$

where χ_{LF} and χ_{HF} are susceptibilities at the low and high frequencies, respectively. Originally, Dearing *et al.* (1996) denoted this parameter by the small letter, χ_{FD} . To avoid confusion between susceptibilities and parameters derived from them, the susceptibilities will henceforth be still denoted by the small letter, χ , while the derived parameters by the capital letter, X . The X_{FD} parameter is simply to use if two operating frequencies are considered. In case of multiple frequencies, the same parameter can in principle be used for various pairs of frequencies, too, but the frequencies under consideration should be indicated in some way. We propose to add to the index FD in brackets also the frequencies under consideration rounded to kHz. For example, the $X_{FD(1,16)}$ parameter means that the operating frequencies are 1 and 16 kHz (in case of the MFK1-FA Kappabridge they are exactly 976 and 15 616 Hz), the $X_{FD(0.5,5)}$ parameter is that calculated from the measurement by the Bartington MS-2 Instrument (frequencies 465 and 4650 Hz).

Sometimes, it is advantageous to work with simple susceptibility difference

$$X_{FV} = X_{LF} - X_{HF}. \quad (2)$$

Dearing *et al.* (1996) calling it the relative loss of susceptibility. It should be noted that although the X_{FD} parameter is the same whether calculated from bulk susceptibilities as in eq. (1) or from mass susceptibilities, the X_{FV} parameter differ. The bulk and mass susceptibilities are related $\chi = \rho\kappa$, where ρ is the rock (soil) density and κ is the mass susceptibility. Then, the $X_{FV_{bulk}}$ parameter is dimensionless, the dimension of the $X_{FV_{mass}}$ parameter being $m^3 \text{ kg}^{-1}$.

All models of the frequency-dependent susceptibility are based on the concept of relaxation time introduced by Néel (1949)

$$\tau = \tau_0 \exp(KV/kT), \quad (3)$$

where V is the particle volume, K is the anisotropy constant, k is Boltzmann constant, T is absolute temperature and $\tau_0 \approx 10^{-10}$ s is a time constant. The relationship between the critical blocking volume (V_b) characterizing the SP/SSD threshold and the operating frequency ($\approx 1/2$ of the relaxation frequency, e.g. Eyre 1997) is then governed by the equation

$$V_b = \frac{kT}{K} \ln \left(\frac{f_0}{2f_m} \right), \quad (4)$$

where V_b is the blocking volume, f_m is the operating frequency and f_0 is a constant. For uniaxial particles whose magnetization can only reverse by coherent rotation, and assuming that the coercivity is independent of particle volume, the anisotropy constant can be related to the microscopic coercivity and saturation magnetization (Worm 1998; Worm & Jackson 1999)

$$K = \frac{\mu_0 H_k M_s}{2}, \quad (5)$$

where μ_0 is permeability of free space, H_k is microscopic coercivity related to macroscopic coercivity (H_c) as $H_k = 2.09H_c$ and M_s is saturation magnetization.

The blocking volume can then be related to the operating frequency

$$V_b = \frac{1}{\mu_0} \frac{2kT}{H_k M_s} \ln \left(\frac{f_0}{2f_m} \right). \quad (6)$$

The susceptibility of ensemble of non-interacting SSD particles, which are in fully blocked state, follow from the Stoner & Wohlfart (1948) theory

$$\chi_{sd} = \frac{2}{3} \frac{M_s}{H_k}, \quad (7)$$

whereas the susceptibility of SP particles, which are in fully unblocked state, is (e.g. Dunlop & Özdemir 1997)

$$\chi_{sp} = \mu_0 V \frac{M_s^2}{3kT}. \quad (8)$$

The susceptibility in between the blocked and unblocked states at the SP/SSD boundary resolves into a component that is in-phase with applied field (χ') and a component that is out-of-phase (χ''). It can be described by the formula introduced by Néel (1949) and transcribed by Egli (2009) as follows:

$$\chi_{sp/sd} = \chi_{sd} \left[\frac{\beta}{1 + i\tau_0\omega e^\beta} + 1 \right], \quad (9)$$

where $\beta = KV/kT$ and $\omega = 2\pi f_m$. The in-phase susceptibility, which is primarily measured by the Kappabridges of the KLY series, the MFK1-FA Kappabridge and the Lakeshore susceptometer, then is

$$\chi' = \chi_{sd} \left[\frac{\beta}{1 + (\tau_0\omega e^\beta)^2} + 1 \right]. \quad (10)$$

Fig. 1 shows the in-phase susceptibility versus particle volume plot for various operating frequencies for magnetite and maghemite. It is obvious that the maximum SP grain susceptibility as well as the blocking volume is the largest for the lowest operating frequency (465 Hz) and decrease with increasing frequency being the smallest at the highest operating frequency (250 kHz). The blocking volumes of maghemite are in general larger than those of magnetite (Table 1).

As the in-phase susceptibility in between the blocked and unblocked states at the SP/SSD boundary depends on the operating frequency, the X_{FD} and X_{FV} parameters depend on the instrument used. If one uses one type instrument only, there is no big problem arising from this situation. On the other hand, big problems may arise if the results obtained by the instruments working at different frequencies (e.g. the MFK1 Kappabridge and Bartington MS-2 Susceptibility Meter) should be compared. This problem could be overcome, if the frequency-dependent susceptibility is corrected for the operating frequencies used. As it follows from eq. (9) and Fig. 1(a) in Egli (2009), the existence of the out-of-phase susceptibility is most characteristic of the transition zone between unblocked and blocked state and χ'' is therefore best suited as frequency dependence parameter. The relationship between the in-phase and out-of-phase susceptibilities is described by the so-called $\pi/2$ -law (e.g. Egli 2009, eq. 18)

$$\frac{\partial \chi'}{\partial \ln f_m} = -\frac{2}{\pi} \chi'' \quad (11)$$

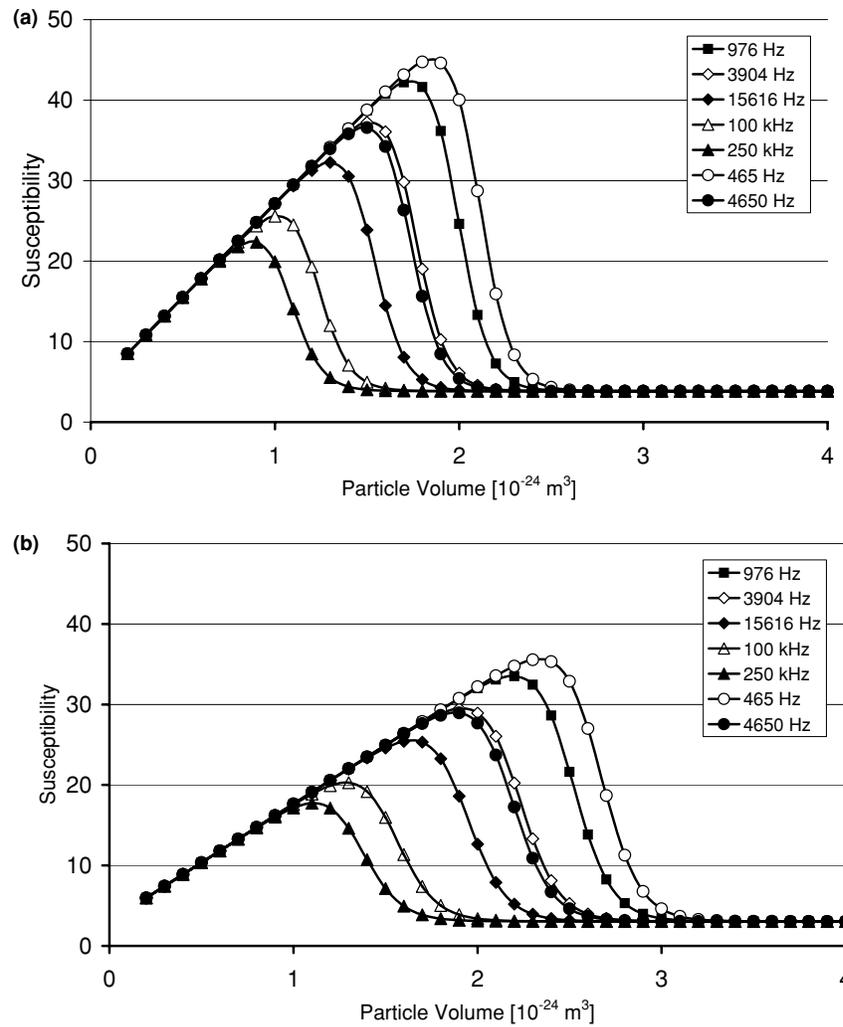


Figure 1. Susceptibility versus particle volume of magnetite (a) and maghemite (b) grains at room temperature (calculated using eq. 10) according to the operating frequencies specified in the legend.

Table 1. Blocking volumes of magnetite and maghemite grains at various operating frequencies at room temperature.

Frequency (Hz)	Magnetite		Maghemite	
	Block volume (10^{-24} m^3)	Diameter (10^{-9} m)	Block volume (10^{-24} m^3)	Diameter (10^{-9} m)
976	2.171	16.09	2.743	17.37
3904	1.942	15.48	2.453	16.73
15 616	1.713	14.85	2.164	16.05
100 000	1.407	13.90	1.177	15.03
250 000	1.255	13.38	1.586	14.47
465	2.292	16.36	2.895	17.68
4650	1.912	14.40	2.415	16.64

from which it follows that the frequency-dependent susceptibility depends on the logarithm of the frequency. Consequently, it appears reasonable to normalize the X_{FD} and X_{FV} parameters as follows:

$$X_{\text{FN}} = X_{\text{FD}} / (\ln f_{\text{mHF}} - \ln f_{\text{mLF}}), \quad (12)$$

$$X_{\text{FS}} = X_{\text{FV}} / (\ln f_{\text{mHF}} - \ln f_{\text{mLF}}). \quad (13)$$

In this case, the frequency dependence is in fact related to the unit difference in logarithms of frequencies. The usefulness of these parameters will be tested by the modelling to be presented later.

THE MODEL CONSTRUCTION

In our modelling, seven operating frequencies are considered, 976, 3904 and 15 616 Hz used in the MFK1-FA Kappabridge, 465 and 4650 Hz used in the Bartington MS-2 Susceptibility Meter and 100 and 250 kHz. In addition, the modelling is made for ferromagnetic fraction consisting only of SP and SSD particles and showing log-normal distribution in grain volumes (as suggested by Eyre 1997). Among magnetic minerals, magnetite and maghemite are considered. The saturation magnetization $M_s = 480 \text{ kA m}^{-1}$ and density $\rho = 5197 \text{ kg m}^{-3}$ is considered for magnetite and $M_s = 380 \text{ kA m}^{-1}$

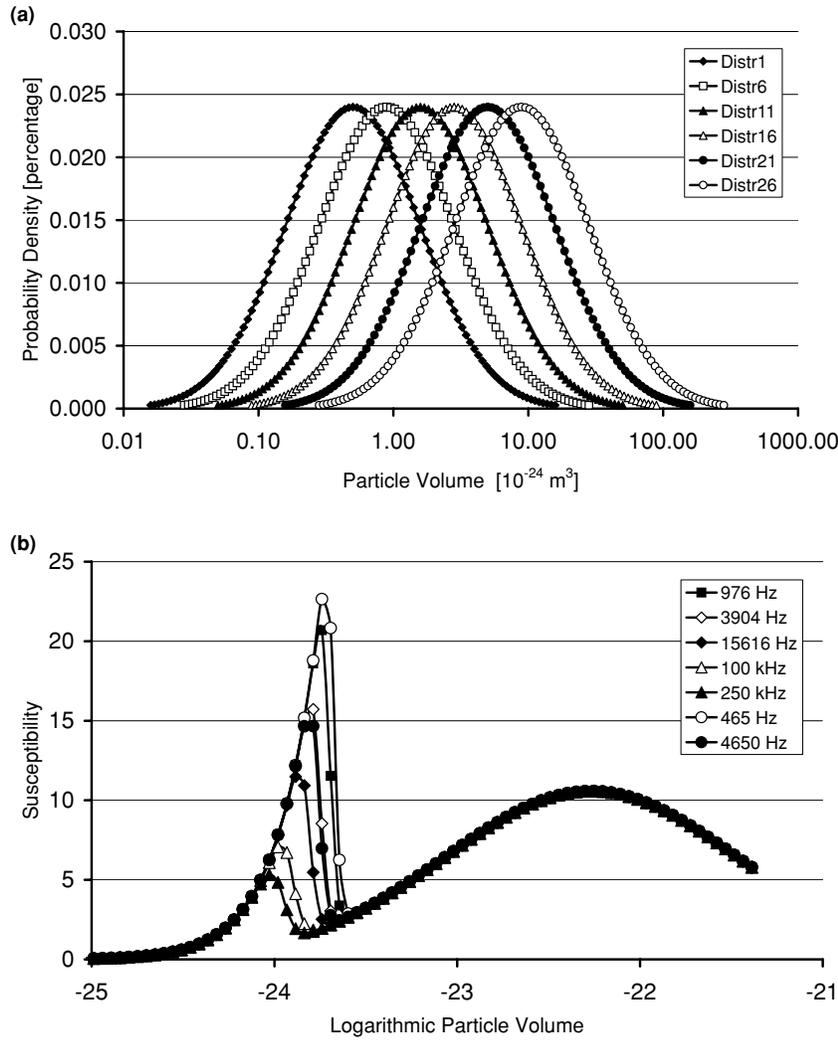


Figure 2. The model log-normal distribution of the volumes of magnetic particles and the frequency-dependent susceptibility distribution. (a) Several log-normal distribution curves of grain volumes and (b) susceptibility distribution of magnetite corresponding to the grain volume distribution curve with $\mu = -23.7$ and $\sigma = 0.8$ variable according to the operating frequency.

and $\rho = 5074 \text{ kg m}^{-3}$ for maghemite (Dunlop & Özdemir 1997). The anisotropy constant is considered $K = 2.5 \times 10^4 \text{ J m}^{-3}$ for magnetite, which corresponds to $M_s = 480 \text{ kA m}^{-1}$ in eq. (5) being similar to the value $K = 2.7 \times 10^4 \text{ J m}^{-3}$ used in the modelling by Dearing *et al.* (1996), and $K = 2.5 \times 10^4 \text{ J m}^{-3}$ for maghemite.

The probability density function of the log-normal distribution is (e.g. Reisenauer 1965)

$$f(V, \mu, \sigma) = \frac{1}{V\sigma\sqrt{2\pi}} \exp\left(-\frac{(\log_{10} V - \mu)^2}{2\sigma^2}\right), \quad (14)$$

where V is the grain volume and μ and σ are the arithmetical mean and standard deviation, respectively, of the logarithms of the grain volume. As the log-normal distribution is defined regardless of the base of the logarithmic function, the common logarithm (with base 10) is used in the models introduced below because of the simple relationship to the grain volumes.

The susceptibility of the whole population of the grains is

$$\chi_{\text{pop}}(\mu, \sigma) = \int_{-\infty}^{+\infty} f(V, \mu, \sigma) V \chi(V) d(\log_{10} V), \quad (15)$$

where $\chi(V)$ in general equals the SSD susceptibility if V is larger than the blocking volume for the frequency under consideration

and it equals the SP susceptibility if V is smaller than the blocking volume; the susceptibility at the SP/SSD boundary follows from eq. (10).

The distribution of susceptibilities is very different compared to the distribution of the grain volumes (Fig. 2). To illustrate the difference, Fig. 2(a) shows several distribution curves of grain volumes, the curves being typically bell-shaped, whereas Fig. 2(b) shows the distribution of the susceptibilities according to the operating frequency for one distribution curve of grain volumes (with $\mu = -23.7$, corresponding volume being $5 \times 10^{-23} \text{ m}^3$, and $\sigma = 0.8$). The resulting susceptibility is given by the overlap of $f(V, \mu, \sigma)\chi_{\text{SD}}$, which is the susceptibility of the blocked particles, and the susceptibility $f(V, \mu, \sigma)\chi_{\text{SP}}$ due to relaxation of the unblocked particles. The log-normal distribution theoretically extends from minus infinity to plus infinity. For practical reasons, however, the grain population volumes were in our modelling considered to span from $\mu - 3\sigma$ to $\mu + 3\sigma$, which encompasses 99.7 per cent of the distribution. The advantage of this simplification is avoiding some physical problems (e.g. grains smaller than mineral lattice cell). The error resulting from using 99.7 per cent of the distribution instead of 100 per cent is evidently negligible with respect to the problems solved by the modelling. The span in grain volumes from

$\mu - 3\sigma$ to $\mu + 3\sigma$ was divided into 100 classes, susceptibility for each class was calculated using the eq. (15) modified for class volume span and the final summation was made through simple adding susceptibilities of individual classes. The arithmetical means (μ) of the logarithms of the grain volumes were considered to vary from -25 (this corresponds to the volume of $0.1 \times 10^{-24} \text{ m}^3$) to -23 (volume of $10 \times 10^{-24} \text{ m}^3$) and the standard deviation (σ) varying from 0.1 to 0.9.

MODELLING RESULTS

Fig. 3 shows variations of the X_{FD} , X_{FV} , X_{FN} , X_{FS} and χ_{LF} parameters with the arithmetical mean (μ) of the logarithms of the grain volumes for a narrow distribution ($\sigma = 0.3$) of magnetite grains; in each plot, each point represents one log-normal distribution curve in particle volumes. From seven frequencies consid-

ered, one can construct too many parameters, which is inconvenient for transparent presentation. For this reason, only those parameters are presented that are based on frequencies of individual instruments.

The curves of all the parameters are mostly bell-like shaped showing low values at very small grains, where they are SP at both frequencies, and at bigger grains that are SSD at both frequencies. In X_{FD} (Fig. 3a) and X_{FV} (Fig. 3b) parameters, the highest values are exhibited at the frequencies 1 and 16 kHz which have the highest difference of the logarithms of frequencies under consideration. They are followed by the frequencies 0.5 and 5 kHz and the lowest values are at the frequencies 100 and 250 kHz, which have on contrary the lowest difference of the logarithms of frequencies. The peak value of the $X_{FD(0.5,5)}$ and $X_{FV(0.5,5)}$ parameters are located rightmost, whereas the peak values of the $X_{FD(100,250)}$ and $X_{FV(100,250)}$ parameters are on contrary located leftmost. In X_{FN} (Fig. 3c) and

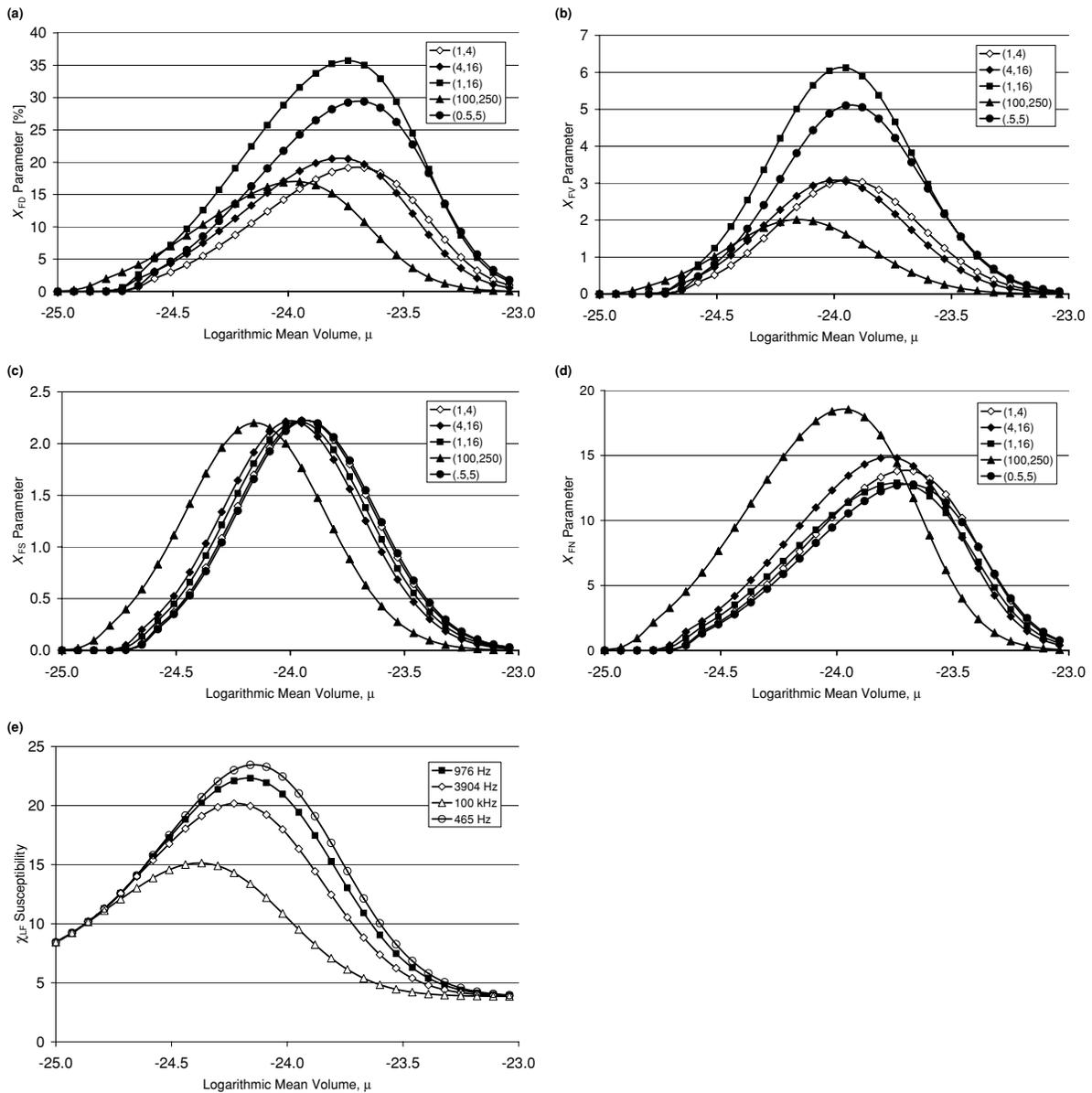


Figure 3. Variation of various parameters characterizing the frequency-dependent susceptibility with logarithmic mean volume (μ) of magnetite grains for narrow log-normal distribution of particle volumes ($\sigma = 0.3$). In legend, frequencies (rounded to kHz) used for parameter calculation are presented in parentheses. (a) X_{FD} parameters, (b) X_{FV} parameters, (c) X_{FS} parameters, (d) X_{FN} parameters and (e) χ_{LF} susceptibility.

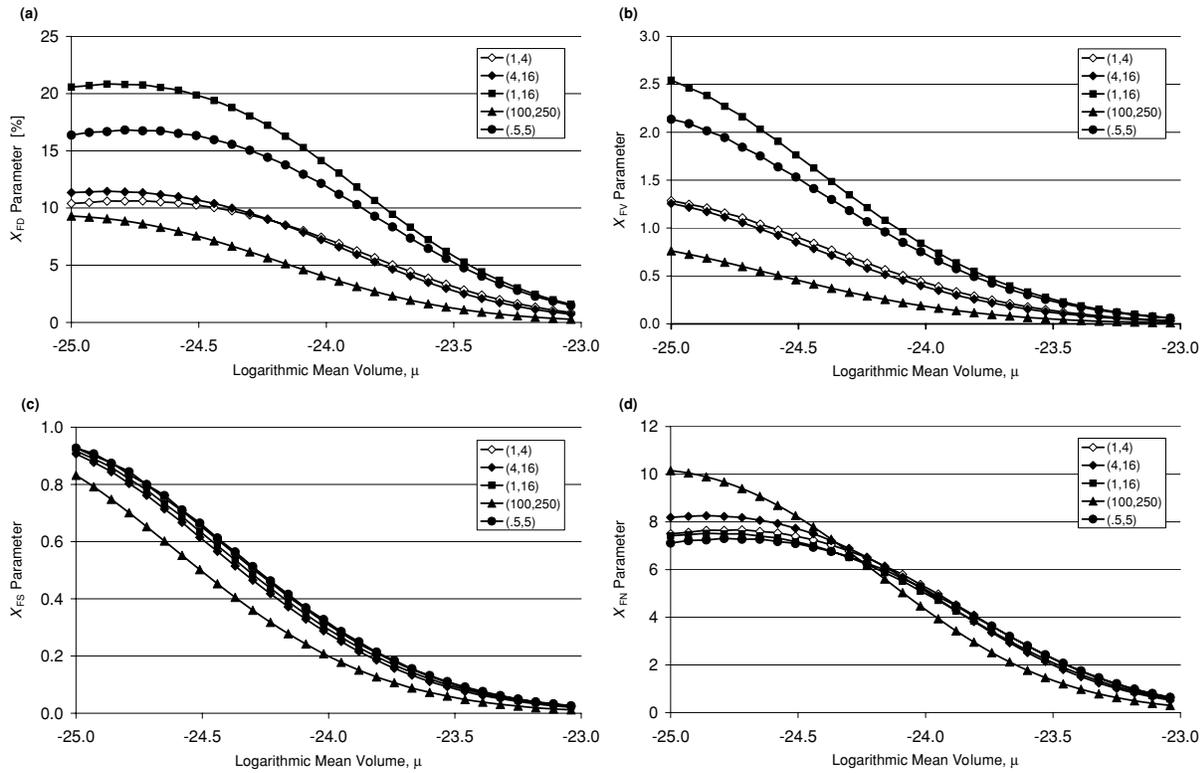


Figure 4. Variation of various parameters characterizing the frequency-dependent susceptibility with logarithmic mean volume (μ) of magnetite grains for wide log-normal distribution of particle volumes ($\sigma = 0.8$). In legend, frequencies (rounded to kHz) used for parameter calculation are presented in parentheses. (a) X_{FD} parameters, (b) X_{FV} parameters, (c) X_{FS} parameters and (d) X_{FN} parameters.

X_{FS} (Fig. 3d) parameters, the curves $X_{FN(1,4)}$, $X_{FN(4,16)}$, $X_{FN(1,16)}$ and $X_{FN(0.5,5)}$ ($X_{FS(1,4)}$, $X_{FS(4,16)}$, $X_{FS(1,16)}$ and $X_{FS(0.5,5)}$) are relatively near one another, whereas the curve $X_{FN(100,250)}$ ($X_{FS(1,4)}$) differ. Consequently, the X_{FN} (X_{FS}) parameters are much more convenient for comparison of measurements made by different instruments than the X_{FD} (X_{FV}) parameters, provided that the frequencies used by different instruments do not differ by several orders in magnitude. It is notable that the peaks in the X_{FD} parameter are located differently than those of the X_{FV} parameter. This can be understood from the distribution of the χ_{LF} susceptibilities (Fig. 3e). The higher susceptibilities at lower grain volumes shift the peaks of the X_{FD} parameter to the right with respect to the of the X_{FV} parameter.

Fig. 4 shows the variation of the X_{FD} , X_{FV} , X_{FN} and X_{FS} parameters with μ for a very wide distribution ($\sigma = 0.8$). The curves have no longer bell-like shape, showing monotonous decrease with increasing grain size. Again, the curves $X_{FN(1,4)}$, $X_{FN(4,16)}$, $X_{FN(1,16)}$ and $X_{FN(0.5,5)}$ ($X_{FS(1,4)}$, $X_{FS(4,16)}$, $X_{FS(1,16)}$ and $X_{FS(0.5,5)}$) are relatively near one another, whereas the curve $X_{FN(100,250)}$ ($X_{FS(100,250)}$) differs.

Fig. 5 shows the variation of the $X_{FD(1,16)}$ parameter with μ for distributions whose width ranges widely (from $\sigma = 0.1$ to 0.8). The curves for relatively narrow distributions are bell-like shaped, with increasing distribution width the curves become more flat, showing monotonous decrease for the wide distribution.

THE EFFECT OF PARAMAGNETIC AND DIAMAGNETIC MINERAL FRACTIONS ON THE WHOLE ROCK (SOIL) X_{FD} PARAMETER

The considerations presented till now concerned the ferromagnetic mineral fraction, consisting only of SP and SSD particles. How-

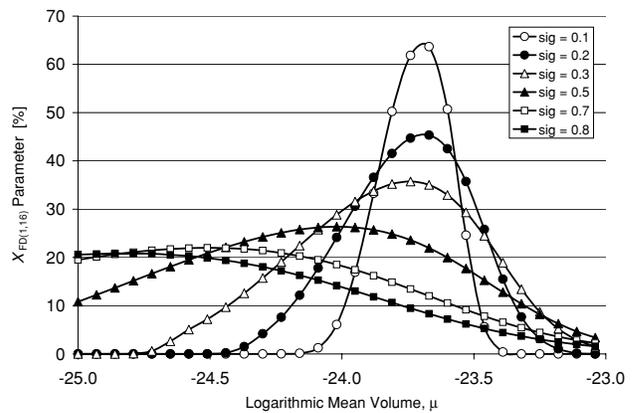


Figure 5. Variation of the X_{FD} parameter with logarithmic mean volume (μ) of magnetite grains for variable widths (σ is denoted as sig in legend) of log-normal distribution of particle volumes.

ever, the rocks, soils and most environmental materials consist also of diamagnetic and paramagnetic minerals and even MD (including PSD) ferromagnetic particles may be present. The whole rock (soil) susceptibility can then be described, with sufficient accuracy, by the following model (Henry 1983; Henry & Daly 1983; Hrouda 2002):

$$X_w = c_d \chi_d + c_p \chi_p + c_f \chi_f, \tag{16}$$

where χ_w is the whole rock (soil) susceptibility, χ_d , χ_p , χ_f are susceptibilities of diamagnetic, paramagnetic and ferromagnetic fractions, respectively, and c_d , c_p , c_f are the respective percentages. The ferromagnetic fraction can be resolved into four subfractions (for illustration, see Fig. 6a). The subfraction 1 is created by the

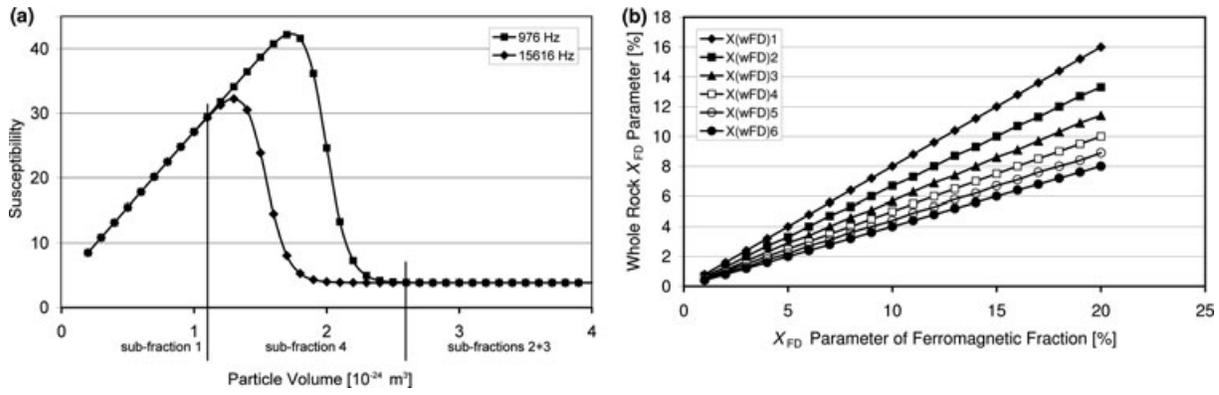


Figure 6. Models of the effect of paramagnetic fraction on the whole rock frequency-dependent susceptibility. (a) Definition of ferromagnetic subfractions (for details see the text) for frequencies 976 and 15 616 Hz. (b) Variation of the X_{wFD} parameter of the model rock (soil) consisting of both ferromagnetic and paramagnetic fractions against the X_{fFD} parameter of the ferromagnetic fraction for several values of the whole rock to the paramagnetic fraction susceptibility. Legend: $X_{\text{(wFD)1}} - \chi_{\text{wLF}}/\chi_{\text{ILF}} = 0.8$, $X_{\text{(wFD)2}} - \chi_{\text{wLF}}/\chi_{\text{ILF}} = 0.7$, $X_{\text{(wFD)3}} - \chi_{\text{wLF}}/\chi_{\text{ILF}} = 0.6$, $X_{\text{(wFD)4}} - \chi_{\text{wLF}}/\chi_{\text{ILF}} = 0.5$, $X_{\text{(wFD)5}} - \chi_{\text{wLF}}/\chi_{\text{ILF}} = 0.4$, $X_{\text{(wFD)6}} - \chi_{\text{wLF}}/\chi_{\text{ILF}} = 0.3$.

grains that are fully unblocked in state at both frequencies (denoted by index *sp*), their susceptibility depends on the grain volume (see eq. 8) and does not depend on the operating frequency. The subfraction 2 is represented by the particles that are fully blocked at both frequencies (index *ssd*), being SSD in state, their susceptibility depends neither on the grain volume (see eq. 7) nor on the operating frequency. The subfraction 3 is due to relatively large MD (PSD) grains (index *md*), the susceptibility depending neither on the grain volume nor on the operating frequency. The subfraction 4 is represented by the grains that are on the transition between fully unblocked and fully blocked state (between SP and SSD, index *mix*). The grains may be SP at low frequency and SSD at high frequency, they may also be SP at both frequencies, but always with $\chi_{\text{LF}} > \chi_{\text{HF}}$, often with $\chi_{\text{LF}} \gg \chi_{\text{HF}}$ (see Figs 1 and 6a).

Then, the whole rock susceptibilities at low and high frequencies are

$$\begin{aligned} X_{\text{WL}} &= c_{\text{d}}\chi_{\text{d}} + c_{\text{p}}\chi_{\text{p}} + c_{\text{sp}}\chi_{\text{sp}} + c_{\text{ssd}}\chi_{\text{ssd}} + c_{\text{md}}\chi_{\text{md}} \\ &\quad + c_{\text{mix}}\chi_{\text{mixLF}} \\ X_{\text{WH}} &= c_{\text{d}}\chi_{\text{d}} + c_{\text{p}}\chi_{\text{p}} + c_{\text{sp}}\chi_{\text{sp}} + c_{\text{ssd}}\chi_{\text{ssd}} + c_{\text{md}}\chi_{\text{md}} \\ &\quad + c_{\text{mix}}\chi_{\text{mixHF}}. \end{aligned} \quad (17)$$

As the susceptibilities of the paramagnetic and diamagnetic mineral fractions as well as of the ferromagnetic mineral subfractions 1–3 are all frequency independent ($\chi_{\text{dLF}} = \chi_{\text{dHF}}$, $\chi_{\text{pLF}} = \chi_{\text{pHF}}$, $\chi_{\text{spLF}} = \chi_{\text{spHF}}$, $\chi_{\text{mdLF}} = \chi_{\text{mdHF}}$), the whole rock (soil) X_{wFV} parameter is

$$X_{\text{wFV}} = c_{\text{mix}}(\chi_{\text{mixLF}} - \chi_{\text{mixHF}}). \quad (18)$$

Consequently, the parameter is primarily controlled by the amount of the particles of the ferromagnetic subfraction 4 (c_{mix}) and by the $\chi_{\text{mixLF}} - \chi_{\text{mixHF}}$ difference, which is constant for a mineral considered. The paramagnetic fraction, diamagnetic fraction and ferromagnetic subfractions 1–3 have no effect on the value of the parameter.

The whole rock (soil) X_{wFD} parameter then is

$$\begin{aligned} X_{\text{wFD}} &= 100c_{\text{mix}}(\chi_{\text{mixLF}} - \chi_{\text{mixHF}})/(c_{\text{d}}\chi_{\text{d}} + c_{\text{p}}\chi_{\text{p}} \\ &\quad + c_{\text{sp}}\chi_{\text{sp}} + c_{\text{mix}}\chi_{\text{mixLF}} + c_{\text{ssd}}\chi_{\text{ssd}} + c_{\text{md}}\chi_{\text{md}}). \end{aligned} \quad (19)$$

Unlike to the X_{wFV} parameter, the whole rock X_{wFD} parameter is controlled not only by the ferromagnetic subfraction 4, but also by all the mineral fractions present in the rock (soil) investigated.

The relationship between the X_{fFD} parameter of the ferromagnetic fraction (X_{fFD} , comprising all four subfractions) and the whole rock X_{wFD} parameter is

$$X_{\text{fFD}} = X_{\text{wFD}}X_{\text{wLF}}/X_{\text{ILF}}, \quad (20)$$

where $\chi_{\text{ILF}} = c_{\text{sp}}\chi_{\text{sp}} + c_{\text{ssd}}\chi_{\text{ssd}} + c_{\text{md}}\chi_{\text{md}} + c_{\text{mix}}\chi_{\text{mixLF}}$.

The susceptibility of diamagnetic minerals is in general very low. For example, in quartz and calcite, the most frequent diamagnetic minerals, the bulk susceptibility is about 15×10^{-5} (SI) and 12×10^{-5} (SI), respectively, (corresponding mass susceptibilities are 5.7×10^{-9} and 5×10^{-9} m³ kg⁻¹), which is very low compared to the susceptibility of common rocks and soils being at least an order of magnitude stronger. Except of limestones, marbles and quartzites, where the diamagnetic fraction constitutes almost 100 per cent of the rock and whose susceptibility can be very low or even negative, the effect of the diamagnetic fraction can be neglected. Then, it is obvious that the $\chi_{\text{wLF}}/\chi_{\text{ILF}}$ ratio in eq. (20) is higher than 1 and the whole rock X_{wFD} parameter is then lower than the X_{fFD} parameter. Fig. 6(b) shows the whole rock (soil) X_{wFD} parameter plotted against the X_{fFD} parameter for several values of the $\chi_{\text{wLF}}/\chi_{\text{ILF}}$ ratio. It is obvious that the increasing presence of the paramagnetic fraction results in decreasing the value of the whole rock (soil) X_{wFD} parameter compared to the X_{fFD} parameter. Consequently, low values of the X_{wFD} parameter do not necessarily indicate low amount of the SP particles, but can also be affected by the paramagnetic and MD ferromagnetic fractions.

The effect of the diamagnetic fraction is weak and can be neglected in the most cases. The contributions of the paramagnetic and ferromagnetic (including MD, PSD, SSD, SP) fractions to the rock (soil) susceptibility can be assessed for example through investigating temperature variation of susceptibility (e.g. Hrouda 1994; Hrouda *et al.* 1997) or hysteresis loops in high fields using vibrating sample magnetometer (e.g. Kelso *et al.* 2002). Provided that the diamagnetic fraction can be neglected and the contribution of the paramagnetic fraction to the rock (soil) susceptibility is known, one is able to calculate the X_{fFD} parameter from the X_{wFD} parameter of the whole rock (soil). However, the X_{fFD} parameter calculated in such a way does not precisely correspond to that of our models considering the SP–SSD range of magnetic particles; the parameter is lowered due to possible presence of very fine particles being SP at both frequencies and due to presence of MD particles.

In some weakly magnetic rocks (soils), the contribution of diamagnetic fraction to the rock (soil) susceptibility can be stronger than that of the paramagnetic fraction and the absolute value of the contribution of diamagnetic fraction can be comparable to the contribution of ferromagnetic fraction to the rock (soil) susceptibility at low frequency. Then, the χ_{wLF}/χ_{fLF} ratio is less than 1 and the whole rock X_{wFD} parameter is higher than this parameter of the ferromagnetic fraction. This should be kept in mind when interpreting the frequency-dependent susceptibility of marly rocks and soils.

THREE-FREQUENCY SYSTEM OF THE MFK1-FA KAPPBRIDGE

The X_{FD} (X_{FV}) parameter, based on susceptibility measurements at two operating frequencies, is the higher the larger is the difference between the logarithms of the operating frequencies. Consequently, it would be advantageous to use very different frequencies, because high X_{FD} (X_{FV}) parameters can be measured relatively precisely. On the other hand, using very different frequencies implies wide span in volumes of magnetic grains and precludes more detail information about grain size distribution. For example, as obvious from Fig. 3(a) and (b), one cannot assign the measured X_{FD} (X_{FV}) value to either left or right branch of the bell-like shape curve or decide whether a particular distribution is narrow or wide. For this reason, it would be desirable to work at more operating frequencies than two, but there are instrumental limits in this respect. Namely, the present author was informed by the designers of the MFK1-FA Kappbridge (Pokorný *et al.* 2006) that there are severe problems in constructing multifrequency high precision instruments based on bridge principle and only limited number of frequencies is practically available. As a compromise, three frequencies were selected in the MFK1-FA Kappbridge, viz. 976, 3904 and 15 616 Hz; the second and third frequencies being four times and 16 times multiples of the basic frequency. Consequently, three X_{FD} or X_{FV} parameters are obtained, viz. $X_{FD(1,4)}$, $X_{FD(4,16)}$, $X_{FD(1,16)}$ or $X_{FV(1,4)}$, $X_{FV(4,16)}$, $X_{FV(1,16)}$. Let us investigate the properties of this system on the above models.

Fig. 7(a) shows the variation of the parameters $X_{FD(1,4)}$ and $X_{FD(4,16)}$ with μ for variable distribution widths of grain volumes ($\sigma = 0.2-0.7$). In narrow distribution ($\sigma = 0.2$), the curves of the parameters $X_{FD(1,4)}$ and $X_{FD(4,16)}$ cross at $\mu = -23.6$. With increasing distribution width the cross point moves towards larger grain size and in wide distribution ($\sigma = 0.7$), the curves of the parameters $X_{FD(1,4)}$ and $X_{FD(4,16)}$ cross at $\mu = -24.0$. If $X_{FD(1,4)} < X_{FD(4,16)}$,

either wide distribution or the left branch of the bell-like curve are indicated.

The relationship between the $X_{FD(1,4)}$ and $X_{FD(4,16)}$ and $X_{FV(1,4)}$ and $X_{FV(4,16)}$ parameters can also be investigated by introducing the following very simple parameter

$$X_R = \frac{\chi_1 - \chi_4}{\chi_4 - \chi_{16}} = \frac{c_{(1,4)}(\chi_{mix1} - \chi_{mix4})}{c_{(4,16)}(\chi_{mix4} - \chi_{mix16})}, \quad (21)$$

where $\chi_1, \chi_4, \chi_{16}$ are the whole rock (soil) susceptibilities at 976, 3904 and 15 616 Hz, respectively, $c_{(1,4)}$ is percentage of the subfraction 4 at 976 Hz and SSD at 3904 Hz, and $c_{(4,16)}$ is percentage of the subfraction 4 at 3904 Hz and SSD at 15 616 Hz. The advantage of the X_R parameter is that it is not affected by any mineral fraction being frequency independent.

Fig. 7(b) shows the variation of the X_R parameter with μ for variable distribution widths of grain volumes ($\sigma = 0.2-0.8$). In relatively narrow distributions ($\sigma = 0.2$ and 0.3) the curves are relatively steep, increasing with increasing μ , whereas in wide distributions ($\sigma = 0.7$ and 0.8) they are flat, but also increasing with increasing μ .

The disadvantage of the X_R parameter is that it is prone to large instability, if the value of the $\chi_1 - \chi_4$ difference or the $\chi_4 - \chi_{16}$ difference is very low, comparable to the error in its determination. The measurement error of the MFK1-FA Kappbridge is better than 0.1 per cent of the measured value for $\chi \geq 1 \times 10^{-5}$ at all three frequencies (Hrouda & Pokorný 2011). It means that the measuring error is about 1×10^{-8} for $\chi = 1 \times 10^{-5}$, about 1×10^{-7} for $\chi = 1 \times 10^{-4}$ and about 1×10^{-6} for $\chi = 1 \times 10^{-3}$. Applying rules for error propagation (e.g. Borradaile 2003), the absolute error in determining $\chi_1 - \chi_4$ (and also $\chi_4 - \chi_{16}$) is about twice the above values. If the value of the $\chi_1 - \chi_4$ ($\chi_4 - \chi_{16}$) difference is near these values, it is better not to calculate the X_R parameter. In this case, it is better to use the $\chi_1 - \chi_4$ versus $\chi_4 - \chi_{16}$ plot, in which the very low values are near the origin.

An example of using the X_R parameter in solving some problems can be presented from the sediments of the Brno Dam located on the Svratka river near the town of Brno and soils in the vicinity of the Vír Dam in West Moravia, also located on the Svratka river, sampled in the drainage area of the Fryšávka river merging into the Svratka river. Fig. 8(a) shows the $X_{FD(1,16)}$ versus κ_1 plot for specimens of both areas; similar spans of the κ_1 mass susceptibility and the $X_{1,16}$ parameter in most specimens indicate similar proportions of magnetic grains. Fig. 8(b) shows the X_R versus $X_{1,16}$ plot. In sediments of the Brno Dam, the values of the X_R parameter are in

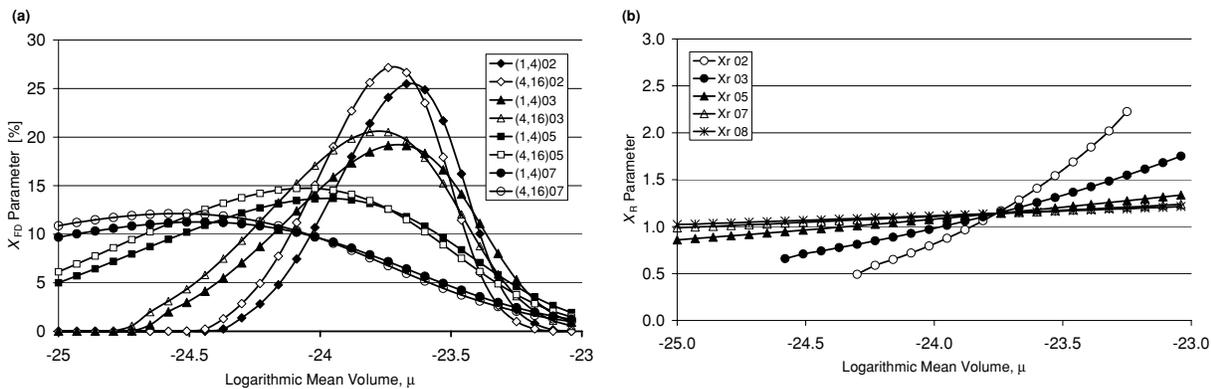


Figure 7. The X_R parameter. (a) Variation of the parameters $X_{FD(1,4)}$ and $X_{FD(4,16)}$ with μ for variable distribution widths of grain volumes ($\sigma = 0.2-0.7$ written in legend behind the parentheses). (b) Variation of the X_R parameter with the width of the log-normal distribution (in legend, numbers behind X_R represent σ).

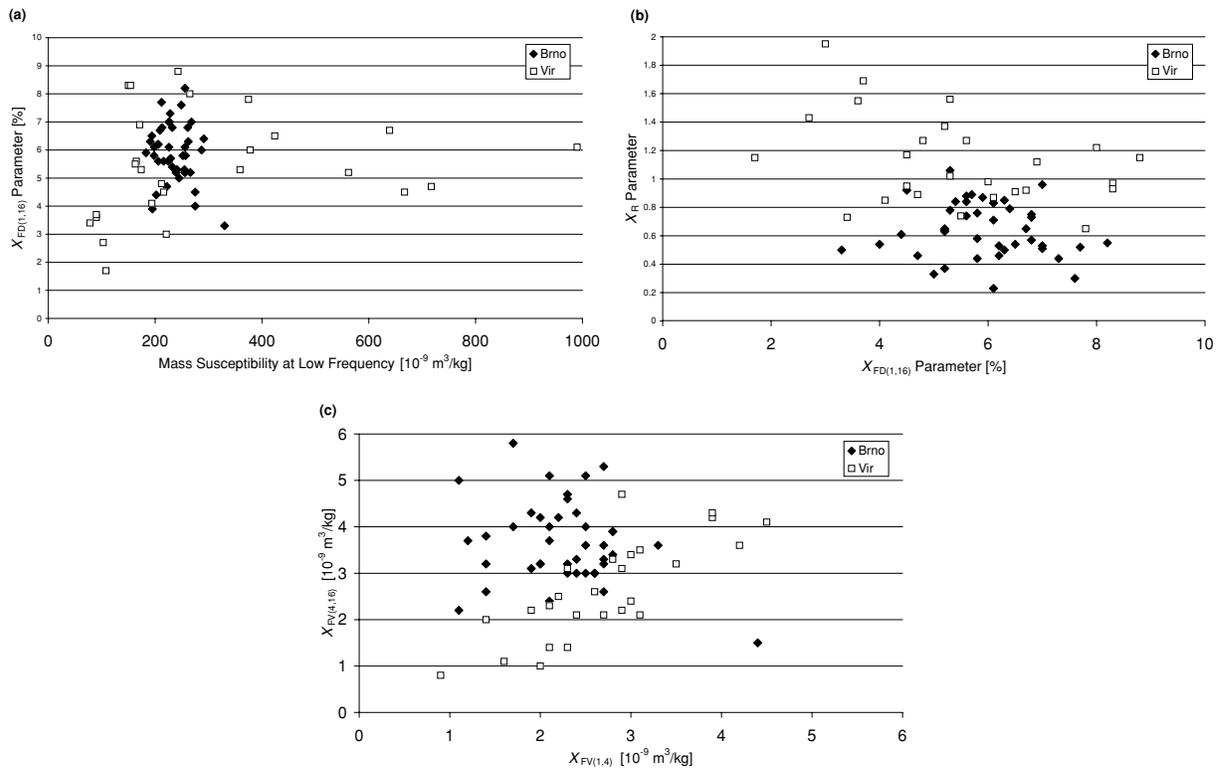


Figure 8. X_R parameter in sediments of the Brno Dam and in soils in the vicinity of the Vir Dam (a) $X_{FD(1,16)}$ versus χ_1 plot, (b) X_R versus $X_{FD(1,16)}$ plot and (c) $X_{FV(4,16)}$ versus $X_{FV(1,4)}$ plot.

almost all cases less than 1 being about 0.6 in average. In soils of the vicinity of the Vir Dam, the X_R parameter is mostly higher than 1. As the $\kappa_1 - \kappa_4$ and $\kappa_4 - \kappa_{16}$ differences are about $5 \times 10^{-9} \text{ m}^3 \text{ kg}^{-1}$, after applying rules for error propagation (e.g. Borradaile 2007) outlined earlier, the error in determining the X_R parameter is about 0.04. This is low enough for interpreting the X_R parameter differences between sediments/soils of both areas as significant. This is confirmed by the $\kappa_1 - \kappa_4$ versus $\kappa_4 - \kappa_{16}$ plot (Fig. 8c), in which plots of both areas are mostly separated clearly. This no doubt indicates differences in grain size distributions of the ferromagnetic subfraction 4 in sediments of both locations. By comparing Fig. 7(c) with Fig. 7(b), one can conclude that the ferromagnetic subfraction 4 is finer in soils in the vicinity of the Vir Dam than in sediments of the Brno Dam. The sedimentological/pedological reasons for this difference must only be searched for.

RELATIONSHIP BETWEEN THE X_{FD} PARAMETERS DETERMINED BY THE MFK1-FA KAPPBRIDGE AND THE BARTINGTON MS-2 SUSCEPTIBILITY METER

The MFK1-FA Kappbridge and the Bartington MS-2 Susceptibility Meter work at different operating frequencies and the X_{FD} (X_{FV}) parameters determined by these two instruments differ, as illustrated by Fig. 3(a) and (b). If one is using one type instrument only, there is no big problem arising from this situation. On the other hand, problems may arise if the results obtained by the Kappbridge and Bartington Susceptibility Meter should be compared. This problem can in principle be solved by using the X_{FN} and X_{FS} instead of X_{FD} and X_{FV} parameters, because the former are almost frequency

independent, being related to the unit difference in logarithms of frequencies.

However, the Bartington instrument uses the operating frequencies differing by decade and has been used by environmental scientists for long time. For this reason, it seems to be advantageous to relate the frequency difference to decade also in the other instruments. Then, the following parameter can be defined as follows:

$$X_{FB} = \frac{\ln 10}{\ln f_{mHF} - \ln f_{mLF}} X_{FD}. \quad (22)$$

In the Bartington instrument $X_{FB} = X_{FD}$, in the other instruments the X_{FB} parameter will differ according to the frequencies used.

Fig. 9 illustrates the results of this approach showing the variations of the $X_{FD(1,4)}$, $X_{FD(4,16)}$, $X_{FD(1,16)}$ and $X_{FD(0.5,5)}$ parameters as well as of the $X_{FB(1,4)}$, $X_{FB(4,16)}$, $X_{FB(1,16)}$ and $X_{FB(0.5,5)}$ parameters with μ for a narrow distribution ($\sigma = 0.3$). It is obvious that the X_{FB} parameters show mutual difference much lower than the X_{FD} parameters. Consequently, it is evident that the X_{FB} parameters are more convenient for comparison purposes than the X_{FD} parameters. Nevertheless, one has to realize that the Kappbridge X_{FB} parameters and the Bartington X_{FB} parameters are interrelated mutually in only approximate way. The reason is that they are derived from different segments of the grain size distribution governed by different blocking volumes following the frequencies used.

IMPLICATIONS FOR MAGNETIC GRANULOMETRY

Our modelling has shown that the X_{FD} parameter is primarily controlled by the amount of the particles on the transition between fully unblocked (SP) to fully blocked (SSD) state being controlled by

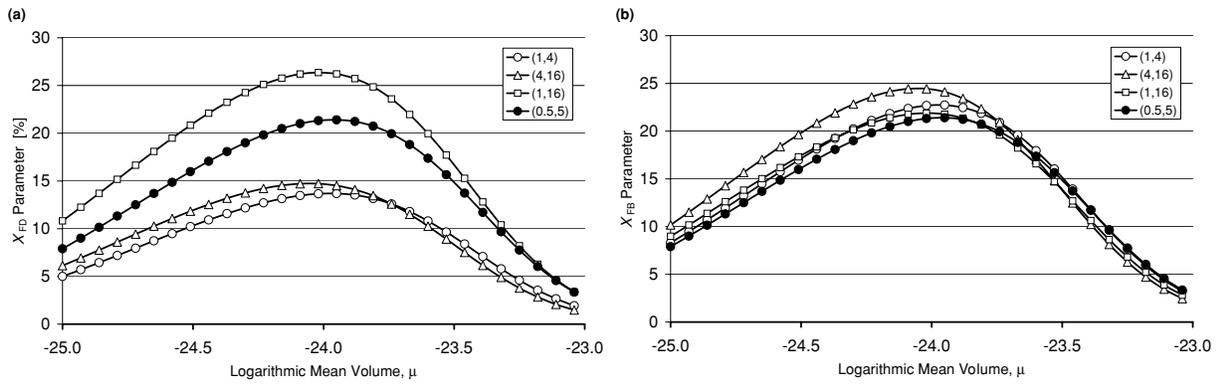


Figure 9. Variation of the MFK1-FA Kappbridge and the MS-2 Bartington X_{FD} parameters with μ for a medium wide distribution ($\sigma = 0.5$). (a) $X_{FD(1,4)}$, $X_{FD(4,16)}$, $X_{FD(1,16)}$, $X_{FD(0.5,5)}$ parameters and (b) $X_{FB(1,4)}$, $X_{FB(4,16)}$, $X_{FB(1,16)}$, $X_{FB(0.5,5)}$ parameters.

the operating frequencies used. Consequently, the X_{FD} parameters obtained at different frequencies on different instruments cannot be processed together or directly compared. Using the X_{FN} parameter instead of the X_{FD} parameter can solve the problem. However, one has to realize that the X_{FN} parameters measured at different frequencies are interrelated in only approximate way, because they are derived from different segments of the grain size distribution governed by different blocking volumes following the frequencies used.

The peak values of the X_{FD} parameter of the models considering only SP and SSD particles are much higher than those measured on rocks and soils. The reasons may be as follows:

1. The paramagnetic, diamagnetic, and even MD (PSD) ferromagnetic mineral fractions, whose susceptibility is frequency independent, may affect the X_{FD} parameter strongly, if they contribute to rock (soil) susceptibility significantly. Except diamagnetic mineral fraction, which can slightly increase the X_{FD} parameter, the other fractions may decrease the X_{FD} values considerably.

2. The SP-SSD fraction is dominated by the SP grains at both frequencies (left-hand branch of the log-normal distribution in grain volumes in narrow distributions) or the fraction is dominated by the SSD grains (right-hand branch of the log-normal distribution in grain volumes in narrow distributions or wide distribution in grain volumes).

The investigation of the frequency-dependent susceptibility at three operating frequencies can help us to decide whether the log-normal distribution in grain volumes is narrow or wide or in case of the narrow distribution, whether the low X_{FD} parameter values are due to the left-hand branch (with dominating very small SP particles) or right-hand branch (with dominating larger SSD particles) of the log-normal distribution.

As the X_{FD} parameter (and also X_{FN} and X_{FB} parameters) can be strongly affected not only by the ferromagnetic particles being on the transition between fully unblocked (SP) to fully blocked (SSD) state, but also by paramagnetic, diamagnetic, and even MD (PSD) ferromagnetic mineral fractions, it is not too convenient for the magnetic granulometry purposes. Using the X_{FV} (X_{FS}) parameter instead of the X_{FD} parameter seems to be much more convenient, because they are not affected by the mineral fractions with frequency-independent susceptibility. The problem is that their values are affected by the amount of the ferromagnetic subfraction 4. Nevertheless, if one is interested in variations of this amount and

the rock (soil) susceptibility is more or less constant, the use of the X_{FV} (X_{FS}) parameter is very convenient for this purpose.

There is a question, often asked by environmental scientists, whether it is possible to determine the concentration of the SP particles in the rock (soil). First of all, one has to realize that the X_{FD} parameter (also X_{FN} , X_{FV} , X_{FS}) is not affected by all SP particles in the specimen, but only by those of the subfraction 4. Then, the concentration of these particles is from the eqs (2) and (18)

$$c_{\text{mix}} = (\chi_{wLF} - \chi_{wHF}) / (\chi_{\text{mixLF}} - \chi_{\text{mixHF}}). \quad (23)$$

The whole rock susceptibilities (χ_{wLF} , χ_{wHF}) are known from measurement, the mineral susceptibilities (χ_{mixLF} , χ_{mixHF}) can be calculated from eq. (10) or roughly estimated from Figs 1 and 6a. It should be noted here that the c_{mix} values are very small in general. For example, for rock (soil) with $\chi_{LF} = 5 \times 10^{-4}$, $X_{FD} = 10$ per cent, and $\chi_{\text{mixLF}} - \chi_{\text{mixHF}} = 30$, $c_{\text{mix}} = 1.67 \times 10^{-6}$ (i.e. 0.000167 per cent). This is because the size intervals defined by the frequencies used are very narrow (see Table 2).

Converting bulk susceptibilities into mass ones ($\kappa = \chi/\rho$, where ρ is density) yields

$$c_{\text{wt-mix}} = \rho_m(\chi_{wLF} - \chi_{wHF}) / \rho_r(\chi_{\text{mixLF}} - \chi_{\text{mixHF}}), \quad (24)$$

where ρ_r is rock density, ρ_m is ferromagnetic mineral density and $c_{\text{wt-mix}}$ is weight percentage of the ferromagnetic subfraction 4.

Our modelling could imply that the grain size of the ferromagnetic subfraction 4 responsible for the frequency-dependent susceptibility can be determined absolutely. The problem is that this determination depends on the mineral constants, like the anisotropy constant and saturation magnetization, used in the modelling and these differ according to the authors and minerals used for their experiments. It is recommended to do this quantitative interpretation with great caution and preferably, to interpret the measured data in terms of relative changes, only.

Table 2. Differences in logarithms of operating frequencies for individual instruments.

Parameter	Log difference	Size interval (10^{-9} m)
$X_{FD(1,4)}$	1.386	16.73–17.37
$X_{FD(4,16)}$	1.386	16.05–16.73
$X_{FD(1,16)}$	2.772	16.05–17.37
$X_{FD(100,250)}$	0.916	14.47–15.03
$X_{FD(0.5,5)}$	2.302	16.64–17.68

CONCLUSIONS

The mathematical models to investigate the frequency-dependent susceptibility in rocks, soils and environmental materials, which were originally developed for two operating frequencies (465 and 4650 Hz possessed by the Bartington Susceptibility Meter), were extended to multiple operating frequencies (976, 3904 and 15 616 Hz possessed by the MFK1-FA Kappabridge and 100 and 250 kHz). The research has led to the following conclusions:

1. The X_{FD} parameter, quantitatively characterizing the frequency-dependent susceptibility, depends not only on the amount of SP particles in the rock (soil), but also on the operating frequencies used for its determination. It is therefore different if obtained by different instruments for the same rock (soil). For comparative purposes, it is recommended to use the X_{FN} or X_{FS} parameter instead of the X_{FD} (X_{FV}) parameter.

2. In the models of frequency-dependent susceptibility, log-normal volume distribution of magnetic particles is considered. The distribution of susceptibilities is very different compared to the distribution of the grain volumes, depending also on operating frequency. The left-hand parts of the susceptibility distribution curves are similar to the curves of χ_{sp} versus V , whereas the right-hand parts of the curves are similar to the log-normal distribution.

3. Even though the blocking volumes, characterizing the SP/SSD threshold, are slightly larger in maghemite than in magnetite, the models of frequency-dependent susceptibility based on log-normal size distribution of magnetic particles virtually do not differ for these two minerals and only magnetite curves are presented.

4. The models are presented as variation curves of the X_{FD} , X_{FN} , X_{FV} , X_{FS} parameters with the logarithmic mean grain volume (μ). The curves have roughly bell-like shape, being very steep in narrow distributions and monotonously decreasing in wide distributions. The peaks of the curves move towards smaller grains with increasing frequency.

5. Paramagnetic fraction tends to decrease the X_{FD} and X_{FN} parameters. Low values of the whole rock X_{FD} (X_{FN}) parameter do not therefore necessarily indicate low amount of SP particles, but can also be indicative of the presence of the paramagnetic fraction.

6. A new parameter X_R is introduced based on susceptibility measurements at three operating frequencies. It is insensitive to dia-, para- and MD ferromagnetic fractions in the rock (soil) and helps us to differentiate between wide and narrow size distributions of SP-SSD particles.

7. The X_{FB} parameter is introduced that originates through normalizing the classical X_{FD} parameter by the difference of natural logarithms of operating frequencies and related to the decade difference between the frequencies. It is convenient for comparison of the Bartington MS-2 Susceptibility Meter data with the MFK1-FA Kappabridge data.

ACKNOWLEDGMENTS

The author thanks an anonymous reviewer for his/her very constructive criticism of the manuscript that was very helpful in improving the paper. Prof. Dr. Josef Ježek is thanked for his help in solving mathematical problems. This work is a part of research projects MSM0021620855 (Ministry of Education and Youth of the Czech

Republic) and P210/10/0249 (Grant Agency of the Czech Republic).

REFERENCES

- Borradaile, G.J., 2003. *Statistics of Earth Science Data: Their Distribution in Time, Space, and Orientation*, 351 pp., Springer-Verlag, Berlin.
- 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.
- Dunlop, D.J. & Özdemir, Ö., 1997. *Rock Magnetism. Fundamentals and Frontiers*, 573 pp, Cambridge University Press, Cambridge.
- Egli, R., 2009. Magnetic susceptibility measurements as a function of temperature and frequency I: inversion theory. *Geophys. J. Int.*, **177**, 395–420.
- Eyre, J.K., 1997. Frequency dependence of magnetic susceptibility for populations of single-domain grains, *Geophys. J. Int.*, **129**, 209–211.
- Heller, F. & Evans, M.E., 2003. *Environmental Magnetism: Principles and Applications of Enviromagnetics*, Academic Press, San Diego, CA.
- Henry, B. 1983. Interprétation quantitative de l'anisotropie de susceptibilité magnétique, *Tectonophysics*, **91**, 165–177.
- Henry, B. & Daly, L., 1983. From qualitative to quantitative magnetic anisotropy analysis: the prospect of finite strain calibration, *Tectonophysics*, **98**, 327–336.
- Hrouda, F., 1994. A technique for the measurement of thermal changes of magnetic susceptibility of weakly magnetic rocks by the CS-2 apparatus and KLY-2 Kappabridge, *Geophys. J. Int.*, **118**, 604–612.
- Hrouda, F., 2002. The use of the anisotropy of magnetic remanence in the resolution of the anisotropy of magnetic susceptibility into its ferromagnetic and paramagnetic components, *Tectonophysics*, **347**, 269–281.
- Hrouda, F. & Pokorný, J., 2011. Extremely high demands for measurement accuracy in precise determination of frequency-dependent magnetic susceptibility of rocks and soils, *Stud. Geophys. Geod.*, **55**, doi:10.1007/s11200-010-0079-6.
- Hrouda, F., Jelínek, V. & Zapletal, K., 1997. Refined technique for susceptibility resolution into ferromagnetic and paramagnetic components based on susceptibility temperature-variation measurement, *Geophys. J. Int.*, **129**, 715–719.
- Kelso, P.R., Tikoff, B., Jackson, M. & Sun, W., 2002. A new method for the separation of paramagnetic and ferromagnetic susceptibility anisotropy using low field and high field methods, *Geophys. J. Int.*, **151**, 345–359.
- Neél, L., 1949. Théorie du trainage magnétique des ferrimagnétiques en grains fins avec applications aux terres cuites, *Ann. Géophys.*, **5**, 99–136.
- Pokorný, J., Suza, P., Pokorný P., Chlupáčová, M. & Hrouda, F., 2006. Widening power of low-field magnetic methods in the investigation of rocks and environmental materials using the Multi-Function Kappabridge Set, *Geophys. Res., Abstracts*, **8**, 04141.
- Reisenauer, R., 1965. *Methods of Mathematical Statistics*, 208pp, State Publishing House of Technical Literature, Praha (In Czech).
- Shcherbakov, V.P. & K. Fabian (2005). On the determination of magnetic grain size distributions of superparamagnetic particle ensembles using the frequency dependence of susceptibility at different temperatures, *Geophys. J. Int.*, **162**, 736–746.
- Stoner, E.C. & Wohlfarth, E.P., 1948. A mechanism of magnetic hysteresis in heterogeneous alloys, *Phil. Trans. R. Soc. Lond., A*, **240**, 599–642.
- Tauxe, L., Mullender, T.A.T. & Pick, T., 1996. Potbellies, wasp-waists, and superparamagnetism in magnetic hysteresis, *J. geophys. Res.*, **101**, 571–583.
- Worm, H.-U., 1998. On the superparamagnetic—stable single domain transition for magnetite, and frequency dependence of susceptibility, *Geophys. J. Int.*, **133**, 201–206.
- Worm, H.-U. & Jackson, M., 1999. The superparamagnetism of Yucca Mountain Tuff, *J. geophys. Res.*, **104**, 25 415–25 425.