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Environmental magnetism: an introduction

M.J. Dekkers
Paleomagnetic laboratory 'Fort Hoofddijk', Faculty of Earth Sciences, Utrecht University, Budapestlaan 17, 3584 CD Utrecht, the Netherlands (e-mail: dekkers@hotspot.geof.ruu.nl; fax: +31 30 253 5030)

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Abstract

In environmental magnetism, the properties of magnetic minerals are used as proxy parameters for many purposes. Examples are paleoclimate analysis, paleoceanographic studies, provenance studies of sediments, studies of anthropogenically-induced pollution, and archeological investigations. Mineral-magnetic techniques are sensitive, require little sample preparation, are rapid, often grain-size indicative, and usually non-destructive. These techniques involve 'bulk' properties which makes them complementary to geochemical micro-analytical techniques. Measurements include the field- and temperature-dependence of various types of induced and remanent magnetizations. Mineral-magnetic methods are continuously being improved. The underlying causal relations between observed mineral-magnetic properties and the processes that led to those properties, are becoming increasingly better understood, and the extended use of such properties as proxy parameters for many processes is foreseen. The following environmental magnetic applications are reviewed: the analysis of paleoclimatic variations in loess and other sediment types, the untangling of sedimentary features in piston cores, and the interpretation of the anthropogenic impact on the environment, in archeological studies and in studies of present-day pollution. The pathway between the provenance area and depositional site is shown to have a crucial impact on the magnetic properties.

Introduction

The discipline of 'environmental magnetism', in which magnetic properties of natural materials are used as proxy parameters for many environmental processes, is based on the same mineral-magnetic principles as paleomagnetism. In addition, the same instrumentation is used. Therefore, the basic principles of paleomagnetism are summarized first, followed by an overview of paleomagnetic methods. Finally, several examples illustrate the environmental magnetic applications.

Natural remanent magnetization

Virtually all rocks contain iron-oxide minerals in trace amounts. These mineral grains can carry a permanent magnetic moment, which is termed the natural remanent magnetization (NRM). Magnetite (Fe_3O_4) is the most important terrestrial magnetic mineral. Ideally, the NRM is locked into the rock during or shortly after its formation. For instance, a lava flow acquires a thermoremanent magnetization (TRM) that is induced by the local geomagnetic field, when its constituent magnetic minerals cool below their respective Curie or magnetic ordering temperatures (T_C). In sediments, detrital magnetic mineral particles are commonly assumed to be the most important source of the NRM. Because freely mobile magnetic particles tend to align along the magnetic field lines, they are deposited with a statistical preference for the field direction, even in small fields like the geomagnetic field. The magnetic minerals are physically immobilized in the sediment at a few centimetres below the sediment-water interface. The resulting remanence is termed post-depositional remanent magnetization (PDRM).

Over geological time, however, rocks do not behave as closed systems and new remanence components may be added to the existing NRM throughout the history of any rock. The original NRM can also be partially or completely erased, for example by dissolution of the magnetic minerals. As a rule, the NRM of a rock is a composite of the primary component, which is imprinted on the rock at or near the time of formation, and any (secondary) components that are subsequently acquired. The primary component can often be isolated from the total NRM by means of stepwise demagnetization, i.e. subjecting a sample to increasingly stronger alternating magnetic fields (alternating field demagnetization) or by applying increasingly higher temperatures (thermal demagnetization) until the NRM is completely removed from the sample. Between each demagnetization step, the residual NRM is measured, and the component that is the most resistant to demagnetization is usually considered to be the primary component. Demagnetization is carried out in a field-free space, i.e. the samples are protected from the ambient geomagnetic field in the laboratory, to prevent the demagnetized grains from acquiring spurious remanence components with no geological meaning. The isolation and recognition of the primary NRM component from a sample collection is central to most paleomagnetic studies. Details of the paleomagnetic method are given by Butler (1992) and more specific information on paleomagnetic and rock-magnetic (or mineral-magnetic) instrumentation can be found in Collinson (1983).

Relaxation time, domain structure and magnetic properties

In order to arrive at a geophysically and geologically meaningful interpretation, the NRM must be: 1) a reliable recorder of the geomagnetic field, and 2) stable over geological time. Remanent magnetism is tied to crystallographic properties and to the shape of magnetic grains; it is thus anisotropic by nature. Thus, to record faithfully the direction of the geomagnetic field and to satisfy criterion 1), the distribution of magnetic grains in the rock should ideally be isotropic. This excludes most metamorphic rocks as well as severely deformed rocks from paleomagnetic observation, although in some deformed rocks the NRM vector can be unstrained (e.g. Cogné & Perroud 1987). Criterion 2) implies that the primary NRM must have a long relaxation time. The NRM in rocks is metastable because any magnetic moment tends to align itself to an ambient magnetic field, because this represents the minimum energy configuration. For example, when the geomagnetic field reverses its direction, the magnetic vector that is recorded by each grain will try to align itself to the new field configuration. The time needed to conform to the new field configuration is related to the height of the energy barrier between the 'old' and 'new' configuration in each individual magnetic particle. If this occurs rapidly (from less than a second to days: laboratory time scale), the relaxation time of a sample is said to be short (NRM = viscous). If it occurs slowly or is negligible with respect to geological time, the relaxation time is said to be long (NRM = stable). After removal of a magnetizing field, a magnetic remanence M_r (expressed per unit volume) decays exponentially with time according to:

$$M_r(t) = M_r(t_0)e^{-t/\tau} \tag{1}$$

where τ is referred to as the relaxation time, the time after which $M_r(t) = M_r(t_0)e$. Provided that the strength of a new magnetic field is low, i.e. if it is comparable to the intensity of the geomagnetic field, this formula also holds as a first approximation when subjecting the sample to a new field. The relaxation time is analogous to the concept of the half-life of decaying radioactive nuclides.

In a rock, there is generally a continuum of relaxation times which extends from below laboratory time scale (minutes to days) to over the age of the Earth (4.56 Ga). Néel (1949) related the relaxation time to magnetic properties of particles as follows:

$$\tau = \frac{1}{C}e^{\frac{\mu_0 v h_c M_s}{2kT}} \tag{2}$$

where C is the frequency factor ($\sim 10^9$ s^{-1}), a constant which depends on how quickly atomic spins can reorient within the magnetic material; μ_0, v, h_c, and M_s are the magnetic permeability, volume, microscopic coercivity, and saturation magnetization per unit volume of the magnetic grain; k is the Boltzmann constant, and T is the absolute temperature. $\mu_0 v h_c M_s$ is referred to as blocking energy or magnetic energy and kT is the thermal energy. The saturation magnetization is the maximum magnetization a grain can acquire, the microscopic coercivity is the field strength which is necessary to overcome the energy barrier between the 'easy directions' in each grain. Easy directions are directions in magnetic particles to which the magnetic vector is constrained, and they are dictated by the crystallographic properties, stress state, and shape of the particles. In magnetite, for example, the easy direction is the [111] direction. Although the microscopic coercivity is not directly measurable, it can be approximated by the macroscopic coercive force or coercive force (B_c), the field strength required to reduce the magnetic moment of a sample (assemblage of grains) to zero after saturation.

Relaxation times and magnetic properties gradually change as a function of domain structure. There are four basic types of such structures. The smallest grains with short relaxation times ($\tau < \sim 150$ s) are termed superparamagnetic (SP). Slightly larger grains are termed single domain (SD). For magnetite the SP/SD threshold size is ~ 0.03 μm. Single domain grains have extremely long relaxation times (a 30% volume increase in small particles increases the relaxation time from a few seconds to longer than the lifetime of the Earth). The SD grain-size range in magnetite is ~ 0.03 to ~ 0.50 μm, depending on the shape of the particles. Still larger grains, which contain a few domains only, are termed pseudo-single-domain (PSD), because their properties resemble those of SD grains; in magnetite the upper threshold of the PSD range is ~ 10 μm. The largest grains ($> \sim 10$ μm) with many domains are referred to as multidomain (MD). The relaxation times of PSD and MD grains decrease with increasing grain size, although at present only approximate trends are known (Equation (2) is valid for SP and SD grains). Even for SD grains, numerical energy-state calculations are mathematically complex and require large computational capacity (e.g. Enkin & Williams 1994).

Environmental magnetism

Since magnetic properties are sensitive to grain-size variations, they are used in environmental magnetism, which was formally defined as a discipline for the first time by Thompson et al. in 1980. The magnetic properties of samples allow the magnetic mineralogy, the concentration of magnetic grains, and the size of these grains to be deduced (Thompson & Oldfield 1986, Verosub & Roberts 1995). Samples are certainly not restricted to hard rocks and unconsolidated sediments: data are also acquired from soils, dusts, organic tissues or peats. This information, in turn, is used as proxy parameters for several environmental purposes, including provenance analysis of sedimentary basins, pollution assessment and paleoclimate studies.

Mineral-magnetic properties and analytical methods

On the atomic scale, magnetism arises from the uncompensated spin moment of the outermost electrons orbiting around a nucleus. Electrons are assigned an orbital moment and a spin moment in quantum mechanics. For the more common magnetic materials the orbital moments of electrons cancel out. In most minerals, the outermost electrons orbit independently of each other yielding a paramagnetic spin configuration. In certain crystalline solids that contain transitional elements in large amounts, the atomic nuclei are sufficiently close to each other to have the orbits of the outermost electrons overlapping with each other. In this situation, the uncompensated spins may line up and their individual magnetic moments can be added to get macroscopic magnetism. This situation can occur in metals, oxides and sulphides. In nature, the transition element of most interest is iron.

Fortunately, the number of magnetic minerals that occur in nature is not large. Most natural magnetic minerals are ferrimagnets or antiferromagnets; only metallic iron is ferromagnetic. In theory, ideal antiferromagnets have no net magnetic moment, but in reality antiferromagnetic minerals have a small net magnetic moment because of (small) imperfections in the crystal lattice. The more common magnetic minerals are listed in Table 1. Magnetites and titanomagnetites are the most frequently occurring terrestrial magnetic minerals. They have the spinel structure. Pure magnetite occurs in igneous and metamorphic rocks but can also be formed by magnetotactic bacteria or indirectly by Fe-reducing bacteria in sediments and water-logged soils. Much mineral-magnetic research has concentrated on the properties of magnetite as a function of Ti content. More elements can substitute for Fe in the spinel structure, for example, Al, Mn and Cr. Aluminum may occur as a minor constituent in spinels in basalts. Manganese or Cr substitution is much rarer. The Mn-endmember jacobsite occurs in carbonatites and the Cr-endmember chromite in a variety of Mg-rich rocks. With the exception of titanomagnetite, the magnetic minerals listed in Table 1 are usually referred to as ‘minor magnetic minerals’. These minerals, however, are being increasingly documented. This applies in particular to the magnetic iron sulphides which can occur in magmatic, metamorphic and sedimentary rocks. Hematite and goethite, with usually some Al-substitution, are common in red beds and in soils. Maghemite is commonly reported in trop-

Table 1. Natural magnetic minerals, Curie (T_C) or Néel temperature (T_N) and magnetic structure below T_C or T_N (in part after Murad 1996). More detailed descriptions, with appropriate references to the literature, of the magnetic properties of most of these phases are given by Hunt et al. (1995a).

Magnetite-ulvöspinel	Fe_3O_4-Fe_2TiO_4	578 °C to –155 °C	ferrimagnetic
Magnetite-hercynite	Fe_3O_4-$FeAl_2O_4$	578 °C to 339 °C	ferrimagnetic
Magnetite-jacobsite	Fe_3O_4-Fe_2MnO_4	578 °C to 350 °C	ferrimagnetic
Magnetite-chromite	Fe_3O_4-Fe_2CrO_4	578 °C to 30 °C	ferrimagnetic
Hematite-ilmenite	α-Fe_2O_3-$FeTiO_3$	675 °C to –170 °C	canted antiferromagnetic–ferrimagnetic(1)
Maghemite	γ-Fe_2O_3	645 °C(2)	ferrimagnetic
Goethite	α-FeOOH	120 °C	antiferromagnetic
Akagenéite	β-FeOOH	26 °C	antiferromagnetic
Bernallite	$Fe(OH)_3$	154 °C	canted(?) antiferromagnetic(3)
Feroxyhite	δ'-FeOOH	177 °C	ferrimagnetic
Pyrrhotite	Fe_7S_8-$Fe_{11}S_{12}$	325 °C	ferrimagnetic–antiferromagnetic(4)
Greigite	Fe_3S_4	350 °C ? (5)	ferrimagnetic
Iron	Fe	770 °C	ferromagnetic
Lepidocrocite	γ-FeOOH	–196 °C	antiferromagnetic(6)
Siderite	$FeCO_3$	–238 °C	antiferromagnetic(6)
Ferrihydrite	$Fe_5HO_8.4H_2O$	–158 °C to –248 °C(6)	speromagnetic(6,7)

(1) Above the Morin transition, hematite has a canted antiferromagnetic structure which results in a macroscopic magnetic moment, its structure becomes ferrimagnetic with increasing Ti-content. As in the titanomagnetite solid-solution series, high Ti-contents yield Curie temperatures below room temperature. (2) Most maghemites invert to hematite before their Curie temperature is reached. (3) The structure of bernallite is not known with certainty. (4) Only monoclinic pyrrhotite (Fe_7S_8) is ferrimagnetic at room temperature, the other pyrrhotite structures become ferrimagnetic on heating above 200 °C. (5) On heating, greigite decomposes before it reaches its Curie temperature. Therefore, the temperature listed should be regarded with great caution. (6) The last three minerals listed are paramagnetic at room temperature but they order magnetically at low temperatures, so their presence can be demonstrated with magnetic methods. Furthermore, on heating a sample above room temperature these paramagnetic minerals chemically alter into magnetic minerals. (7) Speromagnetism is characterized by short-range antiferromagnetic spin coupling. On longer ranges the magnetic ordering is random. The range of ordering temperatures of ferrihydrite is related to its crystallinity.

ical and subtropical soils but rarely in soils in temperate climatic conditions. The reader is referred to Schwertmann (1988) and Fitzpatrick (1988) for an account on how maghemite can form in soils. The occurrence of lepidocrocite, which is occasionally advocated as a precursor of maghemite, seems to be restricted to humid and temperate climatic conditions that lead to reductomorphic pedogenic conditions, which are not in favour of maghemite formation. Dearing et al. (1996a) very recently examined the mechanisms for magnetic enhancement in soils across England which they argued to be dependent on soil type, bed rock, and anthropogenic pollution sources nearby. Metallic iron is extremely rare in nature but has been documented in highly reducing mantle rocks, in serpentinites and in coal beds which were burnt underground. Also, some copper sulphides display magnetic properties but their occurrence is limited to certain low- to medium-temperature mineralizations.

Curie temperatures and hysteresis loops

An important property of magnetic minerals is the Curie or Néel temperature, the temperature above which collective magnetic ordering is overcome by thermal energy and the mineral becomes paramagnetic. Strictly, the collective magnetic ordering temperature for antiferromagnetic materials is termed the Néel temperature, but in practice Curie and Néel temperature are often used interchangeably. Curie temperatures are determined by thermomagnetic analysis, an example of which is shown in Figure 1. On heating, either in air, in vacuum or in an inert atmosphere, chemical alteration of the sample may occur. Repeated thermomagnetic runs serve to distinguish between chemical alteration and magnetic behaviour, which is in principle reversible on cooling. A Curie temperature determination, however, is non-unique: in the case of solid solution, several compositions may yield the same Curie temperature. Therefore, magnetic hysteresis loops (Figure 2) are often determined in conjunc-

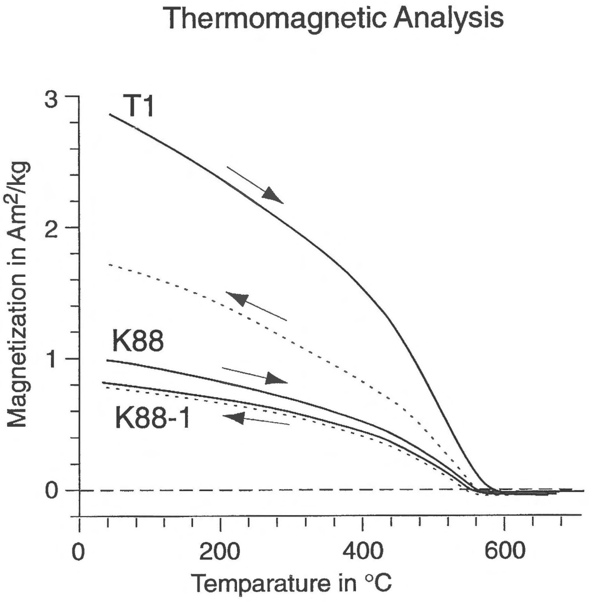

Figure 1. An example of thermomagnetic analysis of a magnetite-bearing fly ash. Heating (cooling) curves are solid (dotted) lines. Sample T1 is distinctly non-reversible: the magnetite oxidizes on heating (in air) which results in a cooling curve that is clearly below the heating curve. Sample K88 is much more reversible, with a second thermomagnetic run (K88-1) being completely reversible.

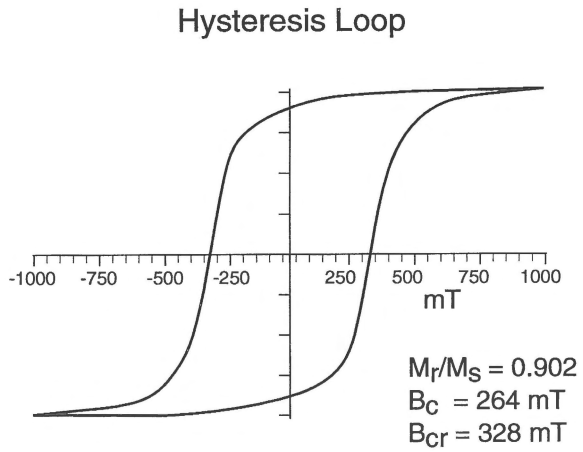

Figure 2. Example of a hysteresis loop for hematite, measured with an alternating gradient magnetometer up to peak fields of 1 T. The squareness of the loop indicates a single-domain structure: M_s = saturation magnetization, M_r = remanent saturation magnetization, B_c = coercive force, B_{cr} = remanent coercive force (see also Figures 3–5 and Appendix 1).

tion with thermomagnetic measurements. A hysteresis loop displays the response of a sample to an applied magnetic field which is measured in that applied field. The loops are indicative of magnetic mineralogy but are also sensitive to subtle changes in grain size which complicates the interpretation of results from natural samples. Classically, the ratios of M_r/M_s and B_{cr}/B_c (for definitions, see Appendix 1) are plotted on a so-called 'Day' plot (Day et al. 1977; Figure 3 is an example). Data usually fall in a narrow band which has been calibrated experimentally with material of known grain size so that grain-size-indicative information can

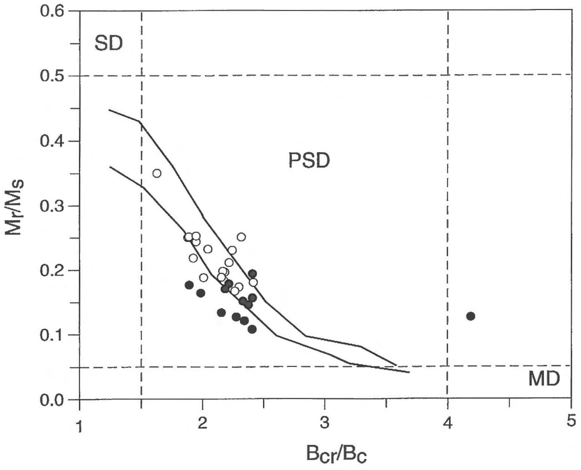

Figure 3. 'Day plot' of M_r/M_s and B_{cr}/B_c ratios. The solid lines represent the experimental calibration of Day et al. (1977), with SD = single domain, PSD = pseudo-single domain, and MD = multidomain. The data points shown are from Icelandic lavas with stable (open circles) and unstable (closed circles) NRM behaviour.

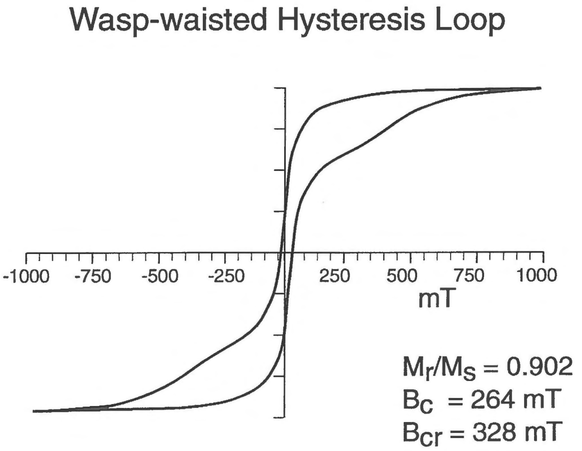

Figure 4. Example of a 'wasp-waisted' hysteresis loop with a thin central part (or 'waist') and much wider outer parts. In this case, the wasp-waistedness is due to the mixture of hematite with a low-coercivity mineral. This example shows the limitations of the 'Day plot' because MD and SP behaviour are difficult to discriminate, and mixtures of magnetic minerals cannot be distinguished. A better understanding of the shape of the hysteresis loop is needed for a more complete interpretation.

be extracted. Roberts et al. (1995) documented empirically that a measured loop is composed of the weighted contribution of each component in the absence of magnetic interactions, i.e. at low concentration ($< 0.1\%$). In many studies, the only parameters used are M_r, M_s, B_{cr} and B_c. It is clear that a wide range of loop shapes is possible and that considerably more information is present in a hysteresis loop than is given by these parameters. Recently, additional methods to retrieve more information from hysteresis loops have been proposed, including unfolding the loop into a pseudo-waveform which can be subjected to Fourier analysis (Jackson et al. 1990), fitting analytical functions to the loops (Von Dobeneck 1996) and decomposing the loops assuming that given fractions of SD and SP material are present in a sample (Tauxe et al. 1996). So-called 'wasp-waisted' loops (Figure 4) have generated much interest (Roberts et al. 1995, Tauxe et al. 1996). Significant magnetic contributions of two (or more) distinct coercivity fractions in a sample generate such loops. Tauxe et al. (1996) modelled loops as consisting of SP and SD contributions and applied the results to submarine basaltic glasses. It is possible to generate all types of hysteresis loops from these two 'endmember distributions', depending on their relative contributions. Von Dobeneck's (1996) procedure cannot be applied to wasp-waisted loops, but 'regularly'-shaped loops can be analysed in detail and subtle differences between sets of loops can be visualised and interpreted.

Low-field or initial susceptibility

Low-field or initial susceptibility (χ_{in}) can be rapidly measured and is frequently used as a proxy for the magnetic mineral concentration. It is often equated to the concentration of magnetite. When the magnetite concentration is $>\sim 1\%$, the paramagnetic contribution of χ_{in} (clay minerals) is negligible compared to the ferrimagnetic contribution of χ_{in}. In sediments and soils, the paramagnetic contribution to χ_{in} is often significant and the ferrimagnetic contribution can only be obtained after subtracting the paramagnetic or high-field susceptibility: the slope of the hysteresis loop measured in a field of approximately 1 Tesla. The low-field susceptibility does not change much with grain size within the SD-MD range; SD grains have only a slightly lower χ_{in} than MD grains. SP grains, however, have a χ_{in} that is remarkably higher (up to 20 times) than that of SD or MD grains. SP grains behave like paramagnetic material because their relaxation times are short. The electron spins are still coupled in an SP grain and the collective spins yield a large magnetic moment when measured in an applied field. This moment is much larger than that which would result from the same amount of isolated electron spins in a paramagnetic configuration (therefore the term 'superparamagnetism' was coined). By increasing the measuring frequency of χ_{in}, the relaxation time(s) of the grains may become longer than the measuring time, and a grain which was SP in a low frequency behaves as SD in a higher frequency, thus distinctly lowering its χ_{in} (Stephenson 1971a, b). Two frequencies (instrumentally dictated) are commonly applied: 470 Hz and 4700 Hz. Dearing et al. (1996b) summarize earlier work and explore the influence of the grain-size distribution within the SP range on the behaviour of χ_{in} at these two frequencies. The maximum susceptibility loss, expressed as a percentage of the measurement at 470 Hz, is calculated at $\sim 16\%$ for magnetite and at $\sim 14\%$ for maghemite. This agrees with experimental data on synthetic and natural samples (e.g. Forster et al. 1994).

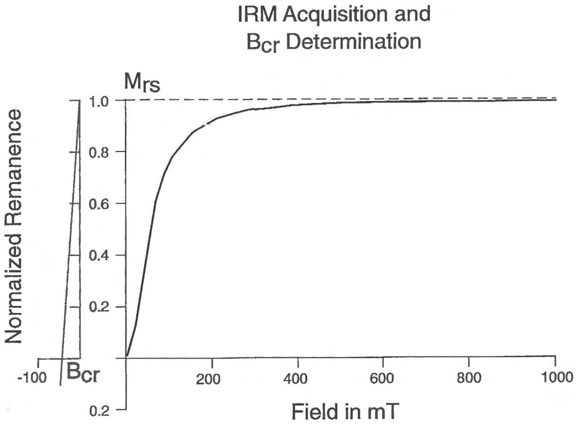

Figure 5. Acquisition curve of an isothermal remanent magnetization (IRM) with B_{cr} determination. After application of a stepwise increasing magnetic field, the remanent magnetization increases until a maximum value is reached, which is termed SIRM, M_{rs} or M_r. After the applied field direction is reversed, the remanence decreases to zero before the remanence direction also reverses. The field value that corresponds to zero remanence (usually determined by interpolation) is termed B_{cr} (remanent coercive force).

Anhysteretic remanent magnetization

A laboratory-induced remanent magnetization which is often used in environmental magnetism studies, is the so-called anhysteretic remanent magnetization (ARM). An ARM is acquired by the action of an asymmetric alternating field applied to a sample (the signal

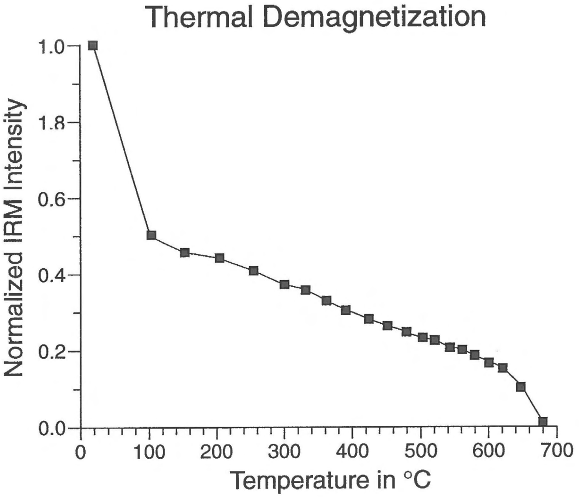

Figure 6. Example of thermal demagnetization of an IRM induced at 2T. Goethite and hematite are difficult to discriminate by their field-dependent properties (both have high coercivity). The magnetically soft part of the IRM was demagnetized by AF treatment at 100 mT before thermal demagnetization. Thermal demagnetization of the remaining IRM separates goethite with a maximum unblocking temperature of ~100 °C from hematite which has a maximum unblocking temperature of ~680 °C.

on analog recording tapes is also an ARM). The alternating field is decreased from a desired peak level to a lower value, usually zero. Usually, standard alternating field demagnetization equipment is used for the induction of ARM. While the alternating field is decreasing, the required asymmetry is obtained by creating simultaneously a direct current (DC) bias field, either by switching off compensation coils or by a dedicated coil generating a small magnetic field comparable to the intensity of the geomagnetic field (~30 to ~60 μT depending on the geographic latitude). For low field values up to ~80 μT, the ARM intensity is linear with the inducing DC bias field. For the low concentrations of magnetic minerals that usually prevail in sediments, ARM is to a first-order approximation linear with concentration. Compared to MD particles, SD particles have high ARM intensities per unit mass which makes ARM an attractive proxy for SD grains. Banerjee et al. (1981) and King et al. (1982) determined a nomogram of ARM versus χ_{in} as a grain-size indicator for granulometric purposes, a procedure which has been used widely since then.

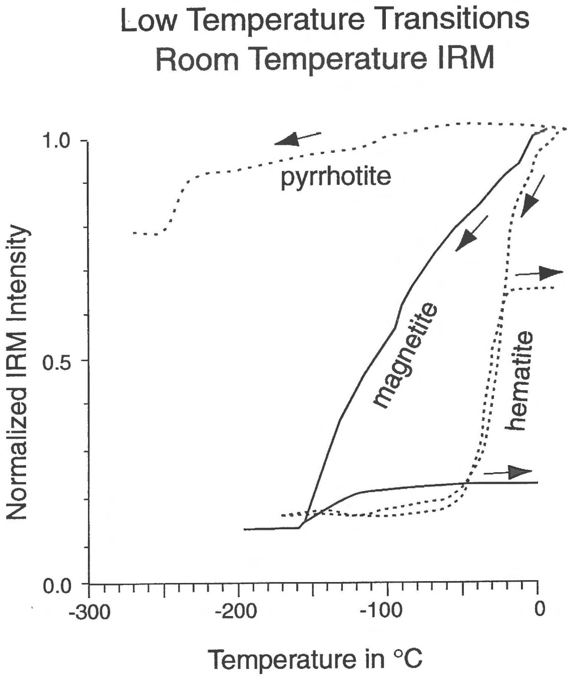

Figure 7. Low-temperature treatment of IRMs induced at room temperature for samples that contain different magnetic minerals. Pyrrhotite (transition at 34 K or –239 °C), magnetite (transition at 118 K or –155 °C) and hematite (transition at 260 K or –13 °C), when sufficiently stoichiometric, can all be identified magnetically in trace amounts (< 0.1%) in bulk samples. Small irregularities in the cooling and warming curves at other temperatures are due to instrumental noise.

Field- and temperature-dependent magnetic properties

Various types of the laboratory-induced remanent magnetizations can also be studied as a function of applied field (Figure 5) or temperature: either above room temperature (thermal demagnetization, Figure 6) or below room temperature ('low-temperature' treatment, Figure 7). Imperfect antiferromagnetic minerals generally have much higher coercivities than ferrimagnetic minerals. It is therefore possible to discriminate between such minerals by measuring the field-dependence at a constant temperature (usually room temperature). The magnetic coercivity of a single mineral also varies with grain size, i.e. with domain structure. Thermal demagnetization provides the distribution of unblocking temperatures for the suite of magnetic minerals present in a sample, although such data may be influenced by chemical alteration during heating (Van Velzen & Zijderveld 1992, 1995). Below room temperature, several magnetic minerals undergo magnetic transitions, changes in magnetic behaviour upon cooling or warming through a specific temperature. The so-called Verwey transition at 118 K is typical of stoichiometric magnetite, whereas hematite and pyrrhotite may be detected by the Morin transition (at 260 K) and the 34-Kelvin transition, respectively. The expression of the magnetic transitions depends on the grain size and chemical composition of the minerals, thus complicating their interpretation. Low-temperature measurements have the advantage that chemical alteration is avoided. More detailed overviews of the magnetic properties of individual minerals are given by O'Reilly (1984), Thompson & Oldfield (1986), Hunt et al. (1995a) and Dunlop & Özdemir (1997).

Origins of magnetic minerals

Natural magnetic minerals

The source of the magnetic minerals in a sample can be either natural or anthropogenically-mediated. First, let us briefly summarize the various natural pathways by which magnetic minerals occur in rocks. In igneous and metamorphic rocks, the magnetic minerals form from a liquid or from precursor mineral phases, respectively. In sediments and soils, the magnetic mineral suite can be a mixture of several components: 1) detrital particles which were eroded and subsequently transported from a source area by eolian, riverine, lacustrine or marine processes, 2) biogenic particles; and 3) authigenic particles that were formed in situ.

Detrital particles

Detrital particles are not simply liberated from a source rock by comminution; they may be physically and/or chemically weathered to some extent before and during transport to the depositional site. Furthermore, some diagenetic alteration usually takes place shortly after deposition as a consequence of the changing geochemical environment. In soils and paleosols, these processes are referred to as pedogenesis and in sediments, post-depositional change is referred to as diagenesis. The amount of cosmogenic detrital particles is insignificant compared to that from other sources.

Biogenic particles

Biomineralization is a widespread phenomenon; for example a major part of the global calciumcarbonate mass is of biogenic origin. Two modes of biomineralization were distinguished by Lowenstam (1981): biologically induced mineralization (BIM) and biologically controlled mineralization (BCM). Iron minerals can be formed by both modes (Frankel & Blakemore 1991). In virtually all sedimentary environments and water-logged soils, bacteria which use iron in their metabolism are known to occur. To the BIM category belong bacteria that use iron as a terminal electron acceptor: magnetite is formed outside the cells (extracellular magnetite) as a consequence of its low solubility product. The crystals are very fine-grained and usually are poorly crystalline. To the BCM category belong bacteria that produce highly crystalline SD magnetite grains (magnetosomes) inside their cells; these are referred to as magnetotactic bacteria. The magnetosomes usually consist of a chain of SD particles. The evolutionary advantage of so-called magnetotaxis (Blakemore 1975) is still debated; with the help of magnetosomes magnetotactic bacteria sense 'up' and 'down' which may enable them to remain in optimal geochemical living conditions. Magnetite magnetosomes are by far most common. Greigite magnetosomes have also been described (Heywood et al. 1990) along with 'magnetosomes' consisting of pyrite (Mann et al. 1990). Bazylinski et al. (1993) reported concurring greigite and magnetite magnetosomes in a bacterium type. The report of pyrrhotite magnetosomes (Farina et al. 1990) is regarded as questionable. The distinctive magnetic properties of these biogenic materials may facilitate discrimination between types of magnetic particles (e.g. Moskowitz et al. 1993, Oldfield 1994). From a quantitative viewpoint, magnetite of extracellular origin is much more important than magnetite from magnetosomes because Fe-reducing bacteria produce extracellular magnetite in vast amounts (grams per day at least in cultured batches). The extracellular magnetite, however, is less resistant to changing chemical conditions and is consequently less important for the paleomagnetic record. In some sediments and in water-logged soils, however, the fossil magnetosomes can be an important fraction of the magnetic minerals.

Authigenic particles

The distinction between authigenesis and biogenesis is somewhat arbitrary. Authigenesis refers to inorganic chemical, rather than biological processes. However, usually at least one of the reactants in an 'inorganic' chemical reaction process is bacterially produced or mediated, in particular when dealing with BIM processes. Bacteria may even be able to reduce iron in smectite (Kostka et al. 1996). The sulphide in the pyrite formation process which usually has a profound impact on the assemblage of magnetic minerals in sediments, is produced by sulphate-reducing bacteria. The sulphate reacts with available ferrous iron to form intermediate monosulphides which usually react completely to form pyrite (e.g. Goldhaber & Kaplan 1974). Greigite and pyrrhotite have been reported as intermediate products in these reactions. They are magnetic and especially greigite is increasingly documented in a wide variety of sediments and soils (e.g. Roberts 1995). The reaction pathway which leads to the formation of pyrrhotite, is not yet clearly established.

Anthropogenic magnetic minerals

Since the industrial revolution in the 19^{th} century, the amount of anthropogenically-produced magnetic particles that have been brought into the environment has increased considerably. Magnetite-containing spherules are common constituents of fly ash and bottom ash which result from combustion of coal and, to a lesser extent, from the combustion of oil (Flanders 1994). Pyritic iron is thought to be the main source of the iron. On heating, the pyrite dissociates to form elemental sulfur and pyrrhotite, which, in turn, dissociates to form sulfur and iron. The iron spherules are then oxidized to magnetite. Metallic iron can be a constituent of blast furnace slags. The magnetic properties of industrial dusts differ from those of natural origin (Oldfield et al. 1985). Polluted samples from the North Sea and the Bay of Biscay have maximum SIRM values and highest SIRM/ARM ratios, approaching those for industrial fly ash. Source areas (Sahara versus South America) for unpolluted dusts from Barbados could also be inferred from mineral-magnetic parameters. Using magnetic methods, Hunt et al. (1984) were able to discriminate between fly ash from power stations and particulate emissions of vehicles. Anthropogenically produced material generally has high SIRM/ARM ratios and comparatively low ARM values (Hunt 1986).

Advantages of mineral-magnetic methods

All rocks and soils contain iron oxides and/or iron sulphides in trace amounts or more. The mineral-magnetic methods can therefore be applied to all of them. In addition, these methods have several analytical advantages over more conventional techniques. The first asset is a simple sample preparation, which consists only of weighing the sample. Most measurements can be performed on powdered material or rock chips. Wet samples can be dried, although magnetic iron sulphides reportedly alter as a consequence of drying in air. Only occasionally does the orientation of powders need to be maintained during measurement. This can be achieved, for example, by fixation in a known amount of epoxy resin. A second advantage lies in the rapidity of data acquisition. The majority of the measurements is rapid. For example, the measurement of the low-field susceptibility takes only a few seconds and that of remanent magnetizations less than one minute. On a state-of-the-art instrument, a complete hysteresis loop can be acquired in about two minutes. Thirdly, most of the instrumentation is comparatively modestly priced.

Furthermore, magnetic measurements are sensitive, especially for strongly magnetic material: 1 ppm of magnetite is easily detected. In addition, magnetic parameters can indicate grain size, particularly in the SP-SD grain-size range. Such ultrafine grains are difficult to identify with other techniques. One can thus incorporate grain-size and concentration-dependent trends in a suite of samples. Another advantage is that the magnetic measurements at room and lower temperatures are non-destructive. Hence, the same material is available for further analysis with any other technique, if desired. Finally, the measurements are done on bulk material, which implies that the data acquired are representative for the samples studied. Magnetic techniques are therefore complementary to geochemical micro-analytical techniques where usually only a fraction of the sample can be analysed.

Several drawbacks should be mentioned. First, the interpretation of a set of complete rock or soil samples is based on the interpretation of the behaviour of one or more trace constituents. This applies, however, for most 'proxy parameters'. Furthermore, the set of magnetic proxy parameters can be blurred by mixed mineral-magnetic assemblages, and experience is often required to enable proper interpretation. Finally, the non-uniqueness of some of the parameters can be a problem. To reduce this problem, a considerable amount of environmental magnetic research is currently devoted to improving interpretational aspects by including other (paleo)environmental and (paleo)climatic indicators and by comparison with other mineralogical and geochemical methods.

The advantages of the environmental magnetic techniques are considerable. Firstly, they save time and analytical costs. Secondly, they furnish grain-size and concentration-dependent information. Thirdly, they can be used to select the most appropriate samples for other complementary analytical techniques. Such techniques in turn are often needed to arrive at a firmer environmental interpretation. Some recent multidisciplinary studies will be reviewed in the next section.

Some examples

Paleoclimate and paleoenvironment studies on loess

Magnetic proxies have been used and are increasingly being used as paleoclimate indicators. Observations on the Chinese Loess Plateau were among the first to point to this potential of mineral-magnetic techniques. The necessary brevity implies that only a selection of the vast amount of literature information can be presented here.

The observation by Heller & Liu (1984) that χ_{in} records measured in loess-paleosol sequences from the Chinese Loess Plateau, which were dated by magnetostratigraphy, closely match the δ^{18}O record of marine sediments, generated a substantial number of studies on the relationship between mineral magnetic proxies and climate. The paleosols formed under relatively warm and humid climate conditions in interglacial and interstadial periods; they have a relatively high χ_{in}. The loess, which has low χ_{in} values, was deposited under cold and fairly dry, glacial conditions. Later research, recently reviewed by Heller & Evans (1995) and by Reynolds & King (1995), showed that at least part of the magnetic signal is caused by magnetite or maghemite particles which are formed in situ by pedogenic processes (e.g. Evans & Heller 1994). The interpretation of the records is complicated by these processes. It is difficult to extract unambiguous information concerning climate. Many workers invoke the presence of SP grains which have been documented by low-temperature techniques or by frequency dependence of χ_{in} (Maher & Thompson 1992, Banerjee et al. 1993, Heller et al. 1993, Verosub et al. 1993, Evans & Heller 1994, Forster et al. 1994, Forster & Heller 1997). The presence of coarse-grained magnetite, which is argued to be of detrital origin, has been demonstrated by direct microscopic observation on magnetic extracts, the presence of the Verwey transition at ~118 K, or by the application of the citrate-bicarbonate-dithionite (CBD) treatment in conjunction with rock-magnetic measurements (Singer et al. 1992, Verosub et al. 1993, Fine et al. 1995, Hunt et al. 1995b, Sun et al. 1995). Forster & Heller (1997) demonstrate that variations in the high-field susceptibility can be attributed to variations in clay minerals. They show that the magnetic enhancement in loess and particularly in paleosols is caused by increased quantities of ferrimagnetic minerals. Further, they suggest different enhancement paths for loesses and paleosols from Hungary, China and Tajikistan which makes the link between mineral magnetic properties and paleoclimate less straightforward.

Although the CBD treatment gives important information on the ultrafine-grained magnetic minerals, various studies do not yield compatible results. Hunt et al. (1995b) showed for synthetic magnetite and maghemite that the commonly applied CBD treatment dissolves fine magnetite and maghemite (< 1 μm) but leaves coarse magnetite (> 1 μm) essentially unaffected. Using less dithionite would enable discrimination between fine-grained magnetite and fine-grained maghemite. Singer et al. (1995), however, showed that the routine CBD treatment of soil samples discriminates between fine-grained pedogenic maghemite and coarse-grained lithogenic magnetite. The discrimination of fine-grained magnetite was equivocal in their study. It may well be that each type of sample or even soil variety requires a taylor-made CBD recipe for a proper discrimination of (maghemitized) magnetite and maghemite in the ultrafine grain-size range. One could vary for example temperature, amount of dithionite, reaction time, and pH.

Mineral-magnetic methods have also been used to infer paleoprecipitation rates. Maher et al. (1994) observed that the present-day rainfall correlates with the ferrimagnetic iron-oxide content in modern soil types of the Chinese Loess Plateau. The lowest ferrimagnetic concentrations (~0.01%) occur under dry conditions with an annual precipitation of ~300 Mm. The iron-oxide concentration gradually increases to ~0.2% for an annual precipitation of ~750 Mm. By applying this observation to ancient loess-paleosol sequences, Maher et al. (1994) inferred paleorainfall conditions from magnetic susceptibility measurements. Based on these observations, they argue that variations in rainfall on the Chinese Loess Plateau would be up to four times larger than those inferred from atmospheric circulation models. This could be caused by spatial inaccuracies in those models which predict a similar variability in rainfall in the area to the northwest of the Chinese Loess Plateau (B.A. Maher, written communication).

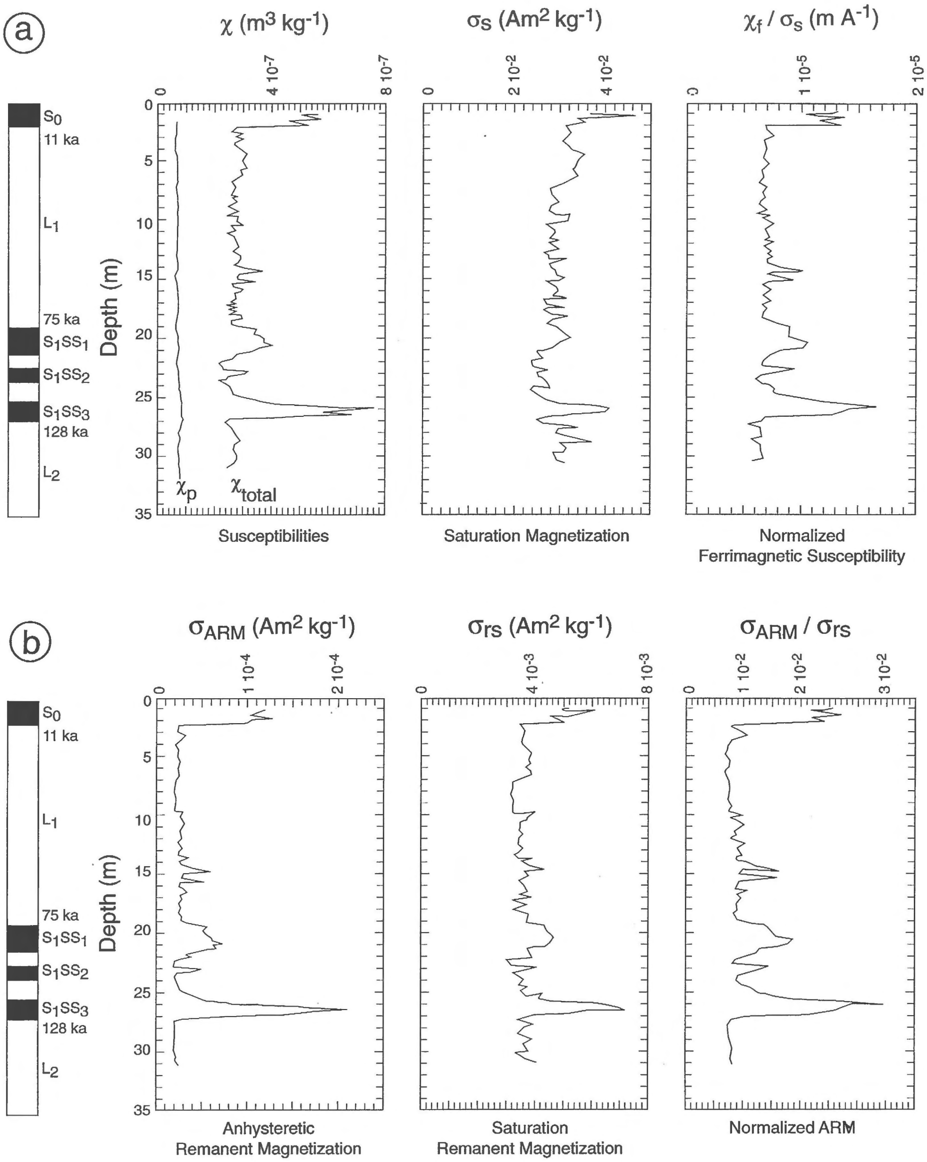

Figure 8. Magnetic properties from the last 140 kyr of a loess-paleosol sequence at Xining, western Loess Plateau, China (simplified after Hunt et al. 1995c). a) Outermost left: Lithological column with ages for the paleosols (black). S = soil; L = loess; numbering starts from the Holocene soil S_0. In the S_1 complex three sub-units are distinguished: SS_1, SS_2 and SS_3. Left-hand panel: Susceptibility. χ_p, the susceptibility of paramagnetic minerals (measured in a high magnetic field), appears to be relatively constant in this profile. χ_{total} refers to the low-field susceptibility. The ferrimagnetic contribution to the low-field susceptibility χ_f equals $\chi_{total}-\chi_p$. Central panel: Saturation magnetization which is used as a normalizer to remove concentration effects. Right-hand panel: The ratio χ_f/σ_s, indicative of SP grains, increases in the paleosols. It is also slightly increased in a zone in L_1, which probably indicates a warmer period in the glacial period in which L_1 was deposited. Note that no soil is visually evident in this zone. b) Outermost left: see a) above. Left-hand panel: ARM intensity expressed on a mass-specific basis (σ_{ARM}). ARM values are higher in soils than in loess. Central panel: SIRM intensity expressed on a mass-specific basis (σ_{rs}). Note that the pattern is noisier than that of the ARM and that is differs from that of σ_s (see a) above). Right-hand panel: Higher ratios of σ_{ARM}/σ_r indicate greater abundances of SD and PSD particles in the soils. Not only the SP fraction, but also the SD and the PSD fractions increase in soils. Subtle paleoclimatic variations can also be traced within and between lithological units, e.g. paleosol S_1 SS_3 is much more developed than S_1 SS_1 and, in particular, than S_1 SS_2. The climate change which led to the formation of the present-day soil, S_0, was apparently sharp because no intermediate values (such as for S_1 SS_3) are present.

The Chinese soils are considered to mature rapidly, in several hundreds to thousands of years, because they contain material that is easily weathered. In contrast, Singer et al. (1992) argued that for some Californian soils, maturation takes up to a million years. Maher et al. (1994) assume that the modern soils have matured to steady-state conditions similar to those of the paleosols; this is the most critical assumption according to Maher (written communication). A further assumption regarding the relationship between ferrimagnetic content and rainfall is that the parent material for each location did not vary with time. Maher et al. (1994) argue that errors in these (debatable) assumptions will have only a limited influence on the paleo-precipitation calculations. The mineral-magnetic characterization of the particles, however, needs further substantiation to better discriminate between lithogenic and pedogenic contributions to the susceptibility, although the major contributor is regarded to be pedogenic (Maher, written communication). A nice example of an extended characterization of the magnetic mineralogy is the study of Hunt et al. (1995c).

Hunt et al. (1995c; see Figures 8a, b) showed that measurement of χ_{in} alone does not allow to distinguish between the effects of concentration, composition and grain-size. By measuring χ_{hf} they corrected for the paramagnetic (clay) contribution. Subsequent normalization of the ferromagnetic component of χ_{in} by the saturation magnetization yields information regarding the amount of SP material, with high values indicating more of this material. This reasoning has been supported by low-temperature measurements (Hunt et al. 1995c). Normalization of the ARM by the saturation isothermal remanent magnetization (SIRM) allows characterization of variations in the SD and PSD grain-size range, because ARM is more 'efficient' in those grains compared to SIRM. They show that comparison of available uncorrected χ_{in} profiles of nearby sites in the Chinese Loess Plateau would imply internally inconsistent paleoclimate information, but that this inconsistency is removed after correction for concentration and grain-size effects.

Paleoclimate and paleoenvironment studies on other sediments

Marine sediments

Apart from the large number of loess-paleosol studies, mineral-magnetic methods have found their way in investigations of other sediments as well. In the 1970s several authors, e.g. Wollin et al. (1978), postulated a linkage between geomagnetic field properties and climate. Kent (1982) and others showed that the postulated linkage was an artifact caused by the magnetic properties of the (marine) sediments because the latter are largely controlled by climate. Robinson (1986) compared various magnetic parameters (NRM, χ_{in}, ARM, IRM and some of their ratios) to other climatic indicators (carbonate content, clay mineral composition, δ^{18}O) in a set of gravity cores from North Atlantic Pleistocene deep-sea sediments, NW of the Azores. He showed that climatic variation could simply be monitored by either χ_{in}, ARM or IRM values which are a function of the climatically controlled carbonate content. In addition, normalization, argued to provide concentration-independent mineral-magnetic parameters, still showed some climate-dependent information. The magnetic variability was explained by different source areas: during glacial periods ice-rafted debris and low carbonate contents prevailed whereas high carbonate contents and eolian input were dominant during interglacial periods. A constant (?) 'background level' was speculated to be biogenic magnetite.

Intermittent sedimentation because of bottom-current erosion or non-deposition in open ocean basins can also be inferred from mineral-magnetic properties. For piston cores from the Bermuda Rise in the North Atlantic and from the Indian Ocean Basin south of Australia, DeMenocal et al. (1988) showed that intermittent sedimentation can be traced by reduced χ_{ARM}/χ_{in} values and a more distinct magnetic fabric compared to adjacent levels in the cores. They used the properties of known short-duration hiatuses as input for a multivariate discriminant analysis model (three variables only) and could identify more zones where sedimentary hiatuses are likely, closely coinciding with the glacial maxima based on δ^{18}O stratigraphy. The authors emphasize that their method should not be applied to areas with variable terrigenous input which could easily mask the subtle signature of the hiatuses.

Another study which interpreted mineral-magnetic variation mainly in terms of variations of eolian input is that of Doh et al. (1988) on a 'giant' piston core from the central North Pacific Ocean, spanning an age interval from Recent to Late Cretaceous. Interestingly, this core showed two paleomagnetically stable segments around a non-stable interval. The youngest stable segment is 0 to 2.56 Ma old and the oldest is older than 55–65 Ma. The latter age is imprecise because microfossils are absent in the sediment. Doh et al. (1988) showed that in the paleomagnetically stable segments, variations in mineral-magnetic parameters reflecting the magnetic hardness, interpreted in terms of a varying contribution of goethite and magnetite, closely correspond to variations in the eolian grain size. In the non-stable interval this relation was absent. This was attributed to either enhanced authigenesis or diagenesis. Intriguingly, no systematic change in the magnetic hysteresis parameters plotted on a Day plot was noted, although B_c and B_{cr} are reported to be lower in the non-stable interval which could indicate the presence of more SP grains.

Later studies (e.g. Canfield & Berner 1987, Karlin 1990a, 1990b, Leslie et al. 1990) showed that as a rule diagenesis affects the supposedly detrital magnetic minerals. The extent of the diagenesis depends on the geochemical environment. More recent studies compared the mineral magnetic properties of Neogene and Pleistocene deep-sea sediments from several ocean basins. Bloemendal et al. (1992) were able to relate variations in selected mineral-magnetic parameters to factors like: 1) sediment lithology, 2) provenance area and pathway of terrigenous input, and 3) diagenetic, authigenic and/or biogenic modifications of the original detrital input. They showed that plots of magnetic parameters are useful in discriminating these processes. Three groups of distributions were distinguished. The first group consisted of 'tightly grouped distributions' where a single main source of magnetic material originates either from bottom currents (intermediate χ_{in} and χ_{ARM} values) or from eolian dust (high χ_{ARM}/χ_{in} and low S-ratio values). The second group has 'broad continuous distributions' where a two-source or a two-pathway magnetic mineral system is assumed to originate from differences between glacial and interglacial periods. The third group has 'bimodal distributions' interpreted to be caused by reductive diagenesis. This latter group is prominent in near-shore and hemi-pelagic sediments.

In a follow-up study on ODP cores from the Owen Ridge (Arabian Sea) Bloemendal et al. (1993) integrated magnetic properties and geochemical data. They documented two diagenetic fronts, one occurring at 1.2–1.5 M and the other (diachronically in different cores) at ~7.5 M below the sediment-water interface. The magnetic signal was dominated by the monsoon-driven eolian influx, although the amplitude of the variations decreases down-core and is low at depths > 12 M below the sediment-water interface. Bloemendal et al. (1993) explained the varying magnetic properties in terms of progressive diagenesis: preferential dissolution of fine grains leading to a coarsening of the grain size, followed by (incongruent) dissolution of these coarse grains at greater depths. Some aspects of the diagenetic processes remain unresolved, however: e.g. the apparent abruptness of the lowermost diagenetic front and the appearance of framboidal pyrite at a much larger depth of ~80 M below the sediment-water interface, seemingly without any influence on the magnetic mineralogy. Most interestingly, Bloemendal et al. (1993) noted that polarity intervals shorter than ~500 kyr are not recorded magnetostratigraphically. They put forward several rather speculative mechanisms, none of which they favour specifically. The mechanisms considered are: 1) reduction in grain size of the magnetic carriers accompanied by a change in domain structure resulting in magnetic resetting, 2) dissolution of magnetite and precipitation of magnetic sulphides (they note that pyrite growth occurs only much deeper in the cores) and 3) the NRM was already carried entirely by the residual magnetic fraction and survived the passage of the diagenetic fronts. Bloemendal et al. (1993) do not mention, however, the mechanism of delayed NRM acquisition (Van Hoof & Langereis 1991). Another example of magnetic properties in fairly rapidly deposited marine sediments is that of Urbat (1996) who analysed the impact of diagenesis in Pleistocene ODP cores (Holes 903A and 904A) off New Jersey with sedimentation rates of several tens of cm/kyr. He showed that an original detrital input signal could still be traced but that it was modified by variable reductive diagenesis.

Assessment of diagenetic processes is important for the understanding of the paleoenvironment but also for the understanding of the NRM. This is illustrated by Dekkers et al. (1994) in a study of a piston core from the central Mediterranean Sea. Cyclic lithology has generated a variety of diagenetic regimes which have a profound influence on the NRM record and, consequently, on its interpretation. Dekkers et al. (1994) used multivariate statistical methods (fuzzy-c-means cluster analysis and non-linear mapping on a combination of mineral-magnetic and geochemical parameters) to recognize grouping in their data set. Two categories of clusters were recognized, one dominated by original lithological properties, the other by diagenesis. Isolation of geomagnetic field information from the latter evidently is doubtful. Recently, Schwartz et al. (1996) argued that many sedimentary records of the paleointensity of the geomagnetic field may still be biased by environmental factors. Studies like these highlight the importance of environmental magnetic investigations for retrieving geomagnetic field information from the sedimentary record.

Because the mineral-magnetic methodology is rapid, recent studies are always based on a fairly large number of samples. Substantial information can be gained from such high-resolution studies. For example Stoner et al. (1995) document subtle changes between the last two deglaciation events, Terminations I and II, as recorded in the Labrador Basin, south of Greenland. Mineral-magnetic parameters show already changes prior to the change in $\delta^{18}O$ which indicates a warmer climate. The changes in magnetic parameters are interpreted as being indicative of the 'degrounding' of the ice-sheet before the actual deglaciation started. Termination I has occurred in a two-step fashion. In contrast, deglaciation was very rapid during Termination II which shows synchronous mineral-magnetic and $\delta^{18}O$ characteristics.

Lake sediments

Lake sediments are traditional targets for the determination of the paleosecular variation of the geomagnetic field because sedimentation rates are generally higher in lacustrine than in marine environments. In the late seventies, Oldfield and his co-workers (e.g. Thompson & Oldfield 1986) started to use the magnetic information in the lacustrine sedimentary record to infer changes in provenance areas which could be tied to changes in paleoclimate. A few recent examples of work on lake sediments are summarized here. Peck et al. (1994) reported on the mineral magnetic properties of slowly deposited Quaternary sediments (0–250 ka) from Lake Baikal (Siberia) and correlated the record to the oxygen isotope time scale derived from marine sediments. They showed that interglacial periods in Lake Baikal were characterized by increased productivity of diatom opal that resulted in fairly low concentrations of magnetic material. Decreased diatom productivity during glacial periods (glaciers never covered the lake during these periods) together with increased eolian deposition resulted in high concentrations of magnetic material. Hematite staining of eolian grains also yielded a magnetic mineralogy which was distinctly 'harder' (has a lower S-ratio, Appendix 1) during glacial periods.

Interestingly, the study of Rosenbaum et al. (1996) on slightly older Quaternary sediments of cores from Buck Lake (Oregon, USA) showed different magnetic properties. They used mineral-magnetic and pollen records to derive the glacial and interglacial periods. In contrast to Lake Baikal, glacial sediments in Buck Lake are characterized by a high S-ratio while the concentration of magnetic minerals is also high compared to interglacial periods. This indicates that understanding of the pathways is crucial for the interpretation of an environmental magnetic record. By correlating trace-element concentrations (immobile elements Ti and Zr) to Fe and magnetic properties, Rosenbaum et al. (1996) showed that the magnetic signal in Buck Lake is mostly detrital, while post-depositional diagenesis has only a minor influence. Therefore, paleoclimate information could still be retrieved from the Buck Lake record. Rosenbaum et al. (1996) advocated peak runoff as the critical factor in generating the observed magnetic properties. Cold-to-warm climatic transitions are recorded as changes in magnetic properties before changes in pollen compositions suggesting that erosional processes (peak runoff) responded more rapidly to climate change than the vegetation in the catchment area.

The pathway can cause considerable changes in mineral-magnetic properties. This is also illustrated by the study of Vlag et al. (1996) of Lac St.Front, a maar lake in the French Massif Central. This lake is ideally suited for provenance and pathway studies because the catchment area only consists of the ancient crater wall. Vlag et al. compared the paleomagnetic properties of the sediments of Lac St.Front to those of nearby Lac du Bouchet (Thouveny et al. 1994) and were able to correlate both records. Glacial periods were characterized by substantially increased concentrations of magnetic minerals. In Lac St.Front, they showed that the mineral-magnetic properties of both glacial and interglacial sediments did not correspond to either those of the bedrock in the catchment area, nor those of the, present-day interglacial, soil bordering the lake. This indicates that weathering of the rock, formation of the soil, the sedimentation itself and post-depositional alteration of the sediments each may induce changes in the observed mineral-magnetic properties.

Mixed magnetic mineralogy and magnetic sulphides

The assemblage of magnetic minerals is usually dominated by magnetite which can be (partially) oxidized to maghemite. The presence of hematite and goethite can be inferred from magnetic hysteresis measurements. In lacustrine and marine sediments, however, magnetic iron sulphides, principally greigite and pyrrhotite, may also be present. In particular, greigite is now regarded as fairly common (Snowball & Thompson

1988, 1990, Krs et al. 1990, 1992, Snowball 1991, Tric et al. 1991, Horng et al. 1992, Roberts & Turner 1993, Reynolds et al. 1994, Jelinowska et al. 1995, Roberts et al. 1996, Torii et al. 1996). This complicates the interpretation of the mineral-magnetic record because the field-dependent behaviour of the magnetic iron sulphides closely resembles that of magnetite (Roberts 1995, Dekkers & Schoonen 1996). When the presence of magnetic sulphides is suspected, one should determine the magnetic behaviour as a function of temperature, either below or above room temperature. Low-temperature determination shows the presence of pyrrhotite by the 34-Kelvin transition. Greigite has no such transition. Because greigite alters on heating, thermal demagnetization or thermomagnetic analysis are discriminative between greigite and pyrrhotite (Reynolds et al. 1994, Roberts 1995, Torii et al. 1996, Dekkers & Schoonen 1997). The presence of magnetic sulphides also complicates the interpretation of the frequency dependence of χ_{in}. For iron oxides, this dependence can be explained in terms of grain-size. For pyrrhotite, however, which has a much higher electrical conductivity than the iron oxides, Worm et al. (1993) documented that effects of eddy currents seriously hamper the interpretation of magnetic susceptibility at frequencies above 1 kHz.

Human impact on the environment

Most industrially-derived dusts contain magnetic particles (Flanders 1994). Industrial dust is clearly distinguishable from natural dust by plotting, for example, the IRM/ARM ratio versus the frequency dependence of the low-field susceptibility (Oldfield et al. 1985). Flanders (1994) collected air-borne dust from tree trunks or other fairly smooth surfaces, or directly from the atmosphere by using sticky surfaces attached to permanent magnets. The second method is much less efficient for collecting material for measurement of hysteresis loops than it is to wipe a smooth tree trunk with a towelette. Wiping of $\sim$0.1 M^2 of a smooth outdoor surface yields plenty of material. The extent of particulate contamination can thus be easily studied. The amount of atmospheric particulate matter of anthropogenic origin usually exceeds the cosmogenic contribution by several orders of magnitude. Surfaces collected in relatively pollution-free non-urban environments contain two orders of magnitude more magnetic material than that inferred from the Earth's bombardment by 10^5 tons of cosmic spherules per year (Flanders 1994). It is therefore only necessary to distinguish the natural terrestrial and the anthropogenic contributions. This can be done by measuring the magnetic properties. Several projects along similar lines are underway in contaminated areas in central Europe. Magnetic susceptibility correlates reasonably well with the heavy-metal content of some fly ashes (Dekkers & Pietersen 1992, Stryszcz 1993) which increases the suitability of magnetic methods for the monitoring of pollution. Mapping of the magnetic susceptibility in various soil profiles in industrialized districts in southern Poland has demonstrated a relationship between magnetic susceptibility in top soil horizons and anthropogenic pollution by coal-burning electrical power plants, and iron and steel works (Stryszcz 1996).

Archeological and historical furnaces are generally used for the determination of the paleosecular variation of the geomagnetic field. Mineral-magnetic techniques, however, have also been applied in archeological studies. Repeated burning in archeological hearths enhances the magnetic susceptibility, a property which can be used in the detection of archeological sites (Tite & Mullins 1971, Von Frese 1984, Sternberg 1987). McClean & Kean (1993) argued iron derived from ferritin, an iron storage protein in plants, to be the source of the iron in magnetite which resulted from the burning of wood. Dalan & Banerjee (1996) used mineral-magnetic methodology in an archeological study of the Cahokia mounds, an ancient native American site in Illinois (USA). By plotting ARM versus χ_{in} of various topsoils, they could distinguish cultivated from undisturbed areas.

Discussion and outlook

Clearly, the interpretation of magnetic measurements as proxy parameters for (paleo)climate and (paleo)environment is not always straightforward. On the other hand, such measurements are rapid, relatively inexpensive, and yield grain-size-dependent information, making them very attractive. To constrain the interpretation, pathway studies are important to trace changes in magnetic properties during the transport of the material from a source area to a depositional site. Climatically driven diagenetic processes usually result in variations of magnetic mineralogy. This magnetic information, in turn, can be interpreted in terms of a variable paleoclimate and paleoenvironment. Understanding paleoclimate requires interdisciplinary work combining geochemical and palynological data with mineral-magnetic information, as illustrated nicely by Rosenbaum et al. (1996). An example combining mineral-magnetic, stable isotope, biostratigraphical, inorganic and organic geochemical data, and sedimentological information, is an ongoing research programme in Utrecht to analyse in detail the paleoclimate and paleoenvironment signal in Early Pliocene lacustrine lignite-marl sequences in Greece.

The application of magnetic methods for paleoclimate reconstruction requires a reliable chronostratigraphy. Integrated magnetostratigraphy, biostratigraphy, cyclostratigraphy, oxygen-isotope stratigraphy and radiometric dating of suitable lava flows or ash layers can be used to construct this time frame. In the aforementioned Utrecht project on the Greek lacustrine lignite-marl sequences the chronostratigraphy is based on magneto-cyclostratigraphy.

The paleomagnetic record may be obscured by diagenetic mobilization of iron. A high-resolution time frame therefore cannot always be established unambiguously. For example, the Matuyama/Brunhes transition at 780 ka is recorded in the Chinese Loess Plateau at seemingly different ages in loess unit 7, at varying depths in soil unit 8, and in loess unit 8, depending on the location (Hus & Han 1992). In marine sediments, geomagnetic reversals may also be recorded 'diachronously' as a consequence of diagenesis (Van Hoof et al. 1993). Paleomagnetic techniques can then be used to estimate the duration of diagenetic effects on the NRM.

The use of magnetic susceptibility as a proxy parameter to delineate suspected polluted areas may be attractive because data are comparatively easily acquired in a cost-effective manner. For a meaningful analysis, however, the human contribution needs to be separated from the natural background signal. This requires additional research to further substantiate the conceptual relations between magnetic properties and elemental pollution. The sorbing capacities of secondary iron oxides and oxyhydroxides in soils to either immobilize or release contaminants, depending on the geochemical environment, make magnetic methods attractive as a first 'screening' for pollution studies. The magnetic and chemical characterization of different pollution sources, e.g. fly ashes, bottom ashes, and tailings of iron and steel works and mines, will further constrain the interpretation of magnetic parameters. Multivariate statistical techniques can be advantageously implemented in this interpretation.

To improve the use of magnetic methods for environmental purposes, particularly for the analysis of grain-size distributions of iron oxides, additional research is required. One should keep in mind that the grain-size-dependent information is often difficult to obtain by other analytical techniques. The underlying processes which have led to the observed properties, also need further attention. Therefore, a considerable research effort is currently being put into developing better interpretational concepts of magnetic hysteresis loops, implementation of temperature-dependent magnetic measurements, and cross-calibration of mineral-magnetic methods with other analytical techniques, like (trace) element geochemistry, various sequential extraction schemes and Mössbauer spectroscopy. Mössbauer spectroscopy at low temperatures can be used for further analysis of ultrafine grains, but is time-consuming. In summary, an increased application of the rapid mineral-magnetic techniques is foreseen.

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Appendix 1

Acronyms of magnetic parameters as used in the present paper

M_s	saturation magnetization expressed per unit volume (Am^{-1}).
σ_s	saturation magnetization expressed per unit mass ($Am^2\ kg^{-1}$).
M_r	remanent saturation magnetization per unit volume (Am^{-1}).
σ_r	remanent saturation magnetization per unit mass ($Am^2\ kg^{-1}$).
B_{cr}	remanent coercive force (mT).
B_c	coercive force (mT).
χ_{in}	low-field or initial susceptibility, the magnetic moment of a sample measured in a low applied magnetic field divided by the field value (per mass-specific units m^3kg^{-1}; volume-specific units are dimensionless).

χ_{hf}	high-field susceptibility, the magnetic moment of a sample measured between two field values higher than the fields needed to saturate the ferrimagnetic contribution to the magnetization. Also referred to as the paramagnetic susceptibility (units are the same as for χ_{in}).
ARM	Anhysteretic Remanent Magnetization (Am^2), acquired by the decay of an asymmetric alternating field (AF) from a peak value to zero (usually). In practice, AF demagnetization equipment is used, a small direct current (DC) bias field is imposed. ARM intensity is linear with the DC bias field for low concentrations of magnetic minerals to field values of $\sim$50 to 80 μT (i.e. to values comparable to the intensity of the geomagnetic field).
σ_{ARM}	ARM intensity per unit mass ($Am^2\ kg^{-1}$).
χ_{ARM}	ARM intensity per unit volume divided by the value of the DC bias field (dimensionless SI units).
(S)IRM	(Saturation) Isothermal Remanent Magnetization. IRM is acquired by imposing a certain DC field to a sample at a constant temperature (usually room temperature), the maximum possible remanence value (after magnetic saturation of the sample) is termed SIRM or M_r.
S-ratio	The ratio of the IRM acquired in a back field (usually 0.3 Tesla) and in a positive field of 1 Tesla. This ratio serves to discriminate magnetite (or other easily saturated magnetic minerals) from hematite and goethite which are not saturated in a field of 0.3 Tesla. To obtain positive values for the S-ratio in most samples, the back-field IRM (which is usually oppositely directed with respect to the orginal IRM acquired in a field of 1 Tesla) is entered negatively. Samples with an S-ratio of 1.0 contain only magnetite; lower values indicate contribution of 'harder' magnetic minerals which are not saturated in a field 0.3 Tesla so that some remanence of the original IRM acquired in a field of 1 Tesla, remains.

For low concentrations (i.e., in the absence of magnetic interactions) mass-dependent parameters are linear with concentration at least to a first-order approximation which is applicable in most natural case studies. By dividing two mass-dependent parameters, a concentration-independent variable is obtained which allows interpretation of grain-size trends. Field values, such as the coercivity parameters, are concentration-independent to a first-order approximation.

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