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Depositional remanent magnetization: Toward an improved theoretical and experimental foundation

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Abstract

The first theoretical predictions for the behavior of magnetic particles in water were that sedimentary magnetizations would be fully aligned with the ambient field, yet redeposition experiments showed a strong (and quasi-linear) dependence on the external field. This empirically observed linearity has served as the fundamental assumption of sedimentary paleointensity studies for decades. We present redeposition experiments which suggest instead that the relationship between depositional remanence (DRM) and applied field may frequently be curved for magnetic fields in the range of the Earth's. Numerical simulations using a flocculation model can explain the redeposition data and suggest that DRM will be significantly non-linear when the flocs are small (several microns). There is a strong dependence of floc size on salinity particularly in low salinity environments. Floc size has a profound influence on the efficiency of DRM, hence low salinity environment may give results with poor reproducibility. The size of the floc in which magnetic particles are embedded is not accounted for in current methods of normalization, yet is the most important parameter. On the bright side, however, it now seems possible to quantitatively explain paleointensity in sedimentary systems opening the door to absolute paleointensity estimates from sediments whose key parameters of floc size distribution and settling times can be constrained.

Keywords: DRM; sedimentary paleointensity; flocculation; depositional remanence

1. Introduction

Direct observation of the geomagnetic field has been carried out for the last 4 centuries; prior to that time, we must cull data from archeological and geological proxies. One of the first motivations for investigating the magnetism of rocks, therefore, was to study the behavior of Earth's magnetic field in the past [1]. Under certain laboratory conditions, sediments have been shown to acquire a remanence that is linearly related to the applied field [e.g., [1–4]]. The assumption behind sedimentary relative paleointensity studies is therefore that the depositional remanent magnetization (DRM) carried by detrital grains, is a linear function of applied field B under natural conditions as well.

In addition to sensitivity to the applied field, DRM is also a strong function of mineralogy, concentration and grain size of the magnetic phases and properties of the non-magnetic matrix (see reviews by [5,6]). This dependence on factors other than the field implies that sedimentary magnetizations must be normalized by some parameter (generally a bulk magnetic property like saturation remanence, anhysteretic remanence or

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magnetic susceptibility) which is intended to compensate for these non-field effects in order to obtain the contribution of the magnetic field (e.g., [7,8]). Yet, the success of normalization relies on a firm theoretical understanding of DRM. However, despite the enormous effort that has been put in to acquiring sedimentary relative paleointensity records, the experimental and theoretical basis for DRM is relatively undeveloped, particularly when compared with the sustained focus on, for example, thermal remanence and absolute paleointensity (see, e.g., [9]). The scatter in relative paleointensity data far exceeds measurement error or errors from stacking of multiple records (see, e.g., [10]) and understanding the possible sources of scatter from a theoretical point of view could lead to an improvement of our understanding of paleointensity variations in the past.

There are several aspects to the problem of translating DRM into a (relative) paleointensity record. One is the physical theory concerning particle alignment in a viscous medium. A second aspect is how to compensate for variations in the magnetizability over the sequence (the choice of normalizer). Finally, there are issues of temporal resolution involving the degree of smoothing and the depth at which the magnetization is fixed. In this paper, we will focus primarily on the first problem which has received little attention, yet lies at the heart of sedimentary paleointensity studies. Once a better theoretical understanding of depositional remanence has been achieved, the problems of how to choose the most appropriate normalizer and the degree of smoothing and lock-in depth can be considered.

2. Theoretical background

Magnetic grains tend to align with the ambient magnetic field as they fall through the water column or re-align after deposition (if freed momentarily from the restraining forces of their neighbors). This initial alignment can be preserved in the sediment the magnetization of which is frequently interpreted in terms of the ancient geomagnetic field (see, e.g., recent review by [6]). We are concerned here not with the myriad possible diagenetic processes (e.g., [11]) nor the physical re-alignment during sedimentary compaction at depth (e.g. [12]) but with the physical process of particle alignment in the syn-depositional environment, generally termed “depositional remanent magnetization”.

The behavior of magnetic particles in a viscous medium has been considered for decades (e.g., [13–15]). A magnetized particle suspended in a fluid is subjected to a hydrodynamic couple generated by fluid shear, a magnetic couple tending to align the magnetic moment with the ambient magnetic field, viscous drag and inertial forces tending to oppose motion and thermally inspired “Brownian” motions. Nagata [13] described the motion of magnetic particles in water with the equation of motion for a magnetic particle with magnetic moment m with angle α with respect to the applied magnetic field B by:

$$I\frac{\mathrm{d}^2\alpha}{\mathrm{d}t^2} = -\lambda\frac{\mathrm{d}\alpha}{\mathrm{d}t} - mB\sin\alpha, \tag{1}$$

where λ is the viscosity coeffcient opposing the motion of the particle through the fluid and I is the moment of inertia. By neglecting the inertial term (which is orders of magnitude less significant than other factors), Nagata [13] solved Eq. (1) as:

$$\tan\frac{\alpha}{2} = \tan\frac{\alpha_o}{2}e^{(-mBt/\lambda)} \tag{2}$$

where α_o is the initial angle between $\mathbf{m}$ and $\mathbf{B}$. Setting $\lambda = 8\pi r^3\eta$ where r is the particle radius and η is the viscosity of water ($\sim 10^{-3}$ kg m^{-1}s^{-1}) the time constant of Eq. (2) over which an inital α_o is reduced to $1/e$ of its value is:

$$\tau = \frac{\lambda}{mB} = \frac{6\eta}{MB} \tag{3}$$

where M is the volume normalized magnetization. The fundamental problem that has plagued DRM theory for over four decades is that this time constant, for almost all reasonable values of M and B, is extremely short. Taking the value of magnetization for single domain magnetite of $M = \sim 4.8\times10^5$ A/m in a field of 50 μT, gives a value of τ of several milliseconds. Even the magnetization for hematite ($M \sim 200$ A/m) results in a τ of less than a second at this field strength. So simple DRM theory predicts that sediments composed of isolated magnetic particles should have magnetic moments that are fully aligned, hence insensitive to changing field strengths.

Ironically, the first measurements of sedimentary paleointensity of Johnson et al. [1] showed a strong dependence of DRM on the applied field. Moreover, the experimentally determined remanent magnetization was more or less linearly related to the field for field strengths in the range of the Earth’s and was orders of magnitude less than the saturation remanence. Much subsequent thought about DRM theory has attempted to reconcile the simple prediction of saturation with the observational fact of a strong and nearly linear field dependence (for low fields) of DRM.

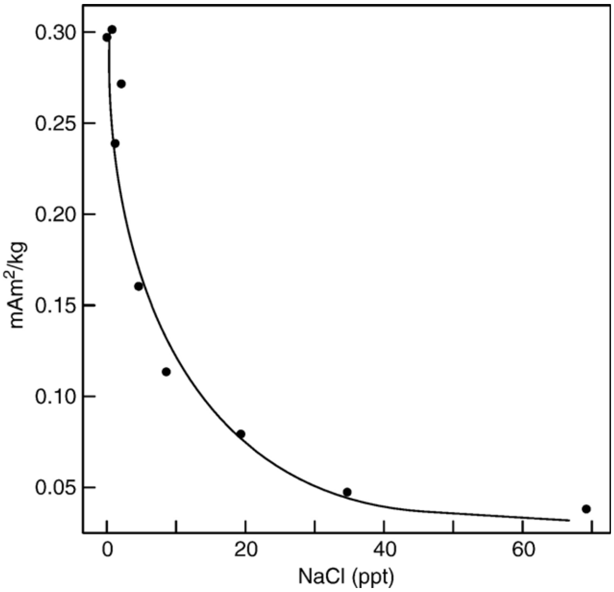

Fig. 1. Effect of NaCl concentration on DRM intensity. Data replotted from [24].

The field dependence of DRM implies a much longer time constant of alignment than that predicted by Eq. (3). There are three ways to accomplish this from a theoretical point of view. First, one can hypothesize a value of M much lower than saturation. Along these lines, Collinson [14] suggested that hematite would have a much lower value of remanent magnetization than saturation remanence because hematite particles would have been magnetized through a chemical process which he claimed would lower M by a factor of 50. He also noted that experimental values of thermal remanences of magnetite are lower than saturation and Stacey [15] used this as evidence for pseudo-single domain magnetite. The problem is, however, that even with values of M in the pseudo-single domain range or even in the range of hematite, the theoretical time constant for alignment is still uncomfortably short.

A second approach for predicting a field dependence of DRM is to call on Brownian motion [14,15] which would act to randomize magnetic moments through thermal agitation. Collinson [14] envisioned a mechanism for DRM that was the result of the ambient field tending to align small magnetic particles opposing a randomizing effect of Brownian motion. Such a remanence would follow the Langevin function for paramagnetic gasses for magnetization as a function of applied field. He suggested that thermal agitation would continue in the pore spaces of the sediment before consolidation. The equlibrium magnetization would then be "frozen" in at some point when the sediment was suffciently dewatered (along the lines envisioned by [16]).

To estimate the size of particles effected by Brownian motion, Collinson [14] used the equation:

$$\frac{1}{2} mB\phi_o^2 = \frac{1}{2} kT, \tag{4}$$

where ϕ_o is the Brownian deflection about the applied field direction, k is Boltzmann's constant (1.38×10^{-23} J/°K) and T is the temperature in kelvin. This equation neglects viscous drag and the momentum of the particles and tends to overestimate the effect of Brownian motion, particularly when the magnetic moments of the particles are small. It is perhaps more useful to consider the viscous drag of a particle (see [17] for a complete derivation) for which we have:

$$\frac{\phi_o^2}{\tau} = \frac{kT}{4\pi\eta r^3},$$

where τ is the time span of observation (say, 1 s). According to this relationship, particles smaller than about a micron will be strongly effected by Brownian motion. Particles that have a substantial magnetic moment however, will be partially stabilized (according to Eq. (4)) and might remain unaffected by Brownian motion to smaller particle sizes (e.g., 0.1 μm). Of course we note here that it is the sub-micron particles of magnetite that most likely give rise to the stable

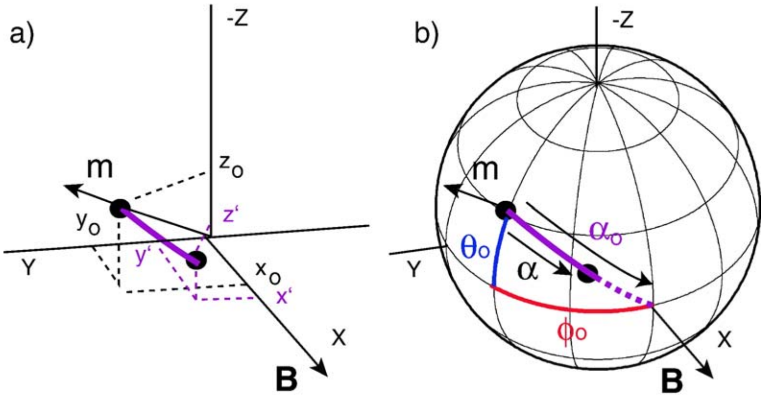

Fig. 2. Coordinate system for numerical simulations. X is the direction of the magnetic field (B) and Y and Z are two other orthogonal axes. a) x_o, y_o, z_o are the Cartesian coordinates of the initial moment direction and x', y', z' are after time t. b) same as a) but expressed in θ, the angle between moment **m** and the X–Y plane and ϕ, the angle between the projection of m in the X–Y plane and the X axis. θ_o, ϕ_o, α_o are the initial angles where α_o is the initial angle between m and the applied field. α is the angle after time t.

magnetizations because larger particles almost certainly have little magnetic stability (see, e.g., [9]) so isolated sub-micron particles of magnetite would be subject to Brownian motion.

Shcherbakov and Shcherbakova [18] took an entirely different approach to the problem of linear dependence of DRM. They ascribed the observed misalignment to sticking of particles together ("coagulation" or "flocculation"), making larger particles with a lower net moment. By reducing M they could increase the theoretical time constant of alignment.

The chemical processes controlling flocculation are described in detail by [19]. Particles suspended in an aqueous solution collide as a result of Brownian motion or turbulence (see [20]). In pure water, they usually separate again whereas if even a small amount of salt is added to the solution, they may stick together forming ever larger agglomerates (flocs). Qualitatively, particles are drawn together by van der Waals forces which are independent of the chemistry of the fluid. In pure water, clay particles like illite are surrounded by a double layer of ions lending an electrical charge to the particles. These charges repulse one another, keeping the clays apart in a stable colloid. The addition of salt (and other electrolytes) interferes with the double layer, thinning it, allowing the van der Waals forces to come in to play and the particles are more likely to stick to one another.

Several papers since the early 90s highlighted the role of water chemistry in controlling depositional remanence [21–24]. We have replotted the data from Katari and Tauxe [24] in Fig. 1 which used NaCl concentration to control floc size of mud slurries; these data confirmed prior results which suggested that particle size resulting from flocculation leads to a decreased DRM.

Katari and Bloxham [25] pursued the role of flocculation in the problem of DRM theory, reminding us that particles of magnetite are unlikely to be in isolation in many natural environments tending to stick to clay particles, as observed in SEM photos by [26]. The tendency to stick is controlled by van der Waals forces under the right conditions of salinity and/or pH (see, e.g., [21,24]).

These small clay particles in turn tend to become incorporated into pellets held together by organic "glue" or become part of larger flocs. Katari and Bloxham [25] argued that the appropriate value of m in Eq. (2) should be the net moment of the floc (or pellet) which could be far less than the moment of an isolated magnetic particle of the same size. This simplest flocculation model of DRM explains the lack of saturation in natural sediments as the effect of the viscous drag on large flocs whose moments are small because they are diluted by non-magnetic "fluff". Higher fields allow more complete alignment and result in the handy field dependence much exploited of late (see, e.g., [6] for a useful recent review).

There are several aspects to the flocculation model which we will consider in turn. We will begin with the Katari and Bloxham [25] version which suggests a practical approach to numerical simulation of DRM. They started with a set of magnetic moments with directions (ϕ_o, θ_o, Fig. 2) drawn from a uniform distribution about the applied field. Then, they extended the equation of motion for a particle in a viscous fluid of Nagata (Eq. (2)) to include the terms ϕ and θ (instead of just α), solving for first θ, then ϕ after settling for time t.

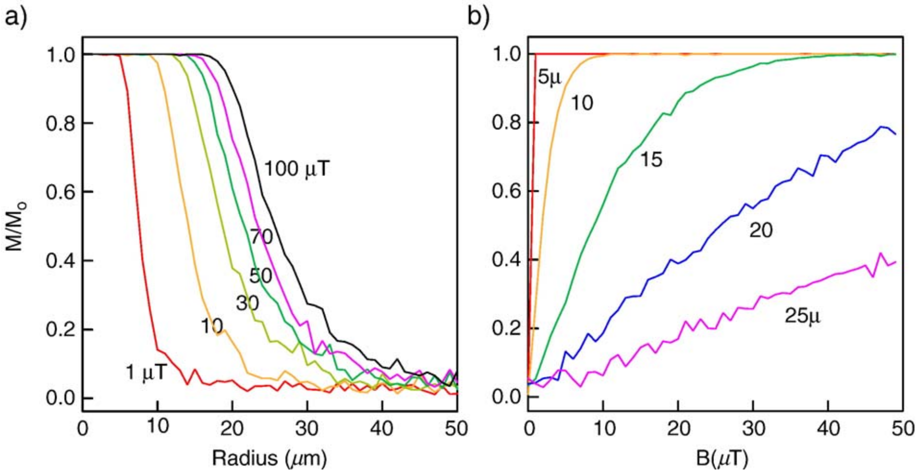

Fig. 3. Results of numerical experiments of the flocculation model using the parameters: $l = 0.2$ m and the viscosity of water, holding m constant at 5 fA m^2. M/M_o is the DRM expressed as a fraction of fully aligned particles. a) Holding B constant and varying r. For a given field strength, particles are either fully aligned or randomly oriented, except for within a very narrow size range. b) Holding r constant and varying B. Small magnetic particles reach saturation at very low fields, while larger particles can have a quasi-linear dependence on B.

Solving for ϕ and θ separately introduces an unrealistic anisotropy in the resulting moment directions. In this paper we develop a slightly modified approach of assuming that the moment moves along the straightest path toward the applied field (see Fig. 2). To find the new coordinates of **m** after time t, we express the angles, ϕ_o and θ_o in their cartesian equivalents (x_o, y_o, z_o) and the angles after time t in terms of x', y', z' (Fig. 2). The moment moves from the original coordinates (x_o, y_o, z_o) toward **B**, so the ratio $y'/z' = y_o/z_o$). Keeping the magnitude of the moment invariant, we have the equations:

$$\frac{y_o}{z_o} = \frac{y'}{z'},$$

and

$$x_o^2 + y_o^2 + z_o^2 = x'^2 + y'^2 + z'^2 = 1.$$

We can calculate the new coordinates (x', y', z') of **m** by:

$$x' = \cos\alpha, \ y' = \sqrt{\frac{1-x_o^2}{1+\frac{z_o^2}{y_o^2}}}, \ z' = y'\frac{z_o}{y_o} \tag{5}$$

Katari and Bloxham [25] rearranged Eq. (2) by replacing time with settling distance, a parameter that is more easily measurable in the laboratory. Because particle settling rates are key parameters in a host of environmental and engineering problems, from silting of harbors to sewage treatment, there has been considerable interest in settling velocity versus floc size and density (see, e.g., [20,27]). Flocs settle in a fluid with a rate that depends on a variety of factors including size, shape, and density of the floc as well as the Reynold's number of the flow regime in which it is settling. To first order, the Gibbs [28] approximation is reasonable for the smallest flocs of interest here and is consistent with the more recent modeling attempts which use the fractal nature of floc formation. The Gibbs settling velocity v (in units of meter per second) is an empirical function of floc radius (in units of meters):

$$v = 1.1r^{0.78} \tag{6}$$

Substituting into Eq. (2) we have:

$$\tan\frac{\alpha}{2} = \tan\frac{\alpha_o}{2}\exp(-mBl/8.8\pi\eta r^{3.78}). \tag{7}$$

The final aspect of the flocculation model is the incorporation of a distribution of particle sizes $f(r)$ [15,25]. The contribution of each size fraction is evaluated separately and normalized by its proportional representation.

In order to explore the predictions of the flocculation model as developed here, we begin with a set of N individual flocs with θ_{oi}, ϕ_{oi} drawn from a uniform distribution. Following Katari and Bloxham [25] we assume initially that each floc has magnetic moment m independent of floc size. Later we will incorporate a more elaborate scheme whereby larger floc moments result from the vector sum of the fundamental flocs from which they are formed (as necessitated by our redepositional data), but for now we will adhere to the simple theory of Katari and Bloxham in its modified

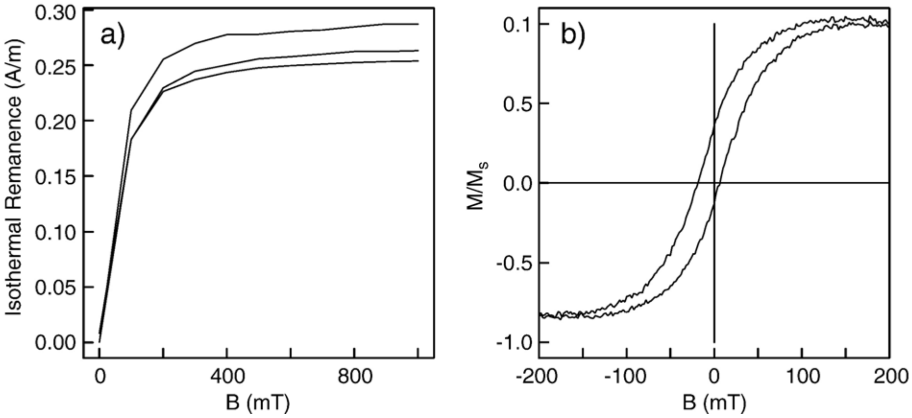

Fig. 4. Rock magnetic behavior of the mud used in our experiments. a) Acquisition of isothermal remanent magnetization for representative examples. b) Typical hysteresis behavior.

form in which we calculate x_{oi}, y_{oi}, z_{oi} from θ_{oi}, ϕ_{oi} and then α using Eq. (5); α can then be converted back to x', y', z' and the net contribution summed for each value of r. The net DRM is the vector sum of the contributions from all the floc moments.

3. Numerical simulations

The flocculation model described in the foregoing can be used to predict DRM intensity behavior, given that settling length l, $\mathbf{B}$, $\mathbf{m}$, and the floc size distribution $f(r)$ are known. For an initial set of simulations, we follow Katari and Bloxham [25], using the viscosity of water, m of 5 fAm^2 (where femto $(f) = 10^{-15}$), and a settling length l of 0.2 m. We have used $N = 1000$. While larger values of N result in smoother models, the conclusions do not change. The first set of simulations assume a single value of r, instead of a distribution of r in order to get a feel for how the model behaves.

Holding B constant (Fig. 3a) and varying r, we see that for small r, full alignment is achieved and the DRM is simply the linear sum of all the individual moments and is independent of changes in B (i.e. it is a saturation DRM or sDRM) [in this case, if the magnetic moments themselves are saturation remanences, M_r or equivalently saturation isothermal remanences (sIRM), the sDRM will equal the sIRM.] Above a certain radius, the particles are unable to align themselves effciently in the time alloted and are essentially random. Increasing B increases the particle size over which saturation is achieved for a given value of m.

Holding r fixed and varying B (Fig. 3b), we see that for r less than about 5 μm, the DRM is essentially saturated, while larger particles display a dependence of DRM on field strength. We note that for most simulations, DRM is only quasi-linear with B, even for fields in the range of 0–50 μT. These simulations show that in the flocculation model, field dependence of DRM is controlled by the fraction of particles that are completely aligned with the field and that this fraction varies with B.

Our DRM model can make specific predictions. If we can control such variables as viscosity, settling time, magnetic moment of the flocs and the magnetic field, we can compare these predictions with redepositional data, thereby evaluating the applicability of the model to sedimentary systems.

4. Materials and methods

Previous studies (e.g., [21–24]) have shown that salinity profoundly affects the DRM acquired for low salinities (< ~20), an effect that was interpreted as the result of changing the floc size distribution (see, e.g., Fig. 1). Here we wish to expand on earlier efforts by measuring DRM not only as a function of floc size but also as a function of applied field. To test the flocculation model of DRM we fabricated settling tubes of 0.3 m in height and 4 cm in diameter of borosilicate. After allowing the mud to settle, the tubes can be inserted into the CTF three axis cryogenic magnetometer housed in the magnetically shielded room at Scripps Institution of Oceanography without the need for drying, freezing or otherwise immobilizing the sample. The magnetometer has a measurement sensitivity of approximately 1 pA m^2 and the tubes have blanks of less than 0.1 nA m^2. We used a set of 12 settling tubes, each with 500 ml of initially de-ionized water to which a known amount of NaCl was added. NaCl concentration in our experiments ranged from 1 to 7 grams NaCl per kilogram water with

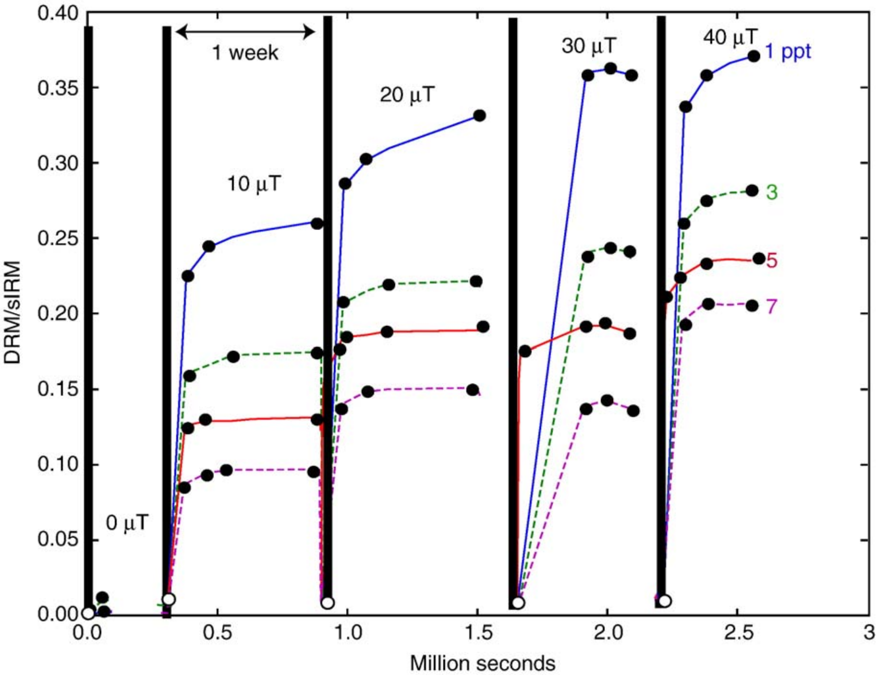

Fig. 5. Results from one settling tube (out of 3) for each NaCl concentration. Tubes were shaken and placed in the fields at times indicated by the heavy vertical bars and measured periodically. DRM is expressed as the fraction of the measured sIRM for each tube.

three tubes of each salinity [please note that grams NaCl per kilogram water is equivalent to salinity quoted as “parts per thousand” or in the dimensionless “practical salinity units”. We will quote NaCl concentrations as concentrations in parts per thousand (ppt)].

All redeposition experiments reported in this paper used hemipelagic mud, similar to the mud used by Katari and Tauxe [24] (Fig. 1), which was obtained from gravity core 149KK taken from 39.918°N, 124.688°W in 1993. The magnetization is likely carried by magnetite based on the acquisition curves of isothermal remanence and hysteresis measurements (see Fig. 4 and [24]).

Samples of 0.3 g of mud were given a saturation isothermal remanence (sIRM) in a 1 tesla impulse field and introduced into the settling tube of a given NaCl concentration. The mud was allowed to settle in vertical magnetic fields ranging from 0 to 40 μT and measured periodically over a period of about a week. The experiments reported here build on those shown in Fig. 1, using very similar mud and an improved experimental design. Because the zero NaCl experiment took many weeks to settle and only 1 ppt NaCl reduced settling time to a week or less, we decided to focus on the interval with the most profound change in DRM intensity as a function of NaCl concentration while avoiding repeating the time consuming zero NaCl experiment.

We estimate the distribution of the resulting floc sizes using a Fluid Imaging Technologies, Inc, FlowCAM available to us at Scripps. The FlowCAM draws a fluid with suspended particles into an imaging tube which illuminates the sample with an LED. Photographs of the flocs are taken with a CCD camera and the images are stored in digital files. Image analysis software estimates the approximate area, length and width of up to 10,000 flocs per run. The areas of individual flocs are estimated from pixel counts made on the photographic images obtained from the FlowCam. These can be converted to equivalent radii (assuming that the flocs are approximately spherical). The lower and upper limits of resolution of this machine are approximately 1 and 100 μm, respectively.

5. Results

We show typical results for tubes of different NaCl concentrations in Fig. 5. In general these results suggest the following:

(1) The higher the NaCl concentration, the lower the net moment (confirming the results of previous efforts [21,22,24] (e.g., Fig. 1).
(2) The higher the salinity, the faster the particles settled, (the tubes with 7 g NaCl per kilogram of

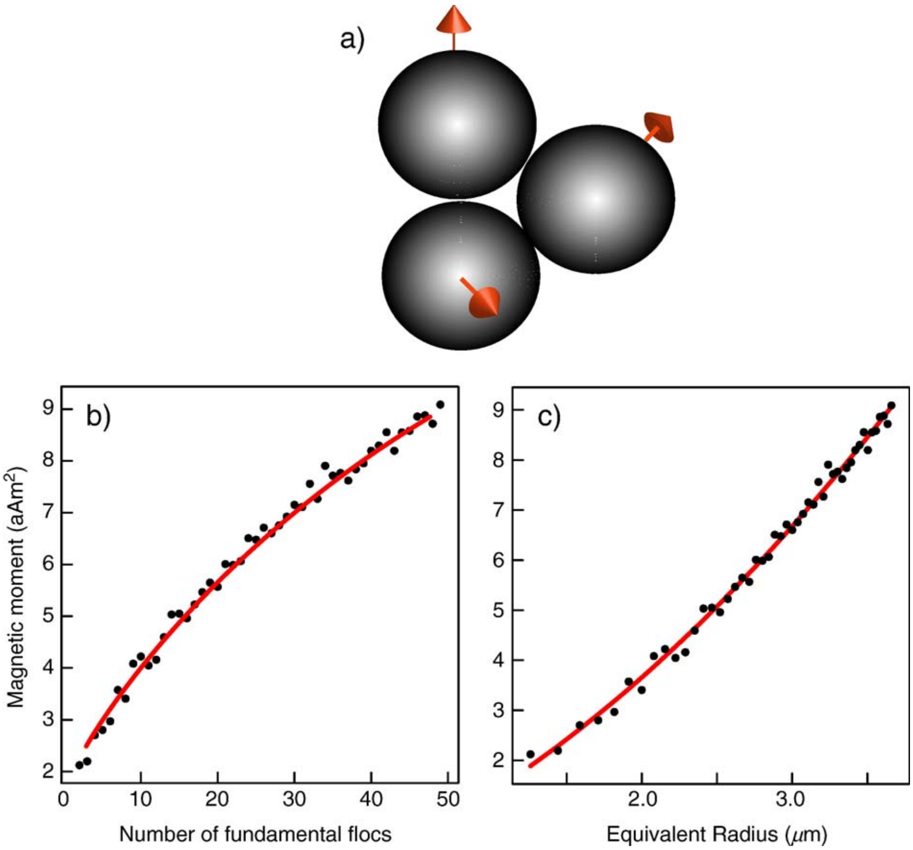

Fig. 6. a) Composite floc built up from individual “fundamental flocs” each with a moment indicated by the arrow. The net magnetic moment of the composite floc is the vector sum of the three moments, much less than the linear sum of the three moments. b) Moment (in atto Am^2; atto $=10^{-18}$) versus number of fundamental flocs in composite floc. c) m versus equivalent radius. Line given by polynomial fit $m=ar^2+br+c$ where $a=3.61\times10^{-7}$, $b=1.2\times10^{-12}$, $c=-2.1\times10^{-19}$ in this case.

water reach a plateau value faster than the tubes with only 1 g NaCl per kilogram water.

(3) In general, the higher the applied field, the higher the DRM, although a saturation DRM appears to be nearly achieved in the 1 ppt NaCl set of tubes by 30 μT.

(4) Most surprisingly, the relationship of DRM to B is far from linear with applied field in all cases.

(5) In the Katari and Bloxham [25] model of DRM, a single magnetic particle is assumed to be embedded in each floc; hence the magnetization of the flocs is independent of floc size. Therefore, the saturation DRM (sDRM) should equal the sum of all the individual flocs, i.e., sIRM in the case of these experiments. The saturation DRM in all of our experiments is well below sIRM. The highest sDRM is some 33% of the sIRM. There is no Katari–Bloxham type model that can account for the results shown in Fig. 5. We consider a more realistic (and successful) model in the next section.

6. Discussion

6.1. Processes in flocculation

It is perhaps not surprising that the simple assumption of constant m independent of r fails. In nature, flocs are built up from so-called “fundamental flocs” to form composite flocs (see Fig. 6a). The fundamental flocs are discrete particles of clay or other constituents of the sediment with the tiny particles of magnetite adhering to them (see, e.g., [26]). In our experiments, the magnetization of each fundamental floc would be the sIRM that had been imparted to the mud prior to its introduction into the slurry (shown as arrows in Fig. 6a). As the fundamental flocs build up into composite flocs by chance encounters while the settling tube is being agitated, the net moment of the composite floc will of course not be the linear sum of all the individual fundamental floc moments, but will be the vector sum. Hence the magnetization of the composite floc will be

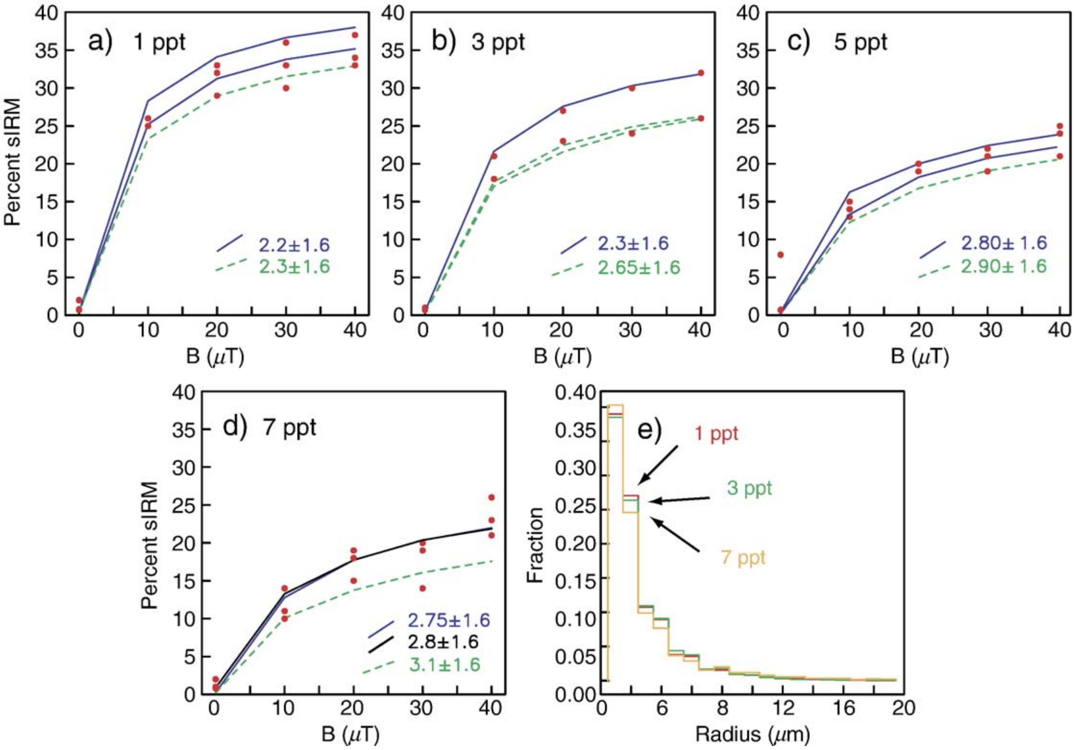

Fig. 7. a)–d) Results of settling experiments as a function of field (*B*) and NaCl concentrations (in parts per thousand). The assumed mean and standard deviations of truncated log-normal distributions for floc radii are shown in the legends and are indicated using the different line styles in the figure. e) Estimated floc radii from images obtained with the FlowCam. The inferred distributions are consistent with those observed, although the observations lack the resolution to distinguish such subtle differences.

somewhat lower than the sIRM and grow with r according to a power law.

In Fig. 6b, we show a simulation of the net moment of flocs of ever increasing size built from fundamental flocs of radius $r_o = 1$ μm, with a magnetization of 0.33 A/m (typical for the sediments used here). Each dot in Fig. 6b is the mean of 10 Monte Carlo simulations whereby directions for N flocs of radius r_o are drawn from a uniform distribution and the moments averaged. Moments are plotted as atto Am^2 (where atto is 10^{-18}) versus the number of fundamental flocs. The equivalent radius is computed by summing the volumes of the N flocs and solving for r, assuming a quasi-spherical composite floc. A polynomial fit to the moment versus equivalent radius is shown as the solid line in Fig. 6b.

When the tubes are placed in the magnetic field, the composite floc magnetic moments are initially randomly oriented. While settling, they will begin to come into alignment with the magnetic field according to the equation of motion outlined in Section 2 of this paper. sIRM was measured on each sample of 0.3 g m of mud prior to dispersal in the water. The density of the mud was 1250 kg m^{-2}. Therefore, we can estimate the moment of a fundamental floc of radius r_o. Using the composite floc model, we can therefore predict the relationship of net moment m versus floc radius for each tube, assuming a value for r_o.

A survey of the literature on flocs in nature (see, e.g., [20,27]) favors a fundamental floc size of around 1 μm. We know it cannot be bigger than this because plenty of flocs are observed that are 1 μm in the FlowCam images. It also cannot be much less than this because the size of a single flake of clay is of the order of a micron [in our model of course this single clay particle would have sub-micron sized grains of magnetite sticking to it]. Our assumed value of r_o can in any case be tested a posteori by our ability to model the data.

In order to model the DRM behavior in our experiments, we must also assume a distribution of r. Each value of r_i can be assigned a value for m using the polynomial fit of the Monte Carlo simulations evaluated for each tube assuming the measured sIRM. In the following, we set r_o to be 1 μm and draw 1000 flocs from log normal distributions of r with various means ($\bar{r}$) and standard deviations (σ). The average settling

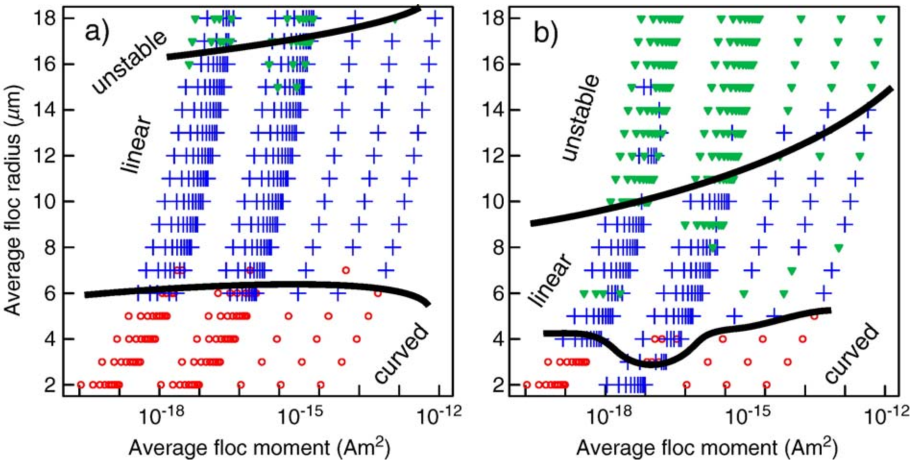

Fig. 8. Results of simulations using different magnetizations and floc distributions. a) Outcomes of many simulations using difference floc size distributions and initial magnetizations. All log normal distributions had a standard deviation of 1.5. "Unstable" results had R^2 values for the polynomial fit of <0.95. b) Same as a) but with standard deviations of 3.0.

length in our tubes is 0.3 m and the viscosity is that of water. Our preferred models and assumed parameters for the floc distributions are shown in Fig. 7a–d. Please note that the assumed floc distributions are entirely consistent with the measured ones shown in Fig 7e, but the latter are too poorly constrained to be used directly.

6.2. Is DRM linear with the applied field?

Fig. 7 shows that we can adequately account for the DRM results using a simple model for floc formation and very few free parameters (r_o, $\bar{r}$, σ). We take this to indicate that the model contains the essential physics in DRM acquisition in our settling tubes. One of the most surprising results of this investigation, from both theory and experiment, is that the relationship of DRM to the applied field may frequently be non-linear, even in the range of field values exhibited by the geomagnetic field.

The *prima facia* assumption in all sedimentary paleointensity studies is that DRM is a linear function of the applied field, yet none of our experiments show linear behavior. In Fig. 3b, we saw that larger floc sizes tended to have a more linear relationship with B than smaller ones. Small floc sizes were predicted to reach saturation in quite low fields. However, particle sizes that are too large are essentially randomly oriented. There is therefore a rather narrow range of particle sizes that can be expected to behave in a linear fashion with the applied field.

Given the importance of the linearity assumption to sedimentary paleointensity studies, we would like to explore numerically under which conditions DRM is likely to be quasi-linear with applied field. To this end, we performed two sets of numerical experiments. In each, we varied m_o, the magnetization of the fundamental floc. We drew log-normal distributions with varying $\bar{r}$ and σ. In the first set of simulations, σ was set to 1.5 and in the second set, it was set to 3.0. For each distribution, we calculated the DRM/sIRM for B ranging from 0 to 100 μT. We fit the simulated DRM/sIRM versus B data with both linear and fourth order polynomials. The ratio of the F statistic for the two fits was used to decide whether the "curved" fit was superior to the "linear" fit given the number of degrees of freedom (5 and 8, respectively) [we realize that an F statistic for simulated "data" has no statistical meaning, but serves as a convenient way of discriminating between essentially linear and essentially curved results].

In Fig. 8a and b we show the fields of "linear" and "curved" behavior as a function of the average composite floc radius and moment for the $\sigma=1.5$, $\sigma=3.0$ distributions, respectively. We also show results that are poorly fit by either model, being highly scattered ($R^2<0.95$); these are labeled "unstable". The results in Fig. 8 show that there is a narrow range of floc sizes exhibiting a quasi-linear relationship with the applied field for fields ranging from 0 to 100 μT. This range is more restricted if the particle size distribution is broader because of the dominance of flocs that are essentially randomly oriented.

6.3. The role of post-depositional remagnetization in the acquisition of DRM

It is likely that the results in this paper will be dismissed by some in the paleomagnetic community

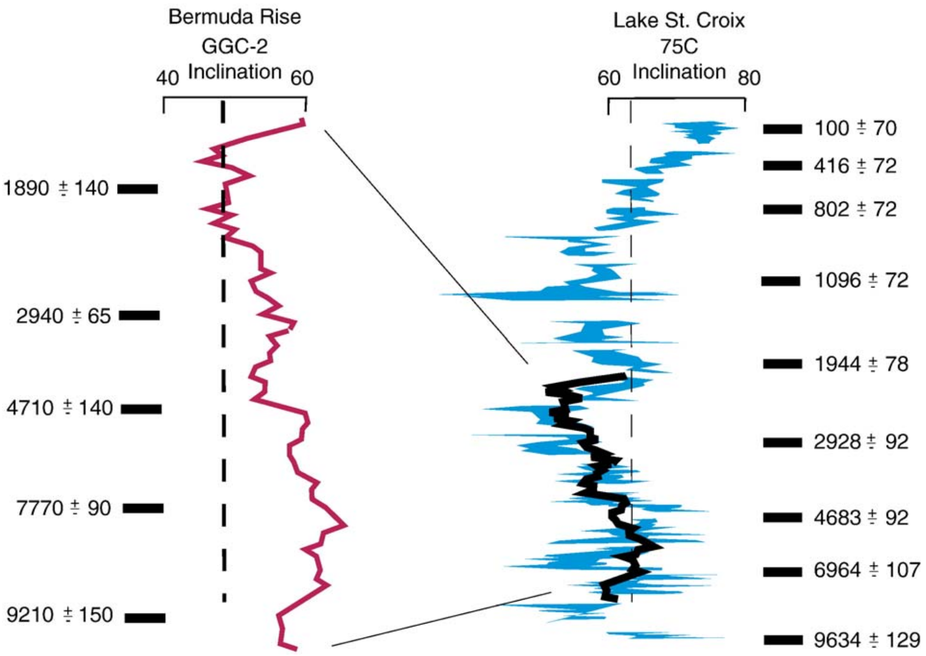

Fig. 9. Inclination and age data from Bermuda rise [34] (shown to the left) and Lake St. Croix (Lund and Banerjee [39]) (data including 95% shaded confidence bounds to right). Correlation using the age constraints in the original papers of Bermuda Rise data to Lake St. Croix shown as heavy line on Lake St. Croix data. Using an appropriate time scale obviates the need for 10–20 cm smoothing window suggested by [34].

because of a wide-spread misconception that DRM is not a sediment/water interface phenomenon, but is acquired many 10 s of centimeters deeper where the role of flocculation would be insignificant. It is therefore important to review the development of our understanding of DRM and assess relevance in the natural world of the theoretical and experimental results presented here.

The "standard model" of depositional remanence (DRM) acquisition that was articulated, for example, by Verosub [29] and Tauxe [5], is that detrital remanence is acquired by locking in different grains over a range of depths leading both to significant smoothing and to a significant offset between the sediment/water interface and the locking in of the remanence. Many practitioners of paleomagnetism adhere to this concept of DRM. This wide-spread belief in a deep lock-in of DRM stems from a series of laboratory redeposition experiments [16,2,30–32], and several oft-cited studies of natural sediments [33–36] all of which suggest a high degree of mobility of magnetic particles after deposition resulting in sedimentary smoothing and delayed remanence acquisition.

As pointed out by Katari et al. [37], the laboratory redeposition experiments were carried out using deionized water or water to which anti-coagulants were added. Therefore flocculation, which occurs ubiquitously in the marine environment, was inhibited and the results of these laboratory experiments are not applicable to DRM acquisition processes in marine sediments.

The paper that is most frequently cited in support of a deep lock-in depth (up to ~16 cm) for marine sediments is deMenocal et al. [33]. This paper assembled records from deep sea sediments with oxygen isotopes and the Matuyama–Brunhes Boundary (MBB) and suggested that higher sedimentation rate cores had younger appearing MBBs, consistent with a downward shift in the magnetic recording of the boundary of up to 16 cm. Tauxe et al. [38] updated the compilation with twice the number of records and, using the same logic as [33], they concluded that, on average, the magnetization is recorded within the top few centimeters. Recently, however, several papers have revived the deep lock-in debate (e.g., [11] and [36]). The former used a complicated lock-in model to explain results not observed anywhere else (substantial reversely magnetized intervals in apparently late Brunhes age equatorial sediments). Channell et al. [36] noted that in North Atlantic drift deposits, the mid-point of the MBB appears a few thousand years "younger" isotopically than the average age estimated by, for example, Tauxe et al. [38]. They suggested that this implied a deep lock-in. However, drift deposits result from the focussing of sediment from a large region. A particular bit of plankton will acquire its isotopic signature in the surface waters of the North Atlantic. When it reaches the bottom, it is transported via bottom currents (see, e.g., Figure 1 in Channell et al. [36]) until it finds a permanent home in the drift. It seems reasonable therefore to infer that the age offset between the isotopic and magnetic ages is in fact the time delay between the surface water and the ultimate deposition in the drift. A deep lock-in depth is not required if sedimentary particles spend time (1–2 kyr) in bottom currents, a reasonable hypothesis given the drift environment.

Another aspect of the deep lock-in notion is that different sized magnetic particles will be fixed at different depths, resulting in smoothing of the paleomagnetic record. The most quoted examples of significant smoothing in natural sediments are those of Lund and Keigwin [34] and Kent and Schneider [35]. Yet on close examination, these claims are also without foundation. Lund and Keigwin [34] proposed a smoothing interval of 10–20 cm in deep sea sediments to explain the apparent lack of correspondence of paleosecular variations (PSV) in cores from the Bermuda Rise with cores from Minnesota. They postulated that the PSV record of Bermuda Rise, western North Atlantic Ocean (see, e.g. Fig. 9, GGC-2) was systematically subdued with respect to the PSV recorded in Lake St. Croix (e.g. Fig. 9, 75C). They supposed that the inferred smoothing of the record stemmed from the observed difference in sediment accumulation rate, the Lake St. Croix record having been deposited at a rate several times that of the Bermuda Rise record. Lund and Keigwin [34] suggested that smoothing the Lake St. Croix data with a 10 or 20 cm moving average window reproduced the Bermuda Rise data with high frequency features smoothed out, and the amplitude of variation significantly reduced. However, they neglected the age constraints provided in the original St. Croix record [39] which are shown on Fig. 9. A substantially better fit of the Bermuda data, shown by the heavy line superposed on the Lake St. Croix data (shown with its 95% confidence bounds), is evident when the available age constraints are used. No smoothing is required to explain the small differences seen which are plausibly the result of the thousands of kilometers of separation between the two sites.

Kent and Schneider [35] showed three records of relative paleointensity across the MBB and interpreted these in terms of sedimentary smoothing. These records came from low and moderate sediment accumulation rates. Hartl and Tauxe [40] augmented the data base of peri-MBB relative paleointensity records using an additional ten records obtained from a wide range of sediment accumulation rates. They showed that the single low-sedimentation rate core of Kent and Schneider (V16-58) most probably had a poorly constrained time scale and that very little, if any, smoothing of sedimentary paleointensity records can actually be observed.

In summary, the laboratory experiments and results from natural sediments do not support a significantly delayed or heavily smoothed process of remanence acquisition in marine sediments. Therefore, the role of flocculation in marine sediments and the implications for methods of proper normalization must be taken seriously.

6.4. Normalization of sedimentary paleointensity records

The flocculation model of DRM has profound implications for the success of relative paleointensity studies. Because few, if any, natural sedimentary sequences are perfectly homogeneous through time, some form of normalization is essential to compensate for changes in the “magnetizability” of the sediments. There has been much discussion in the literature concerning the proper method for normalizing sedimentary remanences [7,41,8,5,42]. Most studies use some form of bulk magnetization such as magnetic susceptibility, anhysteretic remanence (ARM) or IRM. Many use elaborate demagnetization schemes aimed at targeting the normalizor to the magnetic grains carrying the remanence (e.g., [7,42]). The flocculation model, however, implies that the most important factors controlling which grains are responsible for the remanence is the size of the floc in which they are embedded and the length of time during which the flocs settle.

Floc size is controlled by salinity, pH, floc concentration and turbulence. In fresh water, the tendency to flocculate is low, but just a small amount of added salt results in very large changes in DRM for the same sediment and under the same field conditions (see Fig. 1). However, in water with NaCl concentrations greater than about 20 ppt (see [23,24] and Fig. 1) there is little effect of additional NaCl, so changing DRM effciency from variations in salinity is not an issue for marine conditions (salinities over ~30).

The effect of turbulence and particle concentration is most important in flocculating regimes, such as the marine environment (see [20]). As turbulence increases, flocs first tend to grow as the likelihood of chance encounters increases. However, at some point, turbulent shear stresses begin to tear flocs apart and the floc size will begin to decrease. Concentration of sediment in the

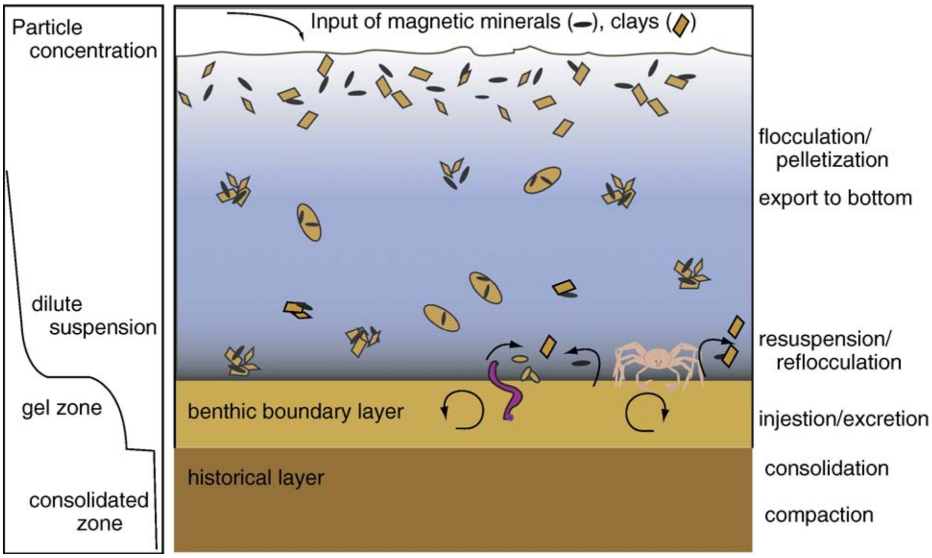

Fig. 10. Schematic drawing of marine sedimentary processes important to DRM acquisition. The left panel is a schematic of particle concentration. Flocs form in zones of high turbulence (surface zone) and high concentration (near the benthic boundary layer). Sediment in the benthic boundary layer is a stable gel unless it is disturbed by benthic currents or the action of the benthic fauna. Some fraction of the benthic boundary layer accretes to the historical layer. DRM is aquired when the gel zone is resuspended, allowing formation of new flocs which settle out over some time interval dependent on how high the resuspension was carried and how large the new flocs are [Little crab drawn by Genevieve Tauxe in animation at http://magician.ucsd.edu/Lab_tour/movs/DRM.mov].

water column also aids floc formation. Larger flocs fall faster, but when the concentration reaches a critical value in the "gel zone", further settling is inhibited until pore fluid can be expelled from the void spaces in the flocs. Constraining settling length is difficult.

DRM is probably not acquired during the long trip from the sea surface to the sea floor because the normalized DRM would be at saturation and not vary with field strength at all. Nor is DRM likely to be acquired in the "gel zone" of the benthic boundary layer where high concentrations of sediment mean that particles are immobilized by contact with neighbors unless there is some shear stress (from, e.g., bottom currents or bioturbation) suffcient to break the van der Waals forces holding them together. For the most part, therefore, it is likely that the magnetization is acquired when sediments are resuspended by bottom currents or bioturbation, perhaps settling through some tens of centimeters until they coalesce again in the benthic boundary layer.

We put these pieces together in the schematic diagram of marine sedimentary processes shown in Fig. 10. The benthic boundary layer is continually being stirred, either from benthic currents or from physical disturbance by creatures. When the sediment is resuspended in this manner, the particles will reflocculate depending on concentration and turbulent shear stresses. The sediment will acquire a DRM that depends on the settling distance and floc size distribution. Some (small) fraction of the sediment in the benthic boundary layer escapes the cycle of resuspension and joins what Katari et al. [37] termed the "historical layer", preserving the DRM acquired when it was last resuspended. Such a DRM may be linearly related to the applied magnetic field, if the floc size allows.

In the view of DRM predicted by our numerical simulations and confirmed by laboratory redeposition experiments, current methods of normalization can only give the crudest estimate of relative paleointensity. It will be quite difficult properly account for changes in DRM effciency because normalization using bulk rock magnetic parameters only compensate for changes in the amount of magnetic material in the sediment and not the size of the flocs in which it is embedded. Moreover, prediction of the equilibrium floc size distribution is extremely complex and will be quite difficult to ascertain from evidence left in the sedimentary record. On the other hand, it may be possible to properly normalize the DRM, if the benthic processes controlling flocculation are approximately invariant with time. Nonetheless, the very large scatter in relative paleointensity data, even in records from nearby cores, may well be the result in the inherent variability in the DRM process. It seems that given the strong dependence of DRM effciency on factors other than the applied magnetic field, only records that have been replicated with nearby cores with a high degree of coherence can be reasonably interpreted as preserving the meaningful amplitudes of (relative) paleointensity variations.

7. Conclusions

- Published theories for DRM acquisition fail to explain the laboratory redeposition data on natural marine sediments under highly controlled conditions. A flocculation model of DRM in which composite flocs are built from fundamental flocs with randomly oriented magnetic moments, however, can explain the data very well.
- Both the flocculation model of DRM and our laboratory redeposition experiments suggest that DRM may often behave in a significantly non-linear fashion with respect to the applied field, even in the range of the Earth's magnetic field.
- DRM is strongly affected by average floc size, which in turn is a strong function of salinity for low salinity waters. Therefore, in low salinity environments such as lakes, relatively small changes of salinity can result in large changes in magnetization.
- A review of the literature finds no compelling support for significantly delayed remanence acquisition through physical re-alignment of magnetic particles well below the sediment water interface in marine sediments; hence the role of flocculation in the acquisition of DRM may be important in marine environments.
- The composite floc model of DRM outlined in this paper suggests that current methods of normalizing DRM records for changes in magnetic grain size and concentration which do not account for changes in floc size will be only partially effective in isolating the geomagnetic contribution to changes in DRM. Changes in floc size and/or settling length may be the cause of the heretofore unexplained large scatter in relative paleointensity records.

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References

[1] E.A. Johnson, T. Murphy, O.W. Torreson, Pre-history of the earth's magnetic field, Terr. Magn. Atmos. Electr. 53 (1948) 349–372.
[2] D.V. Kent, Post-depostitional remanent magnetization in deep-sea sediment, Nature 246 (1973) 32–34.
[3] P. Tucker, Stirred remanent magnetization: a laboratory analogue of postdepositional realignment, J. Geophys. 48 (1980) 153–157.
[4] L. Tauxe, D.V. Kent, Properties of a detrital remanence carried by hematite from study of modern river deposits and laboratory redeposition experiments, Geophys. J. R. Astron. Soc. 77 (1984) 543–561.
[5] L. Tauxe, Sedimentary records of relative paleointensity of the geomagnetic field: theory and practice, Rev. Geophys. 31 (1993) 319–354.
[6] J.-P. Valet, Time variations in geomagnetic intensity, Rev. Geophys. 41 (2003), doi:10.1029/2001RG000104.
[7] S. Levi, S.K. Banerjee, On the possibility of obtaining relative paleointensities from lake sediments, Earth Planet. Sci. Lett. 29 (1976) 219–226.
[8] J.W. King, S.K. Banerjee, J. Marvin, A new rock magnetic approach to selecting sediments for geomagnetic paleointensity studies: application to paleointensity for the last 4000 years, J. Geophys. Res. 88 (1983) 5911–5921.
[9] D. Dunlop, O. Ozdemir, Rock Magnetism: Fundamentals and Frontiers, Cambridge Studies in Magnetism, Cambridge University Press, 1997.
[10] D.G. McMillan, C.G. Constable, R.L. Parker, Limitations on stratigraphic analyses due to incomplete age control and their relevance to sedimentary paleomagnetism, Earth Planet. Sci. Lett. 201 (3–4) (2002) 509–523.
[11] U. Bleil, T. von Dobeneck, Geomagnetic events and relative paleointensity records: clues to high-resolution paleomagnetic chronostratigraphies of late quaternary marine sediments, in: G. Fischer, G. Wefer (Eds.), Use of Proxies in Paleoceanogrpahy: Examples from the South Atlantic, Springer-Verlag, 1999, pp. 635–654.
[12] G.L. Anson, K.P. Kodama, Compaction-induced inclination shallowing of the post-depositional remanent magnetization in a synthetic sediment, Geophys. J. R. Astron. Soc. 88 (1987) 673–692.
[13] T. Nagata, Rock Magnetism, Mazuren, Tokyo, 1961.
[14] D.W. Collinson, DRM in sediments, J. Geophys. Res. 70 (1965) 4663–4668.
[15] F.D. Stacey, On the role of Brownian motion in the control of DRM of sediments, Pure Appl. Geophys. 98 (1972) 139–145.
[16] E. Irving, A. Major, Post-depositional DRM in a synthetic sediment, Sedimentology 3 (1964) 135–143.
[17] W. Coffey, Y. Kalmykov, J. Waldron, The Langevin equation with applications in Physics, Chemistry and Electrical Engineering, Vol. 11 of World Scientific Series in Contemporary Chemical Physics, World Scientific, Singapore, 1996.
[18] V. Scherbakov, V. Scherbakova, On the theory of depositional remanent magnetization in sedimentary rocks, Geophys. Surv. 5 (1983) 369–380.
[19] H. van Olphen, An Introduction to Clay Colliod Chemistry, John Wiley and Sons, New York, 1977.

[20] J. Winterwerp, W. van Kesteren, Introduction to the Physics of Cohesive Sediment in the Marine Environment, Vol. 56 of Developments in Sedimentology, Elsevier, 2004.

[21] R. Lu, S.K. Banerjee, J. Marvin, Effects of clay mineralogy and the electrical conductivity of water on the acquisition of depositional remanent magnetization in sediments, J. Geophys. Res. 95 (1990) 4531–4538.

[22] M. van Vreumingen, The magnetization intensity of some artificial suspensions while flocculating in a magnetic field, Geophys. J. Int. 114 (1993) 601–606.

[23] M. van Vreumingen, The influence of salinity and flocculation upon the acquisition of remanent magnetization in some artificial sediments, Geophys. J. Int. 114 (1993) 607–614.

[24] K. Katari, L. Tauxe, Effects of surface chemistry and flocculation on the intensity of magnetization in redeposited sediments, Earth Planet. Sci. Lett. 181 (2000) 489–496.

[25] K. Katari, J. Bloxham, Effects of sediment aggregate size on DRM intensity: a new theory, Earth Planet. Sci. Lett. 186 (1) (2001) 113–122.

[26] W. Sun, K.P. Kodama, Magnetic anisotropy, scanning electron microscopy, and X-ray pole figure goniometry study of inclination shallowing in a compacting clay-rich sediment, J. Geophys. Res. 97 (1992) 19599–19615.

[27] A. Khelifa, P. Hill, Models for effective density and settling velocity of flocs, J. Hydraulic Res.

[28] R. Gibbs, Estuarine flocs: their size, settling velocity and density, J. Geophys. Res. 90 (1985) 3249–3251.

[29] K.L. Verosub, Depositional and postdepositional processes in the magnetization of sediments, Rev. Geophys. Space Phys. 15 (1977) 129–143.

[30] R. Lovlie, Post-depositional remanent magnetization in a re-deposited deep-sea sediment, Earth Planet. Sci. Lett. 21 (1974) 315–320.

[31] Y. Hamano, An experiment on the post-depositional remanent magnetization in artificial and natural sediments, Earth Planet. Sci. Lett. 51 (1980) 221–232.

[32] Y. Otofuji, S. Sasajima, A magnetization process of sediments: laboratory experiments on post-depositional remanent magnetization, Geophys. J. R. Astron. Soc. 66 (1981) 241–259.

[33] P.B. deMenocal, W.F. Ruddiman, D.V. Kent, Depth of p-DRM acquisition in deep-sea sediments — a case study of the *B*/*M* reversal and oxygen isotopic stage 19.1, Earth Planet. Sci. Lett. 99 (1990) 1–13.

[34] S.P. Lund, L. Keigwin, Measurement of the degree of smoothing in sediment paleomagnetic secular variation records: an example from Late Quaternary deep–sea sediments of the Bermuda Rise, western North Atlantic ocean, Earth Planet. Sci. Lett. 122 (1994) 317–330.

[35] D.V. Kent, D.A. Schneider, Correlation of paleointensity variation records in the Brunhes/Matuyama polarity transition interval, Earth Planet. Sci. Lett. 129 (1995) 135–144.

[36] J.E.T. Channell, J.H. Curtis, B.P. Flower, The Matuyama–Brunhes boundary interval (500–900 ka) in North Atlantic drift sediments, Geophys. J. Int. 158 (2) (2004) 489–505.

[37] K. Katari, L. Tauxe, King, A reassessment of post depositional remanenent magnetism: preliminary experiments with natural sediments, Earth Planet. Sci. Lett. 183 (2000) 147–160.

[38] L. Tauxe, T. Herbert, N.J. Shackleton, Y.S. Kok, Astronomical calibration of the Matuyama Brunhes boundary: consequences for magnetic remanence acquisition in marine carbonates and the Asian loess sequences, Earth Planet. Sci. Lett. 140 (1996) 133–146.

[39] S.P. Lund, S.K. Banerjee, Late quaternary paleomagnetic field secular variation from two Minnesota lakes, J. Geophys. Res. 90 (1985) 803–825.

[40] P. Hartl, L. Tauxe, A precursor to the Matuyama/Brunhes transition — field instability as recorded in pelagic sediments, Earth Planet. Sci. Lett. 138 (1996) 121–135.

[41] D.V. Kent, Apparent correlation of paleomagnetic intensity and climatic records in deep-sea sediments, Nature 299 (1982) 538–539.

[42] L. Tauxe, T. Pick, Y.S. Kok, Relative paleointensity in sediments; a pseudothellier approach, Geophys. Res. Lett. 22 (1995) 2885–2888.