



## Full vector model for magnetization in sediments

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### ABSTRACT

Sediments provide a continuous record of past geomagnetic field variations. Although it is theoretically possible to get both the direction and intensity of the geomagnetic field from sediment records, the mechanism is not fully understood. Previous workers have postulated that flocculation plays an important role in detrital remanent magnetism (DRM). Flocs are porous, loose and highly fragile aggregates of microscopic clay particles and their behavior in a viscous medium is likely to be different than single particles of magnetic minerals. In order to understand the role of flocculation in sediment magnetization, we carried out a set of redeposition experiments at different field intensities and a quasi-constant field inclination of 45°. We present here a simple numerical model of flocculation, incorporating both magnetic and hydrodynamic torques to explain the experimental data. At small floc sizes DRM acquisition is likely to be non-linear in field strengths comparable to the Earth's, but the sediments may be able to record the directions accurately. With increasing floc sizes sediments may retain a record of the intensity that is linearly related to the applied field or a direction parallel to the applied field, but are unlikely to do both at the same time. Also, the majority of the magnetic particles in the sediments may not be contributing significantly towards the net DRM and any bulk normalizing parameter may be unsuitable if the depositional environment has changed over the depositional period.

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### 1. Introduction

Although it is generally accepted that sediments record past variations of the geomagnetic field, there is still no consensus on how sediments get magnetized. The theory of depositional remanent magnetism (DRM) was initially laid out by Nagata (1961) in which individual magnetic grains align themselves with the magnetic field while settling in a viscous medium at low Reynolds number. According to this model, assuming reasonable terrestrial field intensities and magnetic moment of the settling particles, the time for the population of magnetic particles to become substantially aligned with the field is of the order of a few milliseconds. This is true irrespective of field and mineralogy, so, sediments should show saturation remanance. Yet nearly all laboratory redeposition experiments, dating from the first by Johnson et al. (1948), exhibit a strong field dependence of DRM which is nearly linear in fields like the Earth's.

Another aspect of DRM relevant to magnetic recording of the Earth's field is the observation that the remanent inclinations are often anomalously shallow compared to the applied magnetic field. This too has been observed in laboratory redeposition experiments. The first explanation for this inclination "error" was by King (1955).

He proposed that magnetic grains could be divided into two populations; one which were plate-like and presumably would settle with their magnetic moments aligned in the horizontal plane and the other, which were spherical, would align perfectly with the field. The contributions of these two types of grains would give rise to a net shallowing of the inclinations. An alternative explanation was offered by Griffiths et al. (1960), who argued that having two distinct populations of grain shapes were unlikely. They proposed instead a model whereby each individual spherical grain would settle to the bottom where it would encounter a micro-landscape of crests and troughs owing to surface unevenness. If the particles rolled from an aligned position by a random angle there could be a net shallowing of the inclination. A third explanation involving sedimentary compaction was first proposed by Blow and Hamilton (1978) who found a dependence of inclination shallowing with reducing porosity. This model was further extended by Anson and Kodama (1987) who called upon a mechanical model in which the individual magnetic particles, attached to plate-like clay particles, rotate during compaction causing a net shallowing. Arason and Levi (1990) devised a suite of compaction induced inclination shallowing models in which they considered discrete rotation of individual grains in either a rigid or soft matrix and also rotation of magnetic grains attached to clay flakes.

Scherbakov and Scherbakova (1983) first recognized the importance of flocculation in DRM. They pointed out that microscopic clay and sub-micron sized magnetite particles acquire surface charges which make them stick together. In this view, magnetic grains settle

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embedded in loosely aggregated porous bodies of clay. The lower net moment of the flocs would explain the field dependence of DRM. The role of flocculation was further investigated by van Vreumingen (1993a,b). With the help of an array of experiments he explored the role of sediment concentration, type, magnetic mineral concentration on DRM intensity and inclination. He demonstrated a dependence of intensity and inclination shallowing on increasing salinity (which controls the degree of flocculation). At very low salinities, there was only a moderate inclination error. The inclination error increased with increasing salinities. However, intriguingly, at higher salinities, the inclination error began to decline while the intensities remained low. Although the experiments were insightful a number of interpretations were ambiguous. The author was unable to explain the rise of inclination shallowing with a drop in intensity at moderate salinities stating that the phenomenon was “incompletely understood”. More importantly, although the author suggested flocculation to be the driver behind the observed variations in intensity and inclination, a coherent model was not suggested. One of the current authors developed a numerical model for the effect of flocculation on remanent intensities at a range of different field intensities and came to the conclusion that below a critical diameter magnetization in sediments is likely to be non-linear (Tauxe et al., 2006).

So far DRM models incorporating flocculating particles have not been able to quantitatively address both intensity and inclination variation with changing floc size. In this paper we present a series of redeposition experiments with different floc size distributions and field intensities. We confirm the experimental results of van Vreumingen that inclination shallowing can be partly depositional and can vary with the degree of flocculation. We extend the numerical modeling approach of Tauxe et al. (2006) to include processes (viz, hydrodynamic torques) which could give rise to the observed inclination shallowing. Further, with the model we aim to explain some of the ambiguities of earlier studies and make broad predictions about the suitability of depositional environments for paleomagnetic studies. To avoid confusion further on we define DRM strictly as the remanence acquired during the settling of sediments and is not to be confused with post-depositional remanent magnetism (pDRM) which we conceive as the remanence the sediment acquires after it has completely settled, in the presence of an external field due to rotation of individual magnetic particles.

## 2. Methods

We followed the recipe of van Vreumingen (1993a) for creating a synthetic mixture of sediment: we combined kaolinite and illite in the ratio of 2:1. Partially oxidized magnetite powder (W4000; 0.08% by weight) with an average diameter of 50 nm was added to the mixture. The mixture was crushed thoroughly in a rock crusher to ensure homogenization and thorough dispersal of the maghemite particles. A total of 12 samples of 0.9 g of the mixture were given a saturation isothermal remanance (sIRM) in a 700 mT impulse field. Uniformity of the sIRM values ( $11.85 \pm 1.5 \mu\text{AM}^2$ ) ensured between sample homogeneity. The samples were mixed with 300 ml of water and 4 ml of 0.1 M sodium pyrophosphate was added to the mixtures to ensure a deflocculated initial state. van Vreumingen varied salinity to create a range of floc sizes. In our experiment, sodium chloride was added to the sediment slurries to obtain a range of salinities from 0 to 20 ppt in the 12 tubes. Before each settling experiment, the sediment slurries were shaken vigorously for 10 min.

Three pilot studies were done with all 12 tubes in vertical fields of 10, 30 and 60  $\mu\text{T}$ . The tubes were placed in a solenoid generating a uniform field. The settling experiments were carried out inside a 1 m diameter  $\mu$ -metal sheath to cancel external fields. After letting the slurry settle for two weeks, the tubes were carefully taken out and inserted into a CTF three axes cryogenic magnetometer housed in the

magnetically shielded room at Scripps Institution of Oceanography in order to measure their remanence.

Fig. 1 shows the sIRM normalized remanent intensities for the twelve tubes after settling in three different fields. The intensity variations replicate those obtained by van Vreumingen (1993a) in that there is an initial rise in DRM intensity with increasing salinity followed by a pronounced drop in intensity for salinities in excess of about 2 ppt. van Vreumingen argued that isolated magnetic particles would tend to clump together suppressing the net magnetization. Increasing salinity of the solution resulted in increased tendency to flocculate which prevented the magnetic grains from clumping together. He explained the initial rise in DRM intensity with salinity as a result of the magnetic grains becoming increasingly attached to clays, instead of to each other, which would increase the net magnetization. In our experiments, however, we found that the tubes with salinities between 0 and 2.5 ppt were not fully settled and remained cloudy even after two weeks. Hence it is also possible that the rise in intensity was the result of inaccurate normalization. Because of the ambiguity in interpretation, we use results from only fully settled tubes (salinity of 0.3 ppt or more).

To assess the role of post-depositional rotation of magnetic grains (pDRM), we reversed the vertical field direction and measured the intensity after 12 h. Comparison of the results shows that pDRM was negligible in these experiments (Fig. 1).

Our next experiment consisted of placing six tubes with salinities ranging from 3 ppt to 20 ppt in a two axis Helmholtz coil placed within a  $\mu$ -metal sheath. Care was taken to ensure minimum field variations across the tubes. To further offset the effect of field variations, the tubes' positions with respect to the center of the Helmholtz coil were changed between experiments. Experiments were conducted in three fields ( $28.21 \mu\text{T} \pm 0.21$  inclined at  $47.22^\circ \pm 0.68$ ,  $44.10 \mu\text{T} \pm 0.25$  inclined at  $46.41^\circ \pm 1.31$  and  $57.55 \mu\text{T} \pm 0.37$  inclined at  $44.58^\circ \pm 1.42$ ).

Low salinity tubes (marked with smaller, darker circles) exhibit a non-linear field dependence while high salinity tubes are more linear (Fig. 2a). Increasing salt content helps in flocculation and the remanent intensities drop markedly (Fig. 2b). The inclinations show a concomitant drop (increasing inclination error) and subsequently bounce back at higher salinities (Fig. 2c).

The experimental data echo those from a single field intensity experiment observed in van Vreumingen (1993a). We have used a clay composition and salinity range very similar to his experiments and observe a similar drop of intensity in the 4–6 ppt range. This is particularly important because our experimental setup differs

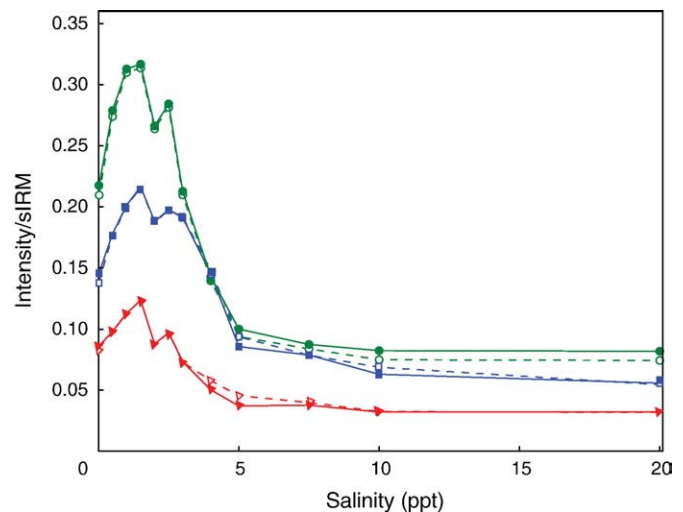
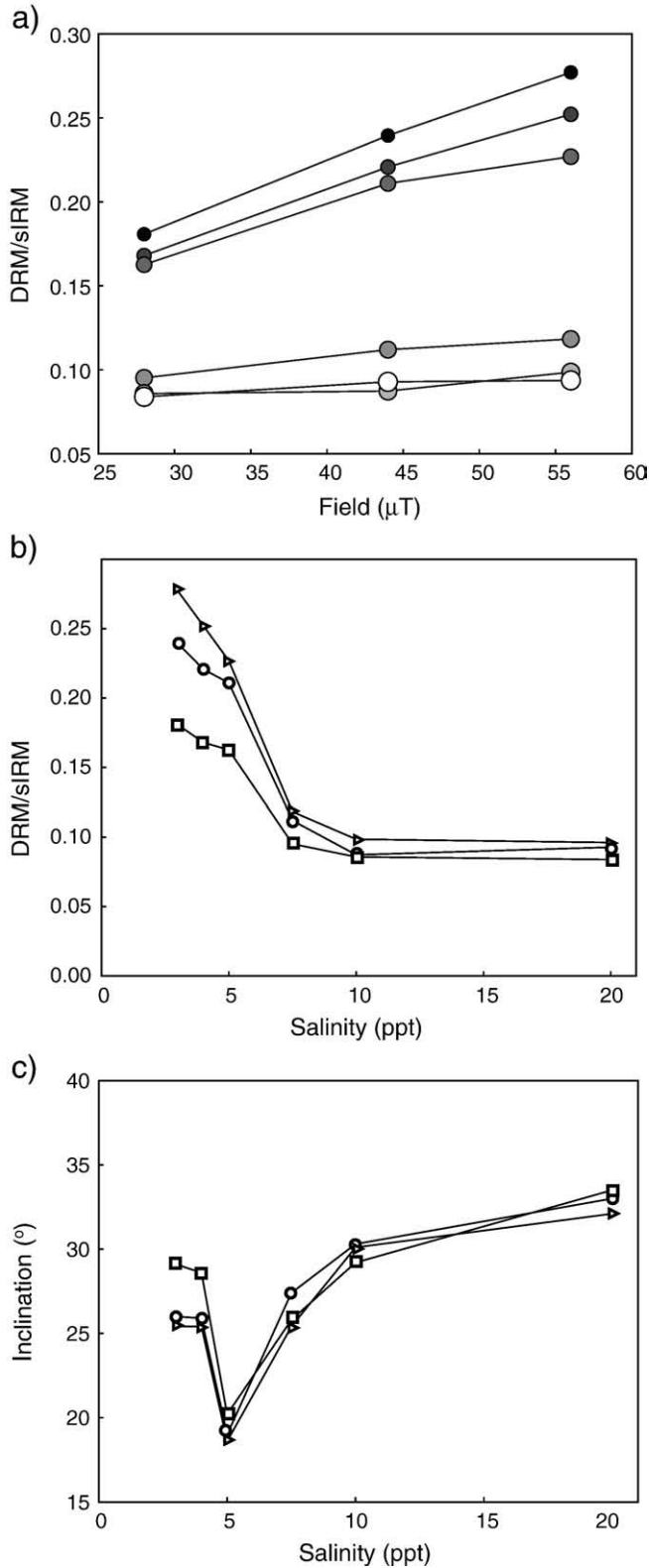


Fig. 1. Normalized intensities from twelve tubes in vertical fields of 10  $\mu\text{T}$  (red triangles), 30  $\mu\text{T}$  (blue squares), 60  $\mu\text{T}$  (green circles). Open symbols are the remanences measured after reversing the field direction for 12 h. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)



**Fig. 2.** Experimental data. a) Normalized intensity as a function of the applied field. Smaller, darker circles indicate lower salinities. b) Normalized intensities as a function of salinity. Squares, circles and triangles indicate field strengths of 28.21  $\mu\text{T}$ , 44.10  $\mu\text{T}$  and 57.55  $\mu\text{T}$ . c) Corresponding inclinations as a function of salinity.

substantially from his. We have used 50 cm long settling tubes while his experiments were conducted in 2 cm plastic cubes. Therefore, the observations of intensity and inclination dependence on salinity are independent of exact experimental setup.

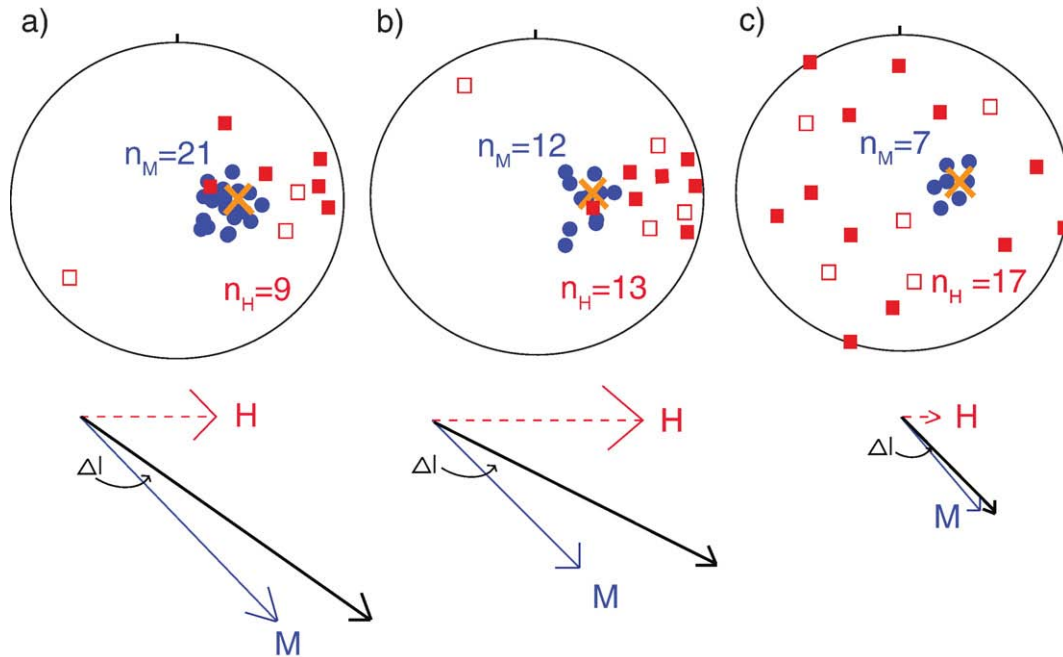
We performed the settling experiments in three field strengths; our results suggest a possible (weak) field dependence of inclination shallowing (inclinations are slightly shallower at higher field strengths). However, this apparent field dependence of inclination shallowing could possibly be ascribed to the fact that the experiments were conducted at slightly different inclinations, with the stronger field strengths having slightly shallower field inclinations. We also note that DRM declinations tracked the field accurately, in keeping with earlier studies. In the following section we formulate a physical model to explain the key observations.

### 3. Model

Flocculated particles are hydrodynamically different from isolated particles. They are porous, loose and highly fragile. Instead of being perfectly spherical as has been hypothesized in models exploring flocculation as an explicit control on DRM (Katari and Bloxham, 2001; Tauxe et al., 2006), they tend to have highly irregular shapes. Studies of rigid ellipsoidal particles have shown that they exhibit complex motion while settling in the fluid column (Field et al., 1997; Belmonte et al., 1998). Although flocs are not rigid ellipsoidal particles it is useful to attempt to model their settling behavior as stemming from slight departures from sphericity. Heslop (2007) attempted such a study and showed that with increasing floc sizes the hydrodynamic torque would increase at a much higher rate than the magnetic torque and for a prolate ellipsoid with a particular aspect ratio there is a critical size beyond which flocs would tend to be dominated by hydrodynamic torques. A different approach was taken by Jezek and Gilder (2006). They considered remanence acquisition of ellipsoidal magnetic particles in a gently creeping viscous flow, conditions which are likely to be found in continental margins and slopes. While the two studies are complimentary to each other, Heslop (2007) showed that even in quiescent and non-sloping conditions like open ocean or lake interiors, the role of hydrodynamic torques could be quite significant.

The proposed model incorporates Heslop's findings into the flocculation model of Tauxe et al. (2006). It is conceptually similar to the model proposed by King (1955) to explain inclination shallowing but differs substantially in the processes involved. Instead of assuming two distinct magnetic grain shape populations, our model separates a continuous distribution of floc sizes into two behavioral groups: one small enough to be responsive to *only* magnetic torques and the other big enough to be governed *chiefly* by hydrodynamic torques. The smaller flocs in the distribution (population *M*, hereafter as *M*) would be dominated by magnetic torques. Their net magnetic moment would be quasi-parallel with the applied field. Larger flocs (population *H*, hereafter as *H*) would be more influenced by hydrodynamic torques. Flocs in *H* first attain hydrodynamic stability and subsequently align with the magnetic field trying to maintain its hydrodynamically stable situation. Therefore, the net declination of *H* would track the field azimuth, but the net inclination would be near zero. The resultant of moments from *M* and *H* could give rise to the observed DRM (Fig. 3). The reader is cautioned that in reality a floc is not expected to show a rotation scheme as envisaged in the model. Instead it is expected to follow a complicated trajectory under the simultaneous influence of magnetic and hydrodynamic torques. But if we consider a large ensemble of flocs then such an approach would give us an 'average' value by ironing out the inconsistencies.

Assuming a distribution of floc sizes, at low salinities (Fig. 3a), the fraction of flocs in *M* is greater than that in *H* because low salinity inhibits flocculation thus keeping the floc sizes small. Most of the flocs align with the field and this results in high DRM intensities and limited inclination shallowing. With increasing salinity (Fig. 3b) the average floc size increases and the contribution of *H* becomes significant, resulting in a net shallowing of the inclination and a reduction in the net moment. With further increase in salinity, net contribution of *H* is strongly reduced (Fig. 3c); their moments are



**Fig. 3.** Schematic of equal area projections of individual floc moments showing the intensity and inclination dependence with increasing floc size. Blue circles represent  $M$  and red squares represent  $H$ . Solid and open squares represent moment directions in the lower and upper hemispheres respectively (see text). Orange cross is the field direction. Arrows show the contributions from  $M$  (blue solid arrow) and  $H$  (red dotted arrow) and the resultant (thick black arrow). a) Tubes with low salinity have small floc sizes and majority of the flocs (blue circles) are dominated by magnetic torques. Very few flocs are dominated by hydrodynamic torques (squares). The horizontal component is thus small, giving a low inclination shallowing ( $\Delta I$ ). b) With increasing floc sizes more flocs are influenced by hydrodynamic torques resulting in a higher net horizontal component. This increases  $\Delta I$ . c) The floc sizes have become so large that most of the flocs are influenced by hydrodynamic torques and they cannot orient themselves as efficiently (as in b) with the field any more. This causes a very small net horizontal moment in the field direction and thus producing only a slight inclination shallowing. The net moment continues to decrease throughout (from a to c) because of less efficient alignment by increasingly bigger flocs. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

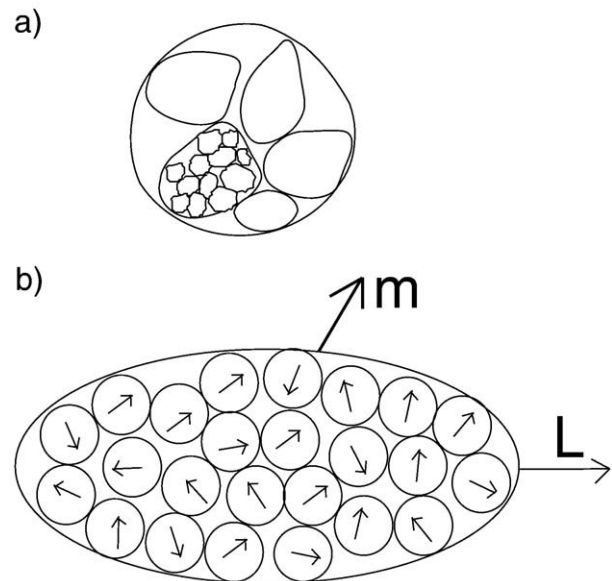
essentially randomly oriented, unable to align with the field at all. The randomization of  $H$  results in both a decrease in inclination shallowing and a reduction in strength of the net moment.

Our conceptual model involves three basic steps; building flocs, partitioning flocs into  $M$  and  $H$ , and finally rotating the flocs in response to the magnetic field. The key to a realistic yet simple numerical model involving such complex processes involves trade-offs in all the three steps. In the following sections we discuss the key assumptions and methods involved in each.

### 3.1. Floc building

Particulate clusters of clay sediments have a tendency to aggregate in a hierarchical fashion. Krone (1986) was the first to suggest a fractal nature of the flocs (Fig. 4a). Clay flocs also tend to have a very open ended structure with high water content. Processes giving rise to fractal floc shapes can be broadly categorized into Diffusion Limited Aggregation (DLA) and Cluster–Cluster Aggregation (CCA). In the former, individual particles execute Brownian motion to collide and stick on to one another. The latter adds one more step of complexity by assuming collisions of clusters instead of single particles. The basic building blocks of any DRM model involving flocculation are the flocs and past workers have approached this problem in a number of ways. The first numerical model dealing with flocculation was by Katari and Bloxham (2001). To build flocs they assumed a single magnetite particle within a larger spherical mass of clay. Tauxe et al. (2006) developed this model by building flocs with micron sized “micro-flocs” each of which had a magnetite embedded in it. Tauxe’s model was closer to Krone’s original thesis of hierarchical nature of flocs without explicit declaration of the processes leading up to such a structure. Shcherbakov and Sycheva (2008) designed very realistic floc structures by explicitly using the CCA and the DLA models of flocculation. Although realistic, such flocs are

difficult to incorporate within the framework of a DRM model because of the lack of studies of their hydrodynamic behavior. Keeping the foregoing in mind, the flocs in our models have been built as in Fig. 4b. Length of the long axis of each floc was drawn out of a lognormal



**Fig. 4.** 2-D cross-sections of flocs. a) Hierarchical structure of flocs as envisaged by Krone (1986). Three level of organization is shown. b) Flocs as envisaged in the current model. Small arrows show the individual moment directions of the magnetite grains embedded in  $1\ \mu\text{m}$  clay balls. Thick arrow shows the net moment ( $\mathbf{m}$ ) of the floc. Long thin arrow is the long axis ( $L$ ) of the ellipsoidal flocs.



distribution. The long axes of the ellipsoidal flocs were uniformly distributed over  $\theta = [-\pi/2, \pi/2]$ . It was assumed that the flocs were prolate spheroids with an axial ratio ( $p = b/a$ ) of 0.5. We further assume that the larger ellipsoidal floc is made up of many small spherical micron sized clay flocs each of which has a magnetic particle embedded in it. To calculate the number of such micro-flocs necessary to form the bigger flocs we assumed a packing density of 0.64, in keeping with random close spaced packing of spheres (Scott and Kilgour, 1969). A final consideration was given to keeping the total mass of magnetic minerals constant in all the simulations. In contrast, Tauxe et al. (2006) kept the number of flocs constant implying an ever increasing magnetic mineral population with increasing mean size of the distribution. This is physically unreasonable. Therefore in our model we have conserved total number of magnetic grains rather than the total number of flocs for each simulation. This translates to having fewer flocs with magnetic grains embedded in them as the mean size of the distribution increases. Given the high clay to maghemite ratio in the experiments we find this assumption to be a better approximation of the experimental conditions. Furthermore, we constrained the maximum number of maghemite grains available to build a floc to 500. Otherwise for distributions with sufficiently large means all the maghemite grains would be used up in building the first few flocs which can be argued to be unnatural. Raising the maximum limit would require more flocs to generate stable results without changing the model results. To calculate the net moment of each floc we use the expression by Rayleigh (1919) which gives the probability density function,  $F$ , of obtaining a resultant magnitude of  $R$ , from the summation of  $N$  randomly oriented vectors distributed uniformly in space.

$$F(R) \approx \frac{3\sqrt{6}R^2}{\sqrt{\pi N^3}} e^{-\frac{3R^2}{2N}} \quad (1)$$

The net moment directions of individual flocs were assumed to be oriented at a random angle with the floc's long axis (Fig. 4b).

### 3.2. Floc partitioning

The current model aims at understanding the average behavior of an ensemble of flocs. It does not take into account the actual trajectory a floc would take when acted upon by hydrodynamic and magnetic forces. This requires a partitioning of the entire population of flocs into  $M$  and  $H$ , which are dominated by either magnetic or hydrodynamic torques. To this end we first consider the hydrodynamic torque ( $\tau^H$ ) on a rigid, ellipsoidal particle (the flocs), with  $a$  as the principle axis and  $b = c$  as the semi-minor axes, in a stationary Newtonian fluid at low Reynolds number. An approximate value for this is given by the following expressions (Kuusela, 2005; Heslop, 2007):

$$\tau^H \approx \frac{16a^4 \rho_f^2 (V_t)^3 G}{\eta} \sin 2\theta \left[ \cos 2\theta \left\{ \left( \frac{\cos \theta}{X^A} \right)^2 - \left( \frac{\sin \theta}{Y^A} \right)^2 \right\} - \frac{\cos 4\theta}{2X^A Y^A} \right] \quad (2)$$

$$\times \sqrt{\frac{\sin^2 \theta}{(Y^A)^2} + \frac{\cos^2 \theta}{(X^A)^2}}$$

where

$$X^A = \frac{8}{3} e^3 \{ -2e + (1 + e^2)L \}^{-1},$$

and

$$X^B = \frac{16}{3} e^3 \{ 2e + (3e^2 - 1)L \}^{-1},$$

are the resistance functions of a prolate ellipsoid with

$$L = \ln \frac{1+e}{1-e},$$

where,

$$e = \sqrt{1 - \left( \frac{b}{a} \right)^2},$$

and  $\rho_f$ ,  $\eta$ ,  $V_t$  are the density, viscosity of water and the terminal velocity of a sphere with radius  $b$ .  $G$  is the geometric factor of Galdi and Vaidya (2001) and for  $p = 0.5$ ,  $G = -0.96$ .  $\theta$  is the angle the long axis of the floc makes with the vertical. The floc is stable (i.e.,  $\tau^H = 0$ ) when  $\theta = \pi/2$ . Eq. (2) is for prolate ellipsoids only. Although a similar expression exists for oblate ellipsoids no expression for the geometric factor,  $G$  is available.

The magnetic torque ( $\tau^M$ ) is given by:

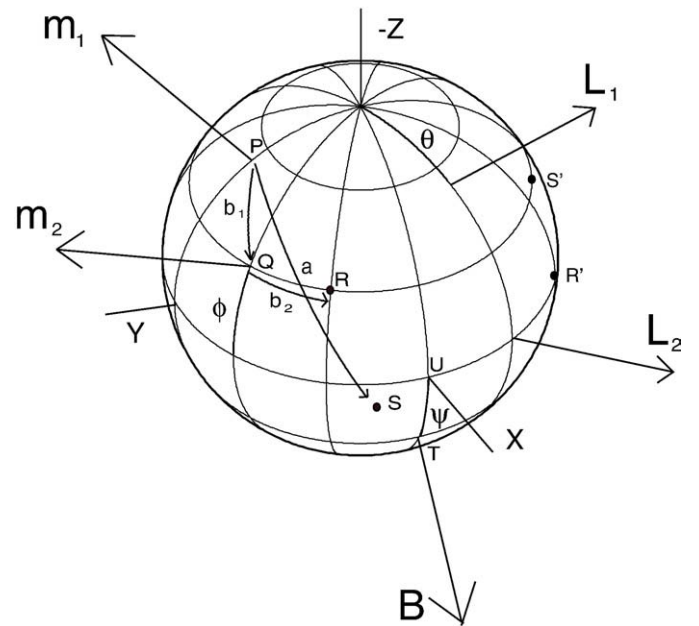
$$\tau^M = \mathbf{m} \times \mathbf{B}, \quad (3)$$

where  $\mathbf{m}$  and  $\mathbf{B}$  are the net magnetic moment and magnetic field vectors respectively.

The flocs are separated into  $M$  and  $H$  depending on the magnitude of the torques  $\tau^H$  and  $\tau^M$  at a random point during their settling. A valid point to consider would be whether the torques should be calculated for a random instance of the flocs or to compare the maximum of the two torques. Since we are looking at the ensemble behavior and not the precise trajectories of individual flocs we believe that either assumption is equally plausible for the purpose.

### 3.3. Floc rotation

If  $|\tau^H|$  is less than  $|\tau^M|$  then the magnetic torque dominate the floc's rotation and it changes its orientation along path a (Fig. 5). If  $|\tau^H|$  is greater than  $|\tau^M|$  then the floc is assumed to be in  $H$  and rotates from



**Fig. 5.** Coordinate system for the model.  $L_1$  and  $\mathbf{m}_1$  are the initial long axis and moment directions.  $L_1$  makes  $\theta = 90^\circ$  with  $Z$  axis.  $B$  is the field direction. Flocs dominated by hydrodynamic torques rotate from  $P$  towards  $T$  in a stepwise fashion.  $L_1$  goes to  $L_2$  and in response  $\mathbf{m}_1$  goes to  $\mathbf{m}_2$  (path  $b_1$ ).  $\mathbf{m}_2$  goes to  $R$  and in response  $L_2$  goes to  $R'$  (path  $b_2$ ). Flocs dominated by magnetic torques take the shortest path towards  $B$  (path  $a$ );  $\mathbf{m}_1$  goes to  $S$  and in response  $L_1$  goes to  $S'$ .

$L_1$  to  $L_2$ , to bring itself to the most stable orientation of  $\theta = \pi/2$ . The moment direction changes from  $m_1$  to  $m_2$  (path  $b_1$ ). The floc undergoes a further rotation due to the magnetic torque acting on it while maintaining its hydrodynamically stable position at  $\theta = \pi/2$  (path  $b_2$ ).

In order to ascertain the role of magnetic field on the flocs we start from the classic equation proposed by Nagata (1961):

$$I \frac{d^2\alpha}{dt^2} = -\lambda \frac{d\alpha}{dt} - \mathbf{mB} \sin \alpha \quad (4)$$

where  $\lambda$  is the viscosity coefficient of water and  $I$  is the moment of inertia and  $\alpha$  is the angle between the moment and the field directions. For non-spherical particles Eq. (4) becomes

$$I \frac{d^2\alpha}{dt^2} = -F_p \lambda \frac{d\alpha}{dt} - \mathbf{mB} \sin \alpha, \quad (5)$$

where  $F_p$  is the Perrin friction factor (Perrin, 1934). For prolate ellipsoidal particles (with an axial ratio of 0.5) we have used an average value of  $F_p = 1.2$ .

Neglecting the inertial term and assuming Gibbs' (1985) empirical relation between floc size and settling velocity we have  $v = 1.1r^{0.78}$ .

Eq. (4) can be solved as in Katari and Bloxham (2001);

$$\tan \frac{\alpha}{2} = \tan \frac{\alpha_0}{2} \exp\left(\frac{-\mathbf{mBl}}{8.8F_p \pi \eta r^{3.78}}\right) \quad (6)$$

where  $l$  is the length of the tube through which the flocs settle,  $r$  is the equivalent radius of the flocs,  $\alpha_0$  and  $\alpha$  are the initial and the final angle between the moment and the field directions.

For particles in  $H$ , constrained to lie in the horizontal plane for hydrodynamic stability, we have:

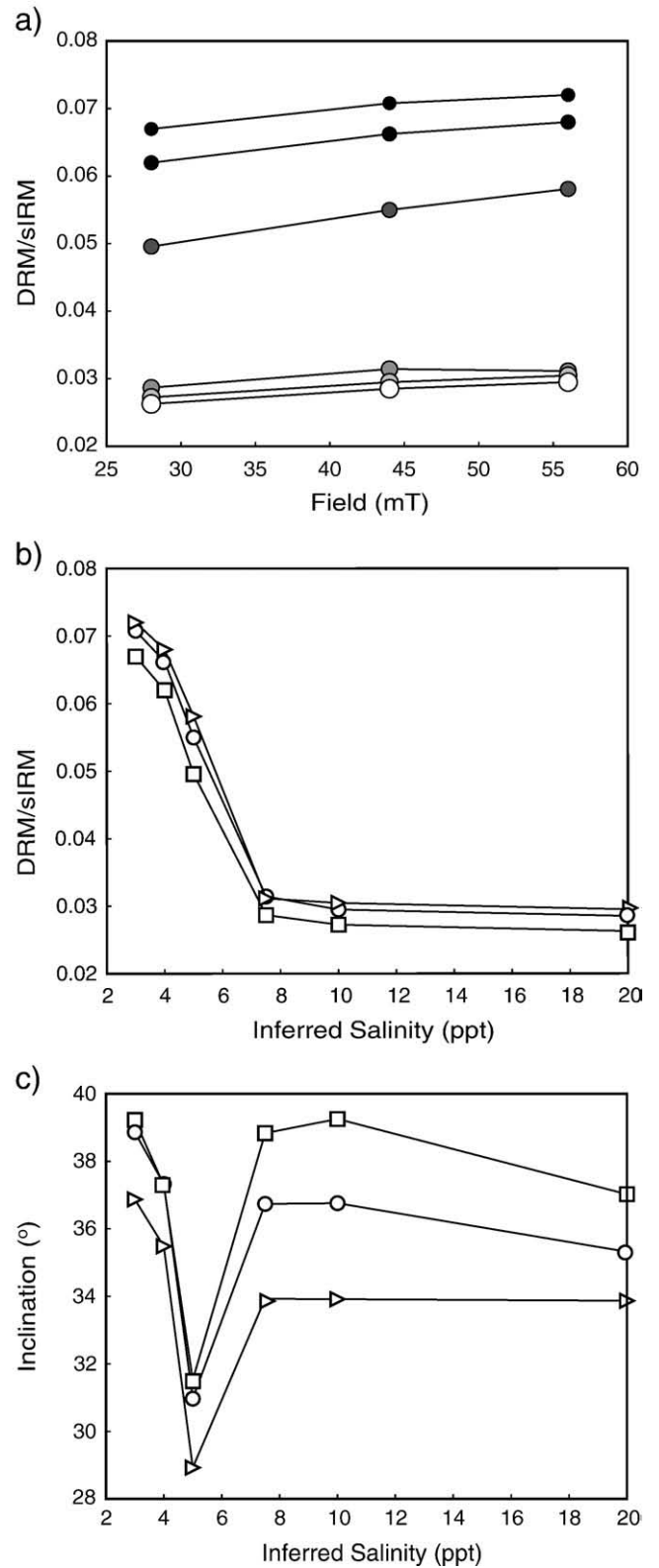
$$\tan \frac{\alpha}{2} = \tan \frac{\alpha_0}{2} \exp\left(\frac{-\mathbf{mB} \cos(\phi) \cos(\psi) l}{8.8F_p \pi \eta r^{3.78}}\right) \quad (7)$$

$\alpha_0$  and  $\alpha$  are the initial and the final angle between the horizontal components of the moment and the field directions.  $\phi$  is the inclination of the floc moment after  $b_1$  and  $\psi$  is the field inclination. The  $\cos(\phi)$  and  $\cos(\psi)$  terms are necessary because in this case we are considering rotation with the flocs long axis constrained to lie in the horizontal plane (Fig. 5). For a more detailed exposition of the governing equations the reader is directed to Tauxe et al. (2006).

#### 4. Results

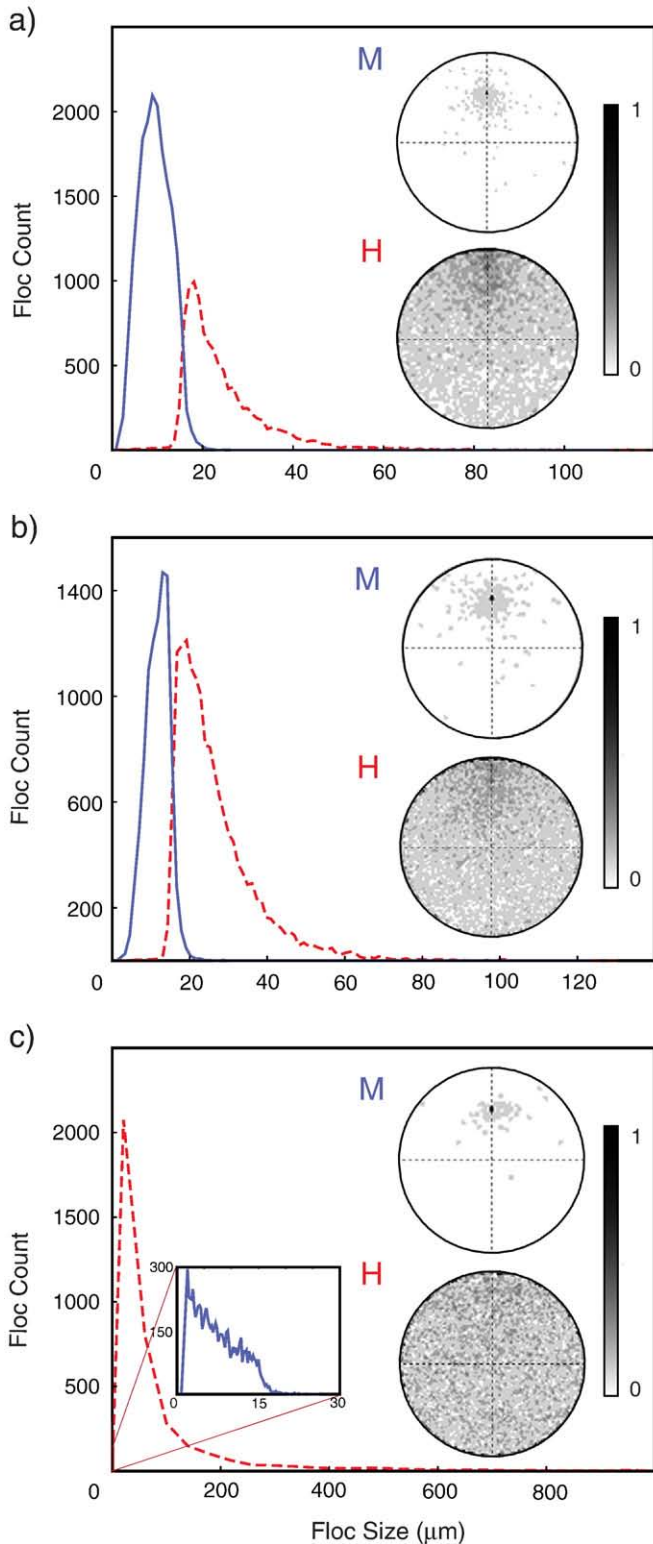
Our model replicates the primary characteristics of the data (Fig. 6). Smaller floc sizes show an increasingly non-linear rise of DRM intensity with field. Moreover, the distinct drop in intensity was accompanied by an increase in the degree of inclination shallowing. The declinations tracked the field azimuth in the experiments as well as the model.

In the model the floc size distribution plays an important role. In model instances with low mean floc sizes and low standard deviations we expect to see the majority of flocs to be dominated by magnetic torques. Also these flocs being small will be very efficiently oriented by the field. The few flocs that are dominated by hydrodynamic torques would show clustering towards the field azimuth because they are still small in size. The moments of these would form a cone around the field azimuth but their total number is not high enough to cause substantial shallowing in inclination (Fig. 7a). With increasing flocculation the mean floc size increases and a greater number of flocs are dominated by hydrodynamic torques. The number of flocs forming the cone increases and that causes the inclination shallowing (Fig. 7b). With very large floc sizes



**Fig. 6.** Modeled results with symbols having the same meaning as in Fig. 2. a) Normalized intensity as a function of applied field. b) Normalized intensity as a function of inferred salinity. c) Inclinations as a function of inferred salinity. For each salinity a lognormal distribution of floc size was assumed. In order of increasing salinities they were ( $\mu$ ,  $\sigma$ ) 2.4  $\mu\text{m}$ , 0.6; 2.5  $\mu\text{m}$ , 0.6; 2.8  $\mu\text{m}$ , 0.5; 3.5  $\mu\text{m}$ , 2.0; 3.6  $\mu\text{m}$ , 2.0; 3.7  $\mu\text{m}$ , 2.0.

most of the flocs are dominated by hydrodynamic torques but because of extremely large sizes there is very little alignment thereby reducing the inclination shallowing (Fig. 7c).



**Fig. 7.** Partitioning of flocs into *M* (blue solid line) and *H* (red dotted line) with a field of  $45 \mu\text{T}$  at  $45^\circ$  towards north for three different floc size distributions; a)  $\mu = 2.4 \mu\text{m}$ ,  $\sigma = 0.6$ , b)  $\mu = 2.8 \mu\text{m}$ ,  $\sigma = 0.5$ , c)  $\mu = 3.5 \mu\text{m}$ ,  $\sigma = 2.0$ . Semi-major axis of the floc is plotted on the abscissa. Insets show the corresponding equal area projections of floc moments for *M* (top) and *H* (bottom). No distinction is made between hemispheres and the plots are normalized by the maximum concentration. High (low) concentration is indicated by darker (lighter) colors and homogeneity (or lack of alignment) of moment direction is indicated by mid tones over the entire projection (as in c). Projections of *M* show small dark areas indicating that majority of moments are well aligned with the field direction. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

We note that although the model replicates the broad character of the intensity variations it is on an average between 2 to 3 times less efficient than the observed data. The reason for such high DRM intensities in the experiments is not understood fully but we propose that this might be because of the way flocs are actually built. In the model we have assumed that maghemite particles are completely randomly oriented within the flocs. This might not be true and the maghemite particles could show some kind of alignment while being incorporated within the flocs. For example, if more than one maghemite particle attaches to a single clay particle, these would be randomly oriented in a plane and not on a sphere as envisaged in the model. Furthermore the clay particles to which the maghemite particles get attached would definitely show some alignment because of their plate-like structures. Modeled inclinations show the same trends as the data although they seem to underestimate the shallowing ( $\sim 10^\circ$ ). The uniformly greater experimental inclination shallowing can be attributed to a flattening of the clay flakes upon deposition. In passing we note that any higher accuracy from a DRM model involving assumptions as made herein although possible is not desirable given the inherent deficiencies in the assumptions (e.g., flocs are not rigid ellipsoids as assumed here). Instead the model results should suffice to understand the relative importance of the processes involved in DRM acquisition and a starting point for more comprehensive models.

## 5. Discussion

An important question to ask is: “What is the likelihood of flocculation affecting the paleomagnetic record?”. Clay particles in water have surface charges on them which attract ions in solution. These build up a mixed double layer of cations–anions with their ratio changing away from the particle surface. Zeta potential is the electric potential in the double layer and is proportional to the width of the double layer or equivalently of the surface charge on the clay particles. With the addition of more salt there is an abundance of ions and the surface charge gets neutralized over a shorter distance which lowers the double-layer width and the absolute value of the zeta potential (Winterwerp and Kesteren, 2004).

By measuring the zeta potential we can estimate the repulsive power of clay layers. As the zeta potential approaches zero, the van der Waals attractive forces become dominant and clay particles will tend to flocculate. The lowering of the zeta potential can be brought about by addition of an electrolyte or by changing the pH of the solution. It has been observed experimentally that zeta potential in kaolinite varies from  $-20 \text{ mV}$  to  $-12 \text{ mV}$  when the salt concentration varies from  $10^{-4} \text{ M}$  to  $0.1 \text{ M}$  salt concentration at constant pH (Vane and Zang, 1997). The dependence of zeta potential on pH is even stronger than on salinity and a charge reversal is observed at  $\text{pH} = 2$  for kaolinite. Bentonite on the other hand shows much less variability with both salinity and pH.

The dependence of zeta potential with salinity and pH has important consequences for the structure of clay flocs. When charges on the negative clay surfaces are not fully neutralized but are weak enough to let an adjacent oppositely charged clay particle come close enough so that van der Waals forces become dominant, clays usually form porous house-of-cards type structures. In such a structure positive edges attach themselves to negative faces. With further lowering of the zeta potential towards zero, the surface charge becomes so low that even similarly charged faces can come in close contact and form dense aggregates of clay flocs (e.g., Olphen, 1977). Environments having a high salinity gradient (viz., estuaries) have shown an increase in floc sizes with salinity (Allersma, 1980; van Leussen, 1999).

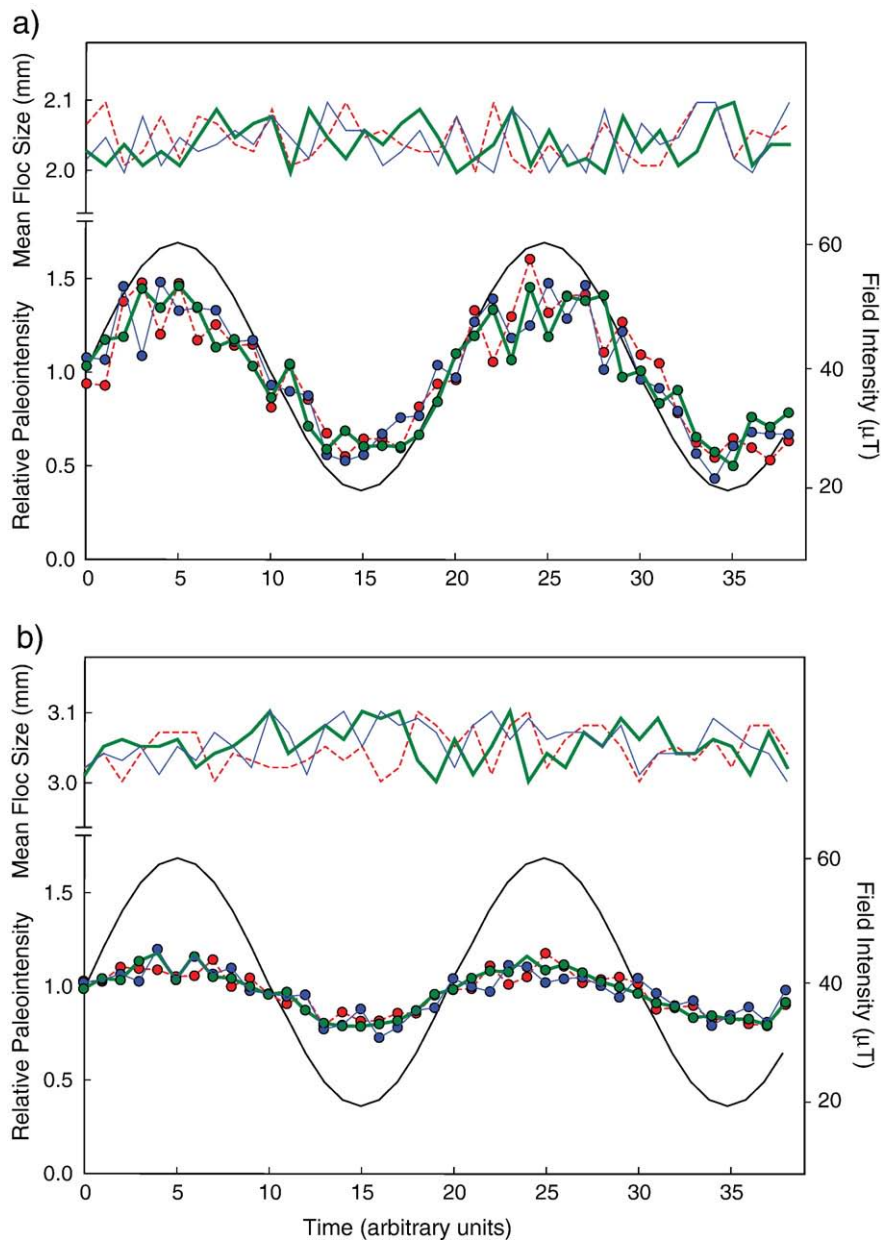
While sticking of particles due to reduction of surface potential is a likely mechanism for clay flocculation, binding of inorganic clays with adsorbed organic polymers (like polysaccharides) are equally important in nature. Polysaccharides are non-ionic and are ubiquitous in the

marine environment as they are produced by organisms like bacteria, algae, filter feeders, etc. They adsorb onto clay particles by strong bipolar forces which are much stronger than van der Waals forces. A long polymer string can attach itself to clay flakes at multiple locations and act as a bridge between such particles (Hunter, 2001; Winterwerp and Kesteren, 2004). Such union of inorganic and organic content of the sediment load gives rise to flocs which can be 10 to 100 times as strong as flocs made of purely inorganic material (Gregory, 1985). Therefore in the open ocean where salinity ranges are small, floc aggregation is likely to be a function of such particulate organic matter. Additionally, changes in sediment concentration and turbulence in the water column affect flocculation. Sediment concentration affects flocculation unidirectionally by promoting greater aggregation because of higher chance of mutual collisions (Dyer, 1989). Turbulence, on the other hand, affects flocculation in a contradictory manner. While turbulent motion promotes aggregation due to

increased number of collisions, turbulent shear breaks down larger flocs into their constituent particles (van Leussen, 1997).

Therefore we can see that a number of mutually independent factors contribute to the distribution of floc sizes observed in nature and it is likely that these factors change considerably across the spectrum of fluvial environments (from glaciomarine lakes to open oceans) and geological time scales to give rise to considerably different floc size distributions. This in turn would play a role in the paleomagnetic recording process (Lu et al., 1990; Katari and Tauxe, 2000; Tauxe et al., 2006).

In order to assess the role of floc size variation on magnetic recording in sediments, we use our model to explore hypothetical scenarios. We simulated two regions each experiencing the same magnetic field variations. The two regions had different mean floc radii, one small (from 2.0  $\mu\text{m}$  to 2.1  $\mu\text{m}$  and  $\sigma=0.3$ ) and one large (from 3.0  $\mu\text{m}$  to 3.1  $\mu\text{m}$  and  $\sigma=1.3$ ). Within each region we ran three simulations of magnetic recording by drawing populations of flocs



**Fig. 8.** Effect of floc size variation and sinusoidal field intensity fluctuations on sediments. The field intensity (black) was varied between 20  $\mu\text{T}$  and 60  $\mu\text{T}$ . Relative paleointensities, normalized by mean values are shown in dotted red, thin blue and thick green lines for the corresponding floc sizes (length of long axis) at the top. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)



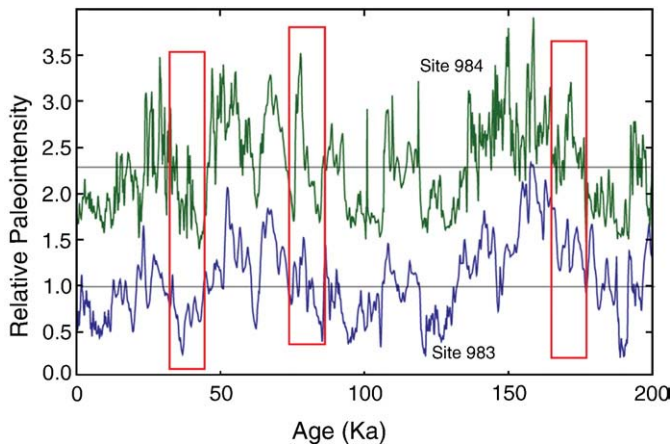
whose mean floc sizes were varied slightly and randomly through time (see mean floc size strips in Fig. 8a and b respectively).

We imposed an input magnetic field which varied sinusoidally between 20  $\mu\text{T}$  and 60  $\mu\text{T}$  to simulate geomagnetic field variation (see smooth black line in Fig. 8a and b corresponding to “field intensity” on right of plots). The inclination was kept constant at 45°. All six records “observed” the same magnetic field, but because of slight differences in floc size distribution, the remanent vectors generated by the model were somewhat different.

The relative paleointensity generally followed the input field variation but was also sensitive to floc size variation. In both scenarios we found that the floc size distribution introduced substantial fluctuations in the relative paleointensity records. The first order amplitude of the record became attenuated with increasing floc size (compare Fig. 8a with b) because larger flocs orient less efficiently with the field. The second order amplitude variations (the minor “wiggles”) decrease substantially because with increasing standard deviation the distribution becomes more positively skewed. The positively skewed distribution makes the proportion of  $M/H$  less variable with increasing mean floc size.

The appearance of minor “wiggles” due to variation of floc sizes has a direct consequence for natural systems. Relative paleointensities from globally distributed sites have been successfully used to build a global stack which is thought to represent the geomagnetic field variation (e.g., Guyodo and Valet, 1996, 1999). But neighboring cores, which should have similar variations in the geomagnetic field have been shown to differ significantly (see Fig. 9). The simulations show that tiny fluctuations in floc size could be the source of such a discrepancy although the magnitude of this fluctuation would depend on actual floc size distribution. Furthermore, small floc size systems would be prone to greater fluctuations with changing floc size. This might also explain the large scatter associated with paleointensity records from freshwater lakes because they are expected to have smaller mean floc sizes (Constable, 1985).

An important aspect of sedimentary paleointensity studies is the assumption of linearity of the remanence with respect to the applied field (Kent, 1973). It is evident from the experiments as well as from the model that this cannot be true for all floc sizes. Tauxe et al. (2006) predicted a narrow range of floc sizes where the DRM response to the field behaves linearly. With our model we can explore the problem further by incorporating the possibility of inclination error into the model: “Are there distinct regimes where the field is linear?”; “Are there regimes where we expect a low inclination error?”



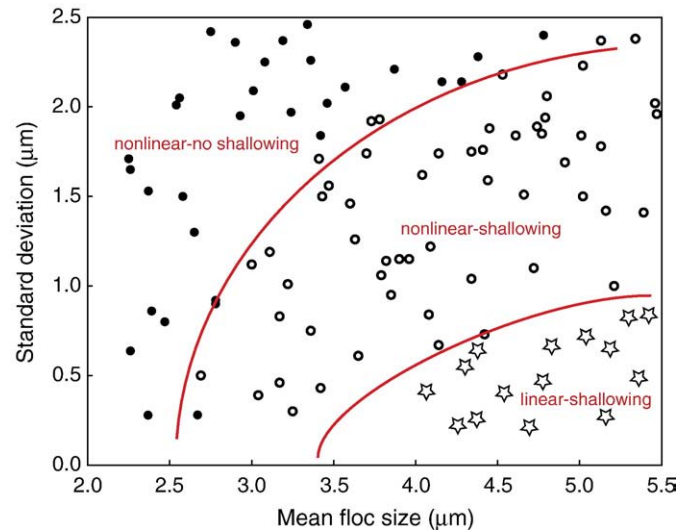
**Fig. 9.** Relative paleointensity (ARM normalized) record from Site 983 and the adjacent Site 984. Red boxes highlight areas showing subtle difference in amplitude and trend of the normalized paleointensity. Data from Channell et al. (1997, 1998, 2004). Figure modified after Tauxe and Yamazaki (2007). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

In order to “map out” the floc distributions whose DRM acquisition was essentially linear with applied field versus those that behaved with a significantly non-linear response, we use the extra sum-of-squares  $F$  test with a critical ratio of 1 and a  $p$  value of 0.05. In our simulation, we draw lognormal populations of varying floc size and standard deviations. For each population we calculated the normalized field intensities at fields ranging from 0 to 100  $\mu\text{T}$  at steps of 10  $\mu\text{T}$  and a constant inclination of 45°. The inclinations were calculated for a representative field of 40  $\mu\text{T}$ .

In general agreement with Tauxe et al. (2006), increasing mean floc sizes makes the DRM response more linear. At low mean floc sizes the flocs tend to have low inclination shallowing and DRM response is largely non-linear (filled circles). With increasing floc sizes the inclination shallowing increases (open circles). A perfectly linear DRM acquisition with field is possible but with a high degree of inclination shallowing (stars). Increasing standard deviation pushes the linear regimes to larger mean floc sizes. At very large standard deviation the DRM response remains largely non-linear (Fig. 10).

At very low salinities we expect to have smaller flocs. Freshwater lakes have salinities typically less than 5 ppt and can be expected to form small flocs. For such flocs the DRM acquisition curve is likely to be non-linear for a large range of standard deviation. It is thus unlikely that lake sediments would give a consistent record of past field variations. Flocculation processes in the oceans are more dominated by the organic content which is capable of producing very large flocs. Depending on the nature of the floc size distribution the sediments might record either no inclination shallowing or linear DRM acquisition (Fig. 10). However, it is unlikely that sediments would be able to capture true field intensities and directions simultaneously.

Both in the experiments and the model, DRM efficiency decreases dramatically as the salinity increases (Figs. 2b and 6b). This is similar to what we observe in natural sediments and is a direct consequence of flocculation; with higher salinities the flocs grow bigger and a higher fraction is dominated by hydrodynamic torques which do not align with the field direction, hence contribute little towards the net DRM. Even the fraction which is dominated by magnetic torques do not orient very well with the field direction. The resultant field



**Fig. 10.** Regimes showing linear/non-linear behavior of DRM intensity acquisition and inclination shallowing. Each point represents a simulation with the given mean size and standard deviation. For each point the simulation was run in fields ranging from 0 to 100  $\mu\text{T}$  at steps of 10  $\mu\text{T}$  at 45° to decide whether DRM acquisition is increasing linearly with the applied field. Corresponding inclination shallowing at field strength of 40  $\mu\text{T}$  was also calculated. Filled circles: non-linear DRM acquisition with  $<5^\circ$  inclination shallowing; Open circles: non-linear DRM acquisition with  $<5^\circ$  shallowing. Stars: Linear DRM acquisition with  $>5^\circ$ .

intensity is thus very low. Based on a similar reasoning, we predict that sIRM normalized paleointensity records from lakes should have a higher average efficiency than those from proximal marine records because sIRM overcompensates for the bulk amount of magnetic material present for larger mean floc sizes. This would provide an independent test for the model.

We come now to another important consideration – the choice of an appropriate normalizer (see, e.g., Tauxe, 1993). An ideal normalizer should account for the total volume of magnetic minerals contributing to the remanence and its efficiency. From our model we see that the total volume of magnetic particles reflected in bulk magnetic parameters like sIRM is but part of the answer. The distribution of floc sizes controls efficiency and is equally or perhaps even more important than bulk content of magnetic particles. For instance, if flocculation partitions higher concentrations of magnetite particles into bigger flocs then the horizontal component is likely to increase, giving rise to greater inclination shallowing. The contribution towards the net magnetization is much less because of their inability to fully align with the field. So, if mean floc sizes were to increase over geological time scales in a particular sedimentary basin, the normalized remanence might decrease in spite of no change in the field intensity or the magnetic content of the sediments.

## 6. Conclusions

A basic premise of our experimental setup was to control flocculation by varying salinity. Salinity variations are most likely to be important in freshwater lakes where a minor change in salinity could affect flocculation dynamics. Seawater salinity is largely constant (35 ppt) and salt concentration is unlikely to play a dominant role in the flocculation processes. More important processes which could affect flocculation in the open ocean include microbial glue coating inorganic particles, clay/carbonate ratio and bottom water turbulence. The conclusions of this study however, are independent of how flocculation takes place. Irrespective of how the flocs form, this study shows that the absolute size distribution of the flocs play an important role in DRM acquisition.

Our simple numerical model serves to capture the major trends in the experimental data but is not meant to be a comprehensive and/or predictive model for DRM acquisition. Instead, this model should be used as a starting point for more complicated models involving precise trajectories of flocs and more realistic redeposition experiment using natural sediments. In spite of the obvious disadvantages in constructing a realistic DRM model the current study shows that hydrodynamic torques could be an important factor in DRM acquisition and outlines the following caveats:

1. Flocculation can explain the full DRM vector obtained from laboratory redeposition experiments.
2. Minute variations in flocculation dynamics may explain the differences observed in paleointensity records from nearby cores.
3. Paleointensity estimates from some glacial or freshwater lakes which show very little flocculation can be difficult because the smaller mean floc sizes could lead to non-linear acquisition of remanence. On the brighter side, these sediments can be ideal recorders for paleosecular variation studies.
4. Marine sediments having large mean floc sizes and standard deviations; these could be suitable for paleointensity or paleosecular variation studies, but a particular region may not be suitable for both. If proper techniques for estimating past floc size distributions become available in the future we might be able to predict the suitability of sediment cores for a particular study.
5. Irrespective of how the magnetic fraction partitions with increasing floc sizes it is important to recognize that not all magnetic grains contribute equally to the remanence. Magnetic grains in large flocs are practically neutral to the ambient field. Therefore any bulk

parameter aimed at normalizing the data is susceptible to error. Depending on the variability of the initial depositional environment this could pose a challenge for interpretation of relative paleointensity records.

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