Quantifying magnetite magnetofossil contributions to sedimentary magnetizations

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A B S T R A C T

Under suitable conditions, magnetofossils (the inorganic remains of magnetotactic bacteria) can contribute to the natural remanent magnetization (NRM) of sediments. In recent years, magnetofossils have been shown to be preserved commonly in marine sediments, which makes it essential to quantify their importance in palaeomagnetic recording. In this study, we examine a deep-sea sediment core from offshore of northwestern Western Australia. The magnetic mineral assemblage is dominated by continental detritus and magnetite magnetofossils. By separating magnetofossil and detrital components based on their different demagnetization characteristics, it is possible to quantify their respective contributions to the sedimentary NRM throughout the Brunhes chron. In the studied core, the contribution of magnetofossils to the NRM is controlled by large-scale climate changes, with their relative importance increasing during glacial periods when detrital inputs were low. Our results demonstrate that magnetite magnetofossils can dominate sedimentary NRMs in settings where they are preserved in significant abundances.

1. Introduction

Magnetotactic bacterial magnetosomes are permanent nano-magnets that, when arranged in chains, provide a means for the bacteria to orient themselves using Earth’s magnetic field (Blakemore, 1975; Blakemore et al., 1980; Kirschvink, 1980a; Simmons et al., 2006). After death, these magnetically ideal single domain (SD) magnetosomes can be incorporated into the sedimentary matrix as magnetofossils. They then have the potential to contribute to sedimentary magnetizations if they maintain an existing alignment (or become aligned after burial) with the ambient geomagnetic field (Kirschvink, 1979; Stolz et al., 1986; Tarduno et al., 1998; Abrajevitch and Kodama, 2009) and if they avoid diagenetic dissolution through burial in anoxic environments (Karlin and Levi, 1983; Canfield and Berner, 1987). Their ideal SD size means that such magnetofossil-based magnetizations should provide a stable, but potentially nonlinear, record of geomagnetic field variations (Tauxe, 1993; Roberts et al., 2012).

Early studies identified magnetofossils in marine sediments and demonstrated that both chains and dispersed particles could carry stable laboratory-induced remanences (Kirschvink and Chang, 1984; Petersen et al., 1986; Stolz et al., 1986; Hesse, 1994). In some sediments, specific magnetofossil-rich horizons have elevated natural remanent magnetization (NRM) intensities with SD-like demagnetization characteristics, which suggests that magnetofossils contribute to their palaeomagnetic record (Tarduno et al., 1998; Abrajevitch and Kodama, 2009). These observations have led to the hypothesis that a biogeochemical remanent magnetization (BgRM), acquired by geomagnetically-aligned magnetosome chains preserving their orientation in the sedimentary record, can contribute to sedimentary NRMs. The consequences of BgRM acquisition would be especially important if the bacteria lived below the surface mixed sediment layer (e.g., Tarduno et al., 1998) and produced an NRM contribution that is offset from the palaeomagnetic signal carried by detrital particles. Alternatively, if magnetotactic bacteria lived in the water column or uppermost part of the sediment column, the post-mortem magnetofossil remains would be reoriented by both geomagnetic torques and a variety of sedimentary processes in the same manner as detrital magnetic particles. These magnetofossils would therefore contribute to a depositional (or postdepositional) remanent magnetization rather than a BgRM.

Testing the contribution of magnetofossils to the palaeomagnetic record has been a challenge until recently because of a lack of rock magnetic techniques that could be used to detect magnetofossils effectively. Instead, time-consuming magnetic extractions and transmission electron microscope (TEM) imaging were necessary to identify magnetofossil particles (e.g., Petersen et al., 1986; Hesse, 1994), which limits the number of samples that can be investigated.
Developments in magnetic “remote sensing” have revealed that magnetofossils are much more widespread in the geological record than was thought previously (Egli, 2004; Egli et al., 2010; Roberts et al., 2012). Mathematical unmixing of laboratory-induced remanence curves demonstrates that a variety of sediment types contain apparent magnetofossil components with narrow size distributions and coercivites consistent with magnetite (Kruiver et al., 2001; Egli, 2004; Abrajевич and Kodama, 2009). First-order reversal curve (FORC) measurements provide a joint measure of the coercivity and interaction field distribution for fine magnetic particle systems (Pike et al., 1999). Thus, FORC diagrams have high diagnostic power, and allow identification of different domain states of particles within mixed magnetic mineral assemblages (Roberts et al., 2000). FORC distributions from magnetosome chains have a clear SD signature with a wide distribution of coercivities, but minimal interactions (Pan et al., 2005; Chen et al., 2007; Carvalho et al., 2009; Roberts et al., 2012; Li et al., 2012). Egli et al. (2010) developed a high-resolution FORC measurement protocol that allows proper quantification of the distribution produced by non-interacting SD particles, the so-called “central ridge” in a FORC diagram. Magnetosomes are flux linked and in combination behave as a single elongated SD particle (Dunin-Borkowski et al., 1998; Muxworthy and Williams, 2006, 2009), so that intact magnetofossil chains will contribute to the FORC central ridge signature. High-resolution FORC distributions are, therefore, an important diagnostic tool for detecting magnetofossils.

While rock magnetic data can provide strong evidence for the presence of magnetofossils, their interpretation is not unique. TEM observations can support the presence of magnetofossils, which have specific compositions, crystal structures, a limited size spectrum and a restricted range of characteristic morphologies (Petersen et al., 1986; Tarduno et al., 1998; Kopp and Kirschvink, 2008). Although magnetic particle extraction and TEM imaging are time consuming, analysis of a small number of samples can provide sufficient evidence to interpret more rapidly acquired rock magnetic data in terms of magnetofossil identification. Using a combination of TEM and rock magnetic techniques, recent work has provided strong support for the common occurrence of magnetofossils in a variety of marine sediments (Roberts et al., 2011, 2012; Chang et al., 2012; Larrasoña et al., 2012; Yamazaki, 2012; Yamazaki and Ikehara, 2012).

Now that the experimental tools needed to identify magnetofossils are available, it is important to quantify their contribution to sedimentary NRM. In this study, we present results from a Quaternary marine sediment core in which magnetofossils appear to be ubiquitous and where their relative contribution to the NRM is primarily modulated by detrital input from the nearby Australian continent. When the relative magnetofossil abundance is high, the sedimentary magnetization becomes more SD-like in its characteristics, therefore providing strong support for a dominant contribution of magnetofossils to the NRM. With the assumption of a two end-member mixing system, the magnetofossil contribution to the overall sedimentary NRM can be determined as a function of age.

2. Geological setting

Core MD00-2361 (113°28.63′E, 22°04.92′S) was recovered ∼41 km off the coast of northwestern Western Australia at a water depth of 1805 m during the Marion Dufresne TIP 2000 expedition (Fig. 1). The summer climate of this region is dominated by the Australian monsoon, with large rainfall episodes causing rivers to flood and transport large volumes of sediment to the ocean (Gingele et al. 2001a). Satellite images have revealed sediment-laden river plumes extending 200–300 km offshore during the northern Australia wet season (Gingele and De Deckker, 2004). The position of core MD00-2361 is ∼160 km from the mouth of the Ashburton River (Fig. 1), in a location expected to receive suspended riverine sediment advected southward by the Leeuwin Current (Spooner et al., 2011, Fig. 1). During the winter months, northwestern Western Australia is dry and the rivers have a reduced carrying capacity, or in some cases they dry out completely (Gingele et al., 2001a). During such dry periods aeolian dust is transported eastward along the so-called Indian Ocean dust path and contributes to the terrigenous flux that reaches the Indian Ocean (Bowler, 1976; McTainsh, 1989; Hesse and McTainsh, 2003). The relative contributions of riverine and aeolian sediment fluxes that reach the Indian Ocean from northwestern Western Australia have been shown to vary on glacial–interglacial time scales. During glacial periods, northwestern Western Australia was relatively cold and arid, with increased aeolian fluxes and reduced riverine fluxes (Hesse, 1997, 2003). In contrast, interglacial periods were characterized by warm and wet conditions with increased river transport and reduced aeolian activity (Gingele et al., 2001b).

3. Materials and methods

Core MD00-2361 is 42 m long; we have analyzed the uppermost 16.5 m of the core in this study. We analyzed 11 continuous 1.5-m long u-channel samples from the upper portion of the core. A detailed late Quaternary palaeoceanographic reconstruction, based on the upper 13.6 m of core MD00-2361, has been published by Spooner et al. (2011).

Magnetic measurements were made at 1-cm intervals with a 2-G Enterprises narrow-access pass-through cryogenic magnetometer (Weeks et al., 1993) at the National Oceanography Centre, Southampton, UK. NRMs were demagnetized in 12 steps up to a maximum field of 100 mT with an alternating field (AF) demagnetizer that is arranged in-line with the magnetometer. NRM demagnetization data were analyzed with the UPMag software of Xuan and Channell (2009) and characteristic remanent magnetization (ChRM) directions were defined using the principal component analysis approach of Kirschvink (1980b). Isothermal remanent magnetizations (IRM) were imparted to the u-channel samples with a 2-G Enterprises off-line pulse magnetizer. The IRM at 900 mT is assumed to represent the saturation isothermal remanent magnetization (SIRM).

Subsamples were taken from the u-channels at 10 cm stratigraphic intervals, and were crushed gently and air-dried. Hysteresis and backfield demagnetization measurements were performed using a Princeton Measurements Corporation vibrating sample magnetometer at the Research School of Earth Sciences, Australian National University. First-order reversal curve (FORC) diagrams (Pike et al., 1999; Roberts et al., 2000) were measured for a selection...
demonstrates that the primary control on the NRM intensity is the (Fig. 2b) follow a similar pattern to SIRM variations (Fig. 2c). This a reliable record of geomagnetic field variations. NRM intensities observed in this portion of the core is not considered to provide of unconsolidated sediments during coring. The fluctuating signal TEM has an EDAX Phoenix retractable X-ray detector and a Gatan search School of Earth Sciences, Australian National University. The aged using a Philips CM300 TEM operated at 300 kV at the Re- Properties Measurement System (MPMS) housed at the Research magnetite particles (Özdemir and Dunlop, 2010). Low temperature measurements were performed using a Quantum Design Magnetic Measurements System (MPMS) housed at the Research School of Earth Sciences, Australian National University. Magnetic particles were extracted for TEM analysis using the procedure of Chang et al. (2012). Magnetic extracts were im- aged using a Philips CM300 TEM operated at 300 kV at the Research School of Earth Sciences, Australian National University. The TEM has an EDAX Phoenix retractable X-ray detector and a Gatan 1024 × 1024 CCD camera.

Spooner et al. (2011) published a planktonic oxygen isotope stratigraphy for the upper 13.6 m of core MD00-2361. For this study their oxygen isotope record was extended to 16.5 m using the same approach and mass spectrometer as described by Spooner et al. (2011). Additional measurements were also made where the original sampling resolution of Spooner et al. (2011) was comparatively low.

4. Results

ChRM inclinations through the majority of the studied interval of core MD00-2361 lie around the direction expected for a normal polarity geocentric axial dipole field (Fig. 2a). At a depth of ∼16.2 m there is a clear transition from reversed to normal polarity. A coherent ChRM inclination pattern is not observed in the uppermost ∼0.5 m of core MD00-2361 probably due to disturbance of unconsolidated sediments during coring. The fluctuating signal observed in this portion of the core is not considered to provide a reliable record of geomagnetic field variations. NRM intensities (Fig. 2b) follow a similar pattern to SIRM variations (Fig. 2c). This demonstrates that the primary control on the NRM intensity is the concentration and composition of magnetic minerals in the sedi- ment rather than palaeomagnetic field strength.

Comparison of the supplemented and extended planktonic oxy- gen isotope stratigraphy for core MD00-2361 with SIRM (Fig. 2c) reveals a low frequency variability that is consistent with that expected for glacial–interglacial cyclicity (Lisiecki and Raymo, 2005). Based on the large-scale oxygen isotope pattern, marine isotope stages can be assigned for core MD00-2361. On the basis of the oxygen isotope stratigraphy for core MD00-2361, the magnetic reversal at ∼16.2 m occurs within marine isotope stage (MIS) 19 and must correspond to the Matuyama–Brunhes boundary (Tauxe et al., 1996; Liu et al., 2008), thus the base of the studied sediment sequence is dated at ∼800 ka (corresponding to a mean sedimentation rate of ∼2 cm/kyr).

Hysteresis ratios for samples from core MD00-2361 lie within the pseudo-single domain (PSD) field in a Day plot (cf. Day et al., 1977, Fig. 3). Using an SD versus multidomain (MD) binary mixing line as a general guide (Dunlop, 2002a, 2002b), it appears that the sediments contain a mixture of SD and coarser grains. On the basis of the Day plot, glacial sediments at 3.15 m contain the finest overall magnetic mineral assemblage. A high content of non-interacting SD particles is confirmed at 3.15 m by a statistically significant central ridge in the high-resolution FORC distribution.
Fig. 4. (a) High-resolution FORC distribution (smoothing factor of 5) for glacial sediments from 3.15 m in core MD00-2361 (the black contour is the 0.05 significance level). The final high-resolution FORC diagram was obtained by stacking 18 measured FORC diagrams, each constructed from 297 FORCs. The sharp central ridge that dominates the diagram is indicative of non-interacting SD particles that reflect the presence of magnetite magnetofossils (Egli et al., 2010; Roberts et al., 2012). (b) TEM image of a magnetic mineral extract from 3.15 m. The image is dominated by magnetite magnetofossils with a variety of morphologies (the larger particles are non-magnetic contaminants, such as carbonates and clays). (c) Low-resolution FORC distribution (207 FORCs, smoothing factor of 5, with 18 repeat measurements stacked) from the same sediment sample with slight vertical spreading at low coercivities. This spread is most likely due to the presence of a minor coarse-grained detrital component.

Fig. 5. (a) High-resolution FORC distribution (smoothing factor of 5) for interglacial sediments from 1.25 m (the black contour is the 0.05 significance level). This FORC diagram was obtained by stacking 8 measured FORC diagrams, each constructed from 297 FORCs. The combination of a sharp central ridge and large vertical spreading indicates a mixed magnetic mineral assemblage with SD particles and coarser PSD/MD grains. The break in the region of significant values at negative $B_u$ values results from the inability of a second-order polynomial surface to fit the data across $B_u = 0$ (Egli, 2013). (b) TEM image of a magnetic extract from 1.25 m with large detrital particles and smaller magnetofossils, which is consistent with the interpretation of the associated FORC distributions. (c) Low-resolution FORC distribution (207 FORCs, smoothing factor of 5, stack of 4 repeat measurements) from the same sample, which confirms the structure of the high-resolution distribution.

(Fig. 4a). While a central ridge FORC signature is characteristic of magnetofossils it is not necessarily an unequivocal indicator of biogenic magnetite particles. TEM images of magnetic mineral extracts taken at 3.15 m demonstrate that the magnetic mineral assemblage is dominated by magnetofossils, both as discrete particles and as apparently collapsed and intact chains (Fig. 4b). The generally dis-aggregated or collapsed nature of magnetofossil chains is probably the result of the sample preparation procedure required to extract magnetic minerals from bulk sediment samples. Energy dispersive X-ray spectra analysis and selected-area electron diffraction performed on individual particles confirm a magnetite composition. The crystal morphology is also as expected for biogenic magnetite particles. Lower resolution FORC distributions place less emphasis on the central ridge and may reveal contributions from larger particles and magnetostatic interactions among particles. The lower resolution FORC distribution for the sample from 3.15 m has a slightly broadened central ridge at low $B_u$ values (Fig. 4c). This broadening could be due to collapsed chains, but given the depositional setting it is more likely to correspond to a minor contribution from coarser detrital (PSD and/or MD) grains.

In the Day plot, the coarsest overall magnetic mineral assemblages are found in interglacial sediments (Fig. 3). The sediments at 1.25 m are early Holocene in age and a high-resolution FORC distribution contains both a central ridge and significant vertical spreading (Fig. 5a). This combination of features indicates a mixed magnetic mineral assemblage containing both non-interacting SD particles and a substantial contribution from coarser PSD/MD grains (Roberts et al., 2000). TEM images of a magnetic mineral extract from a sample at 1.25 m support our interpretation that the FORC distribution reflects a mixture of magnetofossils and larger particles (Fig. 5b), as is also by the lower resolution FORC distribution, which has an essentially identical structure to the high-resolution distribution (Fig. 5c).

Zero-field cycling of a 2.5 T SIRM was performed on the sub-samples from 1.25 m and 3.15 m (Fig. 6). The Verwey transition is apparent in both samples (although it is more pronounced for the sample from 3.15 m), which supports the TEM and FORC evidence for the presence of magnetite magnetofossils in core MD00-2361. The weak nature of the Verwey transition in both samples does, however, indicate that the magnetofossils may be partially oxidized
the magnetic mineral assemblage (Fig. 2c). Studies covering shorter
from core MD00-2361 demonstrates a clear climatic control over
these periods indicate that the contribution of aeolian magnetic
dust reaching the location of core MD00-2361 during glacials, the low SIRM values observed during
interglacial periods, this detrital material must be relatively enriched in magnetic minerals. The
opposite situation occurred during glacial periods, with reduced
SIRM values. If aeolian dust was reaching the location of core
MD00-2361, (chocolate) brown color, while sediments deposited during MIS 11,
rich regolith. Sediments corresponding to MIS 1, 5, 7 and 9 have a
transport fine-grained material that originates inland from iron-
rivers that flow into the Indian Ocean from
northwestern Australia have brown colored water because they
et al., 2011). The rivers that flow into the Indian Ocean from
(Gingele et al., 2001b; Gingele and De Deckker, 2004; Spooner
ence of pigmenting riverine clays during each interglacial interval
(Ginge et al., 2001b; Ginge and De Deckker, 2004; Spooner
the cooling curve containing a strongly suppressed Verwey transition (marked as
time scales have shown that during interglacial periods, precipita-
tions increased over northwestern Western Australia and a greater
riverine sediment load reached the MD00-2361 site (Veeh et al.,
Comparison of the SIRM and planktonic oxygen isotope records
measurement data from this study can be accessed online via
PANGAEA (http://www.pangaea.de/).
5. Discussion
Comparison of the SIRM and planktonic oxygen isotope records
from core MD00-2361 demonstrates a clear climatic control over
the magnetic mineral assemblage (Fig. 2c). Studies covering shorter
time scales have shown that during interglacial periods, precipita-
tion increased over northwestern Western Australia and a greater
riverine sediment load reached the MD00-2361 site (Veeh et al.,
van der Kaars and De Deckker, 2002). This process is illustrated in the studied sediment core by the pres-
eence of pigmenting riverine clays during each interglacial interval
(Ginge et al., 2001b; Ginge and De Deckker, 2004; Spooner
The rivers that flow into the Indian Ocean from
northwestern Australia have brown colored water because they
transport fine-grained material that originates inland from iron-
rich regolith. Sediments corresponding to MIS 1, 5, 7 and 9 have a
(chocolate) brown color, while sediments deposited during MIS 11,
13, 15, 17 and 19 are a lighter beige color, which suggests a rel-
sive shift to greater riverine fluxes in more recent times. Given
the elevated SIRM values during interglacial periods, this detri-
tal material must be relatively enriched in magnetic minerals. The
opposite situation occurred during glacial periods, with reduced
precipitation and a lower detrital mineral flux resulting in dimin-
ished SIRM values. If aeolian dust was reaching the location of core
MD00-2361 during glacials, the low SIRM values observed during
these periods indicate that the contribution of aeolian magnetic
particles to the sedimentary NRM (i.e., particles in the stable SD to
MD range) was minor.
A scenario in which the magnetic properties of the sediments are dominated by magnetofossils and riverine detritus during
interglacial and interglacial periods, respectively, is consistent with
hysteresis, TEM, FORC and low temperature data presented in Figs. 3, 4, 5 and 6. Sediments from 1.25 m (interglacial) contain a mix-
ture of SD magnetofossils and coarser PSD/MD riverine particles,
while sediments from 3.15 m (glacial) are rich in magnetofos-
NRM with only a minor detrital magnetic mineral concentration.
The low temperature cycling data support this interpretation with a
more predominant Verwey transition at 3.15 m (glacial sedi-
ments) compared to 1.25 m (interglacial sediments). This two-end-
member mixing system, with an alternating pattern of glacial (rel-
atively magnetofossil-rich/detritus-poor) and interglacial (relatively
magnetofossil-poor/detritus-rich) sediments, allows quantification
of the contribution of magnetofossils to the sedimentary NRM.
In fine magnetic particle systems many magnetic properties
are a function of grain size. These properties thus provide a
means with which to investigate magnetic mineral assemblages
that contain a mixture of different particle sizes. AF demagneti-
ization curves for a thermoremanent magnetization (TRM) in mag-
etite depend strongly on grain size (see the results of
Argyle et al., 1994 as presented by Dunlop and Ozdemir, 1997). Larger
particles are magnetically soft and give rise to concave-up demag-
etization curves, while finer particles are magnetically harder so
that SD sizes produce sigmoidal demagnetization curves. Detrital
particles in a sediment produce a depositional remanent magneti-
zation (DRM), however, individually they carry a TRM or chemical
remanent magnetization (CRM) from the time of their formation.
In a similar manner, individual magnetofossils will contribute to the
NRM, but they will carry individual CRMs that formed when
they grew through their blocking volume and acquired a stable-
SD magnetization. Magnetite TRMs and CRMs have similar AF de-
magnetization spectra (Kobayashi, 1959), which allows the NRM
contributions carried by magnetofossils and detrital particles to be
compared directly.
To assess the relative contribution of magnetofossils and detri-
tal particles to the NRM of core MD00-2361 the vector difference
sum (VDS, Gee et al., 1993) of each NRM demagnetization curve
was calculated. This involves summing the magnetization vector

Fig. 6. (a) Zero-field cycling from 300 K to 10 K and back to 300 K for a 2.5 T SIRM
from an interglacial sediment sample taken at 1.25 m. (b) Derivatives of the cool-
ing and warming curves shown in (a) calculated with a smoothing spline (de Boor,
1994). The overall structure of the cycling is indicative of oxidized magnetite with
the cooling curve containing a strongly suppressed Verwey transition (marked as
T v). (c) Zero-field cycling of a 2.5 T SIRM from a glacial sediment sample taken at
3.15 m. This sample is relatively enriched in magnetofossils and exhibits a clearer
Verwey transition as demonstrated by the smoothed derivatives in (d).

Fig. 7. (a) Normalized vector difference sum (VDS) AF demagnetization curves for
the 0.5–16.5 m depth interval for core MD00-2361 (1506 curves). For clarity, the
curves are color-coded according to their MDF (blue and red correspond to soft
and hard, respectively). For comparison the AF demagnetization curves are shown
for sized magnetites carrying a TRM imparted in a 0.1 mT direct field (Dunlop and
Ozdemir, 1997). (b) The trajectory (red line) of the first VDS principal component as
a function of MDF. Individual VDS curves (black dots) from core MD00-2361 follow
the principal component closely, which suggests that the sedimentary NRM com-
promises a two-component linear mixing system. The AF demagnetization curves for
sized TRM-bearing magnetites are projected onto the same principal component
(grey points with particle sizes marked) and correspond closely with the sedimen-
tary data from core MD00-2361. (For interpretation of the references to color in this
figure legend, the reader is referred to the web version of this article.)
two-component linear mixing system (Heslop and Roberts, 2012b). The observed spectrum of patterns range from strongly concave up, which is indicative of an NRM carried by coarse grains, to almost straight lines, which are hypothesized to be a mixture of fine (magnetofossil) and coarse (detrital) particles.

To test the hypothesis that the NRM in core MD00-2361 is carried by both magnetofossils and detrital particles, we performed a principal component analysis on the normalized VDS demagnetization curves. VDS magnetizations at a given AF demagnetization step were standardized (zero mean and unit standard deviation) to produce a centered 10-dimensional data distribution. The principal component solution through this multivariate space was then calculated using singular value decomposition (Jolliffe, 2002). Over 85% of the standardized VDS variance is explained by the first principal component (i.e., the leading eigenvector through the centered data), which provides strong support for the presence of a two-component linear mixing system (Heslop and Roberts, 2012b). There is a clear relationship between the position of each VDS curve on the first principal component (represented by the so-called score) and the median destructive field (MDF) of that curve (Fig. 7b). This demonstrates that the principal component is controlled by the shape of the VDS curves. For comparison the AF demagnetization curves for sized magnetites with a TRM imparted in a 0.1 mT direct field (Argyle et al., 1994) were projected onto the first VDS principal component (Fig. 7b). As a function of MDF, the VDS for core MD00-2631 and magnetite TRMs follow slightly different paths, however, there is good correspondence in terms of overlap and trend of the two data sets. Although the first principal component defines an empirical binary mixing line through the VDS data, the locations of the two end-members on the mixing line are unknown. While it is possible to define arbitrary points to act as end-members, for example, the extremes of the data, we have used prior information to define end-members that are consistent with the rock magnetic and TEM analyses presented in Section 4.

While the TRM experiments of Argyle et al. (1994) demonstrate that magnetite demagnetization curves change their form as a function of particle size, the geological setting of core MD00-2361 and mineral magnetic data from this core imply that the VDS curves are a function of the relative NRM contributions of fine SD magnetofossils and coarse PSD/MD detrital particles. To estimate their relative contributions, TRM demagnetization curves for 0.03 × 0.2 μm and 20 μm particles (Fig. 7a) were selected as end-members on the basis of the distribution of the TRM and demagnetization curves for core MD00-2361 along the first VDS principal component (Fig. 7b). Thus, the end-member model assumes that the 0.03 × 0.2 μm and 20 μm TRM demagnetization curves represent the VDS demagnetization of the magnetofossil and detrital components, respectively, for core MD00-2361. It is important to note that for magnetite grain sizes greater than ~10 μm, TRM demagnetization curves are highly similar (Argyle et al., 1994). Therefore, the end-member model will be relatively insensitive to the assumed particle size of the detrital end-member. Additionally, as a result of the geological age and heavy weathering of the northwestern Western Australian hinterland, the riverine flux reaching the Indian Ocean is thought to have maintained a practically constant composition on glacial-interglacial time scales (Gingele et al., 2001b). It is possible that the detrital component contains an SD contribution that would be mistakenly quantified as magnetofossils during the unmixing process. The effects of such misidentification are, however, expected to be minor given that the magnetic min-

deral assemblage of the riverine flux is dominated by coarse-grained particles.

Relative contributions of the two magnetite TRM end-members to each normalized VDS curve for core MD00-2361 were estimated using constrained non-negative least squares (Lawson and Hanson, 1974). Additionally, the field steps on each curve were bootstrapped 1000 times with replacement (i.e., any given field step could be selected more than once in a given bootstrap sample) to provide a numerical assessment of the uncertainties on the estimated relative contributions (Efron and Tibshirani, 1993). The fitting errors between the VDS curves and their representation in the end-member mixing model are shown in Fig. 8a. Across the entire normalized VDS data set the average root mean squared error corresponds to a misfit between the demagnetization curves and the mixing model of <0.04. The estimated relative contributions have a tight linear relationship with the first principal component scores of the normalized VDS curves (Fig. 8b). This indicates a close correspondence between the empirical binary mixing line defined by the first principal component and our supervised unmixing model based on prior knowledge of the end-member compositions.

The relative down-core contribution of the SD end-member has a pattern consistent with the changing grain size of the magnetic mineral assemblage on glacial-interglacial timescales (Fig. 9). During interglacials, the NRM is dominated by a coarse-grained contribution carried by the riverine detrital component. In contrast, during glacial periods, the detrital mineral flux was low, the NRM is dominated (up to ~80%) by an SD-like component, which FORC and TEM analyses indicate to be due to magnetite magnetofossils. This demonstrates that, under appropriate conditions, magnetofossils can make a dominant contribution to the total sedimentary NRM. If the detrital and magnetofossil components acquired their respective NRM contributions at different depths with respect to the sediment–water interface, then it is possible that the two NRM components for any given depth in the core were acquired in different fields. Such a process would bias the unmixing results toward an over-representation of whichever component formed in the stronger field. However, given that the overall pattern of NRM contributions is consistent with the climatically controlled abundances of fluvial and biogenic magnetic particles, it must be concluded that the palaeomagnetic field could only have played a minor role in controlling the relative contributions of the two components to the sedimentary NRM.

The presented unmixing analysis employs normalized VDS and TRM demagnetization curves to provide estimates of the relative contribution of SD magnetofossils to the total sediment VDS (Fig. 9). Because of the different grain sizes of the detrital
and biogenic end-members, however, it is not possible to use component-specific relative palaeointensity parameters, such as NRM/IRM, to determine the relative efficiencies with which the two components acquired their NRM. Llowrie and Fuller (1971) demonstrated that AF demagnetization curves for TRMs and IRMs have different relationships for SD and MD particles. Thus, NRM/IRM for a given component will provide only a compounded representation of both NRM acquisition efficiency and grain size. Additionally, the large glacial-interglacial changes in magnetic mineral concentration mean that the studied sediments are not sufficiently uniform to be suitable for relative palaeointensity analysis (see, Tauxe, 1993).

6. Conclusions

We have analyzed a sediment core from offshore of northwestern Western Australia in which the magnetic mineral assemblage is a climatically controlled two-component mixing system. During glacial periods, the magnetic mineral assemblage of core MD00-2361 is dominated by SD magnetite magnetofossils. Contrastingly, during interglacials, an increased riverine flux of coarse PSD/MD detritus reached the core site, which reduced the relative abundance of magnetofossils. By demarcating the magnetofossil and detrital particle contributions on the basis of their size-dependent demagnetization characteristics, it is possible to estimate the contribution of magnetofossils to the overall sediment NRM.

During glacial periods, the sediments in core MD00-2361 were relatively enriched in magnetofossils that dominate the NRM (reaching contributions as high as ~80%). Interglacial sediments are enriched in detrital minerals and in relative terms the magnetofossil contribution to the NRM decreases (typically below ~30%). While results from core MD00-2361 reveal a clear magnetofossil contribution to the NRM, it is not apparent whether the magnetic signal formed actively (i.e., via a BgRM mechanism with bacteria aligning with the field and chains remaining oriented after death) or passively (i.e., via a post-depositional remanent magnetization with magnetofossils being reworked in the sediment post-mortem and aligning with the field). Whatever the remanence acquisition mechanism, magnetofossils have been shown to be widely preserved in the geological record in a range of sediment types (Roberts et al., 2012), which makes them a potentially important contributor to sediment magnetizations. Our approach provides a method with which to quantify magnetofossil contributions in future assessments of the extent to which biogeochemical remanent magnetizations may be important in the geological record.

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