

# Influence of Magnetofossils on Paleointensity Estimations Inferred From Principal Component Analyses of First-Order Reversal Curve Diagrams for Sediments From the Western Equatorial Pacific

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## RESEARCH ARTICLE

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### Key Points:

- Principal component analyses were applied to first-order reversal curve diagrams from three sediment cores in western equatorial Pacific
- Relative paleointensity (RPI; natural remanent magnetization/anhyysteretic remanent magnetization [ARM]) decreases with increasing proportion of magnetofossils due to their higher ARM acquisition efficiency
- Bending and collapse of biogenic magnetite chains may also influence ARM susceptibility to saturation isothermal remanent magnetization ratio and RPI estimation

### Supporting Information:

Supporting Information may be found in the online version of this article.

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# Influence of Magnetofossils on Paleointensity Estimations Inferred From Principal Component Analyses of First-Order Reversal Curve Diagrams for Sediments From the Western Equatorial Pacific

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**Abstract** Relative abundance of magnetite originated from magnetotactic bacteria (magnetofossils) in sediments may influence relative paleointensity (RPI) estimations of the geomagnetic field, as some studies reported an inverse correlation between RPI and the ratio of anhyysteretic remanent magnetization susceptibility to saturation isothermal remanent magnetization ( $k_{\text{ARM}}/\text{SIRM}$ ), a proxy of the proportion of biogenic to terrigenous magnetic minerals as well as magnetic grain size. This study aims to evaluate the influence of magnetofossils on RPI estimations more selectively using first-order reversal curve (FORC) diagrams. We studied three cores (KR0515-PC4, MD982187, and MR1402-PC1) from the western equatorial Pacific, among which large differences exist in the average natural remanent magnetization intensity normalized by ARM and  $k_{\text{ARM}}/\text{SIRM}$ . Principal component analyses (PCAs) were applied to FORC diagrams measured on bulk specimens from the three cores and silicate-hosted magnetic inclusions extracted from Core MD982187, and three endmembers (EMs) were revealed (EM1: silicate-hosted magnetic inclusions, EM2: other terrigenous, EM3: biogenic). EM3 proportions vary widely among the three cores. The average RPI decreases with increasing EM3 proportion, which is probably caused by higher ARM acquisition efficiency of magnetofossils due to small magnetostatic interactions. EM3 proportion correlates with  $k_{\text{ARM}}/\text{SIRM}$ , which confirms that  $k_{\text{ARM}}/\text{SIRM}$  represents the proportion of biogenic to terrigenous magnetic components. Core MR1402-PC1 has the highest EM3 proportion, and its within-core variation is small. From FORC-PCA applied solely to this core, we infer that the configurations of biogenic magnetite chains such as bending and collapse may also influence  $k_{\text{ARM}}/\text{SIRM}$  and RPI estimations.

**Plain Language Summary** Information on the variations of the geomagnetic field intensity in the past (relative paleointensity [RPI]) is important for understanding, for example, how the geomagnetic field is generated in the Earth's core and whether the geomagnetic field has been related to evolution of life. Marine sediments potentially preserve continuous records of RPI, but it has been suspected that magnetic-mineral composition changes of sediments may induce artificial RPI changes. This study aims to evaluate the influence of fossil magnetite produced by magnetotactic bacteria on paleointensity estimations. We used a technique based on magnetic hysteresis for estimating the proportion of biogenic magnetite to other terrigenous magnetic minerals in magnetic mineral assemblages, which was applied to sediment cores taken from the western equatorial Pacific. This technique enables to focus more on biogenic magnetite compared with previous estimations using a magnetic proxy. We confirmed that apparent RPI values inversely correlate with the proportion of the biogenic to terrigenous magnetic components, which implies that RPI is underestimated at a higher biogenic magnetite proportion. In addition, degradation of chain arrangements of magnetite originally produced by magnetotactic bacteria may also influence RPI. Our results will be useful for improving the reliability of paleointensity estimations.

## 1. Introduction

Information on geomagnetic paleointensity is fundamentally important for better understanding of the behavior and mechanism of the geodynamo, and marine sediments potentially provide continuous records

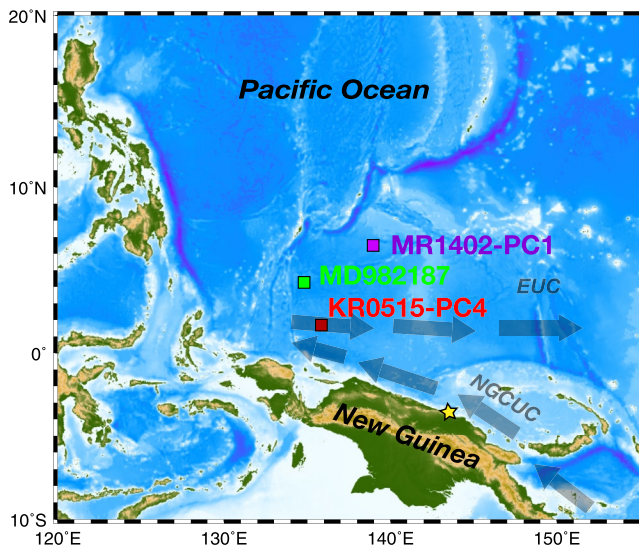
of paleointensity variations (e.g., Tauxe & Yamazaki, 2015). Relative paleointensity (RPI) estimations from sediments are usually based on the normalization of natural remanent magnetization (NRM) intensity with the intensity of laboratory induced remanent magnetization for compensating changes of “magnetizability” within the target sediments (Johnson et al., 1948; Tauxe, 1993). As no artificial remanent magnetization perfectly represents magnetizability, that is, the efficiency of depositional remanent magnetization acquisition, successful RPI estimations were derived from sediments with relatively uniform magnetic properties in general. For the normalizer, anhysteretic remanent magnetization (ARM) has often been used mainly because unlike to isothermal remanent magnetization (IRM), ARM is not sensitive to multi-domain (MD) magnetic grains, which do not carry stable NRM (King et al., 1983; Levi & Banerjee, 1976).

Magnetic mineral assemblages in sediments are usually a mixture of magnetic minerals of different origins such as magnetite originated from magnetotactic bacteria (magnetofossils), terrigenous magnetic minerals, and silicate-hosted magnetic inclusions, and each is thought to have a different paleomagnetic recording efficiency (Chang et al., 2016; L. Chen et al., 2017; Yamazaki et al., 2013). It has become recognized that magnetofossils are ubiquitous in marine sediments, and magnetofossils were often revealed to be the major constituent of magnetic mineral assemblages in deep-sea sediments (Roberts et al., 2011, 2012; Yamazaki & Ikehara, 2012; Yamazaki & Shimono, 2013). Variations in the proportion of magnetofossils to terrigenous magnetic minerals can be estimated from the ratio of ARM susceptibility to saturation IRM ( $k_{\text{ARM}}/\text{SIRM}$ ) for deep-sea sediments in pelagic environments (Egli, 2004; Yamazaki, 2008; Yamazaki et al., 2013, 2020), in which magnetic grain-size changes, another control factor of the  $k_{\text{ARM}}/\text{SIRM}$  ratio (Banerjee et al., 1981; King et al., 1982), are considered to be small because of a long distance from terrigenous source regions. Some deep-sea sediment cores showed an inverse correlation between RPI and the  $k_{\text{ARM}}/\text{SIRM}$  ratio, which raised a concern that changing amounts of magnetofossils in sediments may influence and contaminate RPI estimations (Hoffmann & Fabian, 2009; Sakuramoto et al., 2017; Yamazaki & Yamamoto, 2018; Yamazaki et al., 2013). However, interpretation of the  $k_{\text{ARM}}/\text{SIRM}$  ratio has inherent ambiguity; the ratio responds to both magnetostatic interactions and magnetic grain size. Thus it is difficult to quantitatively evaluate the amount of magnetofossils from the  $k_{\text{ARM}}/\text{SIRM}$  ratio.

In this study, we aim to evaluate the possible influence of magnetofossils on RPI estimations more selectively. For this purpose, we use first-order reversal curve (FORC) diagrams. FORC diagrams have become used widely to detect magnetofossils based on its uniaxial single-domain (SD) behavior with little magnetostatic interactions (Chang et al., 2014; A. P. Chen et al., 2007; Egli et al., 2010; Li et al., 2012; Yamazaki, 2008). By applying principal component analysis (PCA) to FORC diagrams (Harrison et al., 2018; Lascau et al., 2015), it is possible to estimate quantitatively the proportion of magnetization carried by magnetofossils (Channell et al., 2016; Roberts et al., 2018; Yamazaki et al., 2020). It is considered that the FORC-PCA method can reduce the ambiguity of magnetofossil proportion estimations compared with the  $k_{\text{ARM}}/\text{SIRM}$  ratio. Furthermore, information on bending and/or collapse of biogenic magnetite chains in sediments and differences in the morphology of biogenic magnetite can also be obtained from FORC diagrams (Berndt et al., 2020; Chang et al., 2019; Usui & Yamazaki, 2021; Wagner et al., 2021; Yamazaki et al., 2020), which may also influence RPI estimations. In this study, we use three sediment cores taken from the western equatorial Pacific, in which large differences in the proportion of magnetofossils to terrigenous magnetic minerals are expected. Based on the results of FORC-PCA, we show increased underestimation of RPI normalized by ARM with increasing relative abundance of magnetofossils. We also discuss the possible influence of biogenic magnetite chain configurations on RPI estimations.

## 2. Studied Cores and Background

Three piston cores, KR0515-PC4, MD982187, and MR1402-PC1, collected from the West Caroline Basin in the western equatorial Pacific were used in this study (Figure 1). The positions and water depths of these sites are presented in Table 1. Terrigenous particles in this region are considered to be mainly of fluvial origin (Dang et al., 2020; Wu et al., 2013). Riverine input mainly from the Sepik River in the northern part of New Guinea is delivered to this region by the New Guinea Coastal Undercurrent and Equatorial Under Current. The contribution of eolian dust is small in this region (Rea, 1994; Winckler et al., 2016; Wu et al., 2013). Sediments in this area are expected to be suitable for examining the influence of proportional changes in the magnetofossil and terrigenous magnetic components on RPI estimations because it



**Figure 1.** Map of the western equatorial Pacific and locations of the studied cores. Arrows show the New Guinea Coastal Undercurrent (NGCUC) and Equatorial Undercurrent (EUC) that deliver fluvial sediment particles in this region (after Dang et al., 2020). Star indicates the location of the mouth of the Sepik River.

is expected that the proportion of the terrigenous magnetic component varies depending on the distance from New Guinea.

Core KR0515-PC4 consists of greenish gray hemipelagic clay and contains a minor amount of foraminifera (Yamazaki & Horiuchi, 2016). This is one of the four cores used for constructing a stacked RPI curve for the last 400 kyr in the West Caroline Basin (Yamazaki et al., 2008). Core MD982187 consists mainly of hemipelagic clay with calcareous and siliceous microfossils. RPI variations of the last 3 m.y. estimated from this core were published in Yamazaki and Oda (2005). Core MR1402-PC1 consists of light gray calcareous ooze and light brownish gray to grayish yellow brown clayey calcareous ooze. A continuous RPI record from 0.6 to 3.2 Ma was successfully recovered from the core (Sakuramoto et al., 2017). Sedimentation rates of these cores (Table 1) decrease with increasing distance from New Guinea. In Core KR0515-PC4, which is the closest to New Guinea, the average sedimentation rate is about 38 m/m.y. In Core MD982187, sedimentation rates vary from 5 to 20 m/m.y. with an upcore increasing trend. In Core MR1402-PC1, sedimentation rates are 4–5.5 m/m.y. in the most part of the core.

The average RPI estimated from normalizing NRM intensity with ARM intensity differs among the three cores, in particular between Core MR1402-PC1 and the other two cores (Figure 2a). Although the age intervals of the individual paleointensity records are not the same, the time-averaged paleointensity of the geomagnetic field did not vary significantly within the last ca. 3 m.y. (Channell et al., 2009; Yamazaki & Yamamoto, 2018). We thus infer that the difference in the average normalized intensities among the three cores originates from difference in acquisition efficiency of NRM and/or ARM, and that the different efficiencies are caused by variations in magnetic mineral composition as well as other sediment properties among these cores. The average normalized intensity inversely correlates with the  $k_{\text{ARM}}/\text{SIRM}$  ratio (Figure 2a, Table 2). Normalized intensity is the highest in Core KR0515-PC4 and the lowest in Core MR1402-PC1, whereas the  $k_{\text{ARM}}/\text{SIRM}$  ratio is the lowest in the former core and the highest in the latter core. A weak inverse correlation is also observed within individual cores, in particular in Core MR1402-PC1 (insets of Figure 2a). The  $k_{\text{ARM}}/\text{SIRM}$  ratio has been used as a proxy of magnetic grain size (Banerjee et al., 1981; King et al., 1982). As ARM acquisition efficiency increases with decreasing magnetostatic interactions among magnetic grains, and thus the  $k_{\text{ARM}}/\text{SIRM}$  ratio reflects the proportion of magnetofossils in pelagic environments where magnetic grain size (domain state) change is considered to be small (Egli, 2004; Yamazaki, 2008; Yamazaki et al., 2013, 2020). The occurrence of magnetofossils in these cores was confirmed by transmission electron microscopy (Core KR0515-PC4: Yamazaki & Horiuchi, 2016, Core MD982187: Yamazaki, 2017, Core MR1402-PC1: Sakuramoto et al., 2017).

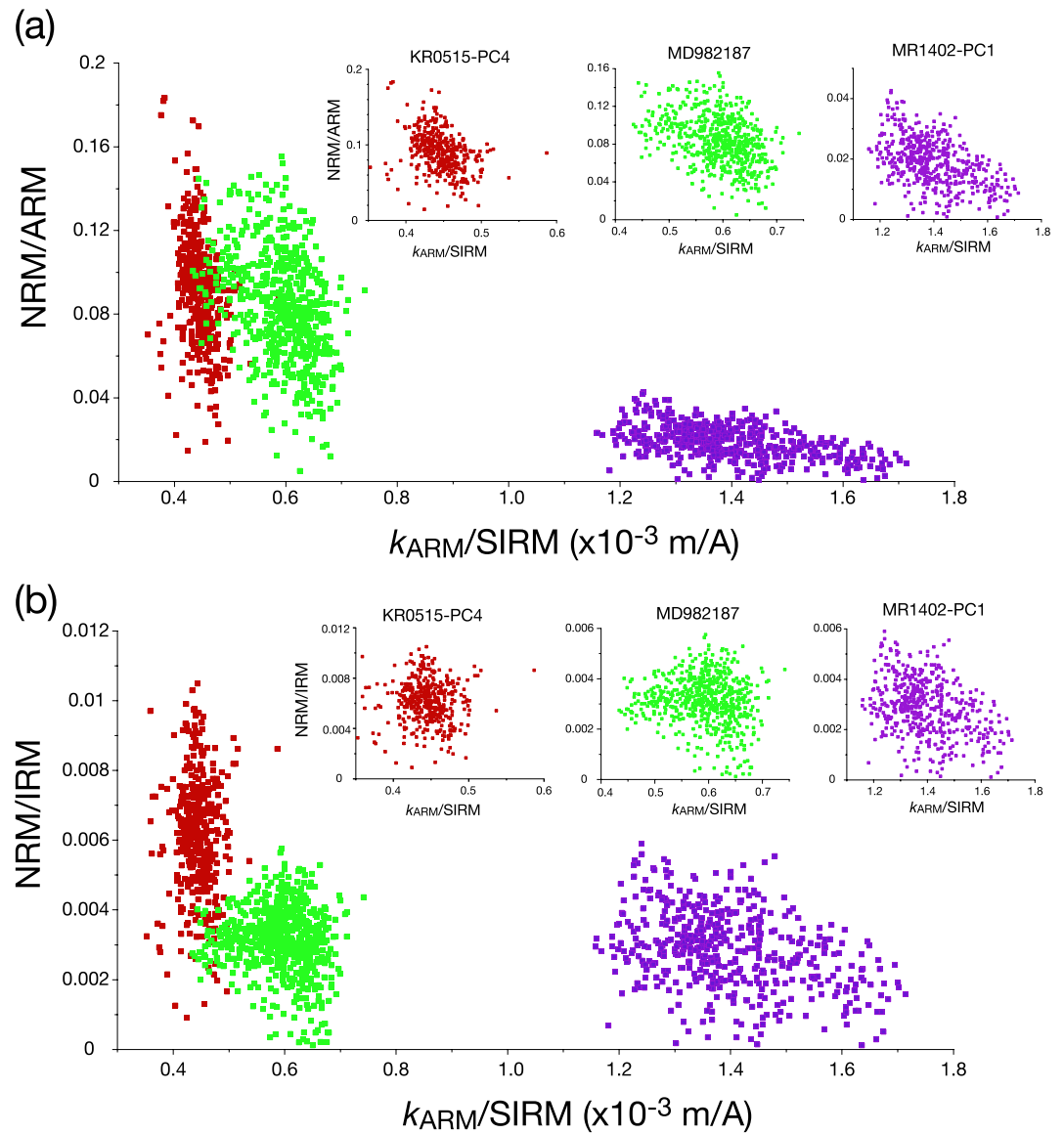
When NRM intensity is normalized by IRM, the normalized intensity still shows differences among the cores, in particular between Core KR0515-PC4 and the other two cores, although the magnitude of the differences is smaller than in the case of ARM normalization (Figure 2b). Within individual cores, Core MR1402-PC1 shows a weak inverse correlation with the  $k_{\text{ARM}}/\text{SIRM}$  ratio (Figure 2b). These observations suggest that RPI estimations normalized by IRM are also influenced by changes in the relative abundance of magnetofossils and terrigenous magnetic minerals when the changes are large.

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**Table 1**  
Information of the Cores Used in This Study

Core ID	Latitude (N)	Longitude (E)	Water depth (m)	Sedimentation rate (m/m.y.)	Reference
KR0515-PC4	1°42.71'	135°50.87'	4,277	38	1, 2
MD982187	4°15.98'	134°49.11'	4,623	5–20	3, 4
MR1402-PC1	6°29.99'	138°56.51'	3,855	4–5.5	5

Note. References: 1: Yamazaki et al. (2008), 2: Yamazaki and Horiuchi (2016), 3: Yamazaki and Oda (2005), 4: Tauxe and Yamazaki (2015), 5: Sakuramoto et al. (2017).



**Figure 2.** Relation between relative paleointensity based on natural remanent magnetization (NRM) intensity normalized by anhysteretic remanent magnetization (ARM) (a) or isothermal remanent magnetization (IRM) (b) intensities and the ARM susceptibility to saturation IRM ( $k_{ARM}/SIRM$ ) ratio for Cores KR0515-PC4 (red), MD982187 (green), and MR1402-PC1 (purple). The insets show the plots of individual cores with the aspect ratios close to one. The data sets used were presented in the references listed in Table 1, except for NRM intensity normalized by IRM of Core KR0515-PC4. The normalized intensities of Cores KR0515-PC4 and MD982187 were calculated as NRM and ARM (or IRM) intensities after AF demagnetization at 30 mT, and those of Core MR1402-PC1 were from NRM versus ARM (or IRM) slopes of stepwise AF demagnetization.

In the following, we evaluate the contribution of magnetofossils in the three cores more selectively using FORC diagrams, and discuss their influence on RPI estimations focusing mainly on those normalized by ARM.

### 3. Methods

#### 3.1. FORC Diagrams

FORC diagrams have become widely used as a method for characterizing quantitatively the constituents in magnetic mineral assemblages. On FORC diagrams, the distributions of coercivity ( $B_c$ ) and magnetostatic interactions ( $B_u$ ) are mapped, from which information on domain states, grain sizes and shapes, and mineralo-



**Table 2**  
Relative Paleointensity (RPI) and Magnetic Properties of the Studied Cores

Core ID	RPI (NRM/ARM)	$k_{\text{ARM}}/\text{SIRM}$ ( $10^{-3}$ m/A)	EM3 (biogenic) proportion (%)
KR0515-PC4	$0.10 \pm 0.02$	$0.44 \pm 0.03$	$8.4 \pm 1.9$
MD982187	$0.065 \pm 0.018$	$0.59 \pm 0.06$	$24.8 \pm 6.0$
MR1402-PC1	$0.017 \pm 0.008$	$1.40 \pm 0.12$	$72.9 \pm 2.9$

Note. Mean and standard deviation ( $1\sigma$ ) are presented. ARM, anhysteretic remanent magnetization; EM, endmember;  $k_{\text{ARM}}/\text{SIRM}$ , ARM susceptibility to saturation isothermal remanent magnetization; NRM, natural remanent magnetization.

gy of magnetic grains, and their spatial distributions can be obtained (Pike et al., 1999; Roberts et al., 2000, 2014). An assemblage of noninteracting SD grains produces a narrow peak elongated along the  $B_c$  axis at zero interaction field ( $B_u = 0$ ) on the diagram, which is called the “central ridge.” Intact biogenic magnetite chains in sediments are expected to behave as isolated noninteracting SD grains, and hence it is considered that the central ridge is diagnostic of magnetofossils in sediments (Chang et al., 2014; A. P. Chen et al., 2007; Egli et al., 2010; Li et al., 2012; Yamazaki, 2008). Magnetic grains of terrigenous origin tend to form aggregates in transport and deposition processes, and thus should have significant magnetostatic interactions. This yields a large vertical spread in the direction of the  $B_u$  axis on FORC diagrams, and hence terrigenous magnetic grains can be discriminated from magnetofossils (Channell et al., 2016; Roberts et al., 2012; Yamazaki & Ikehara, 2012; Yamazaki et al., 2020). Combined with micromagnetic simula-

tions, it was recently revealed that the conditions of biogenic magnetite chains including chain bending and collapse, distance between biogenic magnetites in a chain, and the number of magnetite in a chain significantly influence the distributions of coercivity and magnetostatic interactions (Berndt et al., 2020; Chang et al., 2019).

FORC measurements were conducted using an alternating-gradient magnetometer (Princeton MicroMag 2900) at the Atmosphere and Ocean Research Institute, The University of Tokyo. Dried samples were cut to suitable sizes (tens of milligrams) and stuck to a probe with silicone grease. A total of 165 FORCs were measured, with  $B_c$  between 0 and 100 mT,  $B_u$  between  $-50$  and  $50$  mT, and a field spacing of 1.3 mT. The maximum applied field was 1.0 T, and the averaging time for each measurement point was 200 ms. The numbers of samples measured were 48, 36, and 23 for Cores KR0515-PC4, MD982187, and MR1402-PC1, respectively.

FORCinel software (Harrison & Feinberg, 2008, version 3.06 in 2021) was used for the analyses. The VARI-FORC algorithm of Egli (2013) was used for smoothing ( $S_{c0} = 10$ ,  $S_{b0} = 5$ ,  $S_{c1} = S_{b1} = 10$ , horizontal and vertical lamda = 0.2), which provides smaller smoothing in regions near  $B_u = 0$  and larger smoothing for other regions. PCA was applied to the FORC data set to unmix magnetic components into a linear combination of endmembers (EMs) (Harrison et al., 2018; Lascu et al., 2015). The resolution of PCA grids was set to be 2 mT.

### 3.2. Silicate-Hosted Magnetic Inclusions

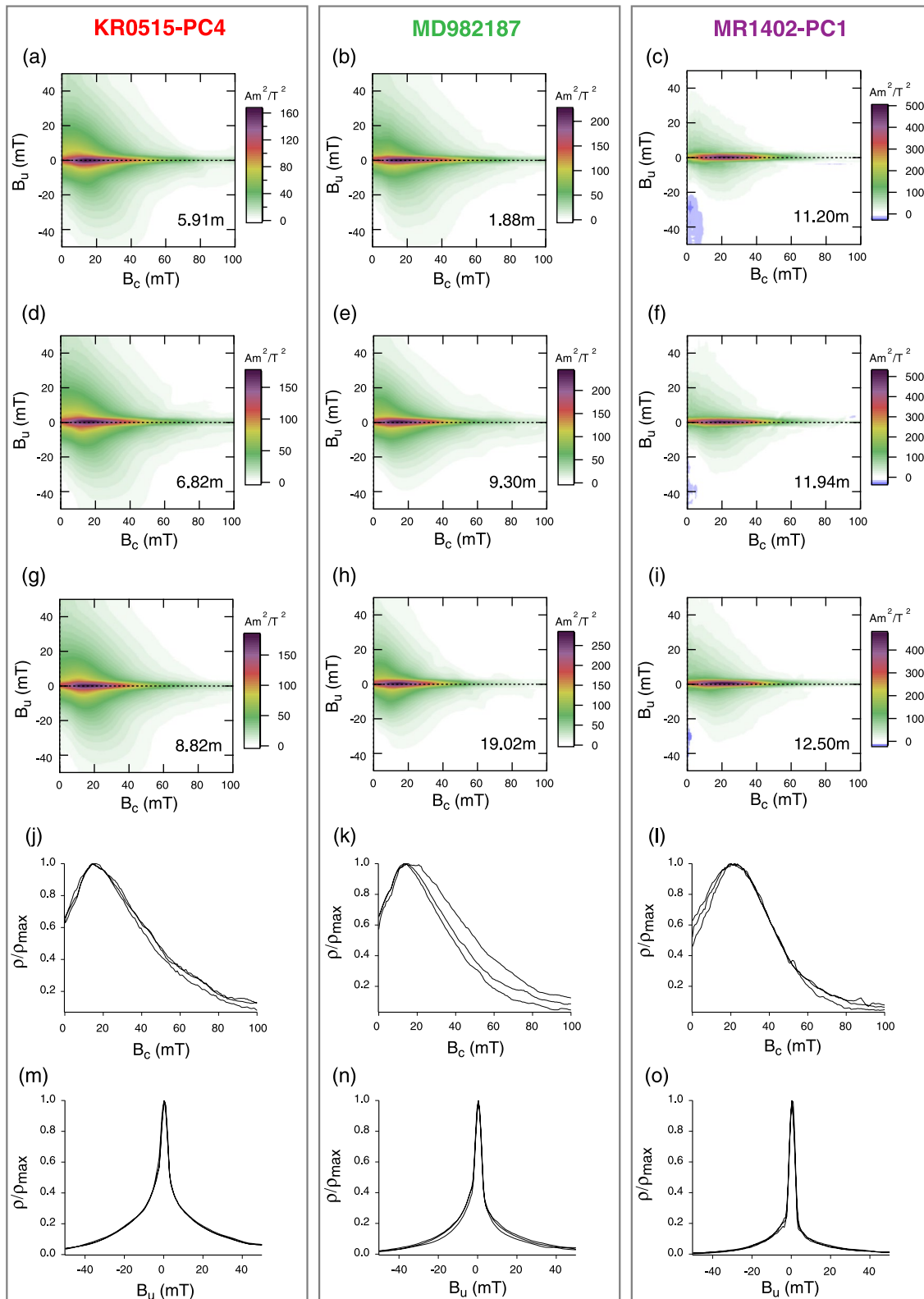
Recently it has been recognized that silicate-hosted magnetic inclusions are widely distributed in marine sediments, and potentially carry NRM (Chang et al., 2016; Usui et al., 2018; Zhang et al., 2018). To evaluate the contribution of silicate-hosted magnetic inclusions to the magnetization of the sediments in this region, we conducted chemical separation of quartz and feldspar from three samples of Core MD982187.

The samples were first treated with 1 N HCl in order to dissolve all carbonates and unprotected magnetic minerals including magnetofossils. Then, sodium pyrosulfate ( $\text{Na}_2\text{S}_2\text{O}_7$ ) fusion technique was applied to separate quartz and feldspars from these samples (Blatt et al., 1982; Clayton et al., 1972; Stevens, 1991; Syers et al., 1968; Usui et al., 2018). Samples were first freeze-dried and heated up gradually to  $460^\circ\text{C}$  with  $\text{Na}_2\text{S}_2\text{O}_7$  and then treated with 3 N HCl and washed with purified water. Next, the residues were heated to  $50^\circ\text{C}$  in 1 M NaOH overnight and washed with purified water. Finally, the residues were freeze-dried.

It is considered that only magnetic minerals remaining in the residues after all the above-mentioned procedures are inclusions in quartz and feldspar. SIRM imparted at a 2.5 T field with a pulse magnetizer was measured before and after the extraction using a spinner magnetometer. FORC measurements were conducted using the same parameters as the untreated samples mentioned above.

## 4. Results

All FORC diagrams from the three cores show the central ridge feature, indicative of noninteracting SD grains (Figure 3). This is considered to represent mainly biogenic magnetite chains because it is considered that terrigenous magnetic minerals tend to form aggregation and hence have strong magnetostatic

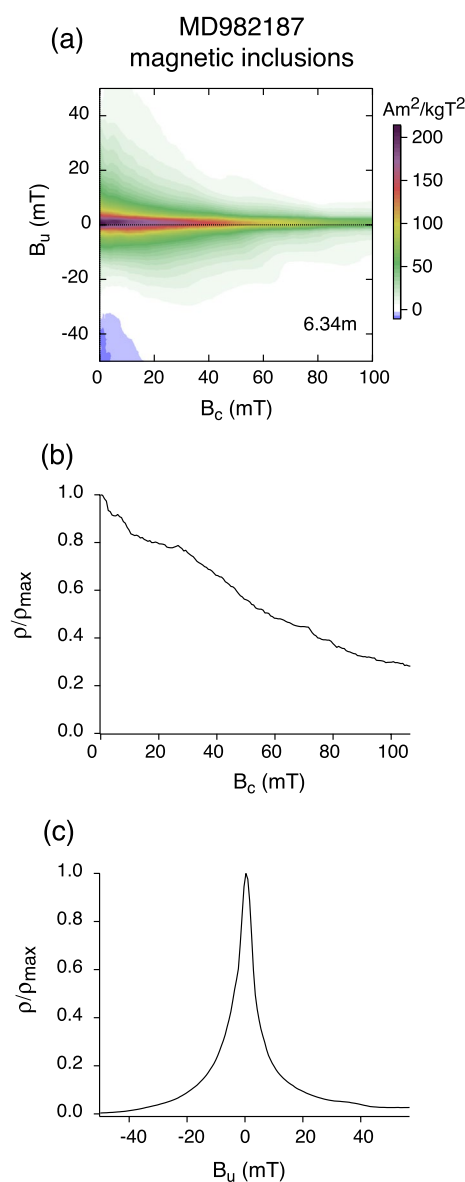


**Figure 3.** Examples of first-order reversal curve (FORC) diagrams measured on samples from the three cores. (a–i) FORC diagrams, (j–l) horizontal profiles along  $B_u = 0$  mT of the three FORC diagrams presented above for individual cores, and (m–o) vertical profiles along  $B_c = 20$  mT.

**Table 3**  
Summary of the Characteristics of Quartz and Feldspar Extracted From Core MD982187

Depth in core (m)	Mass fraction (wt%)	Carbonate-free mass fraction (wt%)	SIRM ( $\times 10^{-3}$ Am <sup>2</sup> /kg)	SIRM fraction (%)
6.34	18.7	27.9	3.63	24.1
7.38	19.4	22.4	7.85	35.0
11.32	12.8	19.9	3.43	25.9

Note. SIRM, saturation isothermal remanent magnetization.



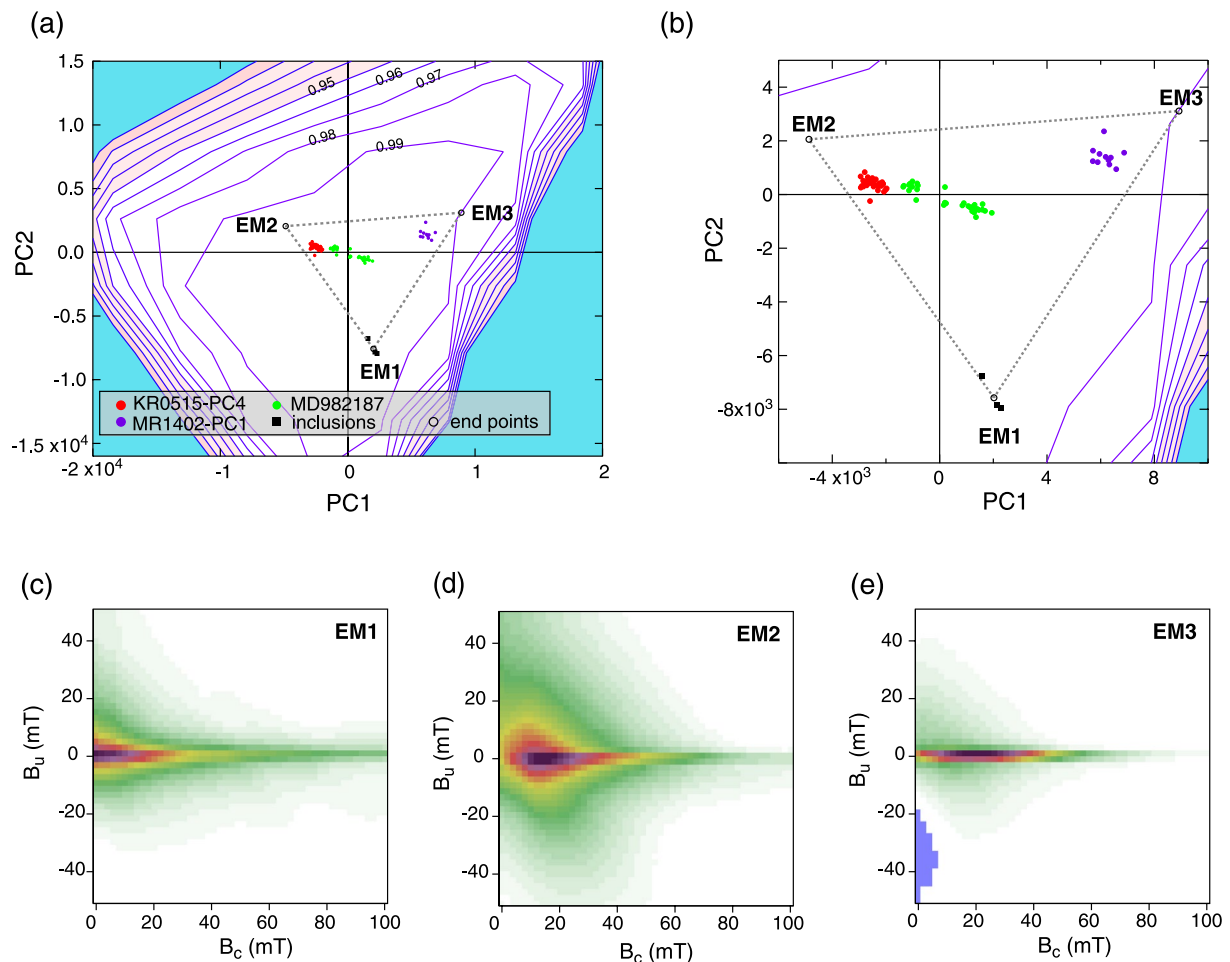
**Figure 4.** (a) Example of first-order reversal curve (FORC) diagrams of silicate-hosted magnetic inclusions extracted from Core MD982187, (b) horizontal profile along  $B_u = 0$  mT of the FORC diagram, and (c) vertical profile along  $B_c = 20$  mT.

interactions. The peak coercivity of the central ridge of each core is about 17 mT (KR0515-PC4), 12–18 mT (MD982187), and 22 mT (MR1402-PC1). FORC distributions with a broad vertical spread are also present, which indicates the contributions of interacting SD or vortex grains. They are probably of terrigenous origin, although magnetofossils may also contribute to the vertical spread when they deviate from a single intact chain arrangement like a collapsed chain. The contribution of the interacting component is the largest in Core KR0515-PC4, while it is the smallest in Core MR1402-PC1, as can be recognized from the vertical profiles in Figures 3m–3o. This will be quantified by PCA below. This is interpreted to reflect decreasing terrigenous inputs with increasing distance from New Guinea. Horizontal profiles along  $B_u = 0$  show that the FORC distributions of Cores KR0515-PC4 and MD982187 extend to a higher coercivity range beyond 80 mT (Figures 3j and 3k). On the other hand, high coercivity tails are small in Core MR1402-PC1 compared with the former two cores (Figure 3l). It is also noted that Core MD982187 shows considerable within-core variations in peak coercivities and high-coercivity tails, whereas the variations are small in other two cores (Figures 3j–3l).

Quartz and feldspar as the residue of the chemical separation from Core MD982187 samples occupy ~13–19 weight percent of the original sediments, and carry ~24%–35% of SIRM (Table 3). Thus the sediments contain a significant amount of magnetic minerals as inclusions. The FORC diagrams of the residues show a relatively narrow peak elongated along the  $B_c$  axis, but the vertical spread is wider than the central ridge of untreated sediments (Figure 4). The vertical spread becomes wider with decreasing coercivity, which would represent a signature of weakly interacting particles (Egli, 2006). Some previous studies suggested from transmission electron microscopy that silicate-hosted magnetic inclusions are mainly in SD size (L. Chen et al., 2017) and have significant magnetostatic interactions (Zhang et al., 2018). The FORC distributions near the origin (Figure 4a) suggest that inclusions of super-paramagnetic (SP) or near the SP/SD boundary are abundant. The relatively small vertical spread of the residues suggests that magnetostatic interactions among inclusions may not be as strong as those of other terrigenous magnetic minerals. The horizontal profiles along  $B_u = 0$  indicate FORC distributions extending to a high-coercivity range beyond 80 mT. Thus it is inferred that silicate-hosted magnetic inclusions carry partly, if not mostly, the higher-coercivity component observed in Cores KR0515-PC4 and MD982187.

PCA was applied to FORC diagrams (FORC-PCA) of the samples from the three cores and the chemical extracts from Core MD982187. First, the data points from all samples were plotted on a principal-component PC1-PC2 plane. The reliability of the data can be examined using the physical metrics of Harrison et al. (2018), which assess the feasibility of reconstructed FORCs from three criteria, saturation (nonexistence of a FORC exceeding the normalized value), monotonicity (nonnegative first derivative of a FORC with respect to the measurement field), and crossing (no intersection of individual FORCs), and the three metrics are combined into a single feasibility metric (Harrison et al., 2018). Samples with the feasibility metric of lower than 0.9 were excluded. The numbers of samples remained are 35, 34, and 11 for Cores KR0515-PC4, MD982187, and MR1402-PC1, respectively. All three samples of silicate-hosted magnetic inclusions passed the feasibility metric test. Low signal-to-noise ratios of



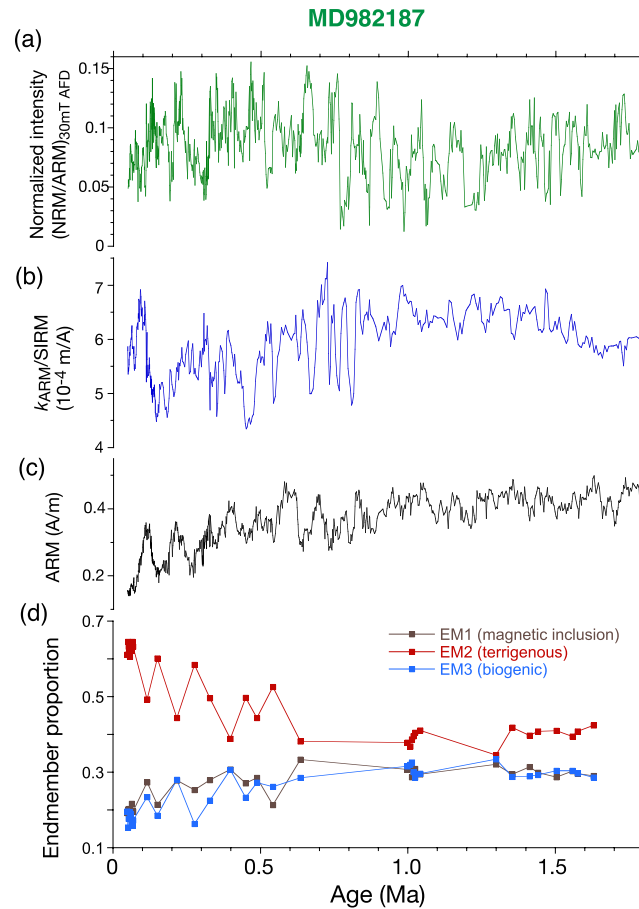


**Figure 5.** Results of principal component analysis applied to first-order reversal curve (FORC) diagrams from the three cores and silicate-hosted magnetic inclusions extracted from Core MD982187. (a) Distribution of FORC data on the principal component PC1-PC2 plane. Red: Core KR0515-PC4, green: MD982187, purple: MR1402-PC1, black: silicate-hosted magnetic inclusions extracted from Core MD982187. Triangular dashed lines define a three-endmember (EM) system. Contours represent the feasibility metric (Harrison et al., 2018). (b) Enlargement of panel (a). (c–e) FORC diagrams of EM1, EM2, and EM3, representing mainly silicate-hosted magnetic inclusions, other terrigenous magnetic minerals, and magnetofossils, respectively.

original FORC data due to weak magnetization are likely responsible for the low feasibility metrics, in particular for Core MR1402-PC1. Then PCA was conducted again using the remaining data (Figure 5). Data variance of ~92% can be explained by the two PCs (PC1: ~71%, PC2: ~21%). Three EMs (EM1, EM2, and EM3) were selected on the PC1-PC2 plane, and the contributions of individual EMs were calculated.

EM1 was selected close to the data points of the chemical extracts. We assume that EM1 reflects silicate-hosted magnetic inclusions. Although distance among silicates that host magnetic minerals in the extracts are different from those in the original sediments, this influence on the FORC diagrams is inferred to be small because magnetostatic interactions among magnetic inclusions within the same silicate host would be more significant than those among different silicates.

EM2 has an elliptical peak centered at  $B_c = \sim 15$  mT on the  $B_u = 0$  axis with a large vertical spread, and does not contain a central ridge (Figure 5d). EM2 bears the signature of interacting SD/vortex grains (Harrison et al., 2018; Muxworthy et al., 2004), and is interpreted to reflect terrigenous magnetic grains except for silicate-hosted magnetic inclusions. EM3 is dominated by the central ridge representing noninteracting SD grains, and considered to be carried mainly by magnetofossils (Figure 5e). The choice of EMs has some arbitrariness; EM2 and EM3 were selected so that the two EMs look distinct with each other and the component with a vertical spread in EM3 is as small as possible. The variations of the data from Core MD982187 are not parallel to the EM2-EM3 mixing line, but fall more in the middle of the space defined by the three EMs



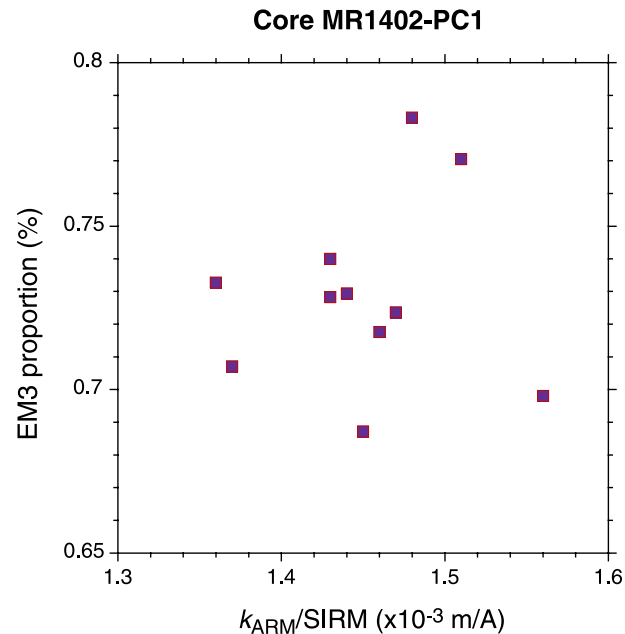
**Figure 6.** (a) Variations of relative paleointensity with age of Core MD982187 based on natural remanent magnetization (NRM) intensity normalized by anhysteretic remanent magnetization (ARM) intensity after alternating-field demagnetization at a peak field of 30 mT (Yamazaki & Oda, 2005), (b) ARM susceptibility to saturation isothermal remanent magnetization ( $k_{ARM}/SIRM$ ) ratio (Tauxe & Yamazaki, 2015), (c) ARM intensity (Yamazaki & Oda, 2005), and (d) the proportions of endmember (EM)1 (gray), EM2 (red), and EM3 (blue).

(Figure 5b). This suggests that magnetic mineral assemblages of these three cores are not a simple mixture of two components like terrigenous versus magnetofossil, and supports the incorporation of another component representing silicate-hosted magnetic inclusions.

In Core KR0515-PC4, the proportion of EM1 (~20%) plus EM2 (~70%) accounts for about 90% of the magnetization, and EM3 accounts only about 10% (Table 2, Figure S1 in Supporting Information S1). Thus magnetic grains of this core are considered to be dominated (roughly 90%) by terrigenous magnetic minerals, although the proportion depends on the choice of EM positions on the PC1-PC2 plane. On the other hand, magnetofossils (EM3) explains ~70% of the magnetization of Core MR1402-PC1 (Table 2, Figure S2 in Supporting Information S1). Within-core variations of the PC scores are minor for Cores KR0515-PC4 and MR1402-PC1. On the other hand, the PC1 score shows some changes within Core MD982187 (Figures 5b and 6).

## 5. Discussion

The results of FORC-PCA indicated that there are large differences in the EM3 (magnetofossil) proportion among the three cores, ranging from 8.4% to 72.9%, and that the variations of the EM3 proportion parallel to the  $k_{ARM}/SIRM$  ratio (Table 2). This confirms that the  $k_{ARM}/SIRM$  ratio reflects the proportion of magnetofossils to terrigenous magnetic minerals in this region. The average normalized intensity (NRM/ARM)

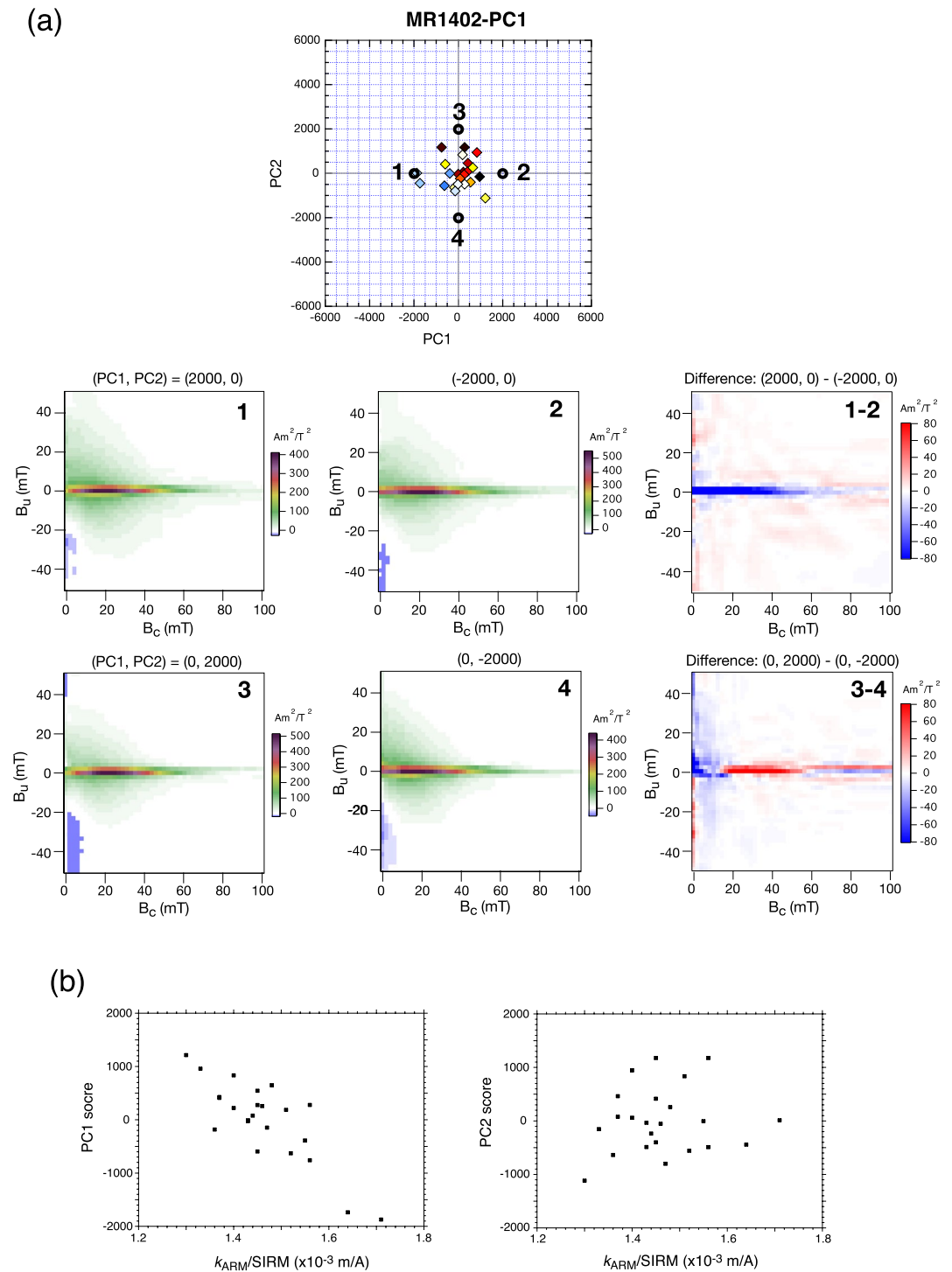


**Figure 7.** Relation between the anhysteretic remanent magnetization susceptibility to saturation isothermal remanent magnetization ( $k_{ARM}/SIRM$ ) ratio and the proportion of endmember (EM)3 in Core MR1402-PC1. EM3 represents mainly of magnetofossils, which was derived from first-order reversal curve-principal component analysis shown in Figure 5.

decreases with increasing EM3 proportion (Table 2). Core MD982187 shows within-core variations of the EM3 proportion (Figure 6). These variations parallel to the  $k_{ARM}/SIRM$  ratio, and the peaks in EM3 and the  $k_{ARM}/SIRM$  ratio broadly correspond to lows in NRM/ARM. These observations indicate that RPI normalized by ARM is influenced by the relative abundance of magnetofossils in sediments, and that underestimation of RPI occurs in sediments with increasing proportion of magnetofossils. This is probably because ARM acquisition efficiency of magnetofossils is higher compared with those of terrigenous magnetic minerals due to small magnetostatic interactions. NRM acquisition might also be influenced by the strength of magnetostatic interactions, but the dependency on magnetostatic interactions for ARM acquisition would be larger than that for NRM, and hence ARM fails to compensate for NRM acquisition efficiency changes caused by variations in the abundance of magnetofossils. ARM is thus not an appropriate normalizer of RPI estimations for sediments with variable amounts of magnetofossils.

FORC-PCA applied to samples from all three cores showed a large proportion of EM3 (~70%) for Core MR1402-PC1, which indicates that magnetic mineral assemblages in this core are dominated by magnetofossils. Variations in the proportion of the biogenic (EM3) to terrigenous components (EM1 plus EM2) within this core are small, whereas the  $k_{ARM}/SIRM$  ratio varies (Figures 2, 5b and S2 in Supporting Information S1). Furthermore, the EM3 proportion does not correlate with the  $k_{ARM}/SIRM$  ratio (Figures 7 and S2 in Supporting Information S1). These observations suggest that the variations of the  $k_{ARM}/SIRM$  ratio in this core are induced by a factor other than the proportion of magnetofossils to terrigenous magnetic minerals.

In order to investigate the cause of  $k_{ARM}/SIRM$  ratio changes, we conducted FORC-PCA using only the samples from Core MR1402-PC1 (Figure 8). All 23 FORC diagrams measured on this core were used for PCA, and no selection based on the feasibility metric was applied although signal-to-noise ratios of the samples are generally low due to weak magnetization. PC1 and PC2 explain ~33% and 22%, respectively, of the data variance. The  $k_{ARM}/SIRM$  ratio inversely correlates with the PC1 score, but does not show correlation with the PC2 score (Figure 8b). When comparing reconstructed FORC diagrams at PC scores (2,000, 0) and (−2,000, 0), the contribution of the central ridge component is smaller at (2,000, 0), whereas a broad background component is a little larger there (Figure 8a). The absence of correlation between the EM3 proportion and the  $k_{ARM}/SIRM$  ratio (Figure 7) suggests that the difference in the FORC diagrams may not be attributable to the increased terrigenous component. We speculate that increased magnetostatic



**Figure 8.** (a) Result of principal component analysis applied to first-order reversal curve (FORC) diagrams from Core MR1402-PC1. Diamonds show the distribution of FORC data on the PC1-PC2 plane. Reconstructed FORC diagrams at the positions 1 through 4 (circles) on the PC1-PC2 plane and the difference of the FORC diagrams between the position 1 (2,000, 0) and 2 (-2,000, 0) (the latter was subtracted from the former), and between the position 3 (0, 2,000) and 4 (0, -2,000) are presented. (b) Relation between the anhysteretic remanent magnetization susceptibility to saturation isothermal remanent magnetization ( $k_{ARM}/SIRM$ ) ratio and PC1 (left) and PC2 (right) scores.

interactions due to bending and collapse of biogenic magnetite chains may be responsible for the increased background component and lower  $k_{\text{ARM}}/\text{SIRM}$  ratio (Chang et al., 2019; Li et al., 2012). Because magnetofossils dominate magnetic mineral assemblages in this core, the  $k_{\text{ARM}}/\text{SIRM}$  ratio may be affected by biogenic magnetite chain configurations rather than the proportion of magnetofossils to terrigenous magnetic minerals. As RPI normalized by ARM decreases with increasing  $k_{\text{ARM}}/\text{SIRM}$  ratios in this core as well (Figure 2a) (Sakuramoto et al., 2017), biogenic magnetite chain configurations may have influenced the RPI estimations. In addition, the comparison of the reconstructed FORC diagrams at PC scores (0, 2,000) and (0, -2,000) suggests that the contributions of extremely low coercivity near the origin increase with lower PC2 scores. These might represent nonchained isolated magnetofossils (Usui & Yamazaki, 2021). On the other hand, the contributions of the broad background component are similar (Figure 8a), which may explain the absence of the correlation between the  $k_{\text{ARM}}/\text{SIRM}$  ratio and the PC2 score (Figure 8b).

The fraction of SIRM carried by the terrigenous components was estimated for each core from the sum of the EM1 and EM2 proportions and total SIRM: 5.79 A/m in average for Core KR0515-PC4, 5.89 A/m for MD982187, and 0.54 A/m for MR1402-PC1. When the differences of the sedimentation rates are taken into account (Table 1), terrigenous magnetic flux is the largest at Core KR0515-PC4 and decreases with the distance from New Guinea as expected. This supports that terrigenous particles in this region are mainly of fluvial origin from the Sepik River in New Guinea. In Core MD982187, the proportion of EM1 ranges from ~20% to 30% (Figure 6d), which is consistent with the observation that silicate-hosted magnetic inclusions extracted from Core MD982187 carry ~24% to 35% of SIRM (Table 3).

The contribution of silicate-hosted magnetic inclusions to the total magnetization of Core MD982187 is much larger than that of pelagic red clay in the South Pacific, ~1% of SIRM (Usui et al., 2018). The mass fraction of quartz and feldspar contained in Core MD982187, ~20% to 28% in carbonate free basis (Table 3), is a little larger than that in the red clay, reported as 2.3%–22.7%. The concentration of magnetic minerals embedded in host silicates seems to be larger in the studied sediments than the red clay, 3.4–7.9 versus  $0.7\text{--}2.0 \times 10^{-3}$  Am<sup>2</sup>/kg as SIRM (Usui et al., 2018), which would reflect differences of source rocks. But these two factors may not be sufficient for explaining the difference in the contribution of silicate-hosted magnetic inclusions to the total magnetization. This suggests that pelagic red clay may contain a larger amount of unprotected magnetic minerals, which may be attached to clay minerals. Our FORC-PCA revealed that the proportion of the magnetization carried by inclusions (EM1) to the magnetization by other terrigenous magnetic minerals (EM2) changes according to the distance from New Guinea; the EM1 proportion is significantly smaller than EM2 in near-shore Core KR0515-PC4, whereas the two are comparable in Core MR1402-PC1 (Figure 5), which is the farthest from New Guinea. This suggests that silicate-hosted magnetic inclusions may be transported for longer distances in this region, whereas unprotected magnetic minerals may deposit earlier.

In this study, we have discussed the influence of the proportional variations of magnetofossils in magnetic mineral assemblages on RPI estimations. For reliable RPI estimations, such influence should be corrected for. To achieve this, it is necessary to understand the differences of magnetization acquisition efficiency of NRM and the normalizes between magnetofossils and terrigenous magnetic minerals in sediments. Furthermore, PCA of the FORC diagrams suggested that the system is not a two-component mixture of magnetofossil and terrigenous magnetic minerals, but that silicate-hosted magnetic inclusions and other terrigenous magnetic minerals may behave differently. Core MD982187 has a larger proportion of EM1 to EM2 compared with that of Core KR0515-PC4 (Figures 6 and S1 in Supporting Information S1). This difference may be related to the observation that Core MD982187 has NRM/IRM values nearly as small as these of Core MR1402-PC1 although its NRM/ARMs are nearly as large as those of Core KR0515-PC4 (Figure 2). Further studies are needed to understand the behavior of individual magnetic constituents for reliable RPI estimations.

## 6. Conclusions

FORC-PCA was applied on three sediment cores from the western equatorial Pacific, among which average RPI normalized by ARM and the  $k_{\text{ARM}}/\text{SIRM}$  ratio differ significantly. Magnetic mineral assemblages in Core KR0515-PC4 are dominated (~90%) by terrigenous magnetic minerals of likely fluvial origin from



New Guinea. Magnetic grains in Core MD982187 are a mixture of magnetofossils and terrigenous magnetic minerals, and magnetic mineral inclusions hosted by quartz and feldspar carry about 30% of the magnetization. Core MR1402-PC1 is dominated by magnetofossils (~70%). RPI based on NRM/ARM decreases with increasing proportion of magnetofossils. This is probably due to that ARM acquisition efficiency of the magnetofossil component is higher compared with the terrigenous magnetic component due to small magnetostatic interactions. The  $k_{\text{ARM}}/\text{SIRM}$  ratio generally increases with increasing magnetofossil component in FORC-PCA. Within Core MR1402-PC1, however, the  $k_{\text{ARM}}/\text{SIRM}$  ratio changes significantly despite little variation in the proportion of magnetofossils. Bending and collapse of biogenic magnetite chains may also influence the  $k_{\text{ARM}}/\text{SIRM}$  ratio and RPI estimations.

## Data Availability Statement

FORC data produced in this study are available in Zenodo repository (<https://doi.org/10.5281/zenodo.5148028>).

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