# Understanding the Role of Biogenic Magnetite in Geomagnetic Paleointensity Recording: Insights from Ontong Java Plateau Sediments

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#### Abstract

Marine sediments can preserve continuous paleomagnetic intensity records. Because different magnetic minerals may acquire remanent magnetizations differently, compositional variations of magnetic mineral assemblages in sediments may hinder extraction of reliable relative paleointensity (RPI) records. To better understand this issue, we conducted a paleo- and rock magnetic study of a sediment core from the Ontong Java Plateau in the western equatorial Pacific Ocean. RPI estimated by normalizing natural remanent magnetization with anhysteretic remanent magnetization (ARM) decreases downcore with an inverse correlation with the ratio of ARM susceptibility to saturation isothermal remanent magnetization. This relationship indicates that the RPI signal weakens as the proportion of biogenic magnetite increases. The NRM-ARM demagnetization diagrams we compiled show concave-down curvature. These observations indicate that the RPI recording efficiency of the biogenic component is lower than that of the terrigenous component when we assume that the magnetizations of the high- and low-coercivity windows are carried dominantly by biogenic and terrigenous components, respectively. This assumption is supported by first-order reversal curve measurements, transmission electron microscope observations, low-temperature measurements, and extraction of silicate-hosted magnetic inclusions from the sediments. Previous studies have suggested that the RPI recording efficiency of biogenic magnetite is higher than that of the terrigenous component, which disagrees with our results. Different concentrations of silicate-hosted magnetic inclusions in different sedimentary environments might explain this contradiction. We concluded that biogenic magnetite contributes to RPI records with lower efficiency than unprotected terrigenous magnetic minerals in the studied sediments. Changing biogenic magnetite proportion distorts ARM-normalized RPI.

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#### 45 Plain Language Summary

Magnetic minerals in marine sediments can preserve the variations over geologic time of the intensity of Earth's magnetic field (paleointensity), which is important information for understanding how the field is generated within Earth's core. Paleointensity can be recorded by magnetic minerals with different recording efficiencies, so it can be difficult to recover reliable paleointensity records from sediments composed of different magnetic minerals. Our study of a sediment core from the western equatorial Pacific Ocean shows that paleointensity in the core is recorded by two main types of magnetic minerals: biogenic magnetic produced by bacteria that align themselves with the Earth's magnetic field, and magnetic minerals produced by weathering of terrigenous sediments. The paleointensity record we obtained from the sediment core weakens with increasing abundance of biogenic magnetite in the sediments. We concluded that biogenic magnetite records paleomagnetic intensity less efficiently than terrigenous magnetic minerals, which conflicts with the results of previous studies. This contradiction might be explained by differences in the concentrations of magnetic minerals embedded in non-magnetic silicate minerals in our study area compared to those in the areas of the previous studies.

#### 60 1. Introduction

61 The geomagnetic field is generated by a self-exciting magnetohydrodynamic system in Earth's core. Systematic analysis of geomagnetic field behavior requires not only directional 62 information but also intensity variations. Studies of relative paleointensity (RPI) provide 63 information about the past behavior of the geomagnetic field and contribute to our understanding 64 of the inner workings of the Earth (Tauxe & Yamazaki, 2015). Moreover, changes of RPI recorded 65 in marine sediments provide reference signals for sediment dating that are independent of 66 conventional marine paleoceanographic proxies, such as the foraminiferal oxygen isotope ratios 67  $(\delta^{18}O).$ 68

69 Marine sediments can preserve continuous RPI records. In paleomagnetic studies of marine sediments, an approximately linear relation between the ambient geomagnetic field and the 70 71 depositional remanent magnetization (DRM) intensity is assumed; however, we lack a solid theoretical foundation for the processes by which marine sediments acquire DRM. Although there 72 has been considerable effort to understand DRM acquisition mechanisms (Mitra & Tauxe, 2009; 73 74 Nagata, 1961; Tauxe et al., 2006), credible extraction of absolute geomagnetic field magnitudes 75 from marine sediments has not yet been achieved. Moreover, the complexity of the composition of marine sediments and their multiple magnetic components can affect the natural remanent 76 77 magnetization (NRM) preserved in marine sediments (i.e., DRM). Post-depositional processes, including sediment compaction and reductive diagenetic alteration, can also affect NRM (Roberts 78 et al., 2013; Tauxe & Yamazaki, 2015). 79

An empirical approach for RPI estimations is to use artificial laboratory-induced magnetization to normalize NRM intensity, thus compensating for magnetic mineral concentration variations (Johnson et al., 1948; Levi & Banerjee, 1976). Homogeneity in magnetic mineralogy

and a narrow range of magnetic grain size are assessed with magnetic hysteresis and bulk rock 83 magnetic parameters to limit non-geomagnetic factors that complicate extraction of RPI signals 84 (Ouyang et al., 2014; Roberts et al., 2013; Tauxe, 1993). However, bulk magnetic properties often 85 fail to identify complexly mixed magnetic-mineral assemblages with different origins and 86 changing proportions, which could influence RPI estimation reliability. Terrigenous magnetic 87 minerals have been recognized as paleointensity signal recorders in RPI investigations for decades. 88 Recently, biogenic magnetite and silicate-hosted magnetic inclusions have also been recognized 89 as stable, single-domain (SD) carriers of NRM, and their roles in RPI recording have been studied 90 extensively (Amor et al., 2022; Chang et al., 2021; Chang, Roberts, et al., 2016; Chen et al., 2017; 91 Hong et al., 2019; Ouyang et al., 2014; Paterson et al., 2013; Roberts et al., 2012; Usui & Yamazaki, 92 2021). 93

94 Biogenic magnetite, which is biomineralized by magnetotactic bacteria (MTB) in marine 95 and lacustrine environments, is commonly found as magnetofossils in sediments (Kirschvink, 1982; Roberts et al., 2012; Yamazaki & Ikehara, 2012). MTB produce magnetite (or greigite) 96 97 crystals with well-controlled SD sizes and shapes (Kopp & Kirschvink, 2008). The common occurrence of biogenic magnetite in the magnetic-mineral assemblages of marine sediments and 98 99 its contribution to RPI recording have been widely recognized, owing partly to improved techniques for decomposing mixed magnetic-mineral assemblages, for example, by using 100 101 isothermal remanent magnetization (IRM) acquisition curves and first-order reversal curve (FORC) diagrams (Egli, 2021; Roberts et al., 2012; Yamazaki, 2008, 2009; Yamazaki & Ikehara, 2012). 102

103 Silicate-hosted magnetic-mineral inclusions are another candidate for preserving 104 paleomagnetic signals in sediments (Chang et al., 2021; Chang, Roberts, et al., 2016; Zhang et al., 2018). Host silicate minerals can protect magnetic-mineral inclusions from iron- and sulfate-105 reducing diagenesis, and the inclusions commonly occur as fine particles. Thus, paleomagnetic 106 signals can potentially be recorded and preserved by silicate-hosted magnetic-mineral inclusions, 107 even in sediments subjected to reductive diagenesis. However, it has been reported that the NRM 108 recording efficiency of magnetic inclusions is lower than that of unprotected terrigenous magnetic 109 minerals, partly because the geomagnetic aligning torque on magnetic inclusions is counteracted 110 111 by the hydrodynamic settling force on oversized non-magnetic host silicates (Chang, Roberts, et 112 al., 2016; Chen et al., 2017; Hong et al., 2019). A recent redeposition experiment on silicate-hosted magnetic inclusions further supported this idea and revealed that non-linear field-dependent NRM 113

acquisition is likely attributable to the magnetic moment distributions of embedded magnetic
inclusions as well as the size and shape of the host-silicate minerals (Chang et al., 2021).

The ratio between the anhysteretic remanent magnetization (ARM) susceptibility ( $k_{ARM}$ ) 116 and saturation IRM (SIRM) (i.e.,  $k_{ARM}$ /SIRM) is often considered to be a grain-size proxy for 117 magnetic minerals in sediments (Banerjee et al., 1981; King et al., 1982). The strength of 118 magnetostatic interactions among magnetic particles can affect this ratio because ARM acquisition 119 efficiency can be influenced significantly by those interactions (Cisowski, 1981; Sugiura, 1979). 120 Biogenic magnetite has little magnetostatic interactions because of its distinctive chain-like 121 122 alignment of magnetite particles, although the strength of interactions may increase in multistranded and collapsed biogenic magnetosome chains (Amor et al., 2022). In contrast, terrigenous 123 124 magnetic minerals tend to have strong magnetostatic interactions because they are more likely to form aggregations. Biogenic magnetosomes are mostly confined within a narrow SD range of grain 125 126 sizes, whereas those of terrigenous magnetic minerals span superparamagnetic (SP) to vortex and/or multi-domain (MD) states. The presence of larger proportions of biogenic magnetite could 127 128 thus enhance ARM acquisition efficiency by the combined influence of differences in grain-size distribution and the strength of magnetostatic interactions, consequently producing a higher 129 130  $k_{\text{ARM}}$ /SIRM ratio. Therefore, the  $k_{\text{ARM}}$ /SIRM ratio can be a proxy for the abundance of biogenic magnetite relative to terrigenous components in magnetic-mineral assemblages, particularly in 131 132 pelagic environments, where the grain-size variability of terrigenous magnetic minerals would be small (Egli, 2004; Yamazaki, 2008, 2012; Yamazaki & Ikehara, 2012; Yamazaki & Shimono, 133 2013; Yamazaki & Solheid, 2011). A reported correlation between k<sub>ARM</sub>/SIRM and RPI for some 134 sediments (Hofmann & Fabian, 2009; Sakuramoto et al., 2017; Yamazaki et al., 2013) may be a 135 universal problem whereby compositional variations of sediments affect the reliability of RPI 136 estimations. This problem needs to be understood and resolved. 137

In this study, we used a sediment core from the western equatorial Pacific Ocean to investigate the influence of compositional variations in sediments on RPI estimations. Variable relative proportions of biogenic and terrigenous components in sediments can have a considerable influence on the RPI records in sediments (Inoue et al., 2021; Roberts et al., 2012; Yamazaki et al., 2013). Some previous research has suggested that biogenic magnetite acquires RPI more efficiently than terrigenous magnetite (Chen et al., 2017; Ouyang et al., 2014). In this study, we sought to further understand how magnetic minerals derived from different sources contribute to the remanent magnetization and RPI recording of sediments due to different proportions ofbiogenic and terrigenous components.

#### 147 **2. Study Materials and Age Model**

The samples used in this study were taken from a piston core acquired from the Ontong 148 149 Java Plateau in the western equatorial Pacific Ocean (Figure 1). The 13.6-m-long piston core MR1402-PC4 (2°03.00'N, 156°06.48'E; water depth 2447 m) consists of Quaternary sediments, 150 which was expected to provide a continuous Pleistocene geomagnetic record. The site lies above 151 the present carbonate compensation depth, which is at a water depth of about 5250 m in the study 152 area (Valencia, 1973). The predominant core sediment is light gray to light olive-gray calcareous 153 ooze. Foraminifera and calcareous nannofossils are the most abundant sediment constituents, and 154 clay minerals are also common. The sediments experienced a moderate to strong bioturbation 155 throughout the core. There is a clear sediment color change from brown to gray about 20 cm below 156 the sediment-water interface, which indicates the Fe-redox boundary (Yamazaki & Solheid, 2011). 157

The core was cut into 1-m-long sections and then split into working and archive halves using a nylon line splitting device onboard R/V *Mirai*. Two rows of discrete samples (7 cm<sup>3</sup> each) were taken continuously from the split-core surface in open-ended plastic cubes. One row was for paleomagnetic and rock-magnetic measurements, the other for oxygen-isotope measurements. Immediately after collection the samples were sealed tightly to prevent dehydration.



Figure 1. Location of core MR1402-PC4 in the western equatorial Pacific Ocean. Bathymetry
 and topography data were obtained from the ETOPO1 Global Relief Model developed by the
 National Oceanic and Atmospheric Administration.

An age model for core MR1402-PC4 was constructed using  $\delta^{18}$ O stratigraphy (Figure 2). 167 About 30 specimens of the planktonic foraminifera Pulleniatina obliquiloculata were picked from 168 the 355-425 µm size fraction of every sample. The foraminiferal shells were gently crushed 169 between two glass plates under the microscope to ensure that all chambers were open. The 170 fragments were transferred to acid-cleaned 500 µL polypropylene microtubes. After 171 172 ultrasonication with Milli-Q water and methanol, a small portion of the fragments was used for  $\delta^{18}$ O analysis. Measurements were conducted with a ThermoFisher Scientific MAT253 mass 173 spectrometer connected to a Kiel IV carbonate device at the National Museum of Nature and 174 Science, Tsukuba, Japan. Isotope values were calibrated using the NBS-19 standard, which was 175 176 analyzed several times in each run for every 7-8 unknown samples. The standard deviations for all samples were less than 0.06‰ (1 $\sigma$ ). The  $\delta^{18}$ O curve for *P. obliquiloculata* was visually fit to 177 the LR04 benthic  $\delta^{18}$ O stack (Lisiecki & Raymo, 2005) at 24 tie points (Figure 2). We estimated 178 an average sedimentation rate of  $\sim$ 1.4 cm/kyr at the core location. 179



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Figure 2. Measured  $\delta^{18}$ O values for core MR1402-PC4 (red), which was correlated with the globally stacked LR04  $\delta^{18}$ O record (Lisiecki & Raymo, 2005) (gray). Red tick marks on the horizontal axis indicate tie points.

#### 184 **3. Methods**

Magnetic susceptibility (k) was measured for all discrete samples by using a Kappabridge
 KLY-4S susceptometer. NRM was measured and subjected to a stepwise alternating-field (AF)

demagnetization at peak fields of 5, 10, 15, 20, 25, 30, 35, 40, 50, 60, and 80 mT. The 187 measurements were performed on alternate samples (290 in total) through core MR1402-PC4 by 188 189 using a pass-through cryogenic magnetometer (Model 760, 2-G Enterprises) at the Center for Advanced Marine Core Research (CMCR), Kochi University, Japan. Principal component analysis 190 (PCA) (Kirschvink, 1980) was applied to the NRM demagnetization data to extract the 191 characteristic remanent magnetization component that reflects past geomagnetic field direction 192 (Figure 3). A range of 5 to 10 demagnetization steps was used for PCA, which was determined by 193 ensuring that the maximum angular deviation (MAD) (Kirschvink, 1980) was minimized. Fitting 194 was not anchored to the origin. Results for samples with  $MAD > 10^{\circ}$  were discarded. After AF 195 demagnetization of the NRM, an ARM was imparted in a 0.1 mT direct current (DC) field with a 196 superimposed decaying AF with a peak field of 80 mT. The ARM was then subjected to stepwise 197 198 AF demagnetization at 10, 15, 20, 25, 30, 35, 40, 50, and 60 mT. Next, an IRM was imparted in a 2.5 T DC field with a pulse magnetizer (MMPM10, Magnetic Measurements) at CMCR, which is 199 200 treated here as SIRM. The SIRM was then demagnetized using the same stepwise peak AFs as the NRM. 201



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Figure 3. (a–d) Examples of stepwise AF demagnetization of NRM for (a, b) the Brunhes Chron,
(c) just above the Matuyama–Brunhes polarity boundary, and (d) within the Matuyama Chron.
Solid symbols are horizontal projections and open symbols are vertical projections of vector end

206 points. N, number of demagnetization steps used for PCA; mbsf, meters below seafloor. (e)

Variations in relative declination (red) and inclination (blue) of NRM. (f) MAD values for PCAfits.

For RPI estimations, we tested both ARM and IRM as normalizers to compensate for variations in sediment magnetizability. First, for each sample, we constructed demagnetization diagrams between NRM–ARM pairs (Figure 4a–4c) and NRM–IRM pairs (Figure 4d–4f) at corresponding AF demagnetization steps. Then, we determined best-fit slopes to represent the RPI for each sample for each normalizer. The demagnetization interval of each best-fit slope was determined based on the conditions that three or more demagnetization steps must be used and that the highest correlation coefficient was chosen (Tauxe et al., 1995; Yamazaki & Yamamoto, 2018).



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217 Figure 4. Examples of RPI estimations from best-fit slopes on (a-c) NRM-ARM

demagnetization diagrams and (d-f) NRM–IRM demagnetization diagrams. AF demagnetization
 steps are indicated by solid circles. mbsf, meters below seafloor.

220  $k_{ARM}$ /SIRM was calculated from the ARM and SIRM values obtained for RPI estimations; 221  $k_{ARM}$  was calculated by normalizing the ARM intensity with the strength of the applied DC field 222 (0.1 mT). To determine the S-ratio, a 2.5 T IRM was first imparted using the impulse magnetizer. Then, a backfield of 0.3 T was imparted in the direction opposite to the initial IRM. IRMs were measured using a spinner magnetometer (SMD88, Natsuhara-Giken) at CMCR. S-ratios were calculated according to the definition of Bloemendal et al. (1992):

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$$S_{-0.3T} = \left(1 - \frac{IRM_{-0.3T}}{IRM_{2.5T}}\right)/2.$$

FORC measurements were made on five samples selected from core MR1402-PC4 at 227 almost equal depth intervals. An alternating gradient magnetometer (AGM) (Flanders, 1988) 228 (MicroMag 2900, Princeton Measurements Corporation) at the Atmosphere and Ocean Research 229 Institute (AORI), The University of Tokyo, was used for these measurements. Samples were first 230 dried overnight at room temperature. Then, samples of about 10 mg were attached to the AGM 231 probe using silicon grease. Assuming that the magnetic minerals in our samples are dominated by 232 SD magnetite, the coercivity  $(B_c)$  range was set to 0–100 mT. The range of the interaction field 233 234  $(B_{\rm u})$  was  $\pm 50$  mT, which determines the upper and lower limits of the FORC diagrams. The saturating field was set to 1 T. The averaging time for each data point was 400 ms (800 ms for 235 some weak samples) and the field increment was set to 2 mT. The number of FORCs was 111. 236 FORC measurement data were analyzed using FORCinel software (Harrison & Feinberg, 2008) 237 with VARIFORC smoothing protocol (Egli, 2013). The parameters used for smoothing were  $S_{c0} =$ 238 239 4,  $S_{b0} = 3$ ,  $S_{c1} = S_{b1} = 7$ , and  $\lambda_c = \lambda_b = 0.1$ .

We obtained low-temperature magnetic measurements of two types on the same five 240 samples that were used for the FORC measurements. For these measurements we used a low-241 242 temperature superconducting quantum interference device susceptometer (MPMS-XL5, Quantum Design) at CMCR. For the first measurement, an SIRM was imparted in a 3 T field at 300 K. 243 244 Samples were then cooled to 10 K and then warmed back to 300 K in a nearly zero field. Magnetization changes were measured during the temperature cycling. For the second 245 246 measurement, samples were cooled from 300 K to 10 K in a nearly zero field, then an SIRM was imparted by a 3 T field. Thermal demagnetization of the SIRM was measured during warming 247 back to 300 K in a nearly zero field. 248

We obtained transmission electron microscope (TEM) observations to complement our rock-magnetic measurements. TEM observation can provide direct morphological and size information about magnetic-mineral assemblages in sediments. Although TEM observation provides advantages compared to indirect rock-magnetic measurements, the results obtained from them remain semi-quantitative and may not faithfully reflect the overall morphology and size distribution of magnetic-mineral assemblages. Moreover, magnetic extraction is required before TEM observation, which can bias the information obtained. Nevertheless, TEM observations provide important information that supplements rock-magnetic measurements.

To extract magnetic minerals, the sediments were first dispersed in distilled water in an 257 ultrasonic bath and circulated with a peristaltic tubing pump for about 24 h. Sodium 258 hexametaphosphate was used as a deflocculant for the suspension. Magnetic minerals were 259 260 extracted from the circulating suspension using a high magnetic field gradient produced by a strong magnet. The magnetic extracts were preserved in ethanol. Finally, a drop of the magnetic extracts-261 262 ethanol suspension was dried on a carbon-coated copper grid in preparation for TEM observation (Yamazaki & Yamamoto, 2018). A TEM (JEM-1400, JEOL) operated at 120 keV at AORI, was 263 264 used for these observations.

To estimate the concentration of silicate-hosted magnetic-mineral inclusions in the studied 265 sediments, and then to assess their contribution to paleomagnetic signal recording, quartz and 266 feldspar were extracted from three selected samples using several geochemical procedures. Freeze-267 268 dried samples were weighed and treated with 1 N HCl to dissolve all carbonates and unprotected magnetic minerals, including magnetofossils. Sodium pyrosulfate (Na<sub>2</sub>S<sub>2</sub>O<sub>7</sub>) fusion was then 269 applied to separate quartz and feldspars from the samples (Blatt et al., 1982; Clayton et al., 1972; 270 Stevens, 1991; Syers et al., 1968; Usui et al., 2018). Next, the samples were freeze-dried and heated 271 gradually to 460 °C with Na<sub>2</sub>S<sub>2</sub>O<sub>7</sub> and then treated with 3 N HCl and washed with purified water. 272 The residues were then heated to 50 °C in 0.5 M NaOH overnight and washed with purified water. 273 Finally, the residues were freeze-dried and weighed again in preparation for further magnetic 274 measurements. We assumed that any magnetic minerals that remained in the residues after these 275 procedures were magnetic-mineral inclusions in quartz and feldspar. We estimated the 276 contribution of silicate-hosted magnetic-mineral inclusions to paleomagnetic signal recording by 277 comparing the SIRM magnitudes before and after extraction. SIRM was imparted using an impulse 278 magnetizer (Model IM10-30, ASC Scientific) with a 2.7 T field and was measured using a spinner 279 280 magnetometer (ASPIN, Natsuhara-Giken) at AORI. To estimate the coercivity distributions of the 281 residues, FORC measurements were conducted using the aforementioned AGM at AORI. FORC measurement parameters were the same as those described above for the untreated samples. 282

#### 283 **4. Results and Interpretation**

#### 4.1. Paleomagnetic Directions

Stepwise AF demagnetization of the NRM generally reveals univectorial behavior, except for the first few demagnetization steps, which probably reflect a soft viscous remanent magnetization overprint that was removed at an AF of 10 mT for most samples (Figures 3a–3d). Paleomagnetic declinations and inclinations were determined by applying PCA (Kirschvink, 1980); MADs of less than 3° were obtained for most samples, except for those near a polarity transition (Figures 3e and 3f). Directional changes at around 100 ka are most likely caused by physical sediment disturbance during coring.

The Matuyama-Brunhes polarity transition appears as a ~180° relative declination flip 292 (Figure 3e, red curve) at about 787 ka. There is a time lag of ~14 kyr relative to the recently 293 proposed age of 773 ka for the Matuyama–Brunhes transition (Channell et al., 2020; Singer, 2014; 294 Suganuma et al., 2018). This difference may be explained by delayed DRM lock-in. From the 295 average sedimentation rate we estimated at the core location (~1.4 cm/kyr), we estimated the DRM 296 depth-lag to be  $\sim 20$  cm. We calculated the average inclination (excluding data at the polarity 297 transition and the interval of physical disturbance) to be -0.54°, and obtained an inclination 298 anomaly of -4.63° from the geocentric axial dipole field inclination (4.09°) at the site latitude. Our 299 result is consistent with the known negative inclination anomaly of about -5° in the western 300 equatorial Pacific (Johnson & Constable, 1997; Yamazaki et al., 2008). The observed high-quality 301 paleomagnetic directional record of this core is a prerequisite for reliable RPI estimation. 302

#### 303 4.2. Estimation of Relative Paleointensity

Before RPI estimation, it is preferable to check if the studied sediments satisfy empirical 304 305 criteria for reliable RPI estimation, including relatively small variations in magnetic mineral concentrations and homogeneities of magnetic mineralogy and grain size (King et al., 1983; Tauxe, 306 1993). A gradual downcore magnetic mineral concentration decrease indicated by decreasing 307 magnetic susceptibility (Figure 5a) reflects magnetic mineral dilution with increasing carbonate 308 content. However, the magnetic concentration varies by less than an order of magnitude, which 309 satisfies the empirical criteria (King et al., 1983; Tauxe, 1993). Variations of magnetic mineralogy 310 311 and grain size can be estimated from bulk magnetic proxies, such as the S-ratio (mineralogy) and

- $k_{\text{ARM}}$ /SIRM (grain size). Sporadic drops of both of these proxies are evident in the studied 312 sediments (Figures 5b and 5c). Because high-coercivity magnetic minerals (e.g., hematite) are 313 more resistive to reductive diagenesis than low-coercivity minerals (e.g., magnetite) (Liu et al., 314 2004; Yamazaki et al., 2003), the lower S-ratios we obtained (Figures 5b) may indicate reductive 315 dissolution of magnetite. Associated magnetic grain size increases indicated by  $k_{\text{ARM}}$ /SIRM drops 316 (Figures 5c) may also be attributed to the reductive dissolution of magnetite because fine grains 317 are expected to be dissolved earlier than coarse grains owing to their higher surface-to-volume 318 ratio (specific surface area). Therefore, in core MR1402-PC4, intervals in which magnetite 319 dissolution might have occurred (shaded in Figures 5a-5e) were excluded from RPI estimations 320
- because they do not meet the empirical criteria of King et al. (1983) and Tauxe (1993).



Figure 5. Variations of (a) magnetic susceptibility (k), (b) S-ratio (S<sub>-0.3T</sub>) and (c) ratio of  $k_{\text{ARM}}$  to

324 SIRM in sediment core MR1402-PC4. Shaded intervals are likely affected by magnetite

325 dissolution due to reductive diagenesis. RPI estimated from NRM intensity normalized by (d)

ARM and (e) IRM. (f) The PISO-1500 paleointensity stack (Channell et al., 2009). VADM,

327 virtual axial dipole moment. Correlations with the RPI records of core MR1402-PC4 are

indicated by the tie lines in panels (d) to (f).

For RPI estimations, NRM intensities normalized by ARM and IRM (Figures 5d and 5e) 329 were obtained from the best-fit slopes of NRM-ARM and NRM-IRM demagnetization diagrams 330 331 (Figure 4). The PISO-1500 paleointensity stack (Channell et al., 2009; Figure 5f) was used as a template to assess the reliability of RPI estimations in this study. The overall patterns of the RPI 332 records estimated from NRM intensity normalized by ARM (Figures 5d) and IRM (Figures 5e) 333 agree reasonably well with the PISO-1500 paleointensity stack of Channell et al. (2009; Figures 334 335 5f). However, RPI normalized by ARM (NRM/ARM) (Figure 5d) has a conspicuous artificial downcore decrease, especially in the lower part of the core, whereas RPI normalized by IRM 336 337 (NRM/IRM) (Figure 5e) does not. Intriguingly,  $k_{ARM}$ /SIRM has an approximate inverse correlation with the RPI normalized by ARM (compare Figures 5c and 5d), indicated by correlation coefficient 338  $\rho = -0.55$  and statistical significance P < 0.01. This approximate inverse correlation is also evident 339 in a biplot of  $k_{ARM}$ /SIRM and NRM–ARM slope (Figure S1), where the amplitude of NRM–ARM 340 slope is subdued with increasing  $k_{\text{ARM}}$ /SIRM. This approximate inverse correlation suggests that 341 the relative proportion of biogenic to terrigenous magnetic components in the sediments increases 342 343 with depth and that ARM fails to compensate for compositional changes. Comparison of the downcore variations of ARM and SIRM (Figure S2) support this interpretation. SIRM decreases 344 consistently with magnetic susceptibility (Figure 5a), reflecting magnetic concentration changes, 345 whereas ARM exhibits a gentler downcore decrease. These observations indicate that changes in 346 ARM acquisition efficiency are responsible for the approximate inverse correlation between the 347 ARM-normalized RPI and the  $k_{ARM}$ /SIRM. Therefore, we consider IRM to be a more appropriate 348 normalizer than ARM for the studied sediments. Inverse correlations between RPI and kARM/SIRM 349 has been reported for sediments from various regions (Hofmann & Fabian, 2009; Sakuramoto et 350 al., 2017; Yamazaki et al., 2013; Yamazaki & Yamamoto, 2018). This could be attributed to 351 352 differences in remanence acquisition efficiency between biogenic and terrigenous components (Yamazaki et al., 2013). 353

We obtained curved slopes in NRM-ARM demagnetization diagrams (Figures 4a-4c). 354 When all NRM-ARM pairs are plotted on a single demagnetization diagram (Figure 6a), a similar 355 356 curved trend is evident; in contrast, NRM-IRM demagnetization diagrams show an approximately linear relationship (Figure 6b). The curvatures suggest that NRM and ARM have different 357 coercivity distributions. ARM acquisition is more efficient for SD biogenic magnetite, whereas 358 terrigenous magnetic minerals are likely to include vortex and/or MD grains. Consequently, 359 biogenic and terrigenous components will have different acquisition efficiencies for NRM and 360 ARM. Therefore, the curvature in NRM-ARM demagnetization diagrams can be attributed to 361 mixtures of biogenic and terrigenous magnetic mineral components. 362

To attempt to separate the different contributions of biogenic and terrigenous components 363 to RPI signals, we re-calculated best-fit slopes on NRM-ARM demagnetization diagrams for two 364 AF demagnetization intervals: a low-coercivity window of 10-25 mT and a high-coercivity 365 366 window of 35–60 mT (Figure 7a–7c), following the methods of Ouyang et al. (2014) and Chen et al. (2017). These coercivity intervals were selected to separate and optimize the fit of curves in the 367 NRM-ARM demagnetization diagrams (average  $R^2 = 0.99$  for the low-coercivity window with 368 standard deviation < 0.02 and average  $R^2 = 0.96$  for the high-coercivity window with standard 369 370 deviation < 0.1).





Figure 6. Demagnetization data for (a) NRM–ARM and (b) NRM–IRM pairs at various (colorcoded) AF demagnetization steps (mT). Dashed lines indicate 1:1 relationships.



Figure 7. Examples of best-fit slopes for (a–c) NRM–ARM demagnetization diagrams
calculated for AF demagnetization intervals of 10–25 mT (red lines) and 35–60 mT (blue lines).

377 RPI signals calculated for the same coercivity windows for (d, e) NRM–ARM slope and (f)

378 NRM–IRM slope. In panel (e) both RPI curves were rescaled by their maximum values.

ARM-normalized RPIs within the high-coercivity window are smaller than those in the 379 low-coercivity window (Figure 7d); however, paleointensity variations for the two windows are 380 concordant after rescaling (Figure 7e). The downcore artificial decreases for the two coercivity 381 windows are also identical. For NRM normalized by IRM (Figure 7f), differences in RPI estimated 382 from the two coercivity windows are smaller than for the ARM normalization, but normalized 383 intensities for the low-coercivity window are still marginally higher than those for the high-384 385 coercivity window. These differences are also evident in Figure 6b, where the low-coercivity data clusters are closer to the 1:1 line, but the high-coercivity data clusters plot below it. 386

#### 387 4.3. FORC Diagrams

Previous research demonstrated that intact magnetosome chains produced by MTB can 388 have similar magnetic properties to those of uniaxial non-interacting SD particles. The latter 389 produces a distinct ridge-like FORC distribution along the  $B_{\rm c}$  axis with almost no vertical spread 390 (known as the central ridge) and a subsidiary negative peak along the negative  $B_u$  axis near  $B_c = 0$ . 391 Hence, its contribution can be isolated easily from magnetic-mineral assemblages using FORC 392 diagrams (Egli, 2021; Egli et al., 2010; Pike et al., 1999; Roberts et al., 2000, 2012, 2014; 393 Yamazaki, 2008, 2009; Yamazaki & Ikehara, 2012). The spread of the central ridge along the B<sub>c</sub> 394 axis represents the coercivity distribution of the magnetic particles and is clearly identifiable even 395 in complex mixtures with other magnetic components. Central ridges are clearly evident in FORC 396 diagrams for the measured samples (Figure 8a). The coercivity distributions of all samples extend 397 398 to about 60 mT or higher, which corresponds to the high-coercivity component of NRM-ARM 399 slopes. The FORC distributions also have a component with a significant vertical spread, with outer contours that diverge toward the  $B_{\rm u}$  axis. This component can be attributed to interacting SD, 400 401 vortex and/or MD particles, which are likely carried by the terrigenous magnetic-mineral 402 component. The central-ridge FORC distributions of biogenic magnetite origin were extracted from the total FORC distributions (Figure 8b) by using the FORCinextras function in the 403 FORCinel software (Harrison & Feinberg, 2008), in which the central ridge signal can be 404 calculated by subtracting a smoothed and extrapolated "ridge-free" background FORC distribution 405 from an original FORC distribution. The detailed mathematical foundation of this approach was 406

explained by Egli et al. (2010). The integrals of these identifiable FORC distributions over 407 specified FORC regions are defined as FORC magnetizations of corresponding magnetic 408 components (Egli et al., 2010). Because the integral of the total FORC distribution over the 409 memory region coincides with the integral of the DC demagnetization curve ( $f_{dcd}$ ), which is 410 alternatively equivalent to the saturation remanent magnetization (M<sub>rs</sub>) (Egli, 2021; Pike, 2003), 411 the ratio between the integral of the extracted central-ridge FORC distribution (M<sub>cr</sub>) and the 412 calculated M<sub>rs</sub> can be used to roughly estimate the relative contribution of the biogenic magnetite 413 414 to the remanent magnetization of the sediments.



416 **Figure 8**. (a) FORC diagrams for five samples from core MR1402-PC4, (b) the central ridge

417 component extracted from each FORC diagram, (c) profiles of interaction field  $(B_u)$  distributions

at coercivity ( $B_c$ ) of 20 mT, and (d) profiles of coercivity distributions at  $B_u = 0$ . In panels (c) and

419 (d), profiles of the central-ridge component (red), the background component (gray), and the

420 total (black) are shown.

We estimated the relative contribution of biogenic magnetite for the low- (10-25 mT) and 421 high- (35-60 mT) coercivity windows separately from the FORC magnetizations of the 422 corresponding  $B_c$  windows (Figure 9a). Biogenic magnetite contribute more to the remanent 423 424 magnetization in the high-coercivity window than that in the low-coercivity window (Figure 9b). The difference in these contributions is greater in the deeper part of the core, where biogenic 425 magnetite in the high-coercivity window contributes up to 40% of the total remanent magnetization. 426 The relative proportion of biogenic magnetite that we calculated should be considered as a 427 428 minimum estimate, because magnetosomes in multi-stranded configurations and in bent or foldcollapsed chains can yield vertical spreads that are nearly symmetrical around the central ridge in 429 430 FORC diagrams (Amor et al., 2022; Berndt et al., 2020; Chang et al., 2019; Inoue et al., 2021; Li et al., 2012; Usui & Yamazaki, 2021), which is counted as the terrigenous component in our 431 calculation. The relative contributions of biogenic magnetite to the remanent magnetization in both 432 coercivity windows increase with depth, which is consistent with the downcore  $k_{ARM}$ /SIRM 433 434 increase, and indicates an increasing relative abundance of biogenic magnetite. These results suggest that the remanent magnetization in the high-coercivity window is carried more by biogenic 435 magnetite. Therefore, the high-coercivity component in RPI estimations corresponds mainly to 436 biogenic magnetite, whereas terrigenous magnetic minerals dominate the low-coercivity 437 component. 438



439

Figure 9. Relative contribution of biogenic magnetite to the remanent magnetization of the sediments. (a) Schematic illustration of the estimation process, which is based on the ratio of the integral of the extracted central-ridge FORC distributions ( $M_{cr}$ ) to the integral of the total FORC distributions over the memory region ( $M_{rs}$ ). Areas within dashed lines define the memory region. (b) Results for five samples from core MR1402-PC4, estimated from the high-coercivity window in red (35–60 mT) and from the low-coercivity window in blue (10–25 mT).

#### 446 4.4. Low-temperature magnetic measurements

447 The Verwey transition (Verwey, 1939) temperature ( $T_v$ ) at around 100 K was observed in 448 zero-field SIRM warming curves (red curves in Figure 10b) for all five analyzed samples. This is easier to recognize in derivative curves (orange curves in Figure 10b). Two distinct peaks at ~95 K and ~115 K (or broad bell-shaped peaks) are observed in the derivative curves. The lower  $T_v$  is reported to be an intrinsic property of biogenic magnetite (Chang, Heslop, et al., 2016; Li et al., 2009; Pan et al., 2005); therefore, the presence of two  $T_v$  peaks suggests that the studied sediments are a mixture of biogenic and terrigenous inorganic magnetites.

The degree of maghemitization can be estimated from the low-temperature cycling curves 454 of SIRM at 300 K (Figure 10a). When temperature decreases from 300 K to  $T_{\rm v}$ , the SIRM of 455 magnetite acquired at 300 K decreases, whereas that of maghemite increases monotonically. Thus, 456 457 low-temperature cycling curves for magnetite with oxidized maghemite surfaces can be considered to be a superposition of the two curves (Özdemir & Dunlop, 2010) and maghemitization can be 458 assessed by comparing the magnetization at 300 K with that below T<sub>v</sub> at, for example, 50 K (Chang 459 et al., 2013; Yamazaki & Solheid, 2011). The magnetization at 300 K is greater than that at 50 K 460 461 in most samples (Figure 10a), indicating a relatively low degree of maghemitization (Chang et al., 2013; Özdemir & Dunlop, 2010), which is consistent with the Fe-redox boundary recognized at 462 463 ~20 cm depth in the core. For the deepest sample (at 853 ka), the magnetization at 50 K is slightly larger than that at 300 K. Considering the low magnetite concentration at that depth, as indicated 464 by the low magnetic susceptibility (Figure 5a), this result might be due to the influence of 465 paramagnetic minerals and the presence of a small residual field in the MPMS during measurement. 466



Figure 10. Results of low-temperature magnetic measurements for five samples from core
MR1402-PC4. Low-temperature cycling curves (a) normalized by SIRM at 300 K (blue); (b)

- zero-field warming curves normalized by SIRM at 10 K (red), with derivative of the curves in
- 471 orange. Verwey transition temperature peaks  $(T_v)$  are also shown in panel (b).

## 472 4.5. TEM Observations

TEM observations were conducted on magnetic-mineral extracts from two samples (at 322 473 474 and 853 ka). In total, more than 2380 magnetofossils were identified in hundreds of TEM images. We recognized magnetofossils of three distinct morphologies: equant (cubo-octahedra and 475 hexagonal prisms where length-to-width ratios are close to one); elongated (hexagonal prisms and 476 elongated octahedra); and bullet-shaped magnetofossils (Akai et al., 1991; Bazylinski et al., 1994; 477 Li et al., 2020; Yamazaki, 2020) (Figure 11). The bullet-shaped morphology is prismatic with one 478 end tapered and curved sides and is easily distinguished from the other magnetofossil types. The 479 elongated type occurs commonly as a hexagonal prism or elongated octahedron. We considered 480 some of the hexagonal prisms (those with length-to-width ratios close to one) to have magnetic 481 properties similar to those of the cubo-octahedra, owing to their small shape anisotropy. However, 482 there are inherent ambiguities in the classification of the three-dimensional morphologies of 483 magnetofossils from projected two-dimensional TEM images (e.g., the axial ratios of 484 magnetofossils vary continuously and at certain angles an elongated octahedron can have a similar 485 two-dimensional projection to that of an equant octahedron); thus, this type of characterization is 486 semi-quantitative. 487



489 Figure 11. TEM images of magnetic extracts from core MR1402-PC4 (a) at 3.82 m (322 ka) and

(b) at 10.57 m (853 ka). Red, blue, and green dots identify bullet-shaped, elongated, and equant

491 magnetofossils, respectively.

The coercivity of intact biogenic magnetite chains is controlled by the shape anisotropy of 492 the magnetite particles as well as by the chain configuration, such as the number of magnetite 493 particles and by the bending angle in a chain structure (Berndt et al., 2020; Chang et al., 2019; Li 494 et al., 2013; Paterson et al., 2013). Nevertheless, the abundance of anisotropic biogenic magnetite 495 shapes in sediments tends to correlate with relatively high coercivities (Lascu & Plank, 2013; Usui 496 497 et al., 2017; Yamazaki, 2012; Yamazaki et al., 2020; Yamazaki & Ikehara, 2012). Our magnetofossil count results for the three morphologies (Table 1) indicate that the anisotropic 498 morphology groups (elongated and bullet-shaped magnetofossils) consist of more than 50% of the 499 magnetofossils in our two samples, which supports the observation from FORC diagrams that the 500 501 central ridge due to biogenic magnetite extends to higher coercivities.

Age of samples (ka)	Bullet-shaped	Elongated	Equant	Total
322	378 (24%)	463 (29%)	734 (47%)	1575 (100%)
853	156 (19%)	291 (36 %)	358 (45%)	805 (100%)

502 **Table 1.** Counts of magnetofossil morphologies from TEM observations.

#### 503 4.6. Silicate-Hosted Magnetic-Mineral Inclusions

The contribution of silicate-hosted magnetic-mineral inclusions to RPI signals was 504 505 assessed by comparison of the SIRM values of the untreated samples with those of the residues of chemical extraction procedures (Table 2). The mass fractions of the residues are less than 2% of 506 507 the untreated sediment samples, and account for less than 7% of SIRM. These results indicate that the contribution of the silicate-hosted magnetic-mineral inclusions to the RPI signal recording is 508 509 smaller than that of biogenic magnetite and unprotected terrigenous magnetic minerals. The magnetite particle concentration within the host silicate minerals can be roughly estimated as 510 follows. Considering that the saturation magnetization of magnetite is 92 Am<sup>2</sup>/kg (O'Reilly, 1984), 511 and if it is assumed that the magnetite particles are all uniaxial SD particles randomly oriented in 512 the host silicate minerals, then, based on Stoner & Wohlfarth (1948) theory, the SIRM of the 513

magnetite particles in the silicates should be  $46 \text{ Am}^2/\text{kg}$ . Compared to the mass-normalized SIRM of the residues in Table 2, the magnetic particle concentration is of the order of 0.01%. Preferred crystallographic orientations are common in exsolved inclusions and can cause variations of remanent magnetization from 25% to 100% as a function of applied field orientation (Feinberg et al., 2006). Therefore, the actual contribution of the inclusions to the RPI signals would be smaller, and the magnetite particle concentration would be in the range 0.001%–0.01%.

520 **Table 2.** Comparison between properties of samples before and after the chemical extraction of

521 quartz and feldspar.

Depth (m)	Age (ka)	Bulk SIRM of untreated samples (10 <sup>-6</sup> Am <sup>2</sup> )	Bulk SIRM of residues (10 <sup>-6</sup> Am <sup>2</sup> )	Magnetization fraction of residues (%)	Mass fraction of residues (%)	Mass-normalized SIRM of untreated samples (10 <sup>-3</sup> Am <sup>2</sup> /kg)	Mass-normalized SIRM of residues (10 <sup>-3</sup> Am <sup>2</sup> /kg)
4.57	378	9.62	0.51	5.25	1.75	1.77	5.32
7.95	649	4.84	0.15	3.03	1.44	0.92	1.94
10.43	843	2.89	0.20	6.78	1.30	0.50	2.61

FORC diagrams for the residues after chemical extraction of quartz and feldspar have a peak near the origin (i.e.,  $B_u = 0$ ,  $B_c = 0$ ) (Figure 12a), which indicates that a large amount of the magnetic inclusions have sizes near the SP/SD boundary (Pike et al., 2001) as well as of SD and vortex state particles. In addition, high-coercivity FORC signals in the residues are smaller than those of the original samples (Figure 8a).

TEM observation was conducted on the residues at 378 ka as a complement to FORC 527 analyses (Figures 12b and 12c). The TEM used here does not have an energy-dispersive 528 spectrometer, so we could not identify the mineralogy of the inclusions. However, magnetic 529 inclusions commonly appear as dark particles in TEM images. The dark particles included in the 530 transparent minerals in Figure 12, which are likely magnetite within silicates, range in size from a 531 few nanometers to tens of nanometers, which is consistent with the SP/SD as well as SD and vortex 532 533 state FORC signatures (Figure 12a). SP particles do not contribute to SIRM in our calculation of 534 the magnetite particle concentration within the host silicate minerals, so the actual magnetite particle concentration would be larger. 535



536

Figure 12. (a) FORC diagram and (b, c) TEM images of silicate-hosted magnetic-mineral
inclusions extracted from core MR1402-PC4 at 4.57 m (378 ka).

#### 539 5. Discussion

Results from FORC measurements indicate that the RPI records of the studied sediments 540 in the high-coercivity window has a larger contribution from biogenic magnetite than those in the 541 low-coercivity window. TEM observations on magnetofossils suggest that elongated 542 magnetofossils could have higher coercivities. Maghemitization processes might contribute to the 543 high-coercivity component. Low-temperature oxidation proceeds inward from the surface of a 544 magnetite grain to produce a fully oxidized maghemite shell that grades gradually into a largely 545 unoxidized magnetite core. This gradient of oxidation degree can induce stresses, and hence the 546 coercivity of a magnetite grain should increase with increasing maghemitization (Cui et al., 1994; 547 van Velzen & Zijderveld, 1995). However, our observarions on low-temperature magnetic 548 549 measurements indicate that the degree of maghemitization is relatively low in the studied sediments. Maghemitization, therefore, is not the likely source of the high-coercivity component. 550 Silicate-hosted magnetic inclusions could be considered a candidate carrier of the high-coercivity 551 component. However, FORC diagrams of the residues after chemical extraction procedures 552 (Figure 12a) show smaller high-coercivity FORC signals than those of the untreated samples 553 (Figure 8a), so the silicate-hosted magnetic inclusions are also unlikely to be the carrier of the 554 high-coercivity signal. We therefore consider biogenic magnetite to be the dominant high-555 coercivity signal carrier with additional unprotected terrigenous magnetic minerals in the studied 556 sediments. 557

The ARM-normalized RPI records estimated from the high- and low-coercivity windows 558 indicate that biogenic magnetite has a lower RPI compared to the terrigenous component (Figure 559 7d). This difference is also evident when RPIs from the high-coercivity window are plotted against 560 those from the low-coercivity window (Figure S3a), where most of the NRM/ARM data fall below 561 the 1:1 line, indicating the higher RPI recording efficiency of the terrigenous component. The 562 NRM intensity normalized by ARM of the terrigenous component is at least 1.5–2.5 times higher 563 than that of the biogenic component. This difference can be explained by RPI underestimations 564 owing to increasing ARM acquisition efficiency with increasing biogenic magnetite proportion. 565 Note that RPI signals from both the high- and low-coercivity windows have identical artificial 566 downcore decreases that correlate with a downcore  $k_{\text{ARM}}$ /SIRM increase (Figures 5c and 7e). These 567 correlations indicate that the RPI signals of the low-coercivity window are also influenced by 568 569 increasing biogenic magnetite proportion in the sediments.

570 Concave-down curvatures in NRM-ARM demagnetization diagrams and distorted RPI variations that correlate with  $k_{\text{ARM}}$ /SIRM indicate that ARM normalization is not appropriate for 571 572 RPI estimation in these sediments, even for the low-coercivity window. However, for NRM normalized by IRM, the difference of the RPI recording efficiencies for biogenic and terrigenous 573 components is lower than that for ARM normalization (Figure S3b), but the low-coercivity 574 terrigenous component still has higher RPI recording efficiency than the biogenic component. The 575 576 terrigenous component is at least 1-1.5 times more efficient than the biogenic component in RPI recording. These observations emphasize that accurate RPI estimations require magnetically 577 homogeneous sediments. 578

579 Chen et al. (2017) used methods similar to ours to investigate the RPI recording efficiency of various magnetic components in sediment cores from the eastern equatorial Pacific Ocean. They 580 used several analyses (including FORC diagrams, IRM unmixing, and TEM observations) to 581 identify two types of dominant stable SD components that correspond to biogenic magnetite in 582 high-coercivity distribution and terrigenous magnetic particles in low-coercivity distribution. They 583 argued that the silicate-hosted magnetic inclusions that dominate the low-coercivity terrigenous 584 component are mainly of SD size and are present at concentrations sufficiently high to be 585 detectable in magnetic measurements. They then compared the ARM-normalized RPIs of the high-586 587 and low-coercivity windows while assuming that biogenic magnetite corresponds to the higher coercivity component and silicate-hosted magnetic inclusions correspond to the lower coercivity 588

component. Their result is contrary to our findings (Chen et al., 2017, their Figure 8): their NRM–
 ARM demagnetization diagrams have convex curvatures, and the biogenic component carries a
 stronger RPI signal.

Possible reason for the contradiction with the study of Chen et al. (2017) might be different 592 concentrations of silicate-hosted magnetic inclusions in sediments from different environments. 593 594 The contribution of eolian dust to sediments in the western equatorial Pacific Ocean site of core MR1402-PC4 is small (Rea, 1994; Winckler et al., 2016; Wu et al., 2013). Terrigenous particles 595 in this region are considered to be mainly of fluvial origin from New Guinea (Dang et al., 2020; 596 597 Wu et al., 2013). The silicate-hosted magnetic inclusion mass-fraction concentration in core MR1402-PC4 is less than 2%, and carries less than 7% of SIRM (Table 2). Therefore, terrigenous 598 magnetic minerals in our study would be mainly unprotected magnetic particles in the interacting 599 SD, vortex, and/or MD states. The core site of Chen et al. (2017) is from a different oceanographic 600 601 regime, and can receive eolian dust from the Americas (Rea, 1994) or from Asia, depending on the position of the intertropical convergence zone (Yamazaki, 2012). If such dust contains more 602 603 silicate-hosted magnetic inclusions and/or less unprotected magnetite per unit volume than the fluvival particles from New Guinea, then silicate-hosted magnetic inclusions might dominate the 604 terrigenous magnetic minerals. In this case, the contradiction can be explained by assuming that 605 the RPI recording efficiency of terrigenous magnetic minerals other than silicate-hosted magnetic 606 607 inclusions is higher than that of biogenic magnetite, whereas the RPI recording efficiency of silicate-hosted magnetic inclusions is lower than that of biogenic magnetites. Some studies have 608 suggested that silicate-hosted magnetic inclusions possess low RPI recording efficiency or even 609 cannot preserve reliable RPI signals (Chang et al., 2021; Chang, Roberts, et al., 2016; Hong et al., 610 2019). Regardless, the apparent contradiction implies that compositional differences in sediments 611 from different geological environments can influence remanent magnetization acquisition 612 613 efficiency. More research is needed to further investigate this issue.

#### 614 6. Conclusions

615 Our paleo- and rock-magnetic study of core MR1402-PC4 sediments from the Ontong Java 616 Plateau in the western equatorial Pacific led us to the following conclusions.

617 1. A downcore increase of the ratio of biogenic to terrigenous magnetic minerals was estimated 618 using  $k_{\text{ARM}}$ /SIRM. The ratio correlates negatively with RPI normalized by ARM, which

- suggests that ARM normalization failed to compensate for compositional changes in thesediments.
- 2. RPIs calculated in high- and low-coercivity windows of NRM–ARM and NRM–IRM
  demagnetization diagrams indicated that the high-coercivity portion has smaller RPI than the
  low-coercivity portion. From FORC diagrams, TEM observations, low-temperature
  measurements, and the extraction of silicate-hosted magnetic inclusions, we showed that
  NRM in the high-coercivity window is carried more by biogenic magnetite, whereas NRM in
  the low-coercivity window is carried more by terrigenous magnetic minerals. Thus, the RPI
  recording efficiency of biogenic magnetite is lower than that of the terrigenous component.
- 3. Previous studies have used similar methods to indicate higher RPI recording efficiency for
   biogenic magnetite, which is contrary to our conclusion. Different magnetic mineral
   components have different RPI recording efficiencies, so different silicate-hosted magnetic
   inclusion concentrations in different sedimentary environments are likely to be responsible
   for the observed differences among these studies.
- 633 Data Availability Statement
- All data produced in this study are available from the Zenodo repository
- 635 (https://doi.org/10.5281/zenodo.6351605).

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Journal of Geophysical Research: Solid Earth

Supporting Information for

## Understanding the Role of Biogenic Magnetite in Geomagnetic Paleointensity Recording: Insights from Ontong Java Plateau Sediments

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# **Contents of this file**

Figures S1 to S3

# Introduction

The following figures are provided to better illustrate some of the statements in our article. Terminologies and acronyms in these figures are not explained or spelled out, since they have been defined in the article. The interpretations of these figures are also presented in the corresponding places in the article.



Figure S1. Relation between kARM/SIRM and NRM–ARM slope.



Figure S2. Downcore variations of ARM (purple) and SIRM (orange).



