Paleoenvironmental significance of magnetofossils in pelagic sediments in the equatorial Pacific Ocean before and after the Eocene/Oligocene boundary

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Key Points:

- Magnetofossils from sediments of eastern equatorial Pacific archive paleoenvironmental change during Eocene-Oligocene transition (EOT).
- Contents of magnetofossils in sediments were decreased as the aeolian particle supply was reduced after the EOT.
- Relatively increased organic matter input induced increase of elongated and bullet-shaped magnetosomes in magnetic particles.

Abstract

Magnetotactic bacteria (MTB), sensitive to redox status, leave fossil bacterial magnetite (magnetofossils) in sediments. The relative contents of cuboctahedral, elongated prismatic, and bullet-shaped magnetofossils archive changes in redox conditions, which indicate paleoenvironmental variations. The Eocene-Oligocene transition (EOT) is a turning point in the Cenozoic climate evolution from greenhouse to icehouse. Global cooling, Antarctic glaciation, and/or a new tectonic structure modified the global ocean and atmosphere circulation mode. In the eastern equatorial Pacific Ocean (EEPO), the organic matter and aeolian supply, which are important for the proliferation of MTB, did not vary synchronously. Here, we study the magnetic particles and environmental magnetism characteristics of samples from EEPO to test the hypothesis that magnetofossil assemblages respond to the dramatic paleoclimatic changes across the EOT. Results show that the abundances of all kinds of magnetic particles are significantly decreased with the reduction of aeolian supply after the EOT. However, the relative abundance of magnetofossils and the proportion of bullet-shaped ones both increased. Simultaneously, organic matter input was enhanced as indicated by: 1) similar productivity but decreased organic carbon consumption, and 2) the increased mass accumulation rates of total organic carbon after the EOT. The enhanced organic matter flux increased redox gradient in pelagic sediment, which together with sufficient iron from aeolian particles, supported the metabolism of MTB, especially those synthesizing bullet-shaped magnetofossils. Therefore, the magnetofossils in the EEPO vary synchronously with the evolution of paleoenvironment before and after the EOT, which thus supports the use of magnetofossil as an effective proxy archiving paleoenvironment.

1 Introduction

Intracellularly synthesized by magnetotactic bacteria (MTB), fossil bacterial magnetites (magnetofossils) are aligned into chains to facilitate the mobility of MTB (Bazylinski & Frankel, 2004; Dieudonné et al., 2019; Lefèvre & Bazylinski, 2013). The single-domain (SD, Petersen et al., 1986; Stolz et al., 1986) magnetofossils have three kinds of morphologies: cuboctahedral, elongated prismatic, and bullet-shaped (Bazylinski & Frankel, 2004; Dieudonné et al., 2019; Kopp & Kirschvink, 2008), which are controlled by the species and genes of MTB (Hesse, 1994; Moisescu et al., 2014). Active metabolism of MTB occurs in the water column and sediments of global lakes or oceans (Bazylinski & Frankel, 2004; Dieudonné et al., 2019; Kopp et al., 2009), and even in hot springs and saline lakes (Dieudonné et al., 2019). Magnetofossils are left in the sediments after the death of MTB, but they are not always preserved — they are easily destroyed, for example, by reductive diagenesis.

The morphology of preserved magnetofossils provides an archive of paleoenvironmental information (Chang et al., 2018; Hesse, 1994; Kopp et al., 2007; Kopp & Kirschvink, 2008; Larrasoaña et al., 2012; Lean & McCave, 1998; Roberts et al., 2011; Usui et al., 2017; Yamazaki, 2012; Yamazaki et al., 2019; Yamazaki &

Kawahata, 1998; Yamazaki & Ikehara, 2012; Yamazaki & Shimono, 2013). Oxic to sub-oxic conditions in and near the oxic-anoxic transition zone (OATZ) and an adequate nutrient supply are indispensable for MTB activity (Bazylinski & Frankel, 2004; Dieudonné et al., 2019; Hilgenfeldt, 2000; Larrasoaña et al., 2012; Roberts et al., 2011; Savian et al., 2016). Previous works indicates that equidimensional magnetofossils (e.g. cuboctahedral) are distributed both in and above the OATZ in sediments, whereas anisotropic ones (e.g. elongated prismatic and bullet-shaped), especially bullet-shaped, concentrate in the zone with less oxic conditions (Chang et al., 2018; Hesse, 1994; Yamazaki et al., 2019; Yamazaki & Kawahata, 1998; Yamazaki & Shimono, 2013). Organic matter brings reducing conditions to pelagic sediments, forming an OATZ with the oxidizing seawater. Adequate amounts of organic matter increase chemical gradient in the sediment and shallow the OATZ. Sedimentary rate influences the input of organic matter into sediments. Oxygen concentration of bottom-water and pore water directly impacts the thickness and depth of OATZ. Therefore, the relative contents of equidimensional and anisotropic magnetofossils imply the thickness and depth of OATZ which is closely correlated to the supply of organic matter (Lean & McCave, 1998; Roberts et al., 2011; Usui et al., 2017; Yamazaki, 2012; Yamazaki & Ikehara, 2012), sedimentary rate (Kopp & Kirschvink, 2008), and oxygen content of bottom-water and pore water (Hesse, 1994; Kopp et al., 2007; Kopp & Kirschvink, 2008; Lean & McCave, 1998). Such conditions are sensitive to the global climate change.

The Cenozoic global climate evolution reached a milestone between the late Eocene and early Oligocene, representing an important step in the greenhouse-icehouse climate switching (Coxall & Pearson, 2007; Miller et al., 2009). This significant turning point is associated with a rapid increase in the oxygen stable isotope composition of benthic foraminifera ($\delta^{18}O_{benthic}$) by ~1.5‰ within ~0.5 Ma (DeConto et al., 2008; Miller et al., 2009). Specifically, the increment is composed by two steps, ~0.5‰ at Eocene-Oligocene transition (EOT, ~33.9 Ma, Miller et al., 2009) and ~1‰ at Oligocene isotope event 1 (Oi-1, ~33.5 Ma, Coxall et al., 2005; Miller et al., 1991), which are more precisely described as EOT-1, 2 and Oi-1a, b, respectively, by high-resolution sampling results from continental shelf of Alabama and southern Indian Ocean (Coxall & Pearson, 2007; Katz et al., 2008; Zachos et al., 1996).

The $\delta^{18}O_{benthic}$ values are controlled mainly by the combined effects of bottom water temperature and the oxygen stable isotope compositions of seawater which is sensitive to the glaciations in polar regions. Many records from both oceans and land suggest a sequence of cooling and then ice sheet growth across this oxygen isotope event (Houben et al., 2012; Katz et al., 2008; Ladant et al., 2014; Lear et al., 2008; Liu et al., 2009). In particular, the $\delta^{18}O_{benthic}$ excursion during the EOT was caused by bottom water cooling with slight ice growth in Antarctica (Eldrett et al., 2009; Plancq et al., 2014; Wade et al., 2012; Xiao et al., 2010), whereas that of Oi-1 was caused by large ice sheet expansion in Antarctica accompanied with a slight drop of bottom-water temperature (DeConto et al., 2008; Grimes et al., 2005; Lear et al., 2004; Petersen & Schrag, 2015; Pusz et al., 2011; Zachos et al., 1996).

Two main reasons have been proposed to cause this climate change event, orbital force and atmospheric CO_2 content. Since the late Eocene, specific orbital configurations leading to relatively cold summer (Coxall et al., 2005; Coxall & Pearson, 2007; Jovane et al., 2004; Zachos et al., 1996) and CO_2 content decreasing to lower than a specific threshold (Coxall & Pearson, 2007; Coxall & Wilson, 2011; Dalai et al., 2006; DeConto & Pollard, 2003; Pearson et al., 2009) have induced some pulses of cooling. Assisted further by the thermal isolation caused by the opening of gateway around Antarctica (Coxall & Pearson, 2007), the above effects resulted in cooling at the EOT and, finally, glaciation at Oi-1. As the ice sheet grew to cover most parts of Antarctica and reached the coastline, new circulation modes of atmosphere and ocean currents were established (Miller et al., 2009; Page et al., 2019), significantly changing the ocean environments and nutrient distribution.

The paleoenvironment in the eastern equatorial Pacific Ocean was changed accordingly. Cooling and deep water from the Southern Ocean modified the nutrient source, which would influence the primary productivity and the amount of organic matters supply into sediment (Miller et al., 2009; 2014, Moore Jr. et al., 2014; Page et al., 2019). Switching from warm age to ice age, the aeolian input in this area was also changed (Rea, 1994). The variations in supplies of organic matters and aeolian particles would influence the redox gradient and the thickness and depth of OATZ in bottom water and shallow sediments as well as the amount of nutrient for MTB, severely influencing the metabolism of the redox-sensitive MTB and altering the magnetofossil aggregates. The corresponding changes, however, has been less investigated. The Ocean Drilling Program (ODP) Site 1218 and the Integrated Ocean Drilling Program (IODP) Site 1333 retrieved excellent continuous samples during this period in the equatorial Pacific Ocean (Lyle et al., 2002a; Pälike et al., 2010). In this work, we study the magnetofossils from before and after the EOT to test their response to the significant global climate change during EOT and Oi-1 by the Sites 1218 and 1333.

2 Materials and Methods

The ODP Site 1218 and the IODP Site 1333 are located in the eastern equatorial Pacific Ocean, with water depths of 4828 m and 4853 m, respectively (Fig. 1, Lyle et al., 2002a; Pälike et al., 2010). The two sites cover sediments from Eocene to Miocene and Site 1218 contains additional Pleistocene sediments (Lyle et al., 2002a; Pälike et al., 2010). Currently, the sulfate contents of pore water are close to seawater value throughout the sedimentary column, indicating that no obvious organic matter reduction reactions are active (Lyle et al., 2002a; Pälike et al., 2010). Samples across late Eocene to early Oligocene from the two sites were collected for this study (Table 1). The ages of samples from the Site 1218 were from Lyle et al. (2002a) and were interpolated with the composite depth, while those from the Site 1333 were based on ages from Pälike et al. (2010) and were interpolated with core depth below seafloor.

Samples contain two main sedimentary phases, nannofossil chalk and biogenic SiO_2 (radiolarian and diatoms, Fig. 2), showing light and dark colors, respectively. From the bottom to the top of the sediment sequence, the phase of Site 1218 changes from a mixture of the two phases, to biogenic SiO_2 before the EOT, to nannofossil chalk after the EOT, while those of the Site 1333 starts with a mixture of phases and is replaced with nannofossil chalk slightly before the EOT (Fig. 2).

2.1 Magnetic measurements

Powder-like samples were dried in an oven at 40 °C and then firmly fixed into standard cubic plastic boxes. The magnetic susceptibility (χ) data were analyzed by an Agico Kappabridge MFK2. Final results were the average value of five continuous tests normalized by weight.

After demagnetization with a 100 mT alternating field in the axial 0.05 mT bias field, the sample boxes were measured using an Agico JR6A for anhysteretic remanent magnetization (ARM) and weight normalized. An isothermal remanent magnetization ($IRM_{1\ T}$) was imparted to the sample in cubic boxes in a 1 T inducing field and subsequently analyzed by an Agico JR6A for $IRM_{1\ T}$ results which were also normalized by weight. Then, the $IRM_{-0.3\ T}$ and $IRM_{1.2\ T}$ were measured using corresponding inducing field. Before each measurement, each powder sample was stirred to eliminate the magnetic orientation induced by previous measurement. Hard isothermal remanent magnetization (HIRM) values were calculated by ($IRM_{1.2\ T}$ + $IRM_{-0.3\ T}$) / 2.

To check for dilution effects caused by the carbonate matrix, magnetic susceptibility and the three remanent magnetization results were normalized on a carbonate free basis (CFB) by the (1 – $CaCO_3$ content) (Hounslow & Maher, 1999). The analysis of CaO content is introduced in section 2.2.

Some samples, separated from the powder in cubic boxes, were mounted firmly into capsules and loaded onto a Lakeshore vibrating sample magnetometer (VSM) to obtain the hysteresis parameters, IRM acquisition curves, and first-order reversal curves (FORC). Hysteresis loops were measured to 500 mT with step of 3 mT. IRM acquisition curves were measured to 1 T with nonlinear 80 steps. The moment of each data point was averaged by the nearby four points and fitted by the IRM-CLG excel file created by Kruiver et al. (2001) to obtain the IRM, IRM contributions, coercivity, and dispersion parameter of each component. 251 first-order reversal curves with step size of 0.8 mT were measured for selected sample with a 500 ms averaging time and processed by VARIFORC (Egli, 2013) using the FORCinel v3.06 software (Harrison & Feinberg, 2008).

All the magnetism analyses were completed in the Centre for Marine Magnetism (CM^2), Department of Ocean Science and Engineering, Southern University of Science and Technology (SUSTech).

2.2 Mineralogical and geochemical analyses

Dried samples were spread onto conductive adhesive tapes which were pasted on standard stages and then slightly pressed to be firmly fixed. After blowing away the loosely stuck particles, the samples were carbon coated and observed under backscatter mode by a Phenom XL G2 scanning electron microscope in the Social Science Center, SUSTech.

About 2 mg powder samples were loaded on stages with silicon tablet background and analyzed by a Rigaku MiniFlex benchtop X-ray diffraction instrument (Cu Kα radiation) in the Social Science Center, SUSTech. The major phase identification and semi-quantification were conducted by the PDXL2 software.

About 0.5 g dried and slightly ground samples were added into 50 ml MQ water and ultrasonic vibrated for 20 min. Then, a magnet-containing Teflon stick with a smooth surface was inserted into the solution which was, afterwards, ultrasonic vibrated for another 10 min. In the process of ultrasonic vibration, some ice made from MQ water was put into the solution to keep the temperature low. After that, the teflon stick was left in the solution for over two hours and, subsequently, transferred into a new bottle full of MQ water. After removing the magnet, the teflon stick was ultrasonic vibrated for 5 min to separate magnetic particles from its surface. The extracted magnetic particles were loaded and clipped between carbon laced double-layer copper grids and analyzed by a Thermo Scientific Talos F200S transmission electron microscope in the Guangzhou Institute of Geochemistry, Chinese Academy of Sciences.

The CaO contents were analyzed in the Qingdao Sibada Instrumental Analysis Co., Ltd.. 40 mg sample powder was reacted with 1.5 ml concentrated hydrofluoric acid and 0.5 ml concentrated nitric acid in a sealed Teflon beaker under 180 °C for 12 hours. After cooled down, the solution was dried under 150 °C. Then, the leachate was treated with 50% nitric acid in the sealed Teflon beaker under 150 °C for 12 hours. The cooled solution was diluted appropriately and analyzed by a Thermo Scientific IRIS Intrepid II XSP ICP-OES.

The contents of total organic carbon (TOC) were measured in the Third Institute of Oceanography, Ministry of Natural Resources. About 1 g sample was reacted with excess 4 mol/dm^3 HCl at room temperature for 24 hours. After rinsed with deionized water to neutral pH and dried in an oven at 60 °C, the residue was weighed and, then, ground into powder and sieved by a 60 mesh sieve. A certain amount of powder was sealed in a tin cup and analyzed by a Sercon Integra2 elemental analysis-stable isotope ratio mass spectrometer (EA-IRMS). The mass accumulation rates of TOC (TOC MAR) were calculated by TOC multiplied by MAR cited from Lyle et al. (2002b) and Coxall et al. (2005) for the Site 1218 and Erhardt et al. (2013) for the Site 1333 with similar ages.

3 Results

3.1 Sediment magnetism

The profiles of magnetic susceptibility, $IRM_{1\,T}$, ARM, HIRM, bulk coercivity, and the ratios of ARM verses susceptibility and $IRM_{1\,T}$ varying with age of the two sites are shown in Fig. 2 and Table S1. The EOT is a significant turning point in all the profiles. In the Site 1218, the values of susceptibility, $IRM_{1\,T}$, ARM, and HIRM before the EOT are sharply higher than those after the EOT, whereas the bulk coercivity and the two ratios reveal an opposite trend. Although the latter three proxies begin to rise only after Oi-1, this could be because of the lack of sample around Oi-1. The carbonate-free basis (CFB) profiles of susceptibility, $IRM_{1\,T}$, ARM, and HIRM reveal the same trend as their un-CFB counterparts, but show small peaks at around 33.2 Ma. Similarly in the Site 1333, the profiles of susceptibility, $IRM_{1\,T}$, ARM, and HIRM show an obvious drop at around the EOT. In contrast, the values of bulk coercivity and the ratios of ARM over susceptibility and $IRM_{1\,T}$ reveal an opposite trend.

The hysteresis parameters of all samples assemble in the PSD area of the Day plot (Fig. 3, Table S2). FORC diagrams of representative samples from before and after the EOT are similar, showing obvious peaks between 30 mT and 40 mT in central ridge accompanied with areas spreading along the B_u axis (Fig. 4). The magnetic intensities are lower in samples after the EOT (Fig. 4). The IRM component analyses detected similar components before and after the EOT (Fig. 5a to c). Six major components are fitted out in the samples from the Site 1218 (Table S3). Mean coercivities are 6.4 mT, 14.8 mT, 37.2 mT, 74.4 mT, 171 mT, and 457 mT, and the corresponding average dispersion parameters (DP) are 0.13, 0.14, 0.19, 0.12, 0.14, and 0.13, respectively. Coercivity component results are similar in the Site 1333 (Table S3). The six major components are 5.8 mT, 14.3 mT, 37.2 mT, 75.1 mT, 168 mT, and 464 mT, while the corresponding mean DP are 0.12, 0.15, 0.19, 0.13, 0.15, and 0.12. The component with average coercivity around 37 mT contributes most gradient (Table S3; Fig. 5a to c). The magnetic signals of biogenic soft (BS, about 37 mT) and hard (BH, about 74 mT) components are plotted against age in Fig. 5. The IRM-CFB values of BS from both sites are slightly decreased after the EOT, while those of BH from the Sites 1218 and 1333 show little change and a small increase, respectively (Fig. 5d and j). Similar to the IRM-CFB values, the IRM contributions of BS are also slightly reduced after the EOT. In contrast, those of the BH are increased from about 4~5% to 8~10% (Fig. 5e and k). In the plots of IRM over susceptibility and $IRM_{1\,T}$, the ratios of BS do not show obvious changes in the Site 1218 before and after the EOT, whereas those in the Site 1333 are elevated after the EOT. The two ratios of BH from both sites synchronously jump onto a higher level after the EOT (Fig. 5f, g, l, and m). In the plots of coercivity and DP variation of BS and BH, no obvious trends are found before and after the EOT (Fig. 5 h, i, n, and o).

3.2 Characteristics of major phases, total organic carbon flux, and magnetic particles

XRD results from the two sites show that most samples are composed of (Mg)-calcite, biogenic SiO_2, smectite, and halite. Additional quartz and barite were detected in samples before the EOT (Table 2 and Fig. 6). An exception is found in the sample A099 of the Site 1218, in which (Mg)-calcite was undetectable. SEM observations agree with XRD results. In the samples before the EOT, nannofossils ((Mg)-calcite)

and biogenic SiO_2 are the common phases and round barite particles in several μm are obvious, whereas the major phase changes into nannofossils with less biogenic SiO_2 in samples after the EOT (Fig. 7).

The TOC contents are overall higher before the EOT (Table 2). The TOC values of the Site 1218 range between 0.043% and 0.461% and from 0.016% to 0.203% before and after the EOT, respectively (Table 2). In the Site 1333, the corresponding ranges are 0.093% to 0.125% and 0.022% to 0.096%, respectively (Table 2). In contrast, the TOC MAR values are higher after the EOT (Fig. 2e and m; Table 2). The trend is obvious in the Site 1333. Before the EOT, the values vary between 1.75×10^{-2} and 3.14×10^{-2} g/cm^2/ka, whereas after the EOT, the values increase to the range of 4.24×10^{-2} to 10.1×10^{-2} g/cm^2/ka, peaking at about 33 Ma (Fig. 2m; Table 2). Although the difference between before and after the EOT in the Site 1218 is not obvious because of the two abnormally high values, there is a rising trend after the EOT in the period between 35.5 and 32.3 Ma, showing an increase from the range between 1.33×10^{-2} and 3.79×10^{-2} g/cm^2/ka to 1.22×10^{-2} and 8.63×10^{-2} g/cm^2/ka (Fig. 2e; Table 2).

Magnetic particles, with sizes of several μm, and magnetofossils, in 10s to 100s nm, were observed in the magnetic extraction particles (Fig. 8a to c). According to the major compositions (Fe, O, and Ti) and electron diffraction patterns, most μm-scale particles are maghemite and magnetite (Fig. 8d and e), while a few hematites were also detected (Fig. 8f). Magnetofossils are commonly cuboctahedral, elongated prismatic, and bullet-shaped, scattering around aeolian particles (Fig. 8a to c) or assembling into chains (Fig. 9a and b). Main mineral phases are magnetite and maghemite according to diffraction patterns at specific zone-axis (Fig. 9c to f). The lengths and axial ratios (width/length) of magnetofossils formed before and after the EOT are distributed in a similar range in the plot of axial ratio vs. length (Fig. 10a and d). Most magnetofossils are with a length around 55 nm (Fig. 10b and e) and an axial ratio between 0.9 and 1 (Fig. 10c and f). There is an increase in the counts of lengths between 90 nm and 120 nm (Fig. 10e) and counts of axial ratios from 0.2 to 0.5 (Fig. 10f) in magnetofossils after the EOT. The abundance of the bullet-shaped magnetofossil after the EOT is over twice as much as their counterparts formed before the EOT (Fig. 10g). Among the cuboctahedral and elongated prismatic magnetofossils, slightly more elongated ones were observed in samples after the EOT (Fig. 10h and i).

4 Discussion

4.1 Variation of magnetic component before and after the EOT

Combining rock magnetism and mineralogy results, we can analyze the changing nature of the magnetic components before and after the EOT. TEM images show that the magnetic particles predominantly consist of pseudo-single-domain (PSD) and SD particles (Moisescu et al., 2014), which correspond to μm-scale component and magnetofossils (Dieudonné et al., 2019), respectively (Fig. 8). As the two sites are

situated far from terrestrial areas and the psephicity of the PSD particles is low (Fig. 8), most PSD particles should be aeolian origin. The FORC diagrams, showing peaks between 30 to 40 mT in central ridge accompanied with spreading areas along the B_u axis (Fig. 4), support this observation and suggest that magnetofossils are a major component. The IRM acquisition curves can distinguish the major magnetic components. Previous work identified the cuboctahedral magnetofossils and the elongated and bullet-shaped ones as biogenic soft (BS) and biogenic hard (BH) components, respectively (Egli, 2004). Their coercivity ranges are approximately 30 to 40 mT (Savian et al., 2016; Usui et al., 2017; Yamazaki & Ikehara, 2012; Yamazaki & Shimono, 2013; Yamazaki et al., 2019) and 60 to 70 mT (Usui et al., 2017; Yamazaki & Ikehara, 2012; Yamazaki et al., 2019). According to TEM observation and FORC results, the magnetic phases with coercivities of about 37 mT and 74 mT are recognized as the BS and BH components. The slightly higher coercivity values for our BH components may be caused by different morphologies, microstructure, or assemblage patterns of the chains of elongated magnetofossils (Berndt et al., 2020; Chang et al., 2019). Other magnetic components of about 6 and 14 mT should be aeolian PSD particles, while those of coercities higher than 160 mT could probably include hematite, as observed in the TEM (Fig. 8f).

Susceptibility, $IRM_{1\,T}$, ARM, and HIRM in bulk samples are sharply decreased after the EOT (Fig. 2), which could be caused by changes in MAR, carbonates, and the content of magnetic particles. The profiles of magnetic results do not change with the variation of MAR profiles (Fig. 2). The synchronous variations in the profiles of magnetic results with their CFB counterparts also exclude the effect of carbonate dilution as the main factor (Fig. 2). Consequently, the intensity drops of magnetic signals after the EOT are likely caused by the reduction in the abundance of aeolian particles and magnetofossils. Reduced supply of aeolian particles after the EOT is indicated by the drop of HIRM (Fig. 2). In the equatorial Pacific Ocean, the source of aeolian particles north and south of Intertropical Convergence Zone (ITCZ) is different. North of the ITCZ, aeolian particles in the equatorial Pacific Ocean originate from Asia and the supply was enhanced during glacials, whereas south of the ITCZ, aeolian particles dominantly originated from the Central and South American, and increased during interglacials (Rea, 1994). Therefore, the equatorial Pacific Ocean might be already south of ITCZ before the EOT. As the glacial age began during the EOT and Oi-1, the aeolian flux from Central and South America was decreased. The decrease of the abundance of magnetofossils should be related with the reduced flux of aeolian particles, because iron is important for the metabolism of MTB (Roberts et al., 2011). But in pelagic red clay in western North Pacific Ocean, the increase of aeolian supply did not raise the abundance of magnetofossils across the EOT (Yamazaki et al., 2020). It is because that organic matter, additional to iron, is also required by the MTB (Roberts et al., 2011). The content of organic matter in pelagic red clay is very low (D'Hondt et al., 2015; Usui et al., 2017; Yamazaki & Shimono, 2013), limiting the proliferation of MTB even though the supply of iron by aeolian particles is increased. In the eastern equatorial Pacific Ocean, the input of organic matter was enough for the metabolism of MTB (section 4.2). Thus, the

reduction of the abundance of magnetofossils was due to the decrease of aeolian supply.

Across the EOT, the bulk coercivity and ratios of ARM/χ and ARM/$IRM_{1\,T}$ increase in various degrees (Fig. 2). Since the coercivity of SD particles are higher than those of PSD particles and ARM represents the signals of fine magnetic particles, the elevated bulk coercivity and ratios imply the relative increase of magnetofossils in the magnetic particles. Magnetic signals of BS and BH components give more details. Although the IRM-CFB and IRM contributions of the BS component are slightly decreased after the EOT, its ratios of IRM over susceptibility and $IRM_{1\,T}$ are little changed in the Site 1218 (Fig. 5f and g) and increased in the Site 1333 (Fig. 5l and m). All those proxies of the BH component from both sites are increased with various degrees after the EOT (Fig. 5f, g, l, and m). Moreover, the statistics of magnetofossils size measurement, the abundance change of bullet-shaped magnetofossils, and comparison of sizes of cuboctahedral and elongated prismatic magnetosomes before and after the EOT suggest relative increase of elongated magnetofossils after the EOT, especially for the bullet-shaped ones (Fig. 10).

4.2 Synchronous variation of magnetofossils to the environmental change

Before the tremendous climate change starting from the EOT, the peaks in the barite accumulation rate (BAR) profiles and abundant barite observed by SEM (Fig. 7) indicate active primary productivity and abundant organic matter imported into pelagic sediments (Fig. 2; Erhardt et al., 2013; Griffith et al., 2010). During the period of the EOT and the Oi-1, cooling, Antarctica glaciation, and the opening of gateways around Antarctica greatly changed the conditions, ocean currents, and nutrient distribution in different oceans, modifying the ecological structure and primary productivity (Coxall & Pearson, 2007; Eldrett et al., 2009; Lear et al., 2004; Zachos et al., 1996). Enhanced primary productivity has been reported in the Southern Ocean (Coxall & Pearson, 2007; Plancq et al., 2014; Salamy & Zachos, 1999). Large changes also occurred in the eastern equatorial Pacific Ocean. Although low BAR values indicate low surface productivity (Erhardt et al., 2013; Griffith et al., 2010), newly established ocean currents transported nutrients from the Southern Ocean to the deep of the equatorial Pacific Ocean (Coxall et al., 2011; Lear et al., 2000; Moore Jr., 2013; Moore Jr. et al., 2014) and triggered the upwelling of deep water after the Oi-1 (Funakawa et al., 2006; Moore Jr. et al., 2014), stimulating not only the peaks in the benthic foraminifera accumulation rate (BFAR) profile during the Oi-1 and at about 33 Ma (Coxall & Wilson, 2011) but also multiple peaks in the profile of diatom over radiolarian ratios in the noncarbonate > 63 μm fraction after the Oi-1 (Moore Jr. et al., 2014). A certain amount of organic matter was still imported into the seafloor, which is also supported by the discrepancy of $\delta^{13}C$ between the planktonic and benthic foraminifera across the EOT (Coxall & Wilson, 2011; Moore Jr. et al., 2014). However, the seawater temperature was much higher before the EOT (Liu et al., 2009), which increased the rates of basal metabolism and nutrient cycling that consumed organic matter (Olivarez Lyle & Lyle, 2006). Therefore, the input of organic carbon into the pelagic sediments of the eastern equatorial Pacific Ocean after

the EOT should be amplified, which is supported by the increased TOC MAR values (Fig. 2e and m; Table 2)

In accordance with the environmental change after the EOT, the proportion of magnetofossils in the magnetic particles increased and, additionally, the percentage of bullet-shaped magnetofossils is also increased (Fig. 2, 5, and 10, Table 3, section 4.1). MTB, especially those producing bullet-shaped magnetofossils, prefer the relatively reducing environment in and near the OATZ (Bazylinski & Frankel, 2004; Dieudonné, et al., 2019; Hesse, 1994; Yamazaki, 2012; Yamazaki et al., 2019; Yamazaki & Shimono, 2013). Organic matter brings reducing conditions to the pelagic sediment with oxidizing conditions. Proper amounts of organic matter supply will increase chemical gradient in the sediment and shallow the depth of OATZ in the sediment (Froelich et al., 1979; Roberts et al., 2011; Usui et al., 2017). Sedimentary rate influences the quantity of organic matter preserved in sediments, which affects the redox gradient of OATZ (Lean & McCave, 1998; Kopp et al., 2007; Kopp & Kirschvink, 2008; Yamazaki & Ikehara, 2012). The variation of oxygen concentration in bottom-water and pore water will directly change the depth of OATZ (Hesse, 1994; Kopp et al., 2007; Kopp & Kirschvink, 2008; Lean & McCave, 1998). The magnetism proxies do not vary with the change of sedimentary rate (Fig. 2). Since the relative contents of magnetofossils are, instead, increased under the influence of oxygen containing water from the Southern Ocean after the Oi-1 (Fig. 2 and Coxall & Wilson, 2011; Miller et al., 2009; Page et al., 2019), the oxygen concentration of the bottom-water was not the main factor that drives the change of magnetofossil assemblages. Therefore, we propose that the change in magnetofossil morphology was mainly controlled by the abundance of organic carbon in the sediment. As indicated by the TOC MAR, the flux of organic carbon was increased after the EOT (Fig. 2e and m). Accordingly, the ratios of ARM over χ and $IRM_{1\,T}$ of the bulk sediment are elevated (Fig. 2). Moreover, the BH IRM CFB, BH IRM contribution, and ratios of BH IRM over χ and $IRM_{1\,T}$ change synchronously with TOC MAR values (Fig. 5). These trends are more obvious in the Site 1333. Reducing conditions provided by the organic matter increased the chemical gradient in the sediment and shallowed the depth of OATZ. Supplied with sufficient iron from aeolian particles, a stable OATZ with increased redox gradient favored the metabolism of MTB, especially for those producing elongated and bullet-shaped magnetosomes, leaving relatively more magnetofossils than other magnetic particles in sediments after the EOT.

5 Conclusions

The large climate change between late Eocene and early Oligocene caused environmental variation in eastern equatorial Pacific Ocean. Magnetic results and mineralogical observation from the Sites 1218 and 1333 indicate relevant alterations after the Eocene-Oligocene transition (EOT): 1) the content of magnetic particles decrease but the major components remain almost unchanged; 2) the percentages of

magnetofossils in magnetic particles and bullet-shaped magnetosomes in magnetofossils are both increased. The content decrease of magnetic particles coincided with the reduction of aeolian supply. Organic matter supply was maintained by the high productivity in surface or deep sea before and after the EOT. However, more organic matter was preserved in sediments after the EOT because of the low basal metabolism caused by lower temperature, which is supported by the increased mass accumulation rates of total organic carbon. Higher organic matter flux increased the redox gradient in the oxic-anoxic transition zone (OATZ). Consequently, the proportions of magnetofossils in magnetic particles and bullet-shaped ones in magnetofossils were elevated. The assemblage patterns of magnetofossils faithfully document the environmental change during late Eocene and early Oligocene, contributing to the hypothesis considering magnetofossils as a proxy for paleoenvironment.

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Figure 1. Map of the eastern equatorial Pacific Ocean. The ODP Site 1218 and IODP Site 1333 are indicated by yellow stars.

Figure 2. The sedimentary phases and the plots of age vs. susceptibility (χ), 1 T isothermal remanent magnetization ($IRM_{1\ T}$), anhysteretic remanent magnetization (ARM), hard isothermal remanent magnetization (HIRM), bulk coercivity, mass accumulation rates of total organic carbon (TOC MAR), and ratios of ARM over χ and $IRM_{1\ T}$ of the ODP Site 1218 across the Eocene-Oligocene transition (EOT) and Oligocene isotope event 1 (Oi-1) (panel 1, a to f), accompanied with the age profiles of high-resolution susceptibility (a, Lyle et al., 2002a), carbon and oxygen stable isotope compositions of benthic foraminifera (g, Coxall et al., 2005), benthic foraminifera accumulation rate (BFAR, h, Coxall and Wilson, 2011), barite accumulation rate (BAR, h, Griffith et al., 2010), diatom/radiolarian ratio (D/R) > 63 μm (h, Moore Jr. et al., 2014), and bulk mass accumulation rate (MAR, h, Coxall et al., 2005), and those of the IODP Site 1333 (panel 2, i to p), compared with the age profiles of high-resolution $IRM_{0.9\ T}$ and ARM (j and k, Yamazaki et al., 2003), carbon and oxygen stable isotope compositions of benthic foraminifera (o, Coxall et al., 2005), barite accumulation rate (BAR, p, Erhardt et al., 2013), D/R > 63μm (p, Moore Jr. et al., 2014), and bulk mass accumulation rate (MAR, p, Erhardt et al., 2013). The χ, $IRM_{1\ T}$, ARM, and HIRM were also calculated on a carbonate free basis (CFB, a to d and i to l). Pink dots beside the sedimentary phases represent the positions of the samples shown in Figs. 4 and 5.

Figure 3. Day plot of the samples from the Sites 1218 and 1333, modified after Dunlop (2002). SD = single-domain. PSD = pseudo-single-domain. MD = multidomain.

Figure 4. First-order reversal curves diagrams of selected samples across the Eocene-Oligocene transition (EOT) and Oligocene isotope event 1 (Oi-1).

Figure 5. Isothermal remanent magnetization (IRM) component analyses of representative samples (panel 1, a to c) and isolated magnetic signals of biogenic soft (BS) and biogenic hard (BH) components vs. age from the Sites 1218 (panel 2) and 1333 (panel 3) across the Eocene-Oligocene transition (EOT) and Oligocene isotope event 1 (Oi-1), including IRM-carbonate free basis (CFB) values (d and j), IRM contributions (e and k), ratios of IRM over χ (f and l) and $IRM_{1\ T}$ (g and m), $B_{1/2}$ values (coercivity, h and n), and dispersion parameters (DP, i and o). The gray lines are corresponding mass accumulation rates of total organic carbon profiles from Fig. 2 (d to g and j to m).

Figure 6. Typical X-ray diffraction profiles and the identified major phases.

Figure 7. Scanning electron microscopy images of selected samples from after (a and c, A128 and B056) and before (b and d, A104 and B062) the Eocene-Oligocene

transition (EOT). The inset in B is the energy-dispersive X‐ray spectroscopy profile of barite. The peaks of Al and Si are from nearby smectite. Brt = barite.

Figure 8. High angle angular dark field transmission electron microscopy (TEM) images of the extracted magnetic particles from A133 (a), A103 (b), and B061 (c), and TEM images of aeolian maghemite from A099 (d), magnetite (e), and hematite from B061 (f) with inset of electron diffraction patterns and energy-dispersive X‐ray spectroscopy profiles. Yellow arrows in (a) to (c) point out the magnetofossil aggregates or chains.

Figure 9. High angle angular dark field transmission electron microscopy (TEM) images of magnetofossil chains from A133 (a) and A103 (b) and high resolution-TEM images of bullet-shaped (c and e, A133 and B059) and cuboctahedral magnetofossils (d and f, A103 and B063) with inset fast Fourier transformation patterns.

Figure 10. Plots of axial ratio (width/length) vs. length (a and d) and histograms of length (b and e) and axial ratio (c and f) of cuboctahedral, elongated, and bullet-shaped magnetofossils before (panel 1) and after (panel 2) the Eocene-Oligocene transition (EOT) and abundance and size comparisons of magnetofossils before and after the EOT (panel 3), illustrating by a column diagram of the percentages of bullet-shaped magnetosomes (g) and box charts of length (h) and axial ratio (i) of cuboctahedral and elongated prismatic magnetofossils. Diamond in (h) and (i) are outlier data points.

Table 1. The depths and ages of selected samples from the Site 1218 and the Site 1333.

Site	Sample	Depth (m)[a]	Age (Ma)[b]
1218B	A128	225.48	32.19
	A129	226.50	32.29
	A130	230.13	32.64
	A131	231.71	32.80
	A132	233.13	32.93
	A133	236.13	33.22
1218A	A098	236.49	33.26
	A099	243.12	33.90
	A100	245.02	34.24
	A101	246.52	34.51
	A102	249.23	35.00
	A103	251.83	35.47
	A104	254.69	35.99
	A105	256.07	36.24
	A106	257.71	36.57
1333B	B053	99.95	32.08
	B054	101.45	32.24
	B055	103.48	32.45
	B056	106.45	32.77
	B057	107.95	32.93
	B058	109.45	33.09
	B059	111.93	33.36
	B060	114.45	33.62
	B061	115.95	33.93
	B062	117.45	34.38
	B063	120.65	35.33

[a] Depth of Site 1218 is composite depth (mcd, Lyle et al., 2002), while depth of Site 1333 is core depth below seafloor (CSF, Pälike et al., 2010).

[b] The ages of Site 1218 samples were calculated according to some ages in the table 15 of Lyle et al. (2002) and interpolation with mcd, while those of Site 1333 samples were based on some ages in the table 24 of Pälike et al. (2010) and interpolation with CSF.

Table 2. Semi-quantification of the main phases, contents of total organic carbon, mass accumulation rates, and mass accumulation rates of total organic carbon of the Site 1218 and the Site 1333.

Sample	Mg-calcite	Bio-SiO_2	Smectite	Quartz	Barite	Halite	TOC[a]	MAR[b]	TOC MAR ($*10^{-2}$)
	(wt%)							($g/cm^2/ka$)	
A128	87	9	2.2			1.8	0.203	0.692	14.1
A129	90	6.3	2.3			1.3	0.037	2.32	8.63
A130	90	2.9	4.9			1.9	0.021	1.62	3.37
A131	93	6	1.1				0.020	0.809	1.59
A132	88	7.4	3.3			1.3	0.026	0.784	2.07
A133	92	4	3.1			0.7	0.016	0.781	1.22
A098	94	3	2.5				0.038	0.814	3.07
A099		75	14	2.6	2.1	6.1	0.069	0.208	1.44
A100	26	52	18	1.8	1.9		0.073	0.232	1.71
A101	67	20	5.5	1.8	1.8	3.5	0.074	0.246	1.83
A102	72	21	5	0.9	0.9		0.056	0.239	1.33
A103	58	17	18.9	2.8	3.2		0.159	0.239	3.79
A104	40	34	12.9	4.2	4	4.5	0.461	0.340	15.7
A105	43	33	16	3.9	5		0.045	0.262	1.19
A106	53	18.1	5.2	23	0.9		0.043	0.423	1.81
B053	94	2.2	3.4			0.9	0.041	1.17	4.82
B054	96	2.4	1.2			0.4	0.048	1.05	5.07
B055	97	1.9	1.1				0.063	0.868	5.46
B056	95	1.3	2.7			1.4	0.080	0.949	7.61
B057	88	7.8	4.5				0.096	1.05	10.1
B058	92	1.9	5.6			0.7	0.042	1.05	4.45
B059	95	4.9	0.3				0.055	0.764	4.24
B060	90	3.4	6.2				0.022	0.508	1.13
B061	91	6	2.6				0.093	0.338	3.14
B062	75	14	4.2	4.4	1.1	1.1	0.125	0.199	2.49
B063	81	16	2.9	0.4	0.7		0.109	0.160	1.75

[a] Total organic carbon (TOC).

[b] Mass accumulation rates (MAR) are cited from Coxall et al. (2005) for A128 to A133 and A098 to A101, Lyle et al. (2002b) for A102 to A106, and Erhardt et al. (2013) for B053 to B063 with similar ages.

Table 3. Counts and ratios of the cuboctahedral, elongated prismatic, elongated rombic, and bullet-shaped magnetofossils of the samples from the Site 1218 and the Site 1333.

	Sample	Cuboctahedral	Bullet-shaped	Elongated rombic	Elongated prismatic	Total
Counts	A133	230	141	478	42	891
	A098	64	51	159	41	315
	A099	40	6	75	12	133
	A103	326	89	544	75	1034
	B055	102	80	314	1	497
	B059	72	20	193	2	287
	B061	204	51	572	26	853
	B063	22	2	72	0	96
Ratios (%)	A133	25.8	15.8	53.6	4.7	
	A098	20.3	16.2	50.5	13.0	
	A099	30.1	4.5	56.4	9.0	
	A103	31.5	8.6	52.6	7.3	
	B055	20.5	16.1	63.2	0.2	
	B059	25.1	7.0	67.2	0.7	
	B061	23.9	6.0	67.1	3.0	
	B063	22.9	2.1	75.0	0.0