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Variation of magnetic properties in sediments from Lake Towuti, Indonesia, and its paleoclimatic significance



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ABSTRACT

We investigated the mineral-magnetic behavior of sediments from Towuti located in the Eastern Sulawesi Ophiolite belt, Indonesia. Rock magnetic analysis supplemented by X-ray diffraction and scanning electron microscopy analysis were performed on sediment core TOW10-9B from the north basin of Lake Towuti to give insights on the environmental and sedimentary processes controlling the magnetic properties of the sediment and its paleoclimatic significance. The results show that the core has three distinct zones of varying magnetic properties. Careful examination demonstrates that these zones correspond to varying levels of iron oxide dissolution and magnetite precipitation that are climatically and environmentally dependent. The magnetically strongest zone is characterized by weak iron oxide dissolution and intense magnetite precipitation, likely driven by changes in the stratification and/or water level of the lake during dry conditions in Marine Isotope Stage 2 (MIS2) period, whereas the two magnetically weaker zones are characterized by signs of dissolution and correspond to relatively wet conditions, respectively, during Marine Isotope Stage 3 (MIS3) and the Holocene. Although our data show that major changes in concentration dependent parameters, such as magnetic susceptibility and saturation isothermal remanent magnetization (SIRM), in Lake Towuti sediment correlate with changes in regional rainfall, many of the concentration changes are more strongly affected by in situ chemical processes than by changes in erosion and terrestrial sediment supply. These findings urge caution in the interpretation of magnetic mineral concentration profiles as indicators of clastic sediment inputs.

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1. Introduction

The magnetic minerals in lacustrine sediments have been widely used to study geo-environmental and paleoclimatic changes (e.g. Williamson et al., 1998; Stockhausen and Zolitschka, 1999; Peck et al., 2004; Asikainen et al., 2007; Wang et al., 2010; Duan et al., 2014). Variations of sedimentary magnetic properties can often reflect hydrologically driven processes such as soil erosion within the catchment area and sediment inputs to lakes (Thompson et al., 1975; Dearing et al., 2001; Duan et al., 2014), providing a valuable proxy for changing rainfall. The magnetic properties of lacustrine sediments, however, can also be affected by redox-related geochemical processes as magnetic minerals are highly reactive and can either be precipitated, or dissolved

in situ (Anderson and Rippey, 1988; Zhang et al., 1996; Vigliotti et al., 1999; Demory et al., 2005). For instance, reductive diagenesis, which commonly occurs in anoxic environments, can reduce the concentration of magnetic minerals (Bloemendal et al., 1993), but on the other hand, it also can stimulate the growth of secondary magnetic minerals (Tarduno, 1995). Although all of the previously mentioned processes are climatically driven, the contribution of each process needs to be disentangled because these processes might vary greatly in different environmental settings.

Lake Towuti is situated on the Island of Sulawesi (or Celebes) in central Indonesia. Its catchment is dominated by ultramafic rocks of the East Sulawesi Ophiolite (ESO) belt, one of the three largest ophiolite complexes in the world (Kadarusman et al., 2004). The humid-tropical climate has led to deep weathering of the ultramafic rocks, producing a magnetically strong lateritic soil rich in chromium, and nickel (Golightly and Arancibia, 1979). Located in the center of the Indo-Pacific Warm Pool (IPWP), Lake Towuti is an important site for paleoclimate research (Russell and Bijaksana, 2012). Previous work by

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Russell et al. (2014) has shown that hydrological variability in this lake, recorded by changes in detrital sedimentation and vegetation, correlate with high-latitude climate forcing. However, the mechanism by which Lake Towuti sediments respond to climate change has not been well determined.

In this study, rock magnetic investigations supported by X-ray diffraction (XRD) and scanning electron microscope (SEM) observations were performed on a sediment core from Lake Towuti in order to understand the environmental processes controlling magnetic signals recorded in Lake Towuti's sediment. Comparison of river sediments, soil, and lake sediment allows us to interpret and contextualize down-core variation in magnetic parameters. Variations in sediment magnetic properties throughout the core were combined with age model presented by Russell et al. (2014) to interpret the climatic causes of magnetic variations and their significance to paleoclimate.

2. Site description and materials

Lake Towuti is the largest lake in the Malili Lake System (MLS), located in the center of the Island of Sulawesi (Fig. 1). The MLS is comprised of five lakes, the three largest of which are interconnected tectonic basins (Lake Matano, Lake Mahalona, and Lake Towuti) while the other two (Lake Lontoa and Lake Masapi) are relatively small satellite lakes. Lake Towuti occurs along the southern side of the Matano fault and is estimated to have formed between 1 and 2 million years ago (Russell and Bijaksana, 2012) as a result of tectonic extension of the Tambalako valley (Van der Meulen, 1970). Lake Towuti is situated at 293 m above sea level, has a surface area of approximately 561 km², and a maximum depth of 203 m (Lehmusluoto and Machbub, 1997). The regional climate is humid-tropical with rainfall of 2737 mm per year (Hope, 2001). The bedrock geology surrounding the MLS is dominated by ultramafic rocks of ophiolitic origin, including hartzburgite, lherzolite, serpentinite, dunite, gabbro and diabase (Kadariusman et al., 2004).

In 2010, the Indonesian Deep Lake Expedition (IDLE) retrieved eleven sediment cores from Lake Towuti using a Kullenberg piston coring system. For this study we analyzed one of these cores, TOW10-9B (length of 11.50 m) which was taken from a water depth of 154 m from the center of the north basin of Lake Towuti. Earlier, based on 23 radiocarbon dates, Russell et al. (2014) showed that core TOW10-9B presents a record since about 60 kyr BP. Moreover, Costa et al. (in press) showed that Al, and K contents in the sediment show similar variations, i.e., relatively low in the middle part of the core and relatively high in both the upper and the lower parts of the core.

3. Methods

Magnetic measurements on core TOW10-9B were carried out in two stages. First, we measured whole-core and unconfined samples to obtain continuous and high-resolution profiles of volume-specific magnetic susceptibility (κ), anhysteretic remanent magnetization (ARM), and SIRM. The measurement of κ was carried out using a Geotek™ Multi-Sensor Core Logger (MSC) with a measurement interval of 1 cm. This parameter approximates the concentration of ferrimagnetic minerals in the core. ARM was imparted by applying a decaying alternating field (peak field 100 mT) with the superimposed steady magnetic field of 0.05 mT. ARM approximates the concentration of remanence carriers, predominantly single domain and fine pseudo-single domain magnetic minerals. SIRM was imparted by exposing the sample to a strong magnetic DC field of 1 T. Both ARM and SIRM were measured using a 2G-Enterprises Cryogenic Magnetometer with a measurement interval of 1 cm.

Second, we performed detailed magnetic measurements on selected discrete samples including mass-specific magnetic susceptibility at low and high frequencies (470 Hz and 4700 Hz), temperature-dependent susceptibility, and magnetic hysteresis analysis. The mass specific magnetic susceptibility was measured using a Bartington MS2 system using an MS2B sensor. The low-frequency mass-specific magnetic susceptibility is denoted χ_{LF} , while that at high frequency is denoted χ_{HF} . These two parameters give rise to a frequency dependent susceptibility (χ_{FD}) that provides a measure of the relative contribution of superparamagnetic (SP) grains in samples, defined as:

$$\chi_{FD}(\%) = \frac{(\chi_{LF} - \chi_{HF})}{\chi_{LF}} \times 100\% \quad (1)$$

Temperature-dependent measurements of mass-specific magnetic susceptibility (χ) were performed using a KLY-2 Kappabridge AC susceptibility meter with an alternating field of 300 A/m and a frequency 920 Hz. The measurements were made while heating the samples in an argon atmosphere to a maximum temperature of 700 °C followed by cooling to 40 °C. These measurements were made to determine the magnetic mineralogy of the samples through observation of the Curie temperature (T_C) and thermally-marked changes in mineralogy (Dunlop and Özdemir, 1997). Magnetic hysteresis parameters were determined by measuring the magnetic hysteresis curves as well as the magnetic backfield curves using a Micromag AGM (Alternating Gradient Force Magnetometer) 2900. Three hysteresis parameters, namely saturation magnetization (M_s), saturation remanence (M_{rs}), and coercivity (B_c) were determined from hysteresis curves, whereas the coercivity remanence (B_{cr}) was determined from the magnetic backfield curve.

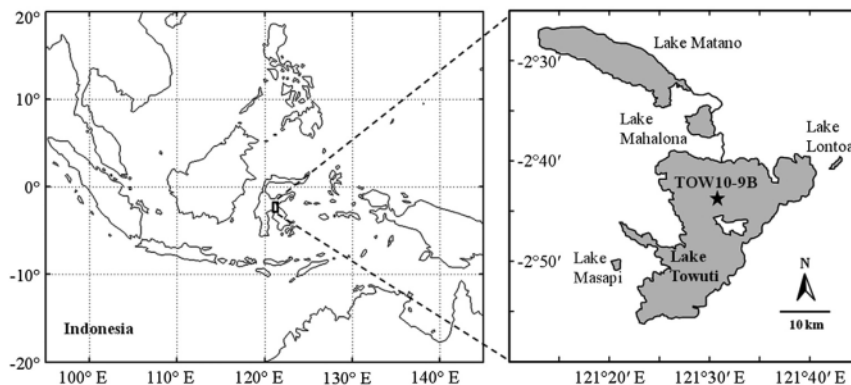


Fig. 1. Map of the Malili Lake System and the coring site TOW10-9B (black star) in Lake Towuti. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

These parameters were used, among others, to infer the magnetic domain state as well as the predominant grain size of ferrimagnetic minerals.

These magnetic measurements were complemented by the analysis to determine the mineralogy of selected samples and by SEM coupled with energy dispersive X-ray (EDX) analyses to image the morphology of extracted magnetic grains and their surface composition. The XRD analysis was conducted using a Phillips X'Pert PRO MRD PW3050/60 with CuK α radiation while SEM–EDX analysis was performed with a JEOL JSM 6510 LA. SEM analyses were undertaken on magnetic grains extracted from six representative samples. The grains were extracted by mixing the samples with alcohol, as it is easier to evaporate to obtain a dry extract and to inhibit oxidation of iron sulfides (Nowaczyk, 2011), following the procedure described in Safiuddin et al. (2011).

The measurements of ARM, SIRM, χ_{LF} , χ_{HF} and hysteresis parameters were conducted in the Paleomagnetic Laboratory at the University of Idaho, whereas κ and temperature-dependent susceptibility properties were measured at the University of Minnesota's LacCore and Institute for Rock Magnetism, respectively. The XRD and SEM–EDX analyses were carried out at the National Nuclear Energy Agency and Institut Teknologi Bandung, respectively, in Bandung, Indonesia.

4. Results

Values of κ , ARM and SIRM throughout core TOW10-9B respectively vary between 81 and 787×10^{-6} SI, 0.13 to 1.33 Am^{-1} , and 1.36 to 49.33 Am^{-1} (Fig. 2). All the parameters show a pronounced stratigraphy that suggests that the core can be divided into three magnetic zones. The first zone (zone I), from 11.43 to 7.26 m, is characterized by relatively low values of κ ($182 \pm 44 \times 10^{-6}$ SI), whereas the second zone (zone II), from 7.25 to 2.81 m, is characterized by high values of κ ($588 \pm 102 \times 10^{-6}$ SI). The third zone, from 2.80 m to the top of the core, is characterized by low to moderate values of κ ($229.12 \pm 74.81 \times 10^{-6}$ SI). Variations of ARM resemble those of κ except in several short intervals where ARM decreases drastically. The average values of ARM are $0.52 \pm 0.09 \text{ Am}^{-1}$ (for zone I), $1.33 \pm 1.09 \text{ Am}^{-1}$ (for zone II), and $0.57 \pm 0.15 \text{ Am}^{-1}$ (for zone III). Although SIRM has larger

fluctuations within each zone, its trends resemble those of κ and ARM. The average values of SIRM are respectively $16.14 \pm 3.50 \text{ Am}^{-1}$, $29.38 \pm 8.01 \text{ Am}^{-1}$, and $17.95 \pm 6.84 \text{ Am}^{-1}$ (for zones I to III).

Temperature-dependent magnetic susceptibility measurements of discrete samples from 1.23 to 1.69 m (representing zone III), 3.49 to 7.10 m (representing zone II), 9.31 m to 11.43 m (representing zone I) suggest a fairly constant magnetic mineralogy through the sediment core (Fig. 3). Curie temperature of about 580°C observed in the samples from zones II and III indicates the predominance of magnetite, whereas the Curie temperature at about 600 to 610°C in the samples from zone I indicates magnetite with high-temperature phases, most likely maghemite that formed during heating. On the other hand, the increases in magnetic susceptibility during cooling suggest neof ormation of a ferrimagnetic mineral (Ortega et al., 2006).

Analyses of temperature-dependent susceptibility also indicate the presence of other magnetic minerals. The sharp increase in magnetic susceptibility observed in selected samples (except in zone I) at 325 to 375°C suggests neof ormation of ferrimagnetic minerals from pre-existing magnetic minerals. Based on the range of formation temperatures there are two probable pre-existing minerals, namely greigite (Fe_3S_4) (Liu et al., 2008) or siderite (FeCO_3) (Ortega et al., 2006). The XRD analyses confirm the presence of magnetite in all samples, while siderite is present in zone II but not in zone I (Fig. 4, red line, zone I; blue line, zone II). Neither pyrite, nor greigite were found in any of the samples, nor are these minerals expected to be present given the extremely low sulfur concentrations present in the Malili Lakes (Crowe et al., 2008). Grains of carbon-rich iron were detected in SEM–EDX analyses, also implying the presence of siderite (Fig. 5), but no iron-sulfide grains were found in the SEM analyses, confirming our XRD results. Thus, it is clear that the sharp increase of magnetic susceptibility during heating at 325 to 375°C is due to the presence of siderite.

Seventeen samples were subjected to magnetic hysteresis measurement to infer changes in magnetic domains through the core. Fig. 6 shows the typical hysteresis curves for representative samples from the three magnetic zones. The blue and red curves are, respectively, the original hysteresis curves before correction for paramagnetic slope and after correction. All the corrected curves saturate below 0.3 T suggesting magnetite (Fe_3O_4) as the predominant magnetic mineral

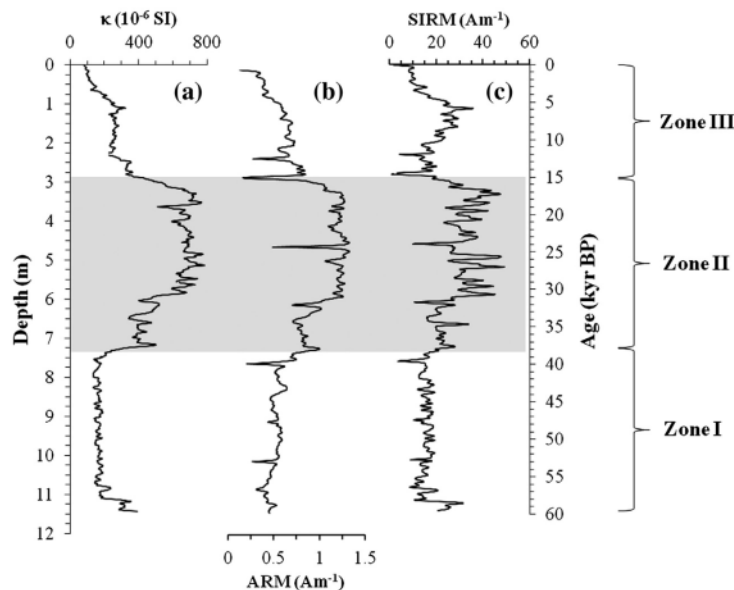


Fig. 2. Profiles of magnetic properties in core TOW10-9B. (a) Magnetic susceptibility (κ), ARM, and (c) SIRM. Gray shading indicates the section with high magnetic concentrations. Age data is after Russell et al. (2014).

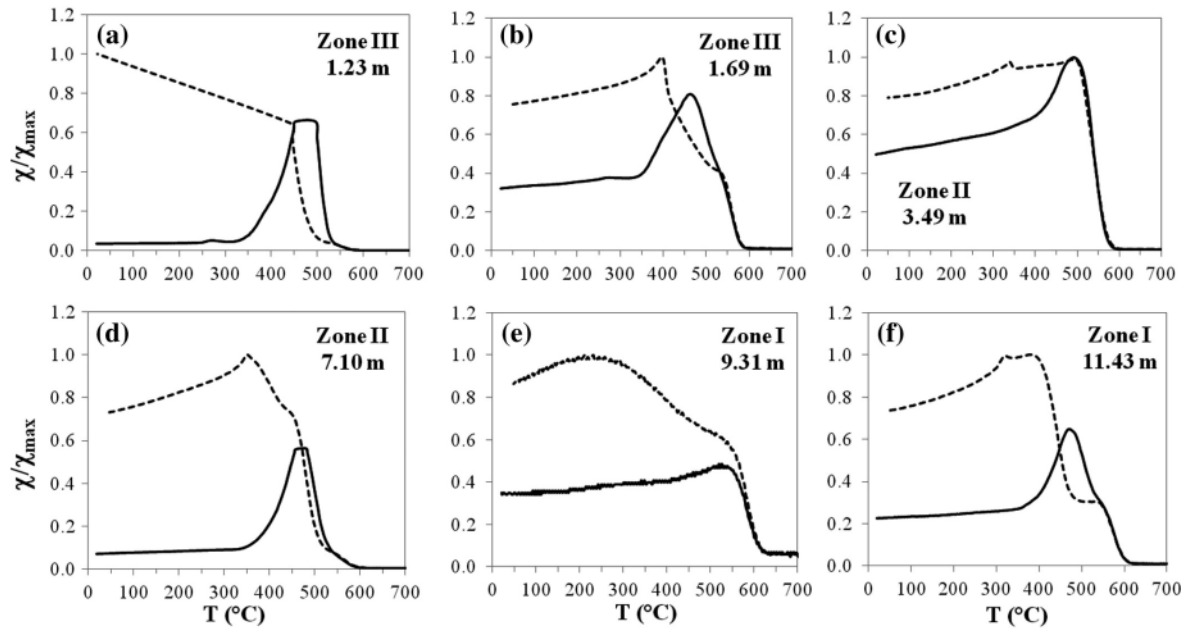


Fig. 3. χ -vs- T curves from six selected samples from core TOW10-9B. (a,b) represent samples from zone III, (c,d) represent samples from zone II, whereas (e,f) represent samples from zone I. The solid line is χ during heating, whereas the dotted line is χ during cooling. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

(Gehring et al., 2009). Moreover, based on their shape, all the corrected curves can be classified as narrow normal hysteresis curves indicating that the magnetic grains are predominantly pseudo-single domain (Tauxe, 2003). The gap between the corrected and uncorrected curves at 0.5 T indicates the relative contribution of the paramagnetic component to the overall magnetization. Compared with the samples from zones I and III, the sample from zone II shows a relatively narrow gap suggesting a smaller contribution of paramagnetic minerals or a larger contribution of ferrimagnetic minerals in zone II.

Fig. 7 shows the variation of hysteresis parameters with depth. Both M_s and M_{rs} show similar variations throughout the core, and

shift from 19.70 to $111.20 \times 10^{-2} \text{ Am}^2 \text{ kg}^{-1}$ for M_s and 3.15 to $15.75 \times 10^{-2} \text{ Am}^2 \text{ kg}^{-1}$ for M_{rs} . B_c and B_{cr} change throughout the core in a similar fashion and vary within the range of 8.74 to 12.67 mT for B_c and 21.05 to 25.74 mT for B_{cr} . The variation of M_s and M_{rs} resembles that of κ , with relatively low values in zones I and III, and high values in zone II. The variation of B_c and B_{cr} , on the other hand, differs from that of κ , with relatively high values in zones I and III, and low values at zone II. While magnetite is the predominant magnetic mineral throughout the core, then variation in κ , M_s , M_{rs} , and SIRM may reflect a variation in magnetite concentration. Thus, zones I and II, respectively have the lowest

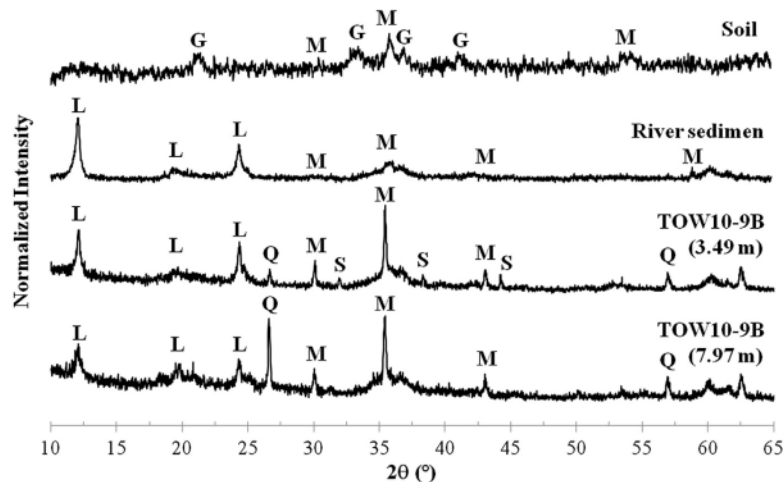


Fig. 4. X-ray diffractogram of selected samples of core sediment, river sediment and soil. Note: M = magnetite; G = goethite; S = siderite; L = lizardite; and Q = quartz. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

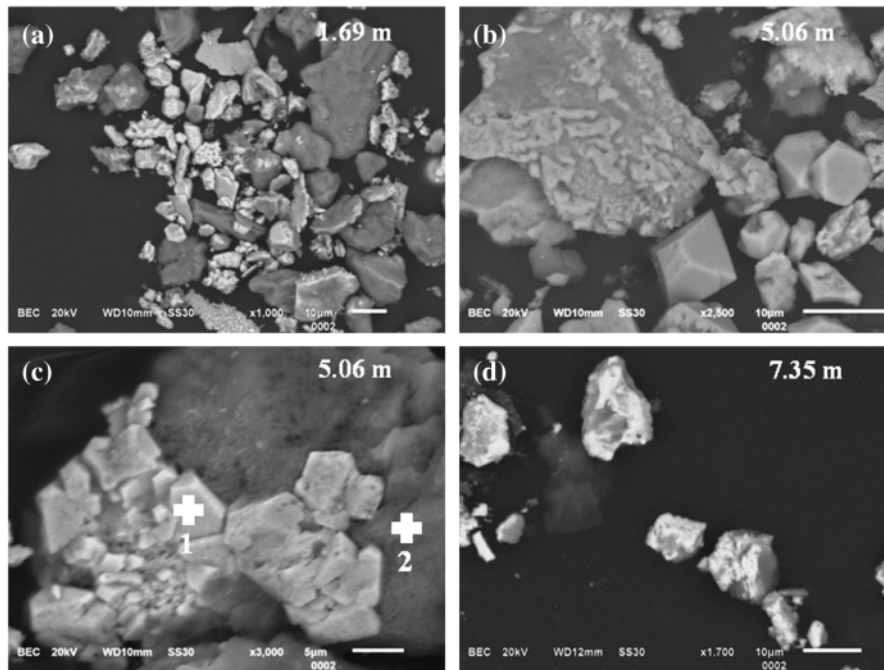


Fig. 5. SEM observations of magnetically extracted grains from sediment samples. (a) Fragmented iron-oxide grains in zone III, (b) bi-tetrahedral iron-oxide grains, most likely magnetite, with well-preserved surfaces in zone II, (c) carbon rich Fe grains, interpreted as siderite, with attached silicates [EDX +01: Fe = 46.31%, C = 36.20%, O = 15.26%; EDX +02: SiO₂ = 40.17%], and (d) magnetite grain with hollows and surface corrosion, probably an effect of magnetite dissolution (zone I).

and the highest concentrations of magnetite, whereas Zone III has low to moderate concentrations.

Magnetite grain sizes can be estimated from their magnetic domains through analyses of hysteresis parameters as well as ARM, SIRM, κ and χ_{FD} . Fig. 8a shows the plot of hysteresis parameters for Towuti samples in a Day diagram (Day et al., 1977) complemented by Dunlop's mixing curve between single domain (SD) and multi-domain (MD) grains (Dunlop, 2002). All the samples fall in the pseudo-single domain (PSD) zone suggesting that the predominant magnetic grains are 0.06 to 10 μm in size (Caitcheon, 1998). Alternatively, the proximity of the samples to the mixing curve implies that the magnetic grains could be a combination of SD and MD grains with a MD proportion between 65 and 75%.

A variation of fine versus coarse magnetic grains could also be inferred from the ARM/ κ , ARM/SIRM, χ_{FD} (Fig. 8b–c). ARM is sensitive to the concentration of SD and fine PSD grains, with higher concentrations of such grains giving higher ARM values (Wang et al., 2008), while κ is more sensitive to concentrations of MD or coarse grains (Dearing, 1999; Rowan et al., 2009) and also SP grains, i.e. magnetic grains 0.03 μm or smaller in size (Gubbins and Herrero-Bervera, 2007). SIRM is a function of grain size in a wide range i.e., ~0.04–400 μm (Evans and Heller, 2003). As SP grains contribute strongly to susceptibility but not to remanence, the presence of abundant SP grains might affect the grain-size interpretation of ARM/ κ . Therefore for the samples containing abundant SP grains, grain-size estimates are better given by ARM/SIRM. A variation of ARM/SIRM shows a relatively higher

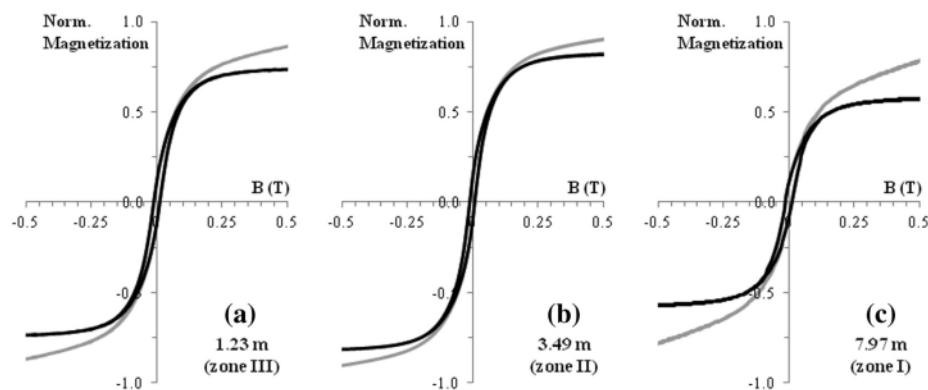


Fig. 6. Hysteresis curves from three representative samples from core TOW10-9B. (a) Sample from zone III, (b) sample from zone II, and (c) sample from zone I. The grey lines are the uncorrected magnetic hysteresis curves, whereas the black solid lines are the hysteresis curves after correcting the paramagnetic component. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

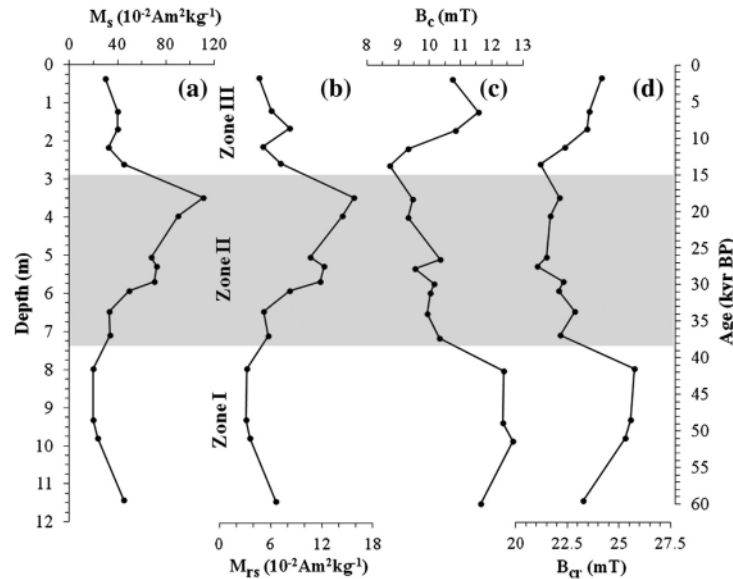


Fig. 7. Variation of hysteresis parameters with depth. (a) Saturation magnetization, (b) saturation remanent magnetization, (c) coercivity, and (d) coercivity remanence. Age data is after Russell et al. (2014).

value at zone II compared with zones I and III (Fig. 8b) suggesting that zone II has a relatively smaller proportion of coarse magnetic grains. As shown in Fig. 8c, zone II has higher values of χ_{FD} suggesting that this zone contains a higher concentration of SP grains.

5. Discussion

5.1. Controls on magnetic parameters in Lake Towuti

The magnetic susceptibility of Lake Towuti is comparable to that in other volcanic/basaltic lakes such as Lake Kinneret in Israel

(Nowaczyk, 2011), Lake Tritrivakely in Madagascar (Williamson et al., 1998), as well as Lake El'gygytyn in Siberia, an impact-crater lake which is also surrounded by volcanic rocks of diverse composition such as basalt and tuffs (Asikainen et al., 2007). Assuming that the magnetic susceptibility in these other lakes' sediments also depends largely on the concentration of magnetite, this would imply similar magnetite concentrations among these systems. In contrast, the concentration of magnetite in Lake Towuti sediment is higher than that in Lake Baikal, Siberia (maximum κ value of $\sim 500 \times 10^{-6}$ SI), which is surrounded by granitic rocks (Demory et al., 2005). Sediments in lakes surrounded by limestone or alluvial deposits, such as Lake Qarun in Egypt (with a

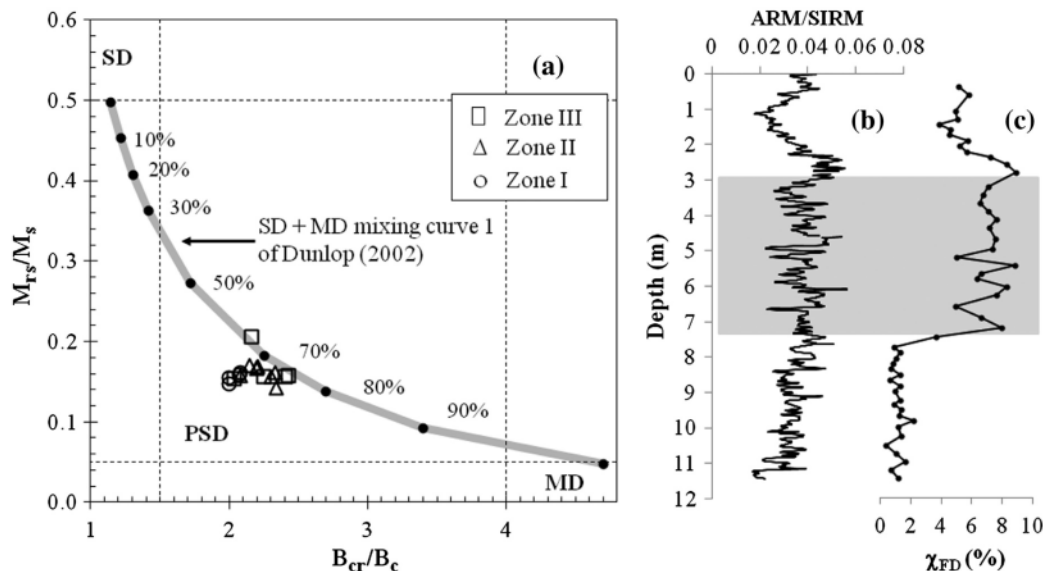


Fig. 8. Estimation of magnetic domain state in selected samples from core TOW10-9B. (a) A Day diagram showing the magnetic particles from the core are dominated by PSD grains; (b) profile of ARM/SIRM showing the relative variation of SD magnetic grains throughout the core, and (c) the change of χ_{FD} with depth indicating the variation of SP grains.

maximum κ value of $\sim 80 \times 10^{-6}$ SI) (Foster et al., 2008) and Lake Hurlig in China (maximum κ value of $\sim 100 \times 10^{-6}$ SI) (Zhao et al., 2010) have much lower concentrations of magnetite compared with Lake Towuti, confirming the primary importance of bedrock type and magnetic mineral supply in controlling magnetic susceptibility of the lake sediment.

In many lakes, variations in mineral magnetic concentration in lake sediments are related to the variation of detrital material influx into the lake. Higher detrital material influx is associated with high mineral magnetic concentration and vice versa (Thompson et al., 1975; Dearing, 1999). Many processes control the influx of detrital magnetic minerals to a lake, including rainfall intensity and variability, vegetation type, river networks, and other processes that control fluvial erosion, transport, and lacustrine sedimentation. In Lake Towuti, fragmented magnetic grains often found in SEM analyses (Fig. 7a) clearly indicate the presence of magnetic minerals that are detrital in origin (Nowaczyk, 2011). The presence of lizardite (Fig. 6) also confirms the allogenic sources of sediment to the lake, as this mineral is almost certainly eroded from the serpentinized rocks to the north.

Detrital influx, however, might not be the sole controlling factor for the variation in magnetic concentration. Russell et al. (2014) observed the Ti concentration in core TOW10-9B. Their interpretation of high Ti as strong rainfall is supported by other proxies such as $\delta^{13}\text{C}_{\text{wax}}$. As Ti concentration is sensitive to redox variation, Ti concentration in sediments, both marine and lacustrine, is often used as an indicator for detrital or terrigenous influx (Boyle, 2001; Ortega et al., 2006). The Ti concentration in core TOW10-9B is inversely proportional to the variations of κ and SIRM (Fig. 9). In zone II, high values of κ and SIRM strongly correlate with relatively low concentration of Ti while the opposite is true in zones I and III. These show that variations of magnetic mineral concentration in core TOW10-9B were affected not only by detrital influx but also by other processes in the lake. Such processes could be classified as dissolution or precipitation of magnetic minerals in varying degrees.

In zone I, the detrital influx is relatively high shown by a relatively high Ti concentration, and thus the relatively low value of κ in this zone could not be due to low detrital influx. Rather, magnetite dissolution in zone I could be inferred from the absence of SP magnetite grains as indicated by low values of χ_{FD} (averaging only 1.2%, Fig. 8c). SP

magnetite grains are easily dissolved because SP grains have a higher surface to volume ratio (Yamazaki et al., 2003; Zheng et al., 2010; Abrajewitch and Kodama, 2011). Magnetite dissolution is also documented by SEM observation from zone I (Fig. 7d). The magnetic grains with surficial pits and hollows can be inferred to be signs of dissolution (Ortega et al., 2006). Magnetic dissolution normally occurs in anoxic conditions followed by the formation of authigenic iron sulfides (Tarduno, 1995; Rowan et al., 2009). No iron sulfides were ever identified by magnetic measurements nor by the XRD and SEM analyses. As shown earlier by Demory et al. (2005) and Ao et al. (2010), the aforementioned observations suggest that magnetic dissolution in zone I occurs in an anoxic but non-sulfidic environment.

Dissolution of iron oxides also occurred in zone II, as indicated by the presence of the authigenic mineral siderite (Williamson et al., 1998). Siderite normally forms in a non-sulfidic environment as soluble iron, which is formed by dissolution through reactions with organic matter (Liu et al., 2012), reacts with alkalinity. Magnetic dissolution in zone II, however, might not be as prominent as in zone I, as indicated by the high value of κ and the abundance of SP grains in this interval. The degree of dissolution in zone II was so weak that it will not affect magnetite, although it might affect more reactive iron-rich non-magnetic minerals (such as hydrous ferric oxides) and iron-oxyhydroxides (such as lepidocrocite and goethite) (Poulton et al., 2004; Liu et al., 2012). The presence of goethite was detected in the XRD data from a soil sample taken from the surrounding area, but not in that from the sediment samples (Fig. 6). This result might indicate that reductive dissolution removed more goethites from the sediment, but it left the magnetite. Indeed, the SEM observation of extracted grains from zone II shows the abundance of well-preserved iron oxides (most likely magnetite) that show no signs of dissolution (Fig. 7b).

The highly magnetic zone II might have experienced magnetite precipitation that amplifies the magnetite concentration above that from the detrital input, producing relatively high values. Magnetite precipitation is inferred both by the relatively high concentration of magnetic minerals (characterized by high value of magnetic susceptibility) and the high proportion of SP grains shown by the high χ_{FD} values (averaging 7%). Magnetite precipitation can also be inferred from the relatively wide spectrum of the magnetite grain size distribution in zone II as compared with that in zone I (Fig. 8b and c), producing fine grain sizes in

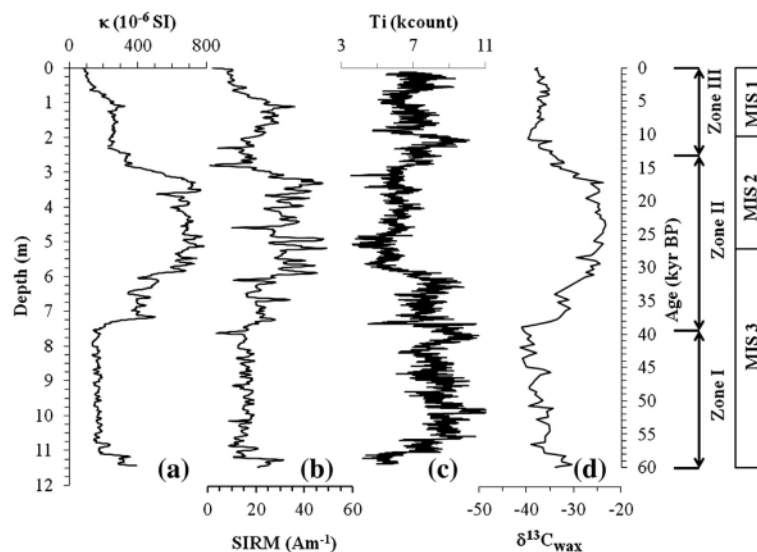


Fig. 9. A comparison between magnetic parameters and paleoclimatic proxies on Lake Towuti sediment. (a) Magnetic susceptibility, (b) SIRM, (c) Ti concentration, and (d) $\delta^{13}\text{C}_{\text{wax}}$. Data shown in (c, d) and age data are after Russell et al. (2014).

addition to coarse grain sizes, the latter of which are likely detrital in origin. In this scenario, magnetite precipitation in zone II occurred in relation to an increase in soluble iron concentration in a water column and in sediment–water interface as iron-bearing minerals from lateritic soil enter the lake in the form of soluble iron as well as the dissolution of hydrous ferric oxide and iron-oxyhydroxide minerals in the sediment. Excess Fe^{2+} in soluble iron can diffuse both in water and sediment to form iron oxide precipitates as it encounters oxidic or suboxic conditions (Karlin et al., 1987). Some of these iron oxide precipitates are probably magnetite, which then enhances the overall concentration of magnetite in the sediment. Recently, Costa et al. (in press) has successfully shown that the degree of oxygenation in the time interval associated with zone II is higher than that in the other zones.

Magnetite precipitation in zone II could also be viewed as biogenic magnetite production through biotic processes. The occurrence of biogenic magnetite in fresh water has been reported (Zhang et al., 2005). Biogenic magnetite could be formed through either biologically induced mineralization (BIM) or biologically controlled mineralization (BCM) (Konhauser, 1998; Frankel and Bazylinski, 2003). In particular, BIM grains were formed at the oxidic–anoxic interface (i.e. suboxic) (Frankel and Bazylinski, 2003). Therefore, the higher oxygen level at zone II (Costa et al., in press) and the absence of iron oxide dissolution (from SEM observation) suggest that the environmental conditions in zone II were probably favorable for biogenic magnetite production and preservation. Moreover, BIM grains are often characterized by a large variation in composition of the Fe phase, including magnetite and siderite, and grain-size distributions (from superparamagnetic to single domain grains) (Frankel and Bazylinski, 2003). This observation might explain the presence of siderite in this zone. The relatively high value of ARM in zone II indicates that the biogenic magnetite is not only SP size but also SD size.

The top-most section of zone III also experienced dissolution of iron oxides. The presence of siderite, inferred from temperature-dependent susceptibility curves (Fig. 3a–b), indicates a reducing environment, and the presence of paramagnetic grains indicated by gaps in the hysteresis curves (Fig. 6a,c) indicates that the dissolution in zone III might not be as intense as that in zone I. Thus, as in zone II, the dissolution of iron oxides in zone III not only initiates the formation of authigenic siderite but also initiates the formation of SP magnetite. Although the magnetite precipitation in zone III was not as intense as that in zone II, the proportion of SP grains in zone III is sizable as shown by the average χ_{FD} value of about 6%.

5.2. Paleoclimatic significance

The variations of magnetic properties in the sediments from Lake Towuti indicate varying levels of reductive diagenesis (in the form of magnetic mineral dissolution) as well as magnetite precipitation. The degree of reductive diagenesis and magnetite precipitation, in turn, depends on depositional conditions that correspond to changes in paleoclimatic or paleoenvironmental conditions. Therefore, the results of this study can provide insight to decipher the magnetic signals of this highly magnetic sediment from an ultramafic area as potential proxy indicators for paleoclimate.

Through analyses of Ti concentration and $\delta^{13}\text{C}_{\text{wax}}$, Russell et al. (2004) showed that the climate around Towuti was dominated by wet conditions during the period of early to mid Marine Isotope Stage 3 (MIS3) and mid Marine Isotope Stage 1 (MIS1) or the Holocene. During late MIS3 and Marine Isotope Stage 2 (MIS2), the climate was dominated by dry conditions, similar to other discontinuous lake records from elsewhere in Sulawesi (Hope et al., 2001; Hope, 2001). Comparing the two parameters above (Ti concentration and $\delta^{13}\text{C}_{\text{wax}}$) with magnetic parameters (mainly χ and SIRM), we find that Ti concentration is negatively correlated with magnetic parameters and $\delta^{13}\text{C}_{\text{wax}}$ is positively correlated with them (Fig. 9), indicating that the low magnetic units of zones I and III are associated with wet conditions during early to

mid MIS3 and Holocene, whereas the high magnetic unit of zone II is associated with dry conditions during late MIS3 and MIS2.

From 60 to ~40 kyr BP, the high degree of magnetic dissolution associated with zone I might be related to high intensity rainfall during wet conditions. High rainfall during this period increased not only the water level of the lake but also the organic material causing an anoxic environment and magnetite dissolution in the sediment (Juveny et al., 1994). The intensity of dissolution decreased at ~39 kyr BP, causing a significant increase in both magnetic susceptibility and SIRM, respectively around 300 and 200%. This transition coincided with lower Ti concentrations implying that during this transition rainfall decreased and that the water column of the lake became more oxidizing. Based on subsequent changes in magnetic signals, this transition marks the beginning of dry conditions in the area surrounding Lake Towuti climaxing during MIS2.

From ~31 to 17 kyr BP, magnetic susceptibility and SIRM are at their highest level. Working respectively with diatom and pollen records from Lake Tondano in North Sulawesi and the pollen record from the Wanda site near Lake Towuti, Dam et al. (2001) and Hope (2001) reported extremely dry conditions in Sulawesi for the same period. This period of dry conditions caused the water level of Lake Towuti to decrease to its lowest level. Low lake levels might allow water to circulate from the surface to the lower part of the lake increasing oxygenation at the sediment–water interface (Groner and Schultze, 2008). This oxygenation in turn might initiate the formation of authigenic magnetic minerals, such as magnetite, as documented by high values of magnetic susceptibility and SIRM.

The transition between MIS2 to MIS1 was marked by a decrease in magnetic susceptibility along with an increase in Ti concentration in sediment implying a significant increase in rainfall around Lake Towuti. From early MIS1 or Holocene to ~5 kyr BP, magnetic susceptibility and SIRM then gradually increase suggesting changes from wet to relatively dry condition. Similar conditions were also reported by Dam et al. (2001) based on analyses of water level fluctuations in Lake Tondano. In the late Holocene, however, magnetic and Ti data indicate a return to wet conditions. It shows an out of phase relationship with Dam's data that indicates that the driest condition during Holocene actually happens in the late Holocene. Wet conditions in Lake Towuti during the late Holocene are likely coincident with a wet phase recorded in southern Indonesia (Griffiths et al., 2009).

6. Conclusions

The sediment of ultramafic-surrounded Lake Towuti is found to be as magnetic as that from volcanic/basaltic lakes. The mineral-magnetic properties of sediments from Lake Towuti are controlled mainly by a combination of changes in the detrital influx and reductive diagenesis in the form of iron oxide dissolution and by magnetite precipitation. The varying levels of dissolution and precipitation in Lake Towuti sediment, specifically the 119 m long core TOW10-9B, the results in three distinct zones with large variations in the mineral-magnetic properties, which correspond to changes in the paleoclimate. Zone I, the weakly magnetic unit in the lower section of the core, is marked by dissolution with minimal precipitation associated with the wet conditions of MIS3. In contrast, zone II, the strongly magnetic unit in the middle section of the core, is characterized by minimal dissolution with intense precipitation associated with relatively dry conditions during MIS2. Zone III, with its low to moderate magnetic values, is characterized by moderate dissolution and precipitation that correspond to relatively wet conditions during MIS1 or the Holocene. In general, the variations of magnetic properties in Lake Towuti sediment essentially depend on environmental processes driven by climate change. Such processes regulated the concentration of magnetic minerals in the sediment. Among the magnetic parameters, SIRM stood out as the most sensitive proxy indicator of paleoclimate with its strong negative correlation with Ti concentration as a proxy indicator of rainfall. Although the variation of magnetic

properties in ultramafic-surrounded lakes, such as Lake Towuti, might serve as reliable proxy indicators for paleoclimate, any interpretation should be done with caution. In these lakes, magnetic parameters are controlled not only by detrital influx, which usually correlates with climate, but also by in situ geochemical processes, which do not always correlate directly with climate.

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