

Tectonic and climatic controls on the Plio-Pleistocene evolution of sediment discharge from Papua New Guinea

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ABSTRACT

This study presents a detailed sedimentary geochemical record of International Ocean Discovery Program (IODP) Site U1489, which was drilled off the northeast margin of Papua New Guinea (PNG). Focusing on the geochemical characteristics and the extent of chemical weathering of siliciclastic component, a total of 183 samples from the upper 84 m of Site U1489 are used to reconstruct the PNG sediment supplies to the tropical western Pacific over the last 4 Ma. Our geochemical results show a 3-stage variability in Site U1489 sediment composition, which are well related to regional tectonic and climatic changes. Stages III (~4.0–3.5 Ma) and II-A (~2.9–1.7 Ma) are characterized by more enhanced and variable contributions from mafic-rock sources than that of felsic-origin. This interval is shown to be accompanied by relatively rapid sediment delivery and more dynamic depositional environments that are possibly related to marginal mixing, coinciding with more frequent PNG tectonic events. In particular, increased contributions derived from ultra-mafic/mafic rocks are recorded between ~3.5 and 2.9 Ma, which is attributed to increased ophiolite input due to arc-continent collision. Accordingly, these results give a consistent but refined timing for an accelerated plate suturing at the eastern margin of PNG. By contrast, Stage I (~1.7–0 Ma) is characterized by a relatively constant background supply of finer particles, showing a persistent increase over time in the intensity of chemical weathering. This increased weathering trend is reported for the first time for the tropical regions of western Pacific, and it is interpreted as indicating a strengthening of the Walker Circulation in the past ~1.7 Ma. Overall, the interpretations of the siliciclastic fraction from Site U1489 presented in this paper provides crucial clues to distinguish the tectonic and climatic influences on the PNG sediment inputs during the Plio-Pleistocene.

1. Introduction

Over the last ~5 million years (Ma) of the Plio-Pleistocene, Earth's climate had undergone a significant cooling, and the middle Pliocene is often taken as a future-analogous warm state (Haywood et al., 2016). The Plio-Pleistocene cooling trend is thought to be stronger in the eastern- than the western- equatorial Pacific (i.e. EEP and WEP) (Fedorov et al., 2015; Tierney et al., 2019), thus causing a gradual intensifying zonal gradient of sea surface temperature (SST) across the Pacific. This west–east SST gradient plays a decisive role in the equatorial air–sea coupled system, i.e., the El Niño Southern Oscillation (ENSO) (Philander, 1983; Wara et al., 2005). Moreover, the different oceanic and climatic conditions of the WEP in the warm Pliocene may have significantly reorganized the circulations and coupled dynamics of

the low- to middle-latitude ocean and atmosphere (Fedorov et al., 2015; Burls et al., 2017; Tierney et al., 2019). However, little is known about the Plio-Pleistocene hydrologic changes over the WEP, mainly because of the short of continuous sedimentary records (e.g. those of Yu et al., 2016, 2018

chemical weathering flux and contribute a prominent proportion of the global CO₂ consumption due to weathering (Hartmann et al., 2009; Hartmann et al., 2014).

Previous studies have shown that, changes in the tectonic configurations of the PNG and WEP regions have profound consequences on not only the regional oceanic (e.g. Pacific-Indian linkage) and climatic states (e.g. Australia aridification, Christensen et al., 2017), but also the biogeochemical cycling along the equatorial Pacific (Wells et al., 1999). Most importantly, for the evolution of the Equatorial Pacific W–E thermal contrast in the last ~8 Ma, the gradual closure

the Indonesian Throughflow to serve as a key connect of the global thermohaline circulation (Gordon, 2005) (Fig. 1a). Changes in such hydroclimate and oceanic variations are proven to be faithfully documented in the marine sediment sequences offshore PNG during the Quaternary (Tachikawa et al., 2011; Wu et al., 2012, 2013; Hollstein et al., 2018; Aiello et al., 2019; Dang et al., 2020a; and references therein), as a result of the tremendous terrigenous detrital material exported from PNG (c.f. Milliman et al., 1999; Walsh and Nittrouer, 2003).

However, over the Plio-Pleistocene, the PNG region experienced drastic tectonic movements, with intensive (micro)plate convergence, arc-continent collision, sea-floor spreading, microplate rotation and magmatism (c.f. Baldwin et al., 2012; Holm et al., 2016) (Fig. 1b). At timescales of million years, tectonic activities and the consequent effects of erosion/weathering are thought to play a critical role in driving the Neogene global cooling (e.g. Molnar and England, 1990; Raymo and Ruddiman, 1992; Hay et al., 2002; Li and Elderfield, 2011). A key driving force for the Neogene cooling and glaciation, was hypothesized to be the arc-continent collision in WEP regions, which could accelerate carbon sequestration through the weathering and erosion of newly-formed mafic rocks (MacDonald et al., 2019). Although, many key aspects about the weathering processes are still under debate, especially the lithologic effect of uplifted landmass on weathering intensity and carbon consumption flux (e.g. Gaillardet et al., 1999; Caves Rugestein et al., 2019; Lu et al., 2020). In the context of the chemical weatherability, the Southeast Asian areas with newly exhumed mafic/ultramafic rock, like those Late Cenozoic terranes of PNG, have the highest

equatorial undercurrents (Fig. 1a, Fine et al., 1994). The North and South Pacific Equatorial Currents (NEC and SEC, respectively) transport sub-surface and intermediate waters from the subtropics into the equatorward western boundary currents, which retroflect via several local eddies into the eastward flowing currents represented by the Equatorial Undercurrent (EUC) (Fig. 1a, Fine et al., 1994). The Indonesian Throughflow (ITF), on the other hand, leaks the Pacific waters into the Indian Ocean (Gordon, 2005). Currents offshore the PNG region includes: (1) monsoon-driven seasonally reversed New Guinea Coastal Current (NGCC), (2) the equatorward flowing New Guinea Coastal Undercurrent (NGCUC) and (3) the equatorward flowing New Ireland Coastal Undercurrent (NICUC), which serve as the major pathways of SEC converging into the Equatorial Pacific (Fig. 1a). The NGCUC originates from the SEC around the Great Barrier Reef, and finally flows along the PNG coast after circulating in the Gulf of Papua (Tsuchiya et al., 1989; Grenier et al., 2011). The NICUC, originating too from SEC but flowing eastern off the New Ireland, ultimately joins half its volume into the NGCUC in the Bismarck Sea and retroflects the other half directly into the EUC (Fig. 1a, Butt and Lindstrom, 1994; Melet et al., 2010). The contribution of NGCUC is estimated to be up to 62% of the EUC at 145°E, and about 50% at 156°E after NICUC has joined in (Slemons et al., 2010). As a result, the NGCUC and NICUC are powerful in delivering the terrigenous matters from the PNG region to the WEP.

2.3. WEP climate

The WEP area experiences the Earth's most expansive and persistent low-level moisture convergence, given the existence and variations of the Inter-tropical Convergence Zone (ITCZ), the South Pacific Convergence Zone (SPCZ) and the ascending limb of the Walker Cell (WC) (Vincent, 1994; Webster et al., 1998; Yim et al., 2016). The meridional displacement of ITCZ over WEP tightly follows the Australasian monsoon and brings slightly more rainfall to the PNG region during austral summer on a seasonal base (Webster et al., 1998). A strong atmospheric link over the PNG and the southwestern subtropical Pacific generates circulation anomalies of the SPCZ (merges with ITCZ over PNG), which further converges the South Pacific northeasterly trade winds and the southeasterlies from Australia, and resulted in abundant rainfall year-round over the PNG region (Kiladis et al., 1989; Vincent, 1994). On the modern inter-annual scale, the shift between El Niño and La Niña events modulates the strength and location of the ascending limb of WC, presumably causing reduced rainfall over PNG during El Niño phases (Yim et al., 2016). On the other hand, shifts of SPCZ in association with ENSO generate more severe consequences to the rainfall over the WEP. During extreme El Niño events, essentially due to a substantial reduction in zonal and meridional SST gradients, the SPCZ shifts equatorward and severe droughts occur over the WEP islands (Cai et al., 2012; Borlace et al., 2014).

3. Materials and methods

3.1. Site U1489 and its chronology

Sediment cores at Site U1489 (02°07.19'N, 141°01.67'E, 3421 m water depth) were drilled during IODP Expedition 363 near the bottom of the western flank of the southern Eauripik Rise, Caroline Sea (Fig. 1). Drilling at Site U1489 penetrated a continuous succession of hemipelagic, carbonate-rich sediment, and the recovered sequences mainly consist of nannofossil ooze and chalk with varying proportions of clay minerals (Rosenthal et al., 2018). For this study, 2-cm-thick half-round samples were collected at a ~ 50-cm interval along the splice (a complete sedimentary sequence of the site, based on patching together the sequences recovered from different holes) from the upper 84 m of Site U1489, resulting in 183 samples in total. A first ¹⁸O-stratigraphic age model for the upper ~84 m sediment succession of Site U1489 was established by benthic foraminifera ¹⁸O analysis on the same samples

as shown in this study (Dang et al., 2020b). Due to the relatively low time-resolution of the sampling strategy, only some major Marine Isotope Stages were recognized in the last ~2.9 Ma, beyond which the age model relies on the paleo-magnetic reversals defined by ship-board studies (Dang et al., 2020b). Based on the estimated sedimentation rates ranging between 1.2 and 3.4 cm/ka, the samples of this study yield a mean time-resolution of ~15–42 ka (Dang et al., 2020b). Given an average uncertainty of <10 ka of the age determinations, variations on a timescale longer than 100 ka (i.e. >0.1 Ma) are considered statistically significant.

3.2. Siliciclastic sediment geochemistry

Detrital, aluminosilicate fraction was isolated by removing all potential biogenic fractions. Noting that some samples in Site U1489 have considerable opal contents (Rosenthal et al., 2018), for each sample, about 200 mg bulk sediments were leached by (1) hydrogen dioxide (10% in volume) to remove organic material, (2) sodium hydroxide solution (1 M) to remove possible biogenic silicon, and (3) hydrochloric acid solution (0.05 M) to remove calcium carbonate. Subsequently, the detritus residues were centrifuged and U_{Wc}/k_{auth} by 4-stage d

the instantaneous intensity of chemical weathering in continents (Li and Yang, 2012).

Neither of the CIA and CIX indexes is sensitive to the lithology of parent rocks. On the other hand, the ICV is calculated by mass fractions of major elements including Fe, Mg, Mn, and Ti (Table 1); and tend to be highest during the weathering of pyroxene and amphibole and decrease with the weathering of more stable minerals like alkali-feldspars (Cox et al., 1995). Therefore, by incorporating heterogeneous sources including metamorphic rocks, the ICV evaluates the compositional maturity of the sediments by taking the parent-rock types into account, making it a more appropriate weathering index for mafic source terrains (Lee, 2002; Price and Velbel, 2003; Borges and Huh, 2007).

4. Results

4.1. Major/minor elements and weathering indexes

To assess the elemental composition and to discount the potential dilution effects, major and minor elements (e.g. Na, Ca, K, Ti) are reported with respect to Al, as well as the ratios of K/Rb and Zr/Rb (Fig. 2). There is a striking similarity between the profiles of Na/Al and Ca/Al, showing a slight increase from ~4.0 to 2.9 Ma, followed by a continuous decrease from ~2.3 Ma till the present (Fig. 2a).

Notably, there is a close correspondence between the ratios of Ti/Al and Zr/Rb, showing generally enhanced values before than after ~1.7 Ma, with highest values between ~2.9 and 1.7 Ma (Fig. 2b, c). The correspondence between the refractory elements (i.e. Zr and Ti) indicates the same first-order control on their secular variations, which must be different from the controlling factor on the much more soluble elements (i.e. Na and Ca) although the changing patterns are quite similar (Fig. 2a, c).

In between, K/Al profile is characterized by a gradual increase over the last ~1.3 Ma (Fig. 2b), displaying a reverse pattern to the decreasing Na/Al and Ca/Al in this interval (Fig. 2a). Moreover, K/Rb ratio exhibit similar yet more spiked variations compared to those of Ti/Al and Zr/Rb (Fig. 2b, c).

Regardless of the different calculations (Table 1; also see Section 3.3), the various indexes of weathering such as CIA, CIX, and ICV exhibit a nearly identical variability (Fig. 2d–f), which generally mirrors those of Na/Al and Ca/Al ratios (Fig. 2a). Although the consistent variations as shown by different indexes confirm the changing trends of weathering intensity (Fig. 2d–f), the absolute CIA values are relatively low (i.e. between 40 and 70), indicating that processes other than weathering and erosion play a role (see Section 5).

4.2. Trace elements including rare earth elements (REE)

With normalization to the upper continental crust (UCC; Rudnick and Gao, 2003), the trace-element distribution of Site U1489 sediments exhibit a clear pattern between mafic and felsic elements (Figs. 3 and 4). The pattern is relatively invariable, showing prominent enrichments in the transition elements of Sc, V, Cr, Ni, and somewhat Co that are typically enriched in mafic igneous rocks, and depletions in the elements of Rb, Y, Zr, Nb, and Th that are more concentrated in felsic rocks (c.f. McLennan et al., 1993) (Fig. 4). Ratios of Ni/Th and Cr/Rb are taken to indicate the relative contributions between mafic and felsic origins (Fig. 3d). The profiles of Ni/Th and Cr/Rb are highly similar, showing enhanced values between ~4.0 and 2.9 Ma, and lowered values from ~2.9 to 1.7 Ma. After a sudden rise at ~1.7 Ma, the ratios gradually decrease (Fig. 3d).

When normalized to the UCC (Rudnick and Gao, 2003), the REE fractionation pattern of Site U1489 sediments show relatively narrow variations, which are characterized by positive Eu anomalies and depletion of light REE (LREE: La to Nd) relative to the middle (M) and heavy (H) REE (MREE: Sm to Gd; HREE: Dy to Lu) (Figs. 3 and 5). Based on these characteristics, useful parameters of REE fractionation are

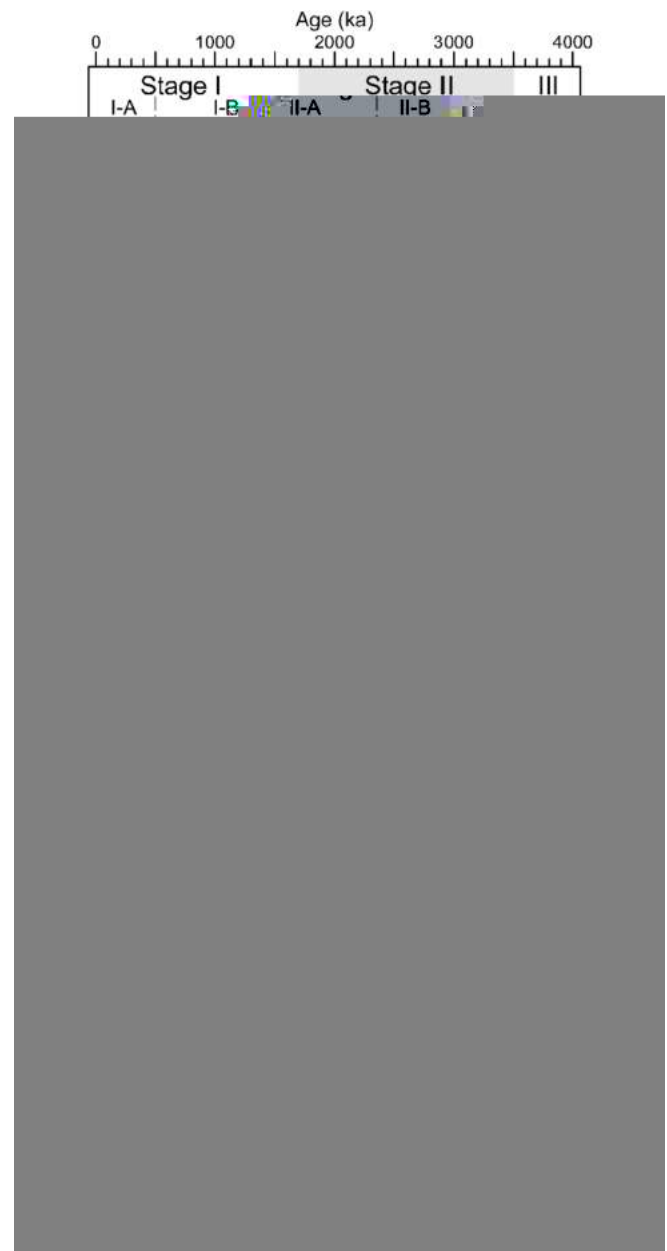


Fig. 2. Major-element composition of siliciclastic sediments of IODP Site U1489.

(a) Elemental ratios of Na/Al (red with plus-signs) and Ca/Al (black with circles). (b) K/Al (blue with plus-sign) and K/Rb (orange with circles). (c) Proxies of sediment grain size: Zr/Rb (green with plus-signs; e.g. Schneider et al., 1997) and Ti/Al (purple with circles; e.g. Boyle, 1983). (d–f) Indexes of weathering intensity: ICV (d, gray; Cox et al., 1995), CIX (e, gray blue; Garzanti et al., 2014) and CIA (f, gray; Nesbitt and Young, 1982). The unit for elemental ratio is g/g. Stage I (1.7–0 Ma) with sub-stages I-A (0.5–0 Ma) and I-B (1.7–0.5 Ma), Stage II (3.5–1.7 Ma) with sub-stages II-A (2.9–1.7 Ma) and II-B (3.5–2.9 Ma), and Stage III (4.0–3.5 Ma) are shown by vertical bars and dashed lines (see Section 4.3). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

extracted, all of which show consistent downcore variations (Fig. 3).

The total, UCC-normalized REE concentrations (i.e. REE, and so on) exhibit a stepwise pattern; after a generally decreasing trend from ~4.0 to 2.3 Ma, the REE persistently increases till today (Fig. 3a). Mirror wise, the Eu anomaly (i.e. Eu/Eu^* , as calculated by $Eu_N / (Sm_N \times Gd_N)^{1/2}$) has large-amplitude variations from ~4.0 to 2.9 Ma, followed by a persistent decrease (Fig. 3a). For the relative enrichment/depletion of

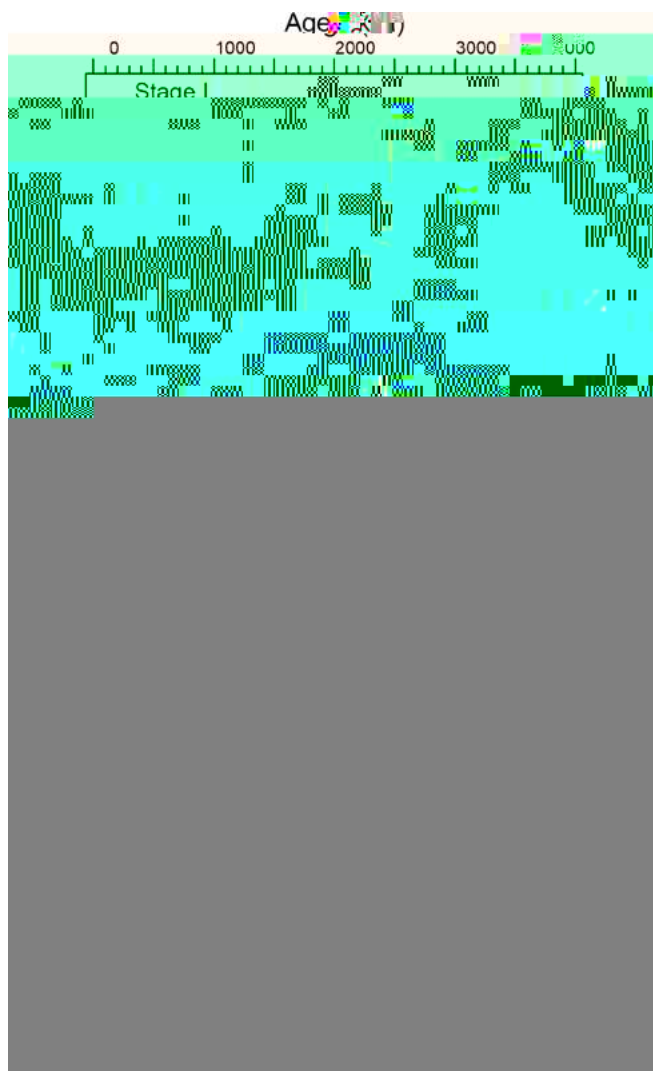


Fig. 3. Trace-element composition of the siliciclastic sediments of IODP Site U1489.

(a) Total REE concentration (REE, black), and the Eu anomaly ($\text{Eu}/\text{Eu}^* = \text{Eu}_N / (\text{Sm}_N \times \text{Gd}_N)^{1/2}$, red). (b) Ratio of light REE to heavy REE (LREE/REE, blue), which is often approximated by La versus Yb (La_N/Yb_N , the subscript N refers to chondrite-normalized, green). (c) Ratios of heavy REE (HREE/REE, brown with plus-signs) and middle REE (MREE/REE, orange with circles) to total REE, respectively. Note that all REE data are normalized to chondrite (McDonough and Sun, 1995). (d) Selected elemental ratios indicative of the mafic versus felsic contributions (c.f. McLennan et al., 1993): Ni/Th (purple with triangles) and Cr/Rb (light green with diamonds). Stage I (1.7–0 Ma) with sub-stages I-A (0.5–0 Ma) and I-B (1.7–0.5 Ma), Stage II (3.5–1.7 Ma) with sub-stages II-A (2.9–1.7 Ma) and II-B (3.5–2.9 Ma), and Stage III (4.0–3.5 Ma) are shown by vertical bars and dashed lines (see Section 4.3). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

LREE, various ratios of LREE/REE, MREE/REE, HREE/REE, and La_N/Yb_N clearly show a three-stage variability (Fig. 3b, c; see Section 4.3). Notably, the LREE/HREE is often approximated by the ratio of La_N/Yb_N . Also note that the subscripts N refers to chondrite normalization (McDonough and Sun, 1995), while subscript C refers to UCC-normalized (Rudnick and Gao, 2003).

4.3. Three stages defined by compositional changes

Taken together, the temporal variations in the geochemical compositions of Site U1489 sediments can be consistently divided into three

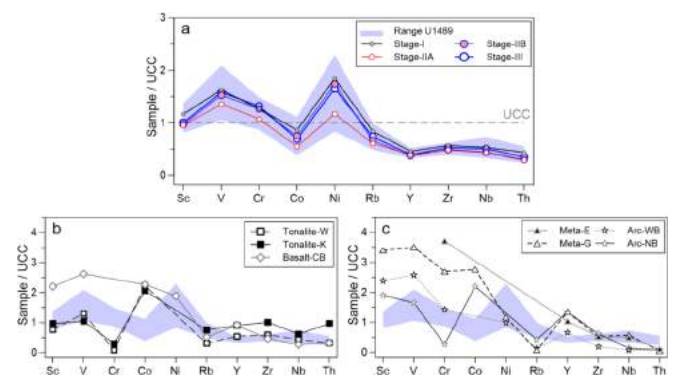


Fig. 4. Trace-element distribution pattern of IODP Site U1489 sediments. (a) Sediment samples of Site U1489, showing the distribution range of all samples (light blue shading; also in b–c) and the average values for the evolutionary stages (lines with symbols). (b) Tonalites (squares; Holm et al., 2015) from Wamum (Tonalite-W) and Kainantu (Tonalite-K), and basalts (diamonds) from the Cloudy Bay (Basalt-CB; Holm and Poke, 2018). (c) Metamorphic rocks (triangles) from Emo (Meta-E; Zirakparvar et al., 2013) and Goropu (Meta-G; Smith, 2013), and arc volcanic rocks (stars) from New Britain (Arc-NB; Holm et al., 2013) and Western Bismarck arc (Arc-WB; Woodhead et al., 2010). Note that the transition elements Sc, V, Co, Cr, and Ni are typically enriched in mafic igneous rocks, whereas Rb, Y, Zr, Nb, and Th are more concentrated in felsic rocks (c.f. McLennan et al., 1993). All trace elements are normalized to mean values of the upper continental crust (UCC; Rudnick and Gao, 2003). See Fig. 1b for the locations of the potential source rocks. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

stages (Figs. 2 and 3), from top to bottom:

Stage I: from ~1.7 to 0 Ma, with sub-stages I-A (~0.5–0 Ma) and I-B (~1.7–0.5 Ma);

Stage II: from ~3.5 to 1.7 Ma, with sub-stages II-A (~2.9–1.7 Ma) and II-B (~3.5–2.9 Ma);

Stage III: from ~4.0 to 3.5 Ma.

Specifically, such a division of the major stages relies mostly on the distinct features of Stage II, characterized by 1) increased ratios of Na/Al, Ca/Al, K/Al, K/Rb, Ti/Al, and Zr/Rb (Fig. 2), and 2) increased Eu/Eu^* values and relatively decreased LREE contents as approximated by La_N/Yb_N (Fig. 3). Besides, Stages I and II are further divided into two sub-stages (i.e. -A and -B), respectively, not only according to the compositional variations in our geochemical results, but also consistent with the regional tectonic and climatic configurations inferred from literature, as discussed below.

5. Discussion

Geochemical composition of marine sediments has a great potential to provide information on sediment provenance, weathering intensity, and transport dynamics. Lithology in provenance areas – mafic versus felsic parent rocks – is the primary source of compositional difference (McLennan et al., 1993; Fralick and Kronberg, 1997). For instance, unweathered felsic rocks (e.g. granites) have higher Al-normalized concentrations of Rb, Y, Zr, Nb, and Th than unweathered mafic rocks (e.g. basalts) (Price and Velbel, 2003; Yang et al., 2008; Wu et al., 2013), because all these elements are relatively incompatible in mantle rocks and thus become concentrated in the crust (McDonough and Sun, 1995; Rudnick and Gao, 2003).

Chemical weathering can exert a significant influence (c.f. Gaillardet et al., 1999). Specifically, areas of intense weathering generate mature sediments characterized by strong cation depletion (e.g. Na^+ , Ca^{2+} , Mg^{2+} , and K^+) and concentration of resistant elements (e.g. Al, Ti, Zr, and Th), whereas limited weathering yields immature sediments that are compositionally similar to their parent rocks (Nesbitt and Young, 1982; Vital and Statterger, 2000; Oliva et al., 2003; Liu et al., 2012). However,

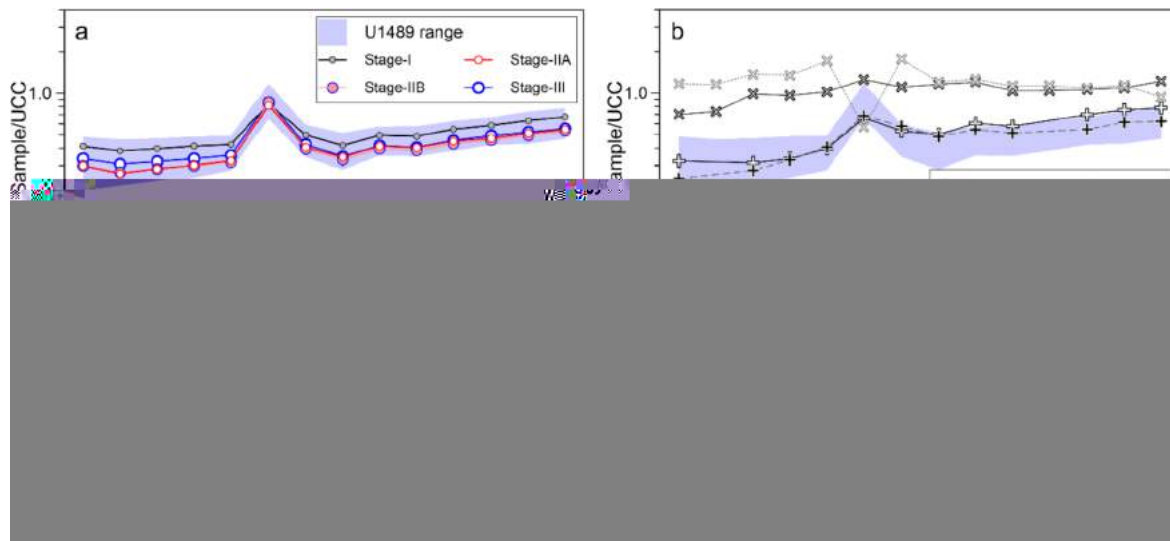


Fig. 5. REE fractionation patterns of IODP Site U1489 sediments in comparison with modern PNG-sourced sediments and potential source rocks. (a) Sediment samples of Site U1489, showing the distribution range of all samples (light blue shading; also in b–d) and the average values for the evolutionary stages (lines with symbols). (b) Modern sediments from two major PNG rivers (crosses; (Hannigan and Sholkovitz, 2001) and from the KX-cores off northern PNG (plus-signs; Wu et al., 2013). (c) Tonalites (squares; Holm et al., 2015) from Wamum (Tonalite-W) and Kainantu (Tonalite-K), and basalts (diamonds) from Manus Islands (Basalt-M; Gena et al., 2001) and the Cloudy Bay (Basalt-CB; Holm and Poke, 2018). (d) Metamorphic rocks (triangles) from Emo (Meta-E; Zirakparvar et al., 2013) and Goropu (Meta-G; Smith, 2013), and arc volcanic rocks (stars) from New Britain (Arc-NB; Holm et al., 2013) and Western Bismarck arc (Arc-WB; Woodhead et al., 2010). All REE data are normalized to mean values of the upper continental crust (UCC; Rudnick and Gao, 2003). See Fig. 1 for the locations of the rivers, KX-cores, and potential source rocks. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

continental weathering is strongly modulated by tectonic movement and climatic conditions (Raymo and Ruddiman, 1992; West et al., 2005; Li and Elderfield, 2011; MacDonald et al., 2019). Tectonic uplift enhances the rates of physical weathering relative to chemical weathering, and contributes to the compositional and mineralogical immaturity of marine sediments via rapid erosion and deposition (Milliman et al., 1999; Walsh and Nittrouer, 2003; Wei and Algeo, 2020).

Changes in the Plio-Pleistocene sediment sequence of Site U1489, as seen from the geochemical results, must be influenced by multiple factors of weathering, tectonism, and associated transportation. As generalized above, such complex processes make it necessary, firstly, to discuss and motivate the use of various geochemical proxies (Section 5.1.1), and to determine the provenance pattern for Site U1489 sediments (Section 5.1.2). Subsequently, the inferred compositional changes in Site U1489 sediments are characterized by a stage-wise pattern (Section 5.2.1), which is tightly related to the major tectonic events that shaped the present PNG (Section 5.2.2). Finally, by deconvolving the influence of tectonism, our results suggest persistently increasing weathering intensity since the Middle Pleistocene over the western tropical Pacific, possibly linked to the strengthening trend of the Walker Cell in the Pleistocene (Section 5.3).

5.1. Geochemical proxies and sediment provenance

5.1.1. Application of geochemical proxies

Variations in the intensity of chemical weathering should result in divergent trends among the detrital elements, related to the varying degrees of elemental mobility. For Site U1489, the observation that the profiles of Na/Al and Ca/Al are similar but different from – sometimes exactly reverse to (i.e. since ~1.5 Ma) – that of K/Al, suggests that the chemical weathering is merely moderate and not sufficiently intense to leach K (Fig. 2a, b). In other words, the weathering should have depleted the elements more soluble than K (e.g. Na, Ca, Mg), while concurrently enriched the elements less or equally soluble than K (e.g. K, Al, Ti, Zr, Th) (Middelburg et al., 1988; Rubin et al., 1993). The moderately intense weathering and the consistent trends are also indicated by the various indexes of CIA, CIX, and ICV, showing a persistent increase in

weathering intensity since at least ~1.7 Ma (Fig. 2d–f). In our case, therefore, the ratios of Ca/Al and K/Al as well as the ICV indexes are chosen to reflect the weathering processes, as ICV serves better as an index for weathering of (ultra-)mafic source terrains than CIA or CIX (see Methods 3.3).

For the highly refractory and insoluble elements, the tight correspondence between K/Rb, Zr/Rb, and Ti/Al ratios points to a different first-order control on their secular changes rather than weathering (Fig. 2b, c). The similarity between Ti/Al and Zr/Rb profiles excludes the major role of sediment provenance changes, as Ti and Zr are respectively enriched in mafic and felsic rocks (Fig. 2c). Instead, these consistent changes can best be attributed to sediment grain size, in response to the varying ratios between silt and clay fractions. The motivation is as follows. Firstly, the silt fraction tends to retain the relatively mobile element K (e.g. K-feldspar) than highly weathered clays (Lange et al., 1966). Instead of forming discrete mineral phases, Rb substitutes readily for K in aluminosilicate minerals, and the enrichment of Rb relative to K is greater in mica compared to the co-existing feldspars (Lange et al., 1966; Dypvik and Harris, 2001). Secondly, much larger amounts of resistant elements like Ti and Zr are concentrated in the silt than clay fractions, as they are typically presented in heavy, refractory minerals (e.g. rutile and anatase for Ti; zircon and sphene for Zr) (Boyle, 1983; Schneider et al., 1997; Taboada et al., 2006). These features have made K/Rb, Zr/Rb, and Ti/Al as widely used grain-size proxy (e.g. Matthewson et al., 1995; Kroon et al., 2000; Dypvik and Harris, 2001; Liu et al., 2004; Sun et al., 2008).

Moreover, this inference is supported by several lines of evidence. First, the strong correlations between Al, Ti, and Zr concentrations indicate a single dominant provenance, thereby precluding the possibility of change in sediment provenance and mixing (see the plots and data in Supplementary File). Second, the similar trends as observed for the ratios normalized to Al between the highly soluble elements (i.e. Na/Al, Ca/Al) and the highly insoluble elements (i.e. Ti/Al, Zr/Al) suggest process other than chemical weathering playing a role (Fig. 2a–c). Third, the use of Zr/Rb and Ti/Al ratios as grain-size proxy is corroborated by their correspondence with the changes in total REE concentrations (Figs. 2 and 3).

Although the REE concentrations may be affected by intense chemical weathering and diagenesis (Nesbitt and Markovics, 1997; Bayon et al., 2020), most studies suggest that sediment provenance play a dominant role in determining REE composition (Condie, 1991; Yang et al., 2002; Wu et al., 2013, 2018; Pham et al., 2019). Here, the REE fractionation pattern and associated parameters (e.g. Eu/Eu^* , La_N/Yb_N) as well as the mafic/felsic element ratios (i.e. Ni/Th and Cr/Rb) are used as tracers of parent rocks and potential sources.

In the following, applications of the geochemical proxies, as motivated above, allow us to explore how these factors including sediment provenance, transport dynamics, and weathering intensity interacted with each other and dominated in the different time intervals.

5.1.2. Provenance of U1489 sediments

Characterized by positive Eu anomaly and relative LREE depletion, the UCC-normalized REE fractionation pattern of U1489 sediments is similar to those of various sediments sourced from PNG, including the particles in northern PNG rivers (e.g. Sepik River), and the marine sediments collected along the northern PNG margin (e.g. core KX15-2) and from the western equatorial Pacific (e.g. core KX21-2) (Fig. 5a, b; also see Fig. 1a for locations). This is consistent with previous studies on the late-Quaternary sediments (Wu et al., 2012, 2013; Dang et al., 2020a; and references therein), suggesting that the northern PNG riverine inputs are the main source of the siliciclastic sediments in the region and that sediment transport is largely controlled by offshore coastal currents (Fig. 1a; also see Section 2).

This interpretation may hold true only for the last ~0.5 Ma, and little is known about the terrigenous inputs from PNG in the geological past with active tectonic events. The comparison of the results from Site U1489 with the REE signatures of rocks that were exposed, dredged, or drilled in the ocean region surrounding PNG can offer further clues (Fig. 5a, c, d; also see Fig. 1b for locations). The comparison shows that, the REE signature of Site U1489 sediments is highly similar to those of the igneous rock outcrops, including the basalts from Manus Islands (i.e. Basalt-M; Gena et al., 2001) and from the Cloudy Bay (i.e. Basalt-CB; Holm and Poke, 2018), and the Tonalites from Wamum and Kainantu (i.e. Tonalite-W and -K, respectively; Holm et al., 2015) (Fig. 5c). By contrast, the overall range of U1489 sediments show a less prominent LREE depletion than those of metamorphic rocks from the southeastern PNG (including Emo, i.e. Meta-E; Zirkparvar et al., 2013; Goropu, i.e. Meta-G; Smith, 2013), and the arc volcanic rocks from New Britain (i.e. Arc-NB; Holm et al., 2013) and from Western Bismarck arc (i.e. Arc-WB; Woodhead et al., 2010). (Fig. 5d).

By and large, we propose that the major source of U1489 sediments is the mafic igneous rocks from the north and east of PNG. Moreover, although the potential input from arc volcanic rocks and metamorphic rocks may be secondary in quantity, it is characterized by a stronger LREE depletion (Fig. 5), and its contribution changes are tightly associated with the different phases of tectonic activity in PNG. In fact, variations in the relative contributions from these sources should have determined the subtle differences between the evolutionary stages and sub-stages recognized at Site U1489 (Fig. 4).

This inference is supported by the trace-element composition. In terms of UCC-normalized trace-element distribution, the siliciclastic sediments of Site U1489 show a consistent pattern, with enrichments of the elements typically concentrated in mafic rocks (e.g. Sc, V, Cr, Ni) and depletions of the elements that are more enriched in felsic rocks (e.g. Th, Nb, Zr, Y, Rb) (McLennan et al., 1993; Yang et al., 2008; Wu et al., 2013; Dang et al., 2015) (Fig. 4). This clearly indicates the predominance of ultra-mafic/mafic source rocks, along with minor felsic-rock source (Fig. 4). Moreover, the felsic contribution appears to be compositionally constant over the last 4 Ma, as indicated by the narrow ranges of variations in Th, Nb, Zr, Y, and Rb (Fig. 4) and by the linear correlation of Th_C vs. Rb_C (note that the subscript C refers to UCC-normalized; Rudnick and Gao, 2003) (Fig. 6d). On the contrary, the much larger compositional variations in the typically mafic-origin elements (Fig. 4) suggest a

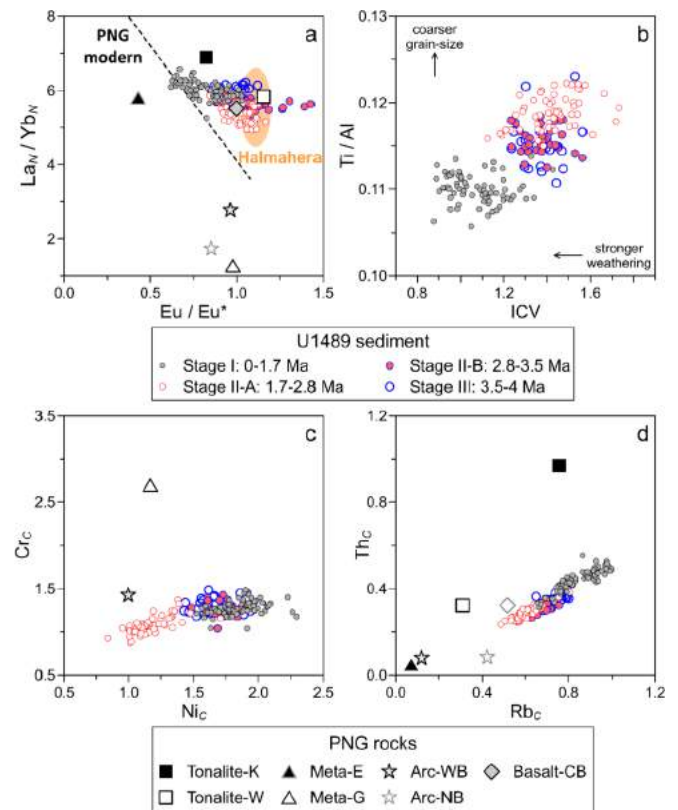


Fig. 6. Plots of extracted proxies for provenance determination of IODP Site U1489 sediments.

(a) La_N/Yb_N vs. Eu/Eu^* (McDonough and Sun, 1995). Gray dashed line shows the regression line as defined by the modern PNG-derived sediments (Wu et al., 2013) and the yellow ellipse shows the range of the Halmahera-Sea sediments (Dang et al., 2015). (b) Ti/Al vs. ICV , distinguishing the influences between transport process (by the grain-size proxy of Ti/Al) and weathering intensity (as proxied by ICV ; also see Fig. 2). (c, d) Plots of Cr_C vs. Ni_C and of Th_C vs. Rb_C (the subscript C refers to UCC-normalized; Rudnick and Gao, 2003), respectively, showing possible compositional changes in the mafic and felsic contributions. The data of Site U1489 are distinguished between the three stages, and the reference data – if available – are also plotted (annotated in Legend). See Fig. 1b for the locations of the potential source rocks. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

complex pattern of the mafic-rock sourced contributions, which must have considerably changed across the stages (Fig. 6c). Although the reference data available so far as plotted in Fig. 4 does not allow us to distinguish the influences between ultra-mafic and mafic rocks, the additions of arc-volcanic and metamorphic rocks is supported by largest compositional variations in Ni, V, and Co that are most enriched in the ultra-mafic minerals such as olivine and spinel (Fig. 4).

5.2. Relations to regional tectonic history

5.2.1. Stage-wise evolution of U1489 sediments

Based on the provenance pattern discussed above, a stage-wise, tectonic-related variations in the siliciclastic supplies to Site U1489 can be identified, via the application of geochemical proxies for provenance, grain size, and weathering intensity (Fig. 6).

Using REE fractionation parameters as provenance proxies (i.e. Eu/Eu^* and La_N/Yb_N), during Stages II and III (before ~1.7 Ma), Site U1489 sediments are dominated by contributions from ultra-mafic/mafic rocks (Fig. 6a). During Stage I (after ~1.7 Ma), the data of Site U1489 approach to the regression line of the modern PNG-derived sediment (Wu et al., 2013), most likely due to a higher contribution of

metamorphic rock sources over time (Figs. 3a, b and 6a). Moreover, plots of UCC-normalized trace elements can shed light on the changes in source-rock types between the three stages as identified (Figs. 3 and 6c, d). The contribution from felsic rocks is minor and the sources of felsic contributions may remain compositionally constant over time, given the linear correlation of Th_C vs. Rb_C ($R^2 = 0.91$, Fig. 6d).

On the contrary, the contributions from mafic rocks should have substantially changed and dominated the variability of geochemical composition of Site U1489, for Stages II and III in particular (Figs. 3 and 6). Specifically, in the plot that represents elements of mafic-origin (i.e. Cr_C vs. Ni_C), there is a systematic trend in the data from sediments deposited during Stage II (Fig. 6c), indicating a single dominant source for that period (~3.5–1.7 Ma). By contrast, the similar range but rather scattered distribution for Stages I and III may indicate multiple types of mafic rock sources (Figs. 3d and 6c).

When looking closely, the features of Stage II are key to decipher the processes related to tectonic movements. The reduced values of both LREE and Eu/Eu^* may have resulted from an increased contribution from mafic rocks like ophiolite or from metamorphic rocks (Zirakparvar et al., 2013) (Fig. 3). Geochemically, the most marked difference between ophiolite and metamorphic rocks is that metamorphic rocks are more enriched in REE (Zirakparvar et al., 2013; Kaczmarek et al., 2015). During Stage-II, the REE decreases (Fig. 3a) and may reflect a relatively limited contribution of metamorphic rocks. However, this inference conflicts with the higher values of the trace-element ratios (i.e. Ni/Th and Cr/Rb) during Stage II-B which suggest an increased input of mafic materials (Fig. 3d). Instead, the changes in REE can be best attributed to coarser grain size of siliciclastic particles, as evidently indicated by the increased ratios of Ti/Al and Zr/Rb (Figs. 6b and 2c).

At a first glance, the apparent contradiction of the geochemical signals in Stage II-A could be explained by somewhat increased influence of felsic materials inputted from the PNG landmasses. However, this case is largely discounted given the linear correlation between Th_C and Rb_C , indicating a relatively constant source of felsic rocks. Alternatively, these puzzling signals of Stage II-A can be reconciled by hypothesizing an increase in grain size of the siliciclastic particles (Figs. 6b and 2c), resulting from changes in the mode and strength of particle transport. This is supported by increased values of Ti/Al and Zr/Rb as grain-size proxies during Stage II-A (Fig. 2c). In fact, it has been shown that sediment grain size is mostly linked to tectonism, with higher values associated with major orogenic events, despite the possible modulations by factors like hydroclimate and vegetation cover (Wei and Algeo, 2020; and references therein).

Taken together, it appears to be the case that enhanced marginal mixing occurred during Stage II-A, leading to not only enhanced fluxes of felsic materials to Site U1489 (Fig. 6c), but also increased transport of coarser sediments over this interval (Figs. 6b and 2c). Besides, the coastal currents might be strengthened due to constriction of the seaway between PNG and the New Britain, causing coarser grain size in U1489 sediments (Figs. 1 and 6b).

The geochemical records of Site U1489 consistently indicate that more mafic materials have been transported to the WEP during Stage II-B (Fig. 3), most likely as a result of the exposure of some ophiolite terrane to the action of weathering by tectonics. Many tectonic reconstructions have shown that the late Pliocene around 4–3 Ma is a period when the Australian Plate moved northward and collided with and elevated the volcanic arcs in Timor-Banda Seas (Cane and Molnar, 2001; Hall, 2009; Karas et al., 2011; Baldwin et al., 2012; Christensen et al., 2017). In the PNG area, in association with the northward subduction of the Australian Plate, a series of tectonic consequences such as arc-continent collision, Bismarck Sea opening, and microplate rotation had occurred along the eastern PNG coast since the Late Pliocene (Holm et al., 2016). In other words, the changes seen here during Stage II-B (i.e. ~3.5–2.9 Ma) should be a direct result of tectonic collision along the eastern margin of PNG (Fig. 3).

To summarize, the three-stage evolution of Site U1489 sediments is

suggested by key REE fractionation parameters and trace-element ratios. In particular, frequent and large tectonic activities should have occurred in Stage II (~3.5–1.7 Ma), leading to enhanced contributions from mafic sources (i.e. Eu/Eu^* , La_N/Yb_N , and mafic/felsic elemental ratios of Ni/Th and Cr/Rb), and also coarser grain size due to more dynamic transport process and sedimentary environment (as seen from enhanced grain-size proxies of Ti/Al and Zr/Rb ratios, and the reduced REE contents).

5.2.2. Refined picture of PNG tectonics

The geochemical results of the marine sediments recovered at IODP Site U1489 (Figs. 3 and 6) provide refined and detailed information and more precise timing of the tectonic evolution of eastern PNG arc-continent collision (Fig. 7, after Baldwin et al., 2012; Holm et al., 2016) since the Mid Pliocene to recent.

Stage III (before ~ 3.5 Ma). The Adelbert-Finisterre Terrane (AFT) and New Britain (NB) might have just started to collide with the mainland PNG (Fig. 7a). This was the time that the NB Trench reactivated subduction, as indicated by a source-rock type shift in the suture zone between the AFT and the mainland PNG between ~3.7–3.0 Ma (Johnson and Jaques, 1980; Abbott et al., 1994; Abbott, 1995; Holm and Richards, 2013). Magnetic evidence also suggests that the Finisterre-Huon Range initially uplifted around 3.5 Ma, and related rifting and extension occurred in association with the rapid opening of the Bismarck Sea (Taylor, 1979; Lee and Ruellan, 2006). Such a transition at ~3.5 Ma is clearly seen in the various provenance indicators in Fig. 3, and consistent with the relatively coarse particles as proxied by Ti/Al and Zr/Rb (Fig. 2c).

Stage II-B (~3.5–2.9 Ma). The arc-continent collision between AFT and mainland PNG might have intensified during this time (Abbott et al., 1994; Abbott, 1995) (Fig. 7b). The collision could have caused the increased supplies of ophiolite-related siliciclastic to Site U1489 as indicated by the largest values of Ni/Th and Cr/Rb (Fig. 3d), while the source areas and depositional environments may remain as the earlier Stage III due to the invariable Eu/Eu^* , La_N/Yb_N , and Ti/Al and Zr/Rb (Figs. 6a, b and 3c).

Stage II-A (~2.9–1.7 Ma). The AFT overthrust the eastern PNG continental margin (Fig. 7c), resulting in magmatism derived from underthrust continental crust (Holm et al., 2016; and references therein). This period is characterized by the typically mafic-sourced sediment composition, i.e., the most prominent positive Eu anomaly and LREE depletion (Fig. 3a–c). Moreover, the occurrence of coarsest sediment particles (as indicated by largest Ti/Al and Zr/Rb values; Fig. 2c) could reflect very dynamic transport processes, possibly with a marginal sediment recycling. Considering the close link between grain size and tectonism (Wei and Algeo, 2020; and references therein), Stage II-A appears to be the major orogenic phase of the eastern PNG mountain ranges.

Stage I (~1.7 Ma to the present). The collision between AFT and PNG gradually relaxed (Fig. 7d), forming the modern-like tectonic arrangement of the eastern PNG. Because of the continued docking of the Finisterre Terrane with the mainland PNG, p Terra??

mainland PNG (Fig. 7) (Abbott et al., 1994; 1995; Holm et al., 2016, and references therein).

At a local point of view, this event of Stage II re-organized the tectonic background of the eastern PNG area and re-shaped the source-rock characteristics of the sediment inputted to the WEP. In terms of the global climate change in association with the Northern Hemisphere Glaciation, the arc-continent collision of PNG region at 3.5–1.7 Ma coincides with the episodes of gradual cooling and climate-state reorganization (Ravelo et al., 2004), possibly supporting the hypothesis of tropical orogenesis on Earth's glacial cooling (MacDonald et al., 2019). The subsequent changes in the U1489 sediment composition took place under such a renewed background condition, and the hydroclimate variations should have played a dominant role in both changes in weathering regime and transport dynamic.

5.3. Climate-driven changes in weathering intensity

Interpretations of weathering intensity rely mainly on the major-element composition of Site U1489. However, the absolute CIA values are erratically low, i.e. between 40 and 70. This discrepancy could be explained by 1) the obscuration by the tectonic-dominated effect as discussed above, 2) a rapid and dynamic sediment delivery as supported by the enhanced ratios of Zr/Rb and Ti/Al as grain-size proxies (Milliman et al., 1999; Wei and Algeo, 2020) (Fig. 2c), or 3) the minor influence of carbonate diagenesis, organic matter as

result of an abrupt warming in the WEP (Wara et al., 2005) with no obvious sign of cooling in the EEP (Liu et al., 2019). An SST warming in the WEP may have effectively enhanced the convective precipitation in the region, and consequently increased the chemical weathering intensity over PNG area.

An increasing weathering over PNG since ~ 1.7 Ma, as observed in U1489 sediment records (Fig. 8a, b), is consistent with the final establishment of the equatorial Pacific sea-air coupled system with an enlarged west-east SST gradient at this timepoint (Wara et al., 2005) (Fig. 8e), and with the continued increase in zonal and meridional SST gradients after ~ 1.7 Ma (Fedorov et al., 2015) (Fig. 8f). From the viewpoint of the meridional transect of Asia-WEP-Australia, the strengthening of chemical weathering intensity in WEP since ~ 1.7 Ma coincided with the long-term trends of aridification in Australia (Christensen et al., 2017) and Central Asia (Zhang et al., 2020), as well as a weakening in Asian summer monsoon (Zhang et al., 2009), possibly suggesting an anti-phased hydro-climatic evolution between the equatorial western Pacific and the mid-latitude continents at both hemispheres (Fig. 8g).

We thus conclude that, although the climate-driven changes should have increased the chemical weathering over PNG from the Pliocene to the latest Pleistocene, the tectonic-controlled evolution of source-rock compositions and sediment dynamics have substantially influenced the composition of the siliciclastic fraction at Site U1489 between ~ 4 and ~ 1.7 Ma. It is only during the last ~ 1.7 Ma, after tectonism in PNG became more quiescent, that the enhanced SST gradients and atmosphere circulation and the resultant strengthening of chemical weathering become prominent, as recorded by the elemental composition of Site U1489 sediments.

6. Conclusions

The long-term tectonic and climate evolutions over the realm where modern Papua New Guinea formed have caused ongoing changes in the terrigenous input to the western equatorial Pacific, with substantial oceanographic and biogeochemical consequences. By h

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