

Early-Pleistocene orbital variability in Northwest Australian shelf s

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Abstract

Paleoclimate proxy records from regions sensitive to humidity/aridity extremes provide valuable insights into natural forcing mechanisms underlying long-term climate variability in the wider region. One such area is Northwest Australia, where the Australian monsoon impacts its northernmost fringes, which are bordered by the Great Sandy Desert inland. Marine sediments from the Australian Northwest Shelf record fluvial run-off and aeolian dust input during the wet and dry seasons. The location is therefore ideal for investigating long-term variability in the Australian monsoon and Northwest Australian dust fluxes over orbital timescales. However, there are few continuous, high-resolution paleoclimate records from the Australian Northwest Shelf spanning the Early Pleistocene, and there is ambiguous orbital phasing even among Late Pleistocene paleoclimate records from the region. Here, we present geochemical and environmental magnetic proxy records of CaCO_3 and dust-flux variability spanning 2.9 to 1.6 Myr ago from International Ocean Discovery Program Expedition 356 Site U1464 on the Australian Northwest Shelf. We establish a new, orbitally-tuned chronology for Site U1464, and observe strong obliquity variability (41 kyr and 54 kyr periodicities) but almost no precession signal in our dust records. We propose that the 41 kyr cycle in Northwest Australian dust fluxes could be a linear response to the East Asian winter monsoon (EAWM) and/or summer inter-tropical insolation gradient (SITIG), whereas the 54 kyr cyclicity might be a non-linear response to obliquity amplitude modulation via the SITIG effect on cross-equatorial flows.

1 **Early-Pleistocene orbital variability in Northwest Australian shelf sediments**

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7 **Key Points:**

- 8 • This study establishes a new orbitally-constrained chronology for IODP Site U1464.
- 9 • Northwest Australian shelf sediments preserve ~400 kyr eccentricity cyclicity in CaCO₃
10 content and obliquity cycles in dust proxy records.
- 11 • Obliquity in Northwest Australian dust fluxes may be related to the East Asian winter
12 monsoon and summer inter-tropical insolation gradient.

13 **Abstract**

14 Paleoclimate proxy records from regions sensitive to humidity/aridity extremes provide valuable
15 insights into natural forcing mechanisms underlying long-term climate variability in the wider
16 region. One such area is Northwest Australia, where the Australian monsoon impacts its
17 northernmost fringes, which are bordered by the Great Sandy Desert inland. Marine sediments
18 from the Australian Northwest Shelf record fluvial run-off and aeolian dust input during the wet
19 and dry seasons. The location is therefore ideal for investigating long-term variability in the
20 Australian monsoon and Northwest Australian dust fluxes over orbital timescales. However,
21 there are few continuous, high-resolution paleoclimate records from the Australian Northwest
22 Shelf spanning the Early Pleistocene, and there is ambiguous orbital phasing even among Late
23 Pleistocene paleoclimate records from the region. Here, we present geochemical and
24 environmental magnetic proxy records of CaCO₃ and dust-flux variability spanning 2.9 to 1.6
25 Myr ago from International Ocean Discovery Program Expedition 356 Site U1464 on the
26 Australian Northwest Shelf. We establish a new, orbitally-tuned chronology for Site U1464, and
27 observe strong obliquity variability (41 kyr and 54 kyr periodicities) but almost no precession
28 signal in our dust records. We propose that the 41 kyr cycle in Northwest Australian dust fluxes
29 could be a linear response to the East Asian winter monsoon (EAWM) and/or summer inter-
30 tropical insolation gradient (SITIG), whereas the 54 kyr cyclicity might be a non-linear response
31 to obliquity amplitude modulation via the SITIG effect on cross-equatorial flows.

32 **1. Introduction**

33 Sediments from the Australian Northwest Shelf (NWS) contain a valuable archive of past
34 Australian monsoon variability, as well as dust fluxes from the Australian interior. Fluvial
35 transport of terrigenous materials to the NWS occurs during the wet season (mainly December to
36 March), primarily from the Ord and Fitzroy Rivers in the far north, and from the De Grey and
37 Fortescue Rivers at the southern margin of the NWS ([Australia's River Basins, 1997](#)) ([Figure 1](#),
38 [Figure S1](#)). In contrast, aeolian dust fluxes to the Australian NWS mostly occur in austral spring
39 to autumn (September to May), when aeolian dust is transported by frontal systems and
40 southeasterly trade winds from Lake Eyre Basin through Northwest Australia to the NWS
41 ([Baddock et al., 2015](#); [Ekström et al., 2004](#); [Gallagher & deMenocal, 2019](#); [McGowan & Clark,](#)
42 [2008](#); [McTainsh, 1989](#); [Strong et al., 2011](#)). This seasonal pattern of precipitation and dust flux

43 is therefore tightly linked to insolation changes and the latitudinal position of the Intertropical
44 Convergence Zone (ITCZ), based on modern observations that seasonal migrations of insolation
45 maxima and the ITCZ are strongly coupled with rainfall and southeasterly trade winds in
46 Northwest Australia (Suppiah, 1992). However, over longer timescales, the relationship between
47 insolation/orbital forcing and Australian monsoon and dust-flux variability is not well known.
48 For example, Australian monsoon strength might either respond to local insolation dominated by
49 precession (Holbourn et al., 2005; Liu et al., 2015; Pei et al., 2021) or to Northern Hemisphere
50 insolation changes driven by precession and obliquity (Magee et al., 2004). These different
51 suggested phasings of the Australian monsoon with orbital forcing could, at least in part, be
52 attributed to fluctuations in global ice volume/sea level. Many studies have reported evidence of
53 increased dust fluxes to the Australian NWS (Courtilat et al., 2020; Hesse et al., 2004; Pei et al.,
54 2021; Stuut et al., 2014, 2019) and decreased Australian monsoonal precipitation (Gallagher et
55 al., 2014; Gallagher & Wagstaff, 2021; Kershaw et al., 2003; Magee et al., 2004; Miller et al.,
56 2018; Stuut et al., 2014, 2019) during glacials and/or stadials, while other studies have
57 interpreted intensified Australian monsoonal precipitation during glacials and/or stadials as a
58 response to southward migration of the ITCZ (Bayon et al., 2017; Denniston et al., 2017; Fu et
59 al., 2017).

60 One of the main issues is that there are few continuous, high-resolution paleoclimate records
61 from this region spanning multiple glacial-interglacial cycles, especially those that extend back
62 to the Early Pleistocene. Of the available high-resolution records, most tend to be focused on
63 periods within the last glacial cycle (the last ~130 kyr), when the effects of millennial climate
64 variability and/or the last deglaciation obfuscate the primary response of Australian monsoon and
65 Northwest Australian dust fluxes to direct orbital forcing (Bayon et al., 2017; Denniston et al.,
66 2017; Fu et al., 2017; Miller et al., 2018). Longer suitable paleoclimate records extending over
67 >1 million years (Myr) exist but are fewer, and their interpretations are more focused on long-
68 term trends (Christensen et al., 2017; Stuut et al., 2019). For example, proxy records of past dust
69 fluxes based on sedimentary Th/K at International Ocean Discovery program (IODP) Site U1463
70 (Christensen et al., 2017) and Log(Zr/Fe) at Ocean Drilling Program (ODP) Site 762 (Stuut et
71 al., 2019) have been used to investigate the Pliocene-Pleistocene evolution of Northwest
72 Australian aridity. These studies found increasingly greater variance in aridity from the Pliocene
73 to Pleistocene.

74 Modelling studies of Australian monsoon variability over orbital timescales are relatively rare.
75 Earlier studies suggested a direct response of Australian summer monsoon precipitation to local
76 insolation (Chappell & Syktus, 1996; Wyrwoll & Valdes, 2003; Wyrwoll et al., 2007), or to
77 Northern Hemisphere insolation via cross-equatorial flow (Miller et al., 2005), but these
78 simulations were limited by modelling capacity at that time, such as low-resolution grids and
79 lacking many feedback processes. More recent model simulations of Australian monsoon
80 variability over orbital timescales suggest that obliquity plays a greater role than expected from
81 its smaller insolation contribution relative to precession, and that higher obliquity could result in
82 stronger summer monsoons in Northwest Australia via a stronger pressure gradient between
83 intensified Siberian High and Australian Low pressure cells (Liu et al., 2015; Shi et al., 2011;
84 Wyrwoll et al., 2007). The interpreted obliquity forcing in simulations, however, is inconsistent
85 with recent proxy observations of monsoonal precipitation in Northwest Australia (Zhang et al.,
86 2020; 2022) and previous understanding of EAWM dynamics (Ding et al., 2002; Sun et al.,
87 2006).

88 We have generated continuous, high-resolution (1 measurement every 0.5 or 1 cm) proxy records
89 of Northwest Australian dust fluxes based on scanning X-ray fluorescence (XRF) and
90 environmental magnetism from IODP Site U1464 (Australian NWS) spanning the last 2.9 Myr.
91 The records show cyclic variability between 2.9 and 1.6 Ma, followed by a marked transition to
92 weak geochemical and magnetic signals in the mid-late Pleistocene. We therefore focus on the
93 2.9-1.6 Ma time interval. This period corresponds to the initial growth and widespread expansion
94 of continental ice sheets across the Northern Hemisphere mid-high latitudes, expansion of
95 Antarctic ice sheets and sea ice, a strengthening in latitudinal (pole-equator) temperature
96 gradients, and consequent strengthening/repositioning of wind bands. Nonetheless, this time
97 interval pre-dates the Mid-Pleistocene Transition (MPT), when the amplitude of glacial-
98 interglacial cycles and attendant ice-climate feedbacks increased. Thus, our records capture dust-
99 flux variability in the Australian monsoon region in detail over multiple orbital cycles during a
100 time of gradual global climate change, prior to the onset of strong ice-climate feedbacks which
101 have been shown to interfere with insolation/orbital forcing of monsoon and dust flux variability.

102 In this paper, we describe materials and methods, and then present a new, orbitally-tuned
103 chronology for IODP Site U1464. Accurate dating of sediment sequences from the Australian
104 NWS over the Plio-Pleistocene has proven to be problematic, due to biostratigraphic

105 diachroneity (Groeneveld et al., 2021), magnetostratigraphic ambiguity (Gallagher et al., 2017a;
106 2017b), and a lack of robust oxygen isotope stratigraphy (Groeneveld et al., 2021). Nonetheless,
107 recent studies have established a revised chronological framework for nearby ODP Site 762 and
108 IODP Site U1463 by combining orbital tuning with biostratigraphic age constraints (Auer et al.,
109 2020; Groeneveld et al., 2021). While these studies incorporated the tuning of benthic $\delta^{13}\text{C}$
110 and/or $\delta^{18}\text{O}$ records, we use the strong eccentricity and obliquity signals in our CaCO_3 % and
111 $\text{Log}(\text{Zr/Rb})$ records for tuning. Finally, we explore the causes of the orbital variability at Site
112 U1464 in the CaCO_3 % and dust-flux proxy records.

113 **2. Materials and Methods**

114 2.1. Site Location & Sampling

115 We use marine cores from IODP Site U1464 (18°03.9'S 118°37.9'E, 264 m water depth, Figure
116 1), which was cored during IODP Expedition 356 (Gallagher et al., 2017a). This site is located
117 on the central Australian NWS at the southern edge of the modern Australian monsoon region
118 (Figure 1), and lies in the main pathway of aeolian dust fluxes from Northwest Australia that are
119 transported by southeasterly trade winds (Bowler, 1976; Gallagher & deMenocal, 2019) (Figure
120 S1). Site U1464 is thus ideally located to study aeolian flux from Northwest Australia over
121 orbital timescales. A suite of 80 u-channels (each measuring 150 x 2 x 2 cm) were sampled at
122 Kochi Core Center, Japan, in January 2019, from continuous core sections spanning the upper
123 108.2 m of Hole U1464D. After non-destructive analyses (see below), the u-channels were sub-
124 sampled every 1 cm downcore into 'cube' sub-samples of $\sim 4 \text{ cm}^3$ (1 x 2 x 2 cm) for further
125 analyses.

126 2.2. Scanning X-ray fluorescence (XRF)

127 All of the Site U1464 u-channels were scanned at 0.5-cm intervals at the Research School of
128 Earth Sciences (RSES), Australian National University (ANU), Canberra, using a third
129 generation Avaatech XRF core scanner. Each u-channel was covered with 4 mm-thick Ultralene
130 film prior to measurement, and then measured at 10 kV with a 0.5 mA current and no filter, 30
131 kV with a 0.5 mA current and Pd-thin filter, and 50 kV with a 0.6 mA current and Cu filter.
132 Count times of 30 s, 20 s and 10 s were used for these three runs, respectively, and three replicate
133 measurements were taken every 75 cm to check reproducibility. Element spectra were processed

134 into element counts using WinAxil software, and reliable element data were obtained for Ca, Sr,
135 Fe, Ti, Zr, Rb and Si. 141 replicate samples are measured three times for estimation of
136 uncertainties, and precision for Ca, Sr, Fe, Ti, Zr, Rb and Si counts in studied interval was 0.2 %,
137 0.4 %, 0.7 %, 4.7 %, 3.3 %, 10.2 % and 1.0 %, respectively.

138 2.3. XRF Calibration

139 To convert the scanning XRF ‘counts’ into element concentrations, 50 of the cube subsamples
140 were chosen to cover a range of lithologies (based on the XRF scan results) and depth intervals.
141 Approximately ~0.2 g of each cube sample was oven-dried and ground with an agate mortar and
142 pestle. Sediment powder samples were soaked and extracted using 2 mL nitric acid and 1 mL
143 hydrochloric acid, and the supernate was diluted for the measurement. Single element
144 concentrations were determined using an Inductively Coupled Plasma Optical Emission
145 Spectrometer (ICP-OES) 5110 by Agilent at the RSES, ANU. High quality control of the ICP-
146 OES measurements was based on 10 certified reference materials (marine sediments BCSS-1,
147 MESS-1 and estuarine sediments 1646). Ca, Sr, Fe and Ti concentrations were well-above
148 detection limits for this method, with measurement precisions of 3.616 ‰, 0.007 ‰, 2.154 ‰
149 and 0.010 ‰, respectively (Table S1).

150 The Ca, Sr, Ti and Fe ICP-OES concentrations were used to convert Ca, Sr, Ti and Fe XRF
151 counts into concentrations using a multivariate log-ratio calibration, following Weltje et al.
152 (2015). This calibration method is more reliable than a basic univariate calibration (i.e., that
153 based on element-by-element regressions), which does not account for physical constraints on
154 compositional data or matrix effects due to the presence of other elements (Weltje et al., 2015).
155 An undefined variable is included in our calibration and is here termed ‘everything else’, hence
156 the relative concentrations of elements Ca, Sr, Ti, Fe and ‘everything else’ sum to 100%. The
157 predictive power of our calibration has been assessed by cross-plotting the reference with the
158 predicted concentrations (Figure S2). High r^2 values for Sr, Ti and Fe indicate a robust
159 calibration, which is strongest for Fe (0.92) and Sr (0.91). Nonetheless, due to calibration
160 problems for high carbonate material (Ellis et al., 2019), the relatively low r^2 value for Ca
161 suggests that the Ca calibration is not reliable. We therefore use the relationship between our
162 $\ln(\text{Ca}/\text{Fe})$ data and shipboard CaCO_3 measurements ($r^2=0.90$) to convert our scanning XRF
163 $\ln(\text{Ca}/\text{Fe})$ record into estimated CaCO_3 content (Table S2), following Liebrand et al. (2016).

164 2.4. Environmental magnetism

165 Magnetic properties were measured on all u-channels at 1-cm intervals using a 2-G Enterprises
166 Model 760 cryogenic magnetometer at the Black Mountain Paleomagnetism Laboratory, ANU.
167 For each u-channel, an anhysteretic remanent magnetization (ARM) was imparted in an
168 alternating field (AF) of 100 mT with a superimposed 0.05 mT bias field and then AF
169 demagnetized in 13 steps from 5 mT to 170 mT. Next, an isothermal remanent magnetization
170 (IRM) was imparted in a 1 T field, which was demagnetized in 13 stepwise AFs from 5 mT to
171 170 mT. The first/last four measurements of each parameter for every u-channel (topmost and
172 lowest 4 cm) were discarded to avoid edge effects impacting the resultant ARM and IRM
173 records.

174 2.5. Spectral analysis

175 To investigate the cyclicity of our time-series, power spectra were obtained using the REDFIT
176 3.8c (Schulz & Mudelsee, 2002) and cross-checked by the Multitaper method (Thomson, 1982).
177 Data were binned (averages in contiguous 0.3 kyr segments) to retain as much information as
178 possible, while avoiding the introduction of spurious serial dependence. Additionally, the cross
179 wavelet algorithm of Grinsted et al., 2004 was employed to investigate the phase relationship and
180 time evolution of specific cycles (records were resampled to 1 kyr bins before wavelet analysis).

181 3. Chronology

182 3.1. Bayesian age-depth model based on biostratigraphic ages

183 An initial chronology was developed for the upper ~126 m of IODP Site U1464 based on
184 shipboard biostratigraphy. For this, nineteen age control-points (Table S3) were used to construct
185 a Bayesian age-depth model using the “Bacon” package (Blaauw & Christen, 2011) (Figure 2a).
186 Cumulative depths below seafloor (CSF-A) for holes U1464B and U1464D were converted to
187 composite splice depths (CCSF-A) before modeling, based on the established linear relationship
188 between CSF-A and CCSF-A (Gallagher et al., 2017a), and a lithologic boundary was imposed
189 in the model at 43 m CSF-A (= 44.87 m CCSF-A), based on the Unit II/I transition in hole
190 U1464D (Gallagher et al., 2017a). Age uncertainties of the U1464 biostratigraphic datums are
191 mainly from Backman et al. (2012), Wade et al. (2011) and Gallagher et al. (2017a), and all dates
192 are close to or within the 99% probability interval of our Bacon age-depth model. The last

193 occurrence of *G. ruber* (pink) given by Thompson et al. (1979) is also consistent with our age
194 model (Figure 2a).

195 3.2. Refined age-depth model based on orbital tunings

196 Additional age control over the interval below ~47 m CSF-A was then investigated using our
197 scanning XRF records, with the goal of orbital tuning (geochemical signals are too
198 weak/ambiguous in the upper ~47 m at this site). Log(Fe/Ca) has been used as a riverine proxy
199 and a tuning target in marine sediments offshore Northwest Australia, while Zr/element ratios
200 have been used as dust proxies (Stuut et al., 2014, 2019; Pei et al., 2021). However, Log(Fe/Ca)
201 at Site U1464 (and other terrigenous element concentrations) appears to primarily reflect CaCO₃
202 variations (Figure 3; see also Section 4.1). Power spectra of our CaCO₃, Log(Zr/Fe) and
203 Log(Zr/Rb) records reveal significant cyclicity at ~413 kyr (long eccentricity) for CaCO₃ based
204 on depth scale and our biostratigraphic Bayesian model, and at ~41 kyr (obliquity) for
205 Log(Zr/Fe) and Log(Zr/Rb) based on our eccentricity tuning model (Figure S3; Figure S4).
206 Therefore, we focus first on tuning Site U1464 CaCO₃, (Section 3.2.1), followed by further fine-
207 tuning (to improve age control) using Log(Zr/Rb) (Section 3.2.2). Finally, we re-ran the Bacon
208 age model using all age constraints with their uncertainties (Section 3.3), thus yielding a new
209 orbitally-constrained, statistically evaluated chronology for Site U1464.

210 3.2.1. CaCO₃ tuning at eccentricity 400 kyr band

211 Spectral analyses revealed significant power in Site U1464 CaCO₃ at frequencies of ~0.0024 kyr⁻¹
212 ¹, equivalent to 413 kyr periodicity (Figure S3a, S3f; Figure S4). This periodicity has previously
213 been used as a tuning target for paleoclimate records from the adjacent IODP Site U1463 (Figure
214 1) (Christensen et al., 2017; De Vleeschouwer et al., 2018; Groeneveld et al., 2021). CaCO₃
215 content in marine sediments is mainly driven by carbonate production and water column/seafloor
216 dissolution (Keil et al., 2017), and its long-term variations have been shown to be paced by
217 Earth's ~400 kyr eccentricity cycle (Kochhann et al., 2016; Liebrand et al., 2016; Moore et al.,
218 1982). The possible mechanism for ~400 kyr eccentricity cyclicity in CaCO₃ at Site U1464 is
219 outlined in Section 4.1. Comparison of Site U1464 CaCO₃ with eccentricity shows that CaCO₃
220 maxima tend to correspond with (413 kyr) eccentricity minima (Figure 4), which is consistent
221 with a negative correlation between insolation minima and enhanced sedimentary carbonate
222 content in previous studies, and *vice versa* (Kochhann et al., 2016; Liebrand et al., 2016). This

223 anti-phasing has also been observed in the benthic and planktonic foraminiferal $\delta^{13}\text{C}$ records of
224 nearby IODP Site U1463 (De Vleeschouwer et al., 2018; Groeneveld et al., 2021), as well as in
225 global-scale benthic and planktonic $\delta^{13}\text{C}$ records (Kochhann et al., 2016; Liebrand et al., 2016;
226 Pälike et al., 2006; Turner et al., 2014; Wang et al., 2010), and likely indicates a ~400 kyr
227 rhythm in the oceanic carbon reservoir. Additional graphic evidence for anti-phasing between
228 Site U1464 CaCO_3 and eccentricity is the cycle-shape of CaCO_3 variations, which display broad
229 troughs and narrow peaks, i.e., pacing with the inverse of the 413 kyr eccentricity cycle (Figure
230 4). The Site U1464 chronology was therefore further constrained by four tie-points between
231 eccentricity minima and CaCO_3 maxima, after trialing different tuning options (Figure S5; Table
232 S4; Supporting Information Text S1). The tie-points all fall within the 99% confidence intervals
233 of the Bacon age-depth model, hence they are consistent with the Site U1464 biostratigraphy
234 (Figure 2a).

235 3.2.2. Log(Zr/Rb) tuning at obliquity 41 kyr band

236 Spectral analyses of the rescaled records after eccentricity tuning revealed significant power in
237 Log(Zr/Rb) (and Log(Zr/Fe) and $\text{IRM}_{\text{IT@AF 170mT}}$) at $\sim 0.024 \text{ kyr}^{-1}$ (equivalent to $\sim 41 \text{ kyr}$
238 periodicity, Figure S3). Several studies have used Zr as a dust proxy in sediments offshore
239 Northwest Australia (Pei et al., 2021; Stuut et al., 2014, 2019), as windblown heavy/coarse
240 minerals (e.g., ZrSiO_4) are enriched in Zr. Conversely, Rb and Fe are preferentially incorporated
241 into riverine fine-grained clays (Pei et al., 2021; Rothwell & Croudace, 2015; Stuut et al., 2014,
242 2019), and thus Log(Zr/Rb) and Log(Zr/Fe) likely reflect aeolian/fluvial (or aeolian) fluxes (see
243 Section 4.2 for further discussion). Bandpass filtering of Log(Zr/Rb) at $0.023\text{-}0.027 \text{ kyr}^{-1}$
244 frequencies reveals close (positive) covariation with obliquity (Figure 5). Recent studies
245 observed an inverse relationship between Australian summer monsoon precipitation proxies
246 (local surface seawater $\delta^{18}\text{O}$ and $\ln(\text{K}/\text{Ca})$ at IODP Site U1483) and obliquity during the Late-
247 Pleistocene (Zhang et al., 2020; 2022), where higher obliquity was associated with less
248 precipitation (hence less vegetation cover and more erodible particles) and *vice versa*. Similarly,
249 a model-based investigation of obliquity forcing suggested stronger southeasterly trade winds
250 during higher obliquity (Bosmans et al., 2015); such a scenario would favour transport of dust
251 particles from Northwest Australia to Site U1464. Based on this obliquity-dust relationship and
252 the close match of obliquity with our filtered Log(Zr/Rb) record, we further constrain our Site

253 U1464 chronology with 32 additional tie-points between minima in obliquity and our filtered
254 Log(Zr/Rb) record (Figure 5; Table S5; Figure 2a).

255 3.3. Refined Bayesian chronology based on biostratigraphy and orbital tunings

256 To consider potential uncertainties of all age constraints, we re-ran the Bacon age model using
257 the 55 age controls, including 17 biostratigraphic ages, 4 eccentricity tuning ties, 32 obliquity
258 tuning ties, the last occurrence of *G. ruber* (pink) and modern age at the seafloor. This refined
259 Bacon age model can account for most of the age constraints within its 99.99% confidence
260 interval, with the exception of four biostratigraphic datums which are clearly outside the model
261 (Figure 2b).

262 Comparison of our Site U1464 chronology with recent chronostratigraphic studies of NWS
263 sediments reveals some inconsistencies. For example, dynamical time warping was employed to
264 align a portion of the Site U1464 wireline natural gamma ray (NGR) record (on depth) to that of
265 neighboring Site U1463 (Groeneveld et al., 2021). Conversion of the dynamically warped U1464
266 depths to CCSF depths and to our age-scale reveals that five (two) points lie outside (inside) our
267 99% confidence intervals (green crosses in Figure S6). Another offset occurs between some re-
268 dated NWS biostratigraphic datums (based on a revised chronostratigraphy for Site U1463;
269 Groeneveld et al., 2021) and the shipboard ages for these datums at Site U1464 (purple crosses
270 on Figure S6). It is difficult to ascertain which chronostratigraphic markers are more/less
271 reliable, especially as dating NWS sediments from IODP Expedition 356 has not been
272 straightforward (Gallagher et al., 2017a; Groeneveld et al., 2021). There appears to be
273 diachroneity among many biostratigraphic datums (Gallagher et al., 2017a; Groeneveld et al.,
274 2021), magnetic intensities (for magnetostratigraphy) are weak (Gallagher et al., 2017a, 2017b),
275 and $\delta^{18}\text{O}$ stratigraphy is either absent or presents a noisier signal compared to deeper ocean sites
276 (Groeneveld et al., 2021; Pei et al., 2021; Stuut et al., 2014, 2019). In addition, wireline logs and
277 scanning XRF records from upper Pleistocene sediments at Site U1463 and U1464 show weak
278 signals (Gallagher et al., 2017a, 2017b; Groeneveld et al., 2021; this study). This may account
279 for uncertainties in dynamic warping between Sites U1463 and U1464, such as observed in
280 Figure S6. However, while our inferred U1464 ages may be up to 200 kyr older at ~60-90 m and
281 ~130 m, shifting our age-depth relationship in that direction would be at odds with the majority
282 of the biostratigraphic datums, as well as with the eccentricity tuning. For the latter, we tried 12

283 different tunings (Figure S5; Table S4; Supporting Information Text S1), but all alternatives
284 result in too much stretching/squeezing (hence unrealistic sedimentation rates and changes) or
285 inconsistent eccentricity-CaCO₃ relationships (Figure S5). Therefore, in light of our (generally)
286 compatible Bayesian model with biostratigraphy and orbital tunings, we consider our chronology
287 to be sufficiently robust for Site U1464, bearing in mind the aforementioned caveats.

288 **4. Results and Discussion**

289 4.1. Carbonate variations at IODP Site U1464

290 The calibrated scanning XRF results show that CaCO₃ and Sr have consistent variations (Figure
291 3), indicative of their marine/biogenic origins (Rothwell & Croudace, 2015). Fe, Ti, Rb and Zr
292 also show similar trends (Figure 3), which likely indicate terrigenous signals as these are typical
293 detrital elements (Rothwell & Croudace, 2015). However, marine and terrigenous variations
294 have an inverse relationship, hence their co-variability may be driven by marine carbonate
295 production, carbonate dissolution, or dilution by terrigenous inputs. Calculated total mass
296 accumulation rates (MARs) appear to be more consistent with CaCO₃ and Sr MARs variations
297 compared to terrigenous element MARs (Figure S7), which suggests that carbonate
298 production/dissolution, rather than terrigenous dilution, predominated in bulk sediment
299 geochemical variations at Site U1464 between ~2.9 and ~1.5 Ma. This inference is consistent
300 with low Al and K concentrations in ICP-OES and scanning-XRF results from Site U1464,
301 which reflect a minor clay mineral-bound fine-grained fraction of riverine runoff on the
302 Australian NWS (Kuhnt et al., 2015; Rothwell & Croudace, 2015; Zhang et al., 2020). Our
303 inference is further supported by the distinct 413 kyr eccentricity signal in Site U1464 CaCO₃
304 and a known ~400 kyr rhythm in the oceanic carbon reservoir (Kochhann et al., 2016; Liebrand
305 et al., 2016; Pälike et al., 2006; Wang et al., 2010). Additionally, the significant 413 kyr signal
306 in Site U1464 CaCO₃ may be attributed to ~2.4-Myr amplitude modulation (AM) (Liebrand et
307 al., 2016), as our study interval falls within the last 2.4-Myr eccentricity minimum (Figure 4);
308 i.e., there is relatively more power in the 400 kyr eccentricity cycle during minima in the 2.4-
309 Myr eccentricity cycle (Laskar et al., 2004).

310 Calcification rates for CaCO₃ shell-forming organisms are closely linked to the CaCO₃
311 saturation state (Ω), which is expressed as: $[\text{Ca}^{2+}][\text{CO}_3^{2-}]/K_{sp}^*$, where K_{sp}^* is the stoichiometric
312 solubility product for CaCO₃ (Feely et al., 2004; Kleypas et al., 1999). Ω depends mainly on the

313 concentration of $[\text{CO}_3^{2-}]$, since the seawater Ca^{2+} concentration is relatively stable over the
314 studied timescale. Natural and culture experiments indicate that most calcifying organisms,
315 especially those with aragonite or high-Mg calcite, may decrease carbonate production in
316 response to a decreased $[\text{CO}_3^{2-}]$, even for $\Omega > 1$ (Beaufort et al., 2011; Feely et al., 2004;
317 Kleypas et al., 1999). In addition, decreased $[\text{CO}_3^{2-}]$ and saturation state could reduce abiotic
318 carbonate precipitation on the NWS (Gallagher et al., 2018; Hallenberger et al., 2019; 2022).
319 CaCO_3 dissolution is unlikely to have occurred at Site U1464 due to its relatively shallow
320 water-depth compared to the lysocline and carbonate compensation depth. CaCO_3 variations at
321 Site U1464 are therefore interpreted to mainly reflect changes in local sea-water $[\text{CO}_3^{2-}]$ and
322 related CaCO_3 production.

323 Percentage carbonate at Site U1464 was relatively elevated at ~ 2.89 - 2.74 Ma, ~ 2.46 - 2.34 Ma,
324 ~ 2.13 - 1.89 Ma and ~ 1.71 - 1.58 Ma (Figure 6). These broad variations are also registered in
325 benthic $\delta^{13}\text{C}$ at IODP Site U1482 offshore Northwest Australia and at Deep Sea Drilling Project
326 (DSDP) Site 593 in the southwest Pacific, but less obviously in benthic $\delta^{13}\text{C}$ at nearby Site
327 U1463 (Figure 6), although 413 kyr variability has been demonstrated in the latter record over
328 ~ 5.2 - 1.7 Ma (Groeneveld et al., 2021). The benthic $\delta^{13}\text{C}$ discrepancy between Site U1463 and
329 U1482/593 may be attributed to different benthic foraminifera species. Epifaunal *P.*
330 *wuellerstorfi* $\delta^{13}\text{C}$ (at Site U1482, Chen et al., 2022 and Site 593, McClymont et al., 2016)
331 usually records bottom-water dissolved inorganic carbon $\delta^{13}\text{C}$ in a positive relationship,
332 however infaunal *Uvigerina* spp. $\delta^{13}\text{C}$ (at Site U1463, Groeneveld et al., 2021) is also
333 influenced by bottom-water dissolved oxygen content and organic matter fluxes to the sea-floor,
334 which can bias its $\delta^{13}\text{C}$ toward more negative values (Mackensen & Schmiedl, 2019). We
335 therefore focus on and interpret the positive covariation between Site U1464 CaCO_3 and Site
336 U1482/Site 593 $\delta^{13}\text{C}$ records, in terms of seawater dissolved inorganic carbon $[\text{CO}_3^{2-}]$ and $\delta^{13}\text{C}$.
337 Considering the water masses influencing these records, the seafloor at Site U1464 (~ 270 m
338 water depth at present) is currently bathed by the Leeuwin Undercurrent (LUC), and its water-
339 masses mainly derive from Sub-Antarctic Mode Water (SAMW) and/or Antarctic Intermediate
340 Water (AAIW) via the Flinders Current offshore South Australia (Herraiz-Borreguero &
341 Rintoul, 2011; Richardson et al., 2019; Wijeratne et al., 2018; Wong, 2005; Woo & Pattiaratchi,
342 2008). Seawater temperature profiles nearby based on *in-situ* and modeling data also show a

343 cold water-mass at ~150-100 m depth, indicating intrusion of a southerly-sourced cold water
344 mass (Ridgway & Godfrey, 2015). SAMW is characterized by high-oxygen content (Herraiz-
345 Borreguero & Rintoul, 2011; Woo & Pattiaratchi, 2008), while AAIW is characterized by a
346 salinity minimum (Wong, 2005; Woo & Pattiaratchi, 2008) and low- $[\text{CO}_3^{2-}]$, thus making it
347 relatively corrosive to carbonate. Profiling float data show that modern SAMW and AAIW are
348 injected northward into the Indian Ocean at ~15°S (Herraiz-Borreguero & Rintoul, 2011;
349 Wong, 2005), and this northward penetration extends to Site U1464. These water masses can be
350 identified in $[\text{CO}_3^{2-}]$, oxygen and salinity profiles from the GLODAPv2.2021 dataset for two
351 transects off Western Australia (Figures S8, S9).

352 *P. wuellerstorfi* $\delta^{13}\text{C}$ at DSDP Site 593 mainly reflects the dissolved inorganic carbon $\delta^{13}\text{C}$ of
353 intermediate water, which is primarily southern-sourced AAIW with minor local contributions
354 (Elmore et al., 2015; McClymont et al., 2016). *P. wuellerstorfi* $\delta^{13}\text{C}$ at Site U1482 records the
355 dissolved $\delta^{13}\text{C}$ content of Indonesian Intermediate Water (IIW), which originates from AAIW in
356 the western Pacific (Figure S8b, c; Figure S9b, c) (Talley & Sprintall, 2005; Zenk et al., 2005;
357 Wong, 2005). Thus, the intrusion intensity of low- $[\text{CO}_3^{2-}]$ AAIW to Sites U1464 and DSDP 593,
358 and indirectly to Site U1482, or changes in AAIW $[\text{CO}_3^{2-}]$, may explain co-variability in the
359 respective CaCO_3 and $\delta^{13}\text{C}$ records (Figure 6). In addition, the proximity of Sites U1464 and
360 U1482 suggests a more direct influence of water masses. At Site U1482, dissolved $[\text{CO}_3^{2-}]$ is
361 high in surface waters due to photosynthesis and decreases with depth as organic matter degrades
362 (Figure S8a, S9a). Dissolved $\delta^{13}\text{C}$ in the water column reflects this biological process, whereby
363 surface/intermediate waters are characterized by heavy/light $\delta^{13}\text{C}$, respectively. Indonesian
364 surface waters feed the south-flowing Leeuwin Current which overlies the Australian NWS;
365 hence, the ventilation and mixing of Intermediate and surface waters at Sites U1482 and U1464
366 could account for similar trends in their respective $\delta^{13}\text{C}$ and CaCO_3 % records (Figure 6a). This
367 interpretation is in line with hypotheses that ocean circulation played an important role in the
368 ~400 kyr eccentricity forcing of the global oceanic carbon reservoir (e.g., Wang et al., 2010).

369 4.2. Aeolian inputs to IODP Site U1464

370 From our scanning XRF results, Zr, Fe and Rb values can be used to isolate the aeolian
371 component from the total terrestrial composition at Site U1464. $\text{Log}(\text{Zr}/\text{Fe})$ has been used at
372 ODP Site 762 (Figure 1) as a proxy for aeolian dust from Northwest Australia (Stuut et al.,

373 2014, 2019). Log(Zr/Fe) can indicate the transport pathway and grain-size of terrigenous
374 particles, since the wind-blown fraction is enriched in Zr due to its presence in coarser and/or
375 heavier grains, whereas Fe is enriched in run-off sediments (Stuut et al., 2014, 2019). We also
376 use Log(Zr/Rb) as a dust proxy, as Rb is preferentially incorporated into riverine fine-grained
377 clays (Pei et al., 2021; Rothwell & Croudace, 2015). The Log(Zr/riverine element) ratio is
378 therefore used to estimate the relative contribution of aeolian versus riverine inputs.

379 Isothermal remanence magnetism (IRM) has also been used to approximate past aeolian dust
380 fluxes in marine cores (Larrasoana et al., 2003; Grant et al., 2022), where the IRM remaining
381 after AF demagnetization at 170 mT ($IRM_{1T@AF\ 170mT}$) reflects the relative contribution from
382 magnetic minerals with a coercivity of remanence larger than 170 mT (i.e., imperfect
383 antiferromagnetic minerals, such as hematite) (Verosub & Roberts, 1995). Hematite is generally
384 thought to be preferentially formed in dry and hot environments, and then transported to marine
385 sediments via aeolian dust. Because hematite concentration is affected by variations in the
386 concentrations of other constituents, the variations of $IRM_{1T@AF\ 170mT}$ may simply reflect
387 dilution by carbonate. To eliminate this dilution effect, CaCO₃-free $IRM_{1T@AF\ 170mT}$ was
388 calculated based on the following formula: $CaCO_3\text{-free } IRM_{1T@AF\ 170mT} = [IRM_{1T@AF\ 170mT}] / (1 -$
389 $[CaCO_3])$, where $[CaCO_3]$ ranges from 0 to 1. The long-term consistency of magnetic
390 susceptibility, ARM, IRM_{1000mT} , $IRM_{1T@AF\ 170mT}$ with CaCO₃-free $IRM_{1T@AF\ 170mT}$ indicates that
391 their variations represent the changes in magnetic mineral concentrations, despite some
392 differences in short-term variations (Figure S10), however it is not straightforward to use the
393 magnetic susceptibility, ARM, IRM_{1000mT} as aeolian dust proxies. Thus, we omit to use these
394 magnetic parameters in this study. Broadly consistent long-term trends and higher frequency
395 co-variability in the obliquity band between our magnetic hematite proxy and geochemical dust
396 proxies (Figure 7a-c, Figure S11, note reversed y-axes; see Supporting Information Text S2)
397 corroborate their interpretation as aeolian dust proxies at Site U1464.

398 For comparison, a Log(Zr/Fe) record is available for ODP Site 762 offshore Northwest Australia
399 (Stuut et al., 2019) (Figure 1; Figures 7e). In terms of absolute values, Log(Zr/Fe) at Site U1464
400 has higher values than that at ODP Site 762 during the same period. Discrete samples from
401 modern river beds and dune sediments of Northwest Australia showed that Log(Zr/Fe) values of
402 dune sediments are generally greater than -1, while those in river beds are typically less than -1
403 (Stuut et al., 2019). If modern end-member values are applicable over the Late Pliocene/Early

404 Pleistocene, it implies that Site U1464 Log(Zr/Fe) values mostly reflect dune end-members
405 (based on a 21-point running average (Figure 7c), although some individual data-points are less
406 than -1), while ODP Site 762 Log(Zr/Fe) values reflect riverine sources (Figure 7e). At first
407 glance this observation appears counter-intuitive, as Site 762 is located further offshore than Site
408 U1464 (Figure S1) and the proportion of riverine (aeolian) components in off-shore sediment
409 tends to decrease (increase) with distance from river mouths (Stuut et al., 2019). However, ODP
410 Site 762 is more proximal to outflow from the Fortescue, Ashburton and Lyndon-minilya rivers
411 than Site U1464 (Figure S1), which, in contrast, lies directly in the path of the Northwest
412 Australian dust belt (Figure S1, Bowler, 1976). An additional/alternative explanation is that Site
413 U1464 lies on the shelf slope at ~260 m water depth, while ODP Site 762 is further offshore at
414 1360 m water depth, so prevailing currents and sediment dynamics/transport differ between the
415 sites. Inconsistent trends between Site U1464 and Site 762 Log(Zr/Fe) records may therefore be
416 explained by their contrasting locations and associated differences in the proportion of
417 riverine/dust components reaching each site. Chronological offsets/uncertainties could also
418 explain some discrepancies between the records, although it is hard to explain such differing
419 trends by chronology alone (e.g., the records are almost anti-phased between ~2.9 and ~2.4 Ma).

420 Better agreement is observed between our IODP Site U1464 Log(Zr/Fe) record and a downhole
421 natural gamma radiation (NGR)-derived Th/K record from neighboring IODP Site U1463
422 (Christensen et al., 2017) (Figure 1; Figure 7d), taking into account the chronological
423 uncertainties/offsets between these sites. The Th/K record has been interpreted as a dust proxy
424 (Christensen et al., 2017), and potentially synchronous dust maxima can be identified between
425 our dust records and the Site U1463 Th/K record (dashed lines in Figure 7a-d). Nonetheless,
426 there are uncertainties in NGR-derived K and Th measurements (De Vleeschouwer et al., 2017).
427 One limitation is that the wireline NGR system integrates counts from a 40-cm long core, which
428 causes significant smoothing of signals during measurements. Also, the algorithm that produces
429 accurate estimates of K and Th content relies on different density measurements (gamma ray
430 attenuation, moisture and density), and these density measurements can contribute additional
431 errors to the K and Th estimates. The above uncertainties may explain some of the offsets
432 between our U1464 dust proxy records and the Site U1463 Th/K record.

433 4.3. Obliquity-paced Northwest Australian dust fluxes

434 Spectral analyses of our dust records reveal significant peaks corresponding to three obliquity
435 frequencies (at $\sim 0.024 \text{ kyr}^{-1}$ (41 kyr), $\sim 0.034 \text{ kyr}^{-1}$ (29 kyr) and $\sim 0.019 \text{ kyr}^{-1}$ (54 kyr), based on
436 [Hinnov \(2000\)](#) and [Mélise et al. \(2001\)](#)) using the Redfit method ([Figure S3c-e](#)). These three
437 frequencies are also observed in the Multitaper method ([Figure S3h-j](#)), showing that these three
438 obliquity frequencies can be identified using different approaches. Furthermore, the position and
439 relative amplitude of the 0.019 kyr^{-1} peak, and to a lesser extent the 0.034 kyr^{-1} peak, are
440 relatively insensitive to our obliquity tuning and Bacon age modelling ([Figure S3](#); [Figure S12](#)),
441 thus underscoring the reliability of our spectral analyses and the dominance of obliquity in Site
442 U1464 dust-flux proxy records. Spectral power in the precession band is less pronounced, and
443 limited to $\sim 19 \text{ kyr}$ periodicity ([Figure S3](#)). Interestingly, the 0.019 and/or 0.034 kyr^{-1} frequencies
444 have also been detected in Australian monsoon records ([Holbourn et al., 2005](#); [Kershaw et al.,](#)
445 [2003](#); [Liu et al., 2015](#); [Zhang et al., 2020, 2022](#)) and in aeolian grain size records from other
446 Indo-Pacific regions ([Clemens & Prell, 1990](#); [Hovan et al., 1991](#); [Pisias & Rea, 1988](#)), which
447 suggests that these frequencies may be a consistent feature of large-scale atmospheric
448 circulations over the Plio-Pleistocene ([see Section 4.4 below](#)).

449 The spectral peak in Site U1464 dust proxies at $\sim 41 \text{ kyr}$ could be explained by a linear response
450 to obliquity forcing, since this is the dominant frequency of obliquity. However, the amplitude of
451 the $\sim 54 \text{ kyr}$ peak in our dust proxies is much higher than that of the 41 kyr peak, contrary to the
452 obliquity spectrum ([Hinnov, 2000](#); [Laskar et al., 2004](#); [Mélise et al., 2001](#)); the 54 kyr peak may
453 therefore reflect a non-linear response to obliquity forcing or a heterodyne of other orbital
454 frequencies. To investigate the evolution of these two obliquity periods ($\sim 0.024 \text{ kyr}^{-1}$ and ~ 0.019
455 kyr^{-1}) in our time-series, we calculated bandpass-filters of $0.023\text{-}0.027 \text{ kyr}^{-1}$ and $0.017\text{-}0.021 \text{ kyr}^{-1}$
456 1 in $\text{Log}(\text{Zr/Rb})$, and found that the 0.024 kyr^{-1} frequency dominated during the $\sim 2.9\text{-}2.24 \text{ Ma}$
457 interval, while the 0.019 kyr^{-1} frequency became dominant after $\sim 2.1 \text{ Ma}$ ([Figure S13b, c](#)). This
458 frequency shift from 0.024 to 0.019 kyr^{-1} might be attributed to $\sim 1.2\text{-Ma}$ obliquity AM, which
459 decreased during the interval of $2.24\text{-}2.1 \text{ Ma}$ ([Figure S13a](#)) ([Hinnov, 2000](#); [Laskar et al., 2004](#)).
460 The transformation from AM into frequency modulation (FM) seems consistent with previously
461 proposed FM hypotheses, in which a high-frequency carrier can be modulated by a low-
462 frequency modulator to create sidebands based on $F_{\text{Carrier}} \pm F_{\text{Modulator}} = F_{\text{Sideband}}$ ([Rial, 1999](#); [Rial](#)
463 [& Anaclerio, 2000](#)). In this case, the 0.019 ($1/54$) kyr^{-1} peak in our observation coincides with

464 the sideband of a 0.024 ($1/41$) kyr^{-1} carrier modulated by a 0.005 ($1/171$) kyr^{-1} modulator; that is
465 $0.024 - 0.005 = 0.019$, in which the ~ 0.005 ($1/171$) kyr^{-1} originates from the obliquity AM
466 (Hinnov, 2000; Mélice et al., 2001). Similarly, the 0.034 ($1/29$) kyr^{-1} peak may also be the
467 sideband of a 0.024 ($1/41$) kyr^{-1} modulated by the 0.005 ($1/171$) kyr^{-1} (i.e., $0.024 + 2 \cdot 0.005 =$
468 0.034). The specific origins of these obliquity signals will be further discussed in the following
469 section.

470 4.4. Origin of obliquity signals in Northwest Australian dust records

471 Here we explore possible origins of the dominant obliquity-scale variability in our dust proxy
472 records, and what this reveals about climate forcing of Australian dust fluxes over longer orbital
473 timescales. Two key climate forcings to consider are i) high-latitude climate forcing via ice-sheet
474 fluctuations and/or the EAWM, and ii) direct low-latitude insolation forcing via the SITIG
475 (summer inter-tropical insolation gradient). Both of these mechanisms show strong obliquity
476 periodicities and have been proposed to explain the presence of obliquity cycles in low-latitude
477 monsoon and aeolian dust fluxes (Beck et al., 2018; Ding et al., 2002; Hennekam et al., 2022;
478 Leuschner & Sirocko, 2003; Liu et al., 2015; Stuut et al., 2019; Sun et al., 2006).

479 4.4.1. High-latitude climate forcing: Glacial cycles and the EAWM

480 We first note that it is difficult to disentangle the potential influences of glacial cycles and the
481 EAWM, because intensity variations in the EAWM over orbital timescales were closely
482 associated with (Northern Hemisphere) ice-sheet fluctuations (Ding et al., 2002; Sun et al.,
483 2006). Nonetheless, ice sheets and EAWM intensity could affect Northwest Australian dust
484 fluxes by different mechanisms. For example, the extent of large Northern Hemisphere ice sheets
485 determines pole-equator temperature gradients and the position and strength of latitudinal wind
486 bands (Clark et al., 1999; Ganopolski et al., 1998), and we could consider this as an ‘indirect’
487 forcing of Northwest Australian dust fluxes, as it pertains to global atmospheric changes.
488 Additionally, Northern Hemisphere ice sheets could indirectly influence NWS dust variations via
489 sea-level fluctuations, as the distance between coastline and Site U1464 might control fluvial
490 inputs (Stuut et al., 2019) and consequently influence dust content via fluvial dilution. In
491 contrast, the EAWM is directly coupled to the Australian monsoon via cross-equatorial flows
492 (Liu et al., 2015; Suppiah, 1992; Suppiah & Wu, 1998). Accordingly, past changes in Northwest
493 Australian dust fluxes may have been influenced indirectly by ice-sheet forcing via large-scale

494 atmospheric circulations and sea-level fluctuations, or directly by EAWM intensity via local
495 cross-equatorial flows.

496 We compare our dust records to global sea level (= ice volume) and to a stacked mean grain-size
497 of quartz (MGSQ) record from the Chinese Loess Plateau (Figure 8a-d), to investigate whether
498 the obliquity signal in the Site U1464 dust fluxes reflects an indirect influence of Northern
499 Hemisphere ice sheets or a direct influence of EAWM intensity. Clearly, our Australian dust-flux
500 reconstructions show no consistent phasing with ice volume, even if both have high common
501 power at the 41 kyr obliquity band (Figure 8a, b; Figure S12a). In addition, our geochemical
502 results show that terrestrial sediments are mainly composed of aeolian dust at Site U1464 and
503 indicate that fluvial input was minor. The palaeodepth estimation (>300 m water depth during
504 the Early Pleistocene) (Gallagher et al., 2017a) and absence of potential river sources at Site
505 U1464 further suggest that this region was unlikely to receive significant amounts of fluvial
506 siliciclastic input. Accordingly, the 41 kyr cycles in our dust proxies were unlikely influenced by
507 fluvial dilution caused by eustatic sea-level fluctuations. From these observations, we can rule
508 out the ‘indirect’ forcing of ice volume, consistent with previous studies about the Indian
509 summer monsoon based on Arabian Sea sediments (Clemens et al., 1991a; Clemens & Prell,
510 2003; Leuschner & Sirocko, 2003) and the (Early Pleistocene) North African monsoon based on
511 eastern Mediterranean Sea sediments (Hennekam et al., 2022; Reichert, 1997). Although there is
512 no linear correlation between global ice volume and Northwest Australian dust fluxes, ice
513 volume might exert an influence indirectly via the EAWM, given that the EAWM was
514 influenced by North Hemisphere ice sheets (Ding et al., 2002; Sun et al., 2006).

515 Comparison between Site U1464 Log(Zr/Fe) and the stacked MGSQ record reveals that
516 Northwest Australian dust flux had a similar, but anti-phased, long-term trend with EAWM
517 intensity (Figure 8c). In general, decreased dust fluxes off Northwest Australia correspond to a
518 stronger EAWM, and *vice versa*. A straightforward explanation is that enhanced (diminished)
519 EAWM intensity would favor a stronger (weaker) Australian summer monsoon, expanded
520 (reduced) vegetation cover, less (more) erodible particles in source areas, and dampening
521 (strengthening) of southeast trade winds which transport dust from the Australian interior
522 towards the NWS. Three peaks associated with obliquity frequencies are present in both records
523 over the 1.6-2.9 Ma interval (Figure S12b, d-f); however, cross-wavelet analysis suggests an
524 ambiguous relationship between Site U1464 dust fluxes and EAWM variability at obliquity

525 frequencies (Figure 8d). This inconsistency may be an artifact of age model uncertainties and
526 assumptions in both records. For example, different obliquity phasings were assumed in their
527 orbital tuning. The stacked MGSQ chronology involves an assumed 8-kyr lag with respect to
528 obliquity (Ding et al., 2002; Sun et al., 2006), while we assumed zero phase lag between Site
529 U1464 dust fluxes and obliquity in this study. We did, however, apply an age uncertainty of ± 20 -
530 kyr for the obliquity tuning tie-points in our Bacon age model. The 8-kyr lag for the MGSQ
531 record is based on SPECMAP (Imbrie et al., 1984); hence, it is based on relatively large
532 amplitude glacial-interglacial cycles of the Late Pleistocene, whereas a smaller lag should be
533 expected for the Early Pleistocene. Equally, there may be an ice-volume effect on our dust
534 records, via the EAWM, in which case a lagged obliquity tuning may be more appropriate.
535 Therefore, although we cannot objectively compare obliquity-band phasing of our dust records
536 with the stacked MGSQ record, we nonetheless propose EAWM intensity as a possible origin of
537 the obliquity cycles observed in our Site U1464 dust-flux time series, given the similarity of their
538 long-term trends and obliquity-band spectral peaks, and a known physical mechanism (cross-
539 equatorial flow) linking the two monsoon regions.

540 4.4.2. Low-latitude SITIG forcing

541 The SITIG (i.e., the difference in summer insolation between 30° N and 30° S) is a low-latitude
542 insolation forcing which contains notable precession and obliquity components (unlike, for
543 example, 30° N or 30° S insolation time-series), and has been invoked as a potential driver of
544 East Asian (Beck et al., 2018), Indian (Leuschner & Sirocko, 2003) and African monsoon
545 (Reichert, 1997) variability over orbital timescales. This monsoon-forcing mechanism has been
546 corroborated by model simulations and has a physical basis as described by previous studies
547 (Beck et al., 2018; Bosmans et al., 2015; Mantsis et al., 2014). In this mechanism, a stronger
548 SITIG can drive an intensified winter Hadley circulation and thus a stronger cross-equatorial
549 moisture transport into the summer hemisphere, and this has been validated for seasonal
550 (Schwendike et al., 2014) and orbital timescales (Beck et al., 2018; Bosmans et al., 2015;
551 Mantsis et al., 2014).

552 Obliquity plays a key role in modulating the SITIG and meridional heating gradients. Several
553 modelling studies have demonstrated that when the SITIG is stronger during higher obliquity,
554 high-pressure anticyclones in the Southern Hemisphere (which form the descending limb of the

555 winter hemisphere Hadley cell) are strengthened, while high-pressure anticyclones in the
556 Northern Hemisphere (which form the descending limb of the summer hemisphere Hadley cell)
557 are weakened (Beck et al., 2018; Bosmans et al., 2015; Mantsis et al., 2014; Schwendike et al.,
558 2014). Thus, during a stronger obliquity-induced SITIG, intensified cross-equatorial winds and
559 moisture transport into the Northern Hemisphere is consistent with intensified southeasterly trade
560 winds and a more arid Australian interior, and *vice versa*. Northwest Australian dust fluxes can
561 therefore respond immediately to changes in the SITIG via these dynamics. Since muted
562 precession and large obliquity are present in Site U1464 dust proxy records (Figure S3), the
563 summer half-year (21 March - 20 September), instead of one single month, was calculated in the
564 SITIG as follows: $SITIG = I_{\text{March-September}}(30^\circ \text{ N}) - I_{\text{March-September}}(30^\circ \text{ S})$, where $I_{\text{March-September}}(30^\circ$
565 $\text{ N or } 30^\circ \text{ S})$ is sum of monthly insolation from 21 March to 20 September at $30^\circ \text{ N or } 30^\circ \text{ S}$. This
566 calculation can amplify obliquity and mute precession (Figure S12c), and is hence more
567 consistent with our observations. There is reasonable agreement between Site U1464 dust fluxes
568 and SITIG before 2.55 Ma and after 2.35 Ma (Figure 8e, f), in both frequency and amplitude
569 variability. Between 2.55 and 2.35 Ma the records appear to be offset, i.e., there appears to be a
570 phase lag of dust flux relative to the SITIG. Given that we assumed no phase lag in our obliquity
571 tuning (based on the near-direct mapping of the filtered $\text{Log}(\text{Zr/Rb})$ onto obliquity prior to
572 tuning, Figure 5), comparison of the SITIG to our dust proxy record cannot include interpretation
573 of phase relationships. Nonetheless, the agreement through most the records in both amplitude
574 and dominant frequency is consistent with SITIG forcing of Northwest Australian dust fluxes
575 through the Early Pleistocene.

576 An additional/alternative consideration is that the 54 kyr obliquity cycle in our dust records
577 might be amplified by a nonlinear response to the AM of obliquity (and hence to the resultant
578 SITIG). For example, the long-term mean of the SITIG AM narrowed down to a lower value
579 during ~2.24-2.1 Ma due to the 1.2-Myr obliquity AM (Figure S13a), and the decreased
580 amplitude of SITIG variability, amplified by internal climate feedbacks, could have shifted the
581 sensitivity of Northwest Australian dust fluxes to SITIG forcing from a 41 kyr cycle to a
582 prolonged 54 kyr cycle (Figure S13b, c). On one hand, smaller amplitude in the SITIG would
583 reduce the relative strength of winter Hadley circulation and associated cross-equatorial flows
584 between SITIG maxima and minima, thereby weakening the 41 kyr effect on dust fluxes over
585 Northwest Australia. On the other hand, smaller SITIG amplitude after ~2.1 Ma would make the

586 171 kyr AM cycle comparable or stand out with respect to the 41 kyr cycle, which would
587 explain the presence of 171 kyr cycles in our dust records (Figure S13a, d, e). The 54 kyr
588 spectral peak is dominant over the 41 kyr peak in our dust records, especially after ~2.1 Ma
589 (Figure S13b, c, d, e), and may have been amplified by a FM mechanism based on $0.024 (1/41) -$
590 $0.005 (1/171) = 0.019 (1/54)$ (Rial, 1999; Rial & Anaclerio, 2000).

591 4.4.3. Insights from other Indo-Pacific dust records

592 Orbitally-driven changes in the intensity of the EAWM, the SITIG, or both, may underpin the
593 dominant obliquity cycles observed in Site U1464 dust-flux reconstructions. For further insights,
594 we therefore turn to other Indo-Pacific dust records. Interestingly, dust-flux time-series from Site
595 RC27-61 in the northwestern Arabian Sea (Clemens & Prell, 1990; Clemens et al., 1991a), Site
596 RC11-210 in the central Equatorial Pacific (Pisias & Rea, 1988) and Site V21-146 in the
597 Northwest Pacific (Hovan et al., 1991) all contain cycles at 54 and/or 29 kyr, in addition to 41
598 kyr, similar to our Site U1464 dust records. Australian monsoon records also show these three
599 obliquity cycles (Holbourn et al., 2005; Kershaw et al., 2003; Liu et al., 2015; Zhang et al., 2020,
600 2022), while these spectral peaks are not present in the sea level time-series spanning this
601 timeframe (Figure S12a). These findings imply that our Site U1464 dust-flux reconstruction
602 likely reflects a component of low-latitude, Indo-Pacific climate variability that is independent of
603 ice-sheet related variability.

604 The long-term variability of Walker circulation and sea surface temperature (SST) gradients in
605 the tropical Pacific and Indian Ocean (i.e., ENSO-like) might amplify the 29 kyr cycles via
606 resonance with obliquity (Beaufort et al., 2001; Pisias & Rea, 1988). The 29 kyr period was
607 dominant in easterly trade wind intensity and equatorial divergence in the central Equatorial
608 Pacific during the Late Pleistocene, based on aeolian grain-size and relative abundance of two
609 radiolarian species, respectively (Pisias & Rea, 1988). This 29 kyr cycle was also found as a
610 secondary period in: central Equatorial Pacific SST (Pisias & Rea, 1988); southern subtropical
611 Indian Ocean SST (Clemens et al., 1991a, b); wind strength of the southwest monsoon in the
612 northwestern Arabian Sea (Clemens & Prell, 1990; Clemens et al., 1991a, b); and, primary
613 productivity associated with the tropical Indo-Pacific thermocline (Beaufort et al., 2001). This
614 non-primary 29 kyr variability inherent in these records strongly suggests that the resonance

615 between obliquity and tropical Indo-Pacific atmosphere-ocean dynamics may play a key role in
616 amplifying the 29 kyr variability in our dust records.

617 **5. Conclusions**

618 We present CaCO₃ and aeolian dust proxy records (Log(Zr/Rb), Log(Zr/Fe) and CaCO₃-free
619 IRM_{1T@AF 170mT}) for IODP Site U1464 on the Northwest Shelf of Australia from the Early
620 Pleistocene (~1.6 - 2.9 Ma). A new, orbitally-tuned chronology was constructed for Site U1464
621 using a Bayesian model of biostratigraphic datums in conjunction with eccentricity- and
622 obliquity-tuning of our CaCO₃ and Log(Zr/Rb) timeseries.

623 A ~400 kyr (eccentricity) cyclicity is present in the CaCO₃ record similar to benthic δ¹³C records
624 from the wider region, where the benthic records are typically indicative of intermediate water
625 δ¹³C, and where CaCO₃ and δ¹³C minima are associated with maxima in the 400 kyr eccentricity
626 cycle (and *vice versa*). These observations are consistent with a ~400 kyr pacing of the oceanic
627 carbon reservoir via ocean ventilation and large-scale circulation.

628 Obliquity frequencies dominate the dust proxy records at Site U1464. Three obliquity cycles (41
629 kyr, 54 kyr and 29 kyr) are present and we explore their possible origins in terms of low- *versus*
630 high-latitude forcing mechanisms. The 41 kyr signal in Northwest Australian dust fluxes may
631 originate from fluctuations in the intensity of the EAWM and/or the SITIG, both of which are
632 directly driven by obliquity variations. In contrast, the 54 kyr signal in the dust-flux records
633 might result from a non-linear response to AM of obliquity and attendant SITIG variability. The
634 resonance between obliquity and tropical Indo-Pacific atmosphere-ocean dynamics may also be
635 an amplifying mechanism of secondary obliquity frequencies, especially for the 29 kyr cycle,
636 which is detected in other paleoclimate proxy records from the Indo-Pacific region.

637 **Figure 1.** Precipitation (land), sea surface temperature (SST, ocean) and 850 hPa wind patterns of the Australian-
638 Indonesian region for average austral summer (DJFM, top) and winter (JJAS, bottom). Precipitation data (1901-
639 2013) is from the GPCP dataset (Schneider et al., 2011); SST data (1870-2017) is from the HadISST1 dataset
640 (Rayner et al., 2003); Wind data (1948-2017) is from the NCEP/NCAR Reanalysis (Kalnay et al., 1996). Brown
641 dash lines denote the Pilbara heat low (Suppiah, 1992). Red star marks IODP Site U1464 (this study). Core sites
642 discussed in this study are indicated with black circles: IODP Site U1482 (Chen et al., 2022), IODP Site U1463
643 (Christensen et al., 2017), ODP Site 762 (Stuut et al., 2019), and DSDP Site 593 (McClymont et al., 2016). Detailed
644 drainage basins (number 1-16) in the black rectangle are shown in Figure S1.

645 **Figure 2.** Bacon age-depth models for IODP Site U1464 (red). (a) Bacon age-depth model based on biostratigraphic
 646 ages. Black, blue, green, yellow and white dotted lines indicate the model's 99.99%, 99%, 95%, 90% and 80%
 647 probability intervals, respectively. (b) Refined Bacon age-depth model based on all biostratigraphic ages and
 648 tunings. Gradual darker grey shadings indicate the model's 99.99%, 99%, 95%, 90% and 80% probability intervals,
 649 respectively. Red line is the median probability. Red solid circles are biostratigraphic age control-points (Table S3).
 650 Blue solid circles are four tie-points between eccentricity and the CaCO₃ record (Table S5). Yellow solid circles are
 651 32 tie-points between obliquity and the filtered Log(Zr/Rb) record (Table S5). Black solid circle is the last
 652 occurrence of *G. ruber* (pink).

653 **Figure 3.** IODP Site U1464 bulk geochemistry from ~1.6 to 2.9 Ma. (a) Log(Fe/Ca); (b) CaCO₃; (c) Sr; (d) Fe; (e)
 654 Ti; (f) Rb and (g) Zr. Note that (b-e) show calibrated element concentrations, and (f, g) show scanning-XRF element
 655 counts. Bold lines (grey lines) in a-g are 21-point running averages (original data).

656 **Figure 4.** IODP Site U1464 eccentricity tuning. (a) Site U1464 CaCO₃ (gray: original time-series; purple: 21-point
 657 running average) on initial Bacon age model. (b) Eccentricity (green, Laskar et al., 2004). (c) Eccentricity (green)
 658 and Site U1464 CaCO₃ (gray: original time-series; purple: 21-point running average) rescaled after eccentricity
 659 tuning. Tie-points are indicated (dotted lines) for visual clarity. Cycle numbers represent 413 kyr eccentricity cycles,
 660 counted back from the present. (CaCO₃ y-axes are reversed for visual comparison).

661 **Figure 5.** IODP Site U1464 obliquity tuning. (a) Site U1464 Log(Zr/Rb) (gray: original time-series; green: 21-point
 662 running average) and its 0.023-0.027 band-pass filter (magenta) on the eccentricity tuning age model. (b) Obliquity
 663 (black, Laskar et al., 2004). (c) Obliquity (black) and the 0.023-0.027 band-pass filter (magenta) after obliquity
 664 tuning. 32 ties between obliquity minima and minima in the 0.023-0.027 filtered Log(Zr/Rb) are indicated (dotted
 665 lines) for visualization. Red triangles in (a) represent four eccentricity tuning tie-points.

666 **Figure 6.** IODP Site U1464 carbonate content and regional benthic $\delta^{13}\text{C}$ records. (a) Site U1464 CaCO₃ (purple) and
 667 IODP Site U1482 *P. wuellerstorfi* $\delta^{13}\text{C}$ (blue, Chen et al., 2022). (b) Site U1464 CaCO₃ (purple) and IODP Site
 668 U1463 *Uvigerina spp.* $\delta^{13}\text{C}$ (green, Groeneveld et al., 2021). (c) Site U1464 CaCO₃ (purple) and DSDP Site 593 *P.*
 669 *wuellerstorfi* $\delta^{13}\text{C}$ (cyan, McClymont et al., 2016). The eccentricity tuning is used for the age model of Site U1464
 670 CaCO₃ to validate the ~400 kyr eccentricity tuning. Red triangles represent four eccentricity tuning tie-points for
 671 Site U1464 CaCO₃. Blue, green and cyan triangles represent age constraints for Site U1482 *P. wuellerstorfi* $\delta^{13}\text{C}$,
 672 Site U1463 *Uvigerina spp.* $\delta^{13}\text{C}$ and Site 593 *P. wuellerstorfi* $\delta^{13}\text{C}$, respectively.

673 **Figure 7.** Dust proxy records from the Australian NWS. CaCO₃-free IRM_{IT@AF 170mT} (a, red), Log(Zr/Rb) (b, green:
 674 21-point running average), and Log(Zr/Fe) (c, blue: 21-point running average) records at Site U1464. Wireline-
 675 derived Th/K record at Site U1463 (d, Christensen et al., 2017) and Log(Zr/Fe) record at Site ODP762 on Auer et al.
 676 (2020)'s updated age scale (e, orange: 9-point running average, Stuut et al., 2019). Dust maxima events are indicated
 677 in dash lines for visual alignment.

678 **Figure 8.** Comparison of Northwest Australian dust fluxes with global sea level, EAWM and SITIG from 1.6 to 2.9
 679 Ma. (a) Site U1464 Log(Zr/Rb) and global sea level (Rohling et al., 2021) and (b) their cross wavelet spectrum. (c)

680 Site U1464 Log(Zr/Rb) and a stacked MGSQ record (Sun et al., 2006) and (d) their cross wavelet spectrum. (e) Site
681 U1464 Log(Zr/Rb) and the SITIG summer half-year (Laskar et al., 2004) and (f) their cross wavelet spectrum. The
682 30° N-30° S SITIG is shown in e (black) and the 0.023-0.027 filter of Log(Zr/Rb) is also shown in e (red line). In b, d
683 and f, the cone of influence and 5% significance level are indicated by opaque shading and bold black contours,
684 respectively. Arrows pointing to the right indicate an in-phase relationship.

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694 **Data Availability Statement**

695 Geochemical and environmental magnetic data from IODP Site U1464 were generated in this
696 study. New data are archived in the Zenodo database (Zhao & Grant, 2023).

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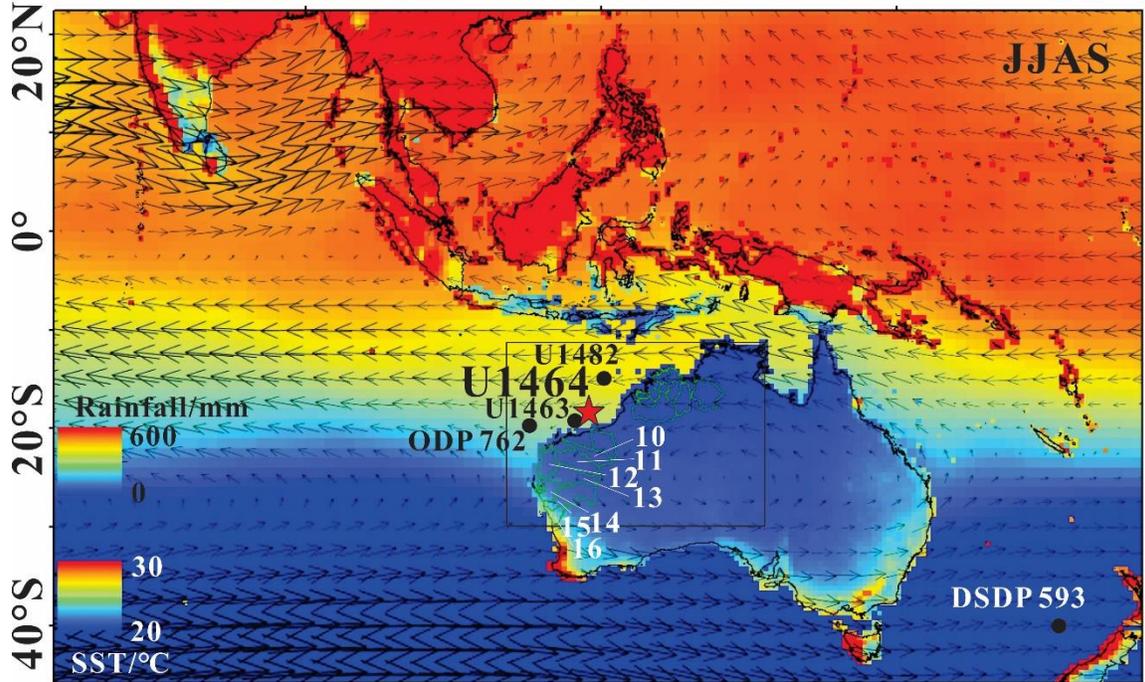
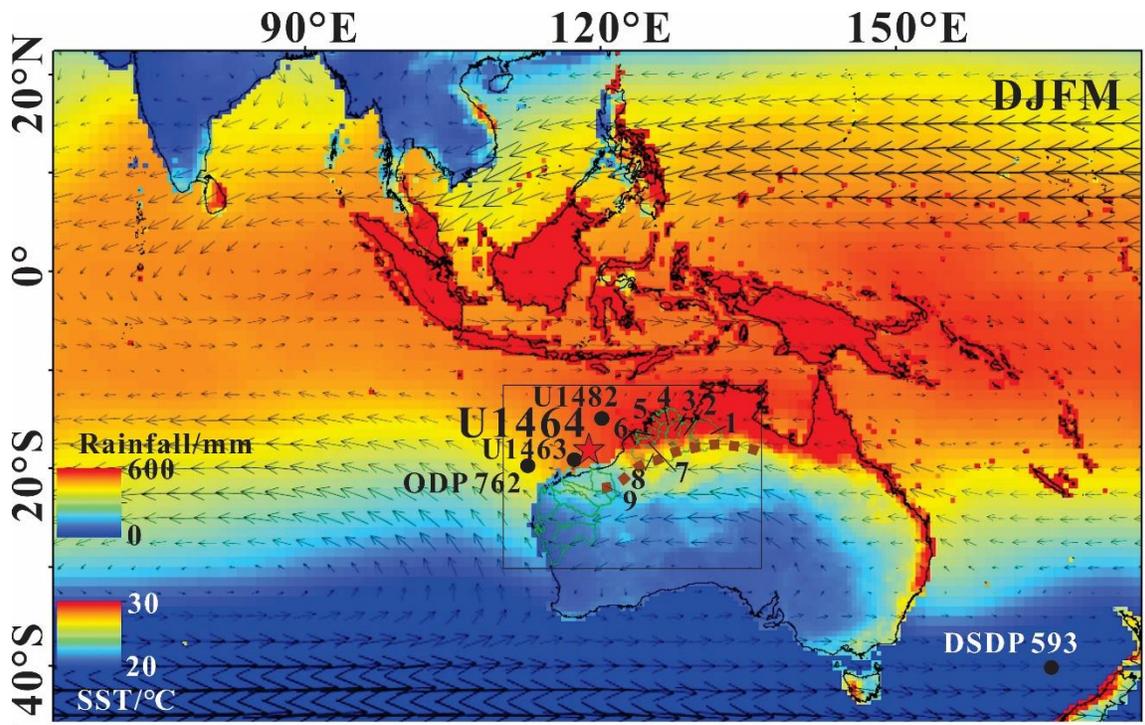
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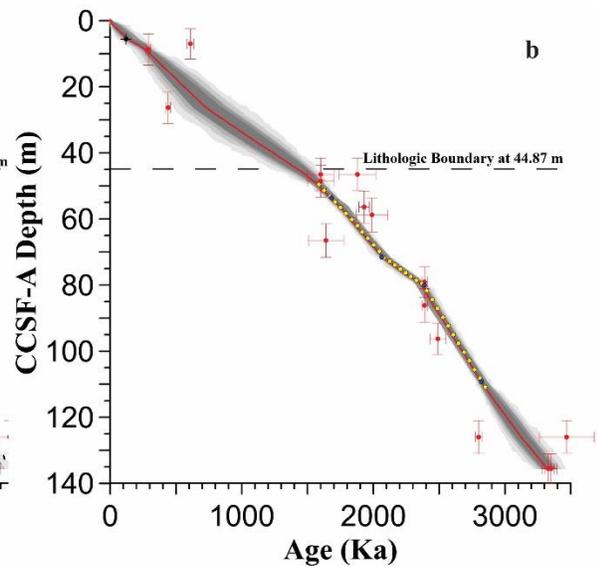
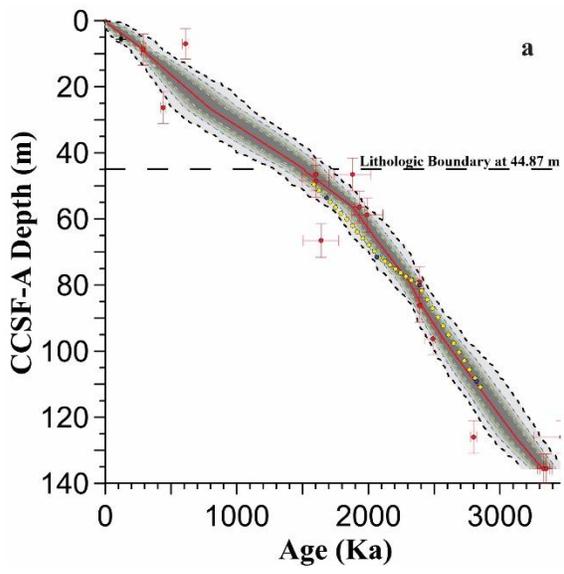
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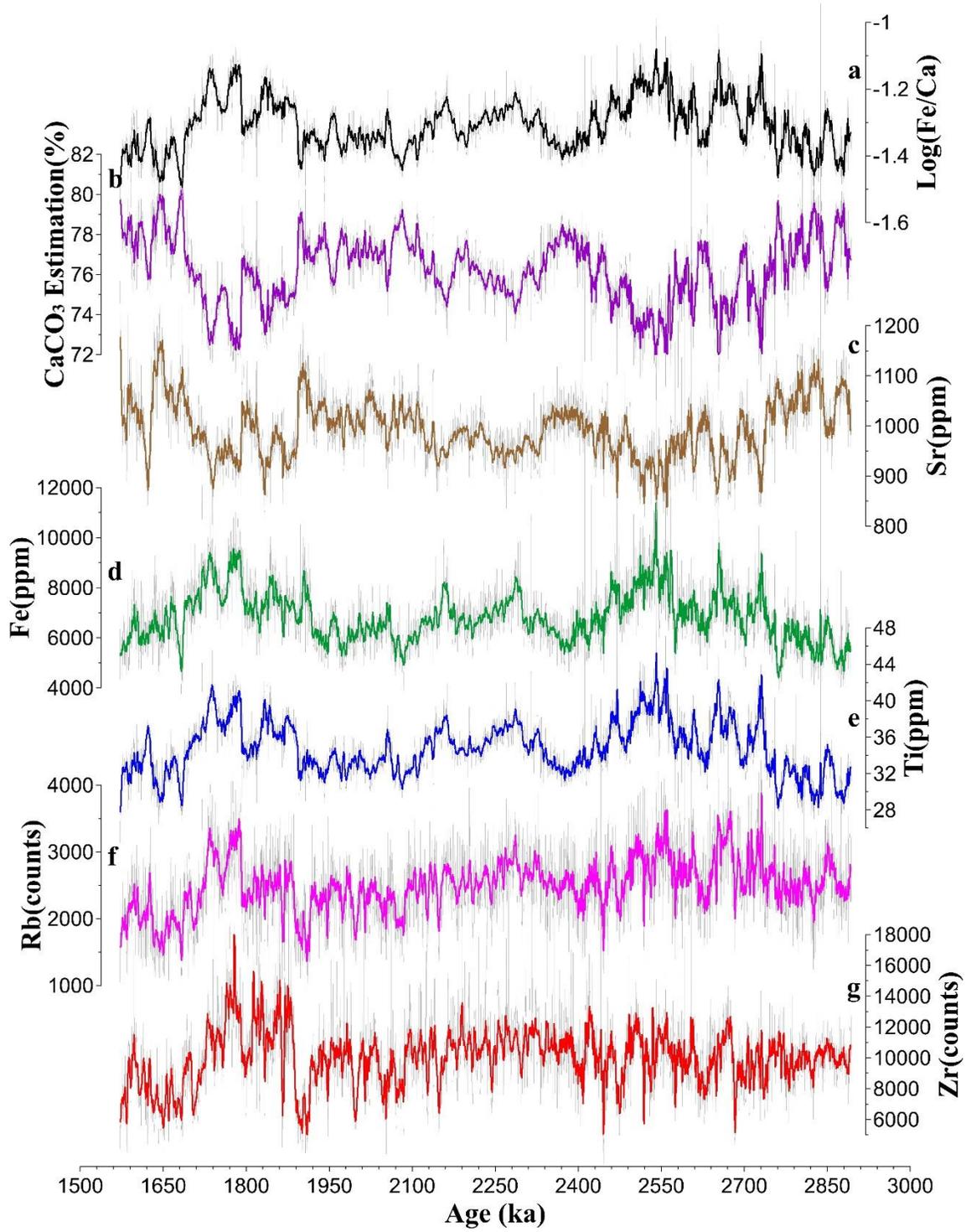
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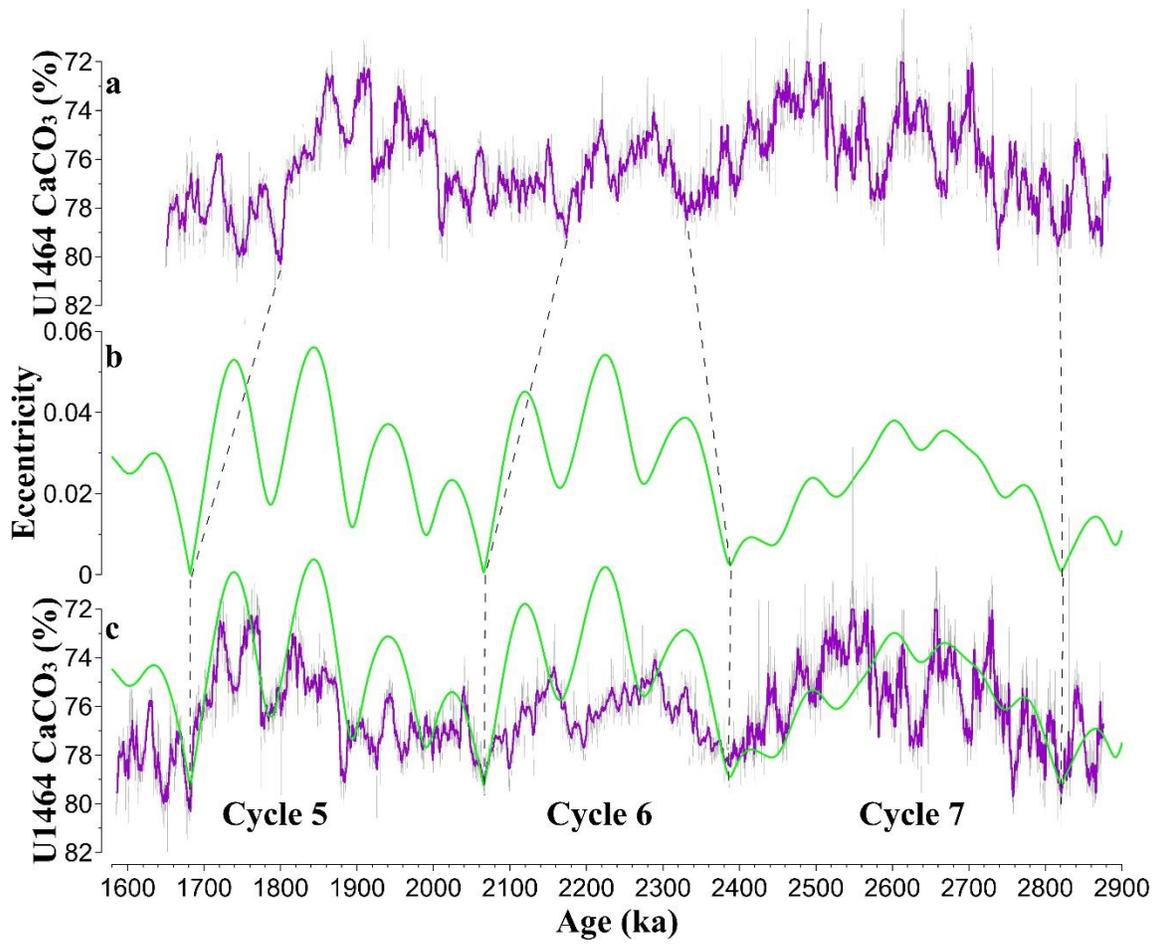
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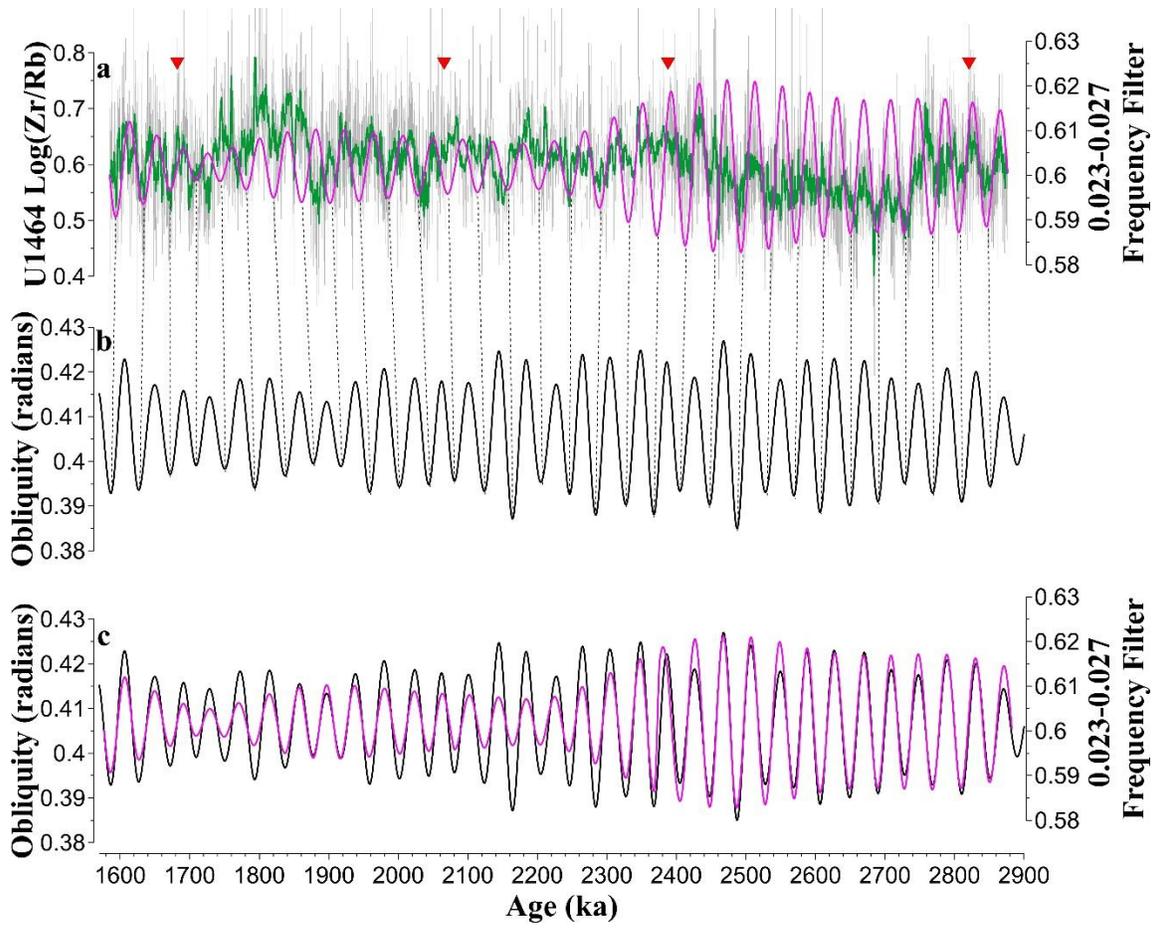
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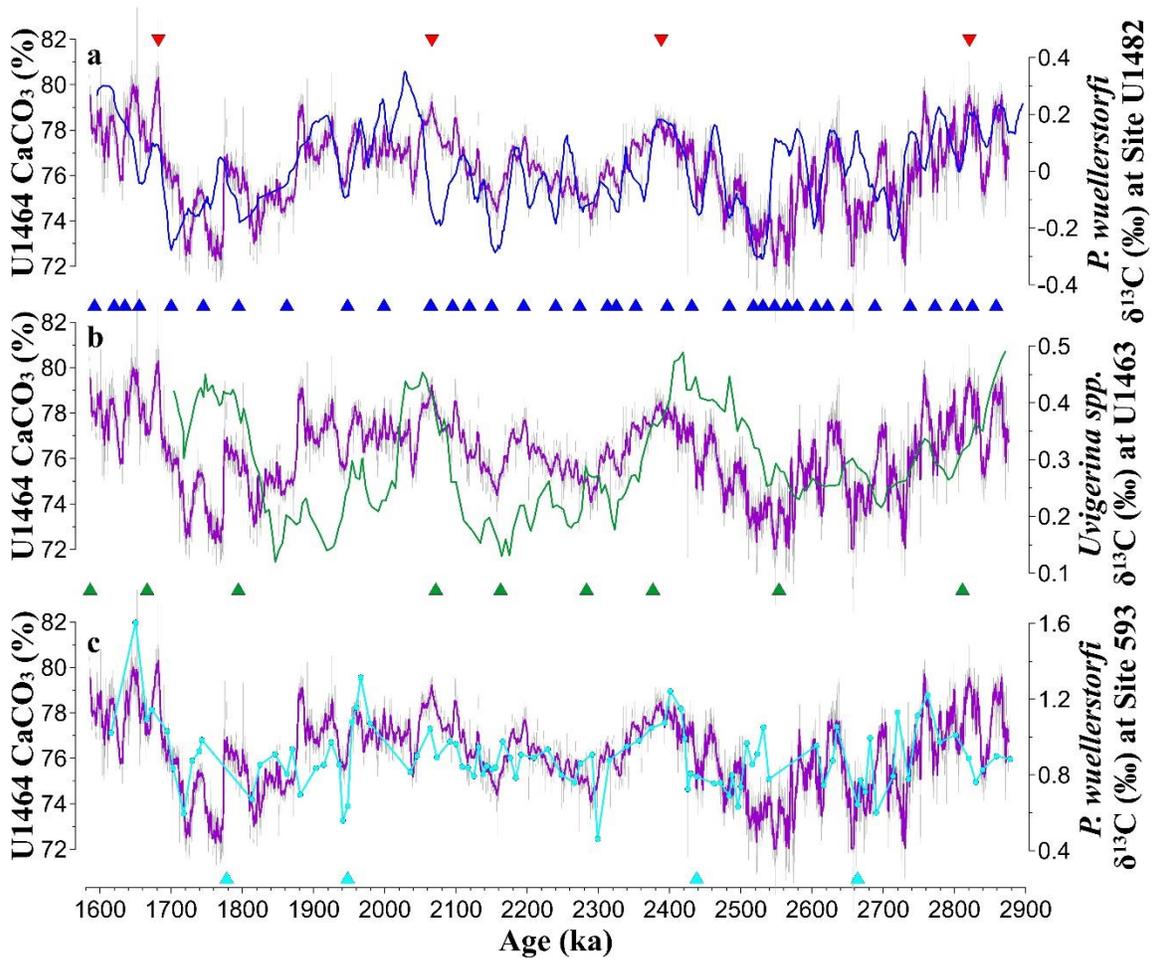


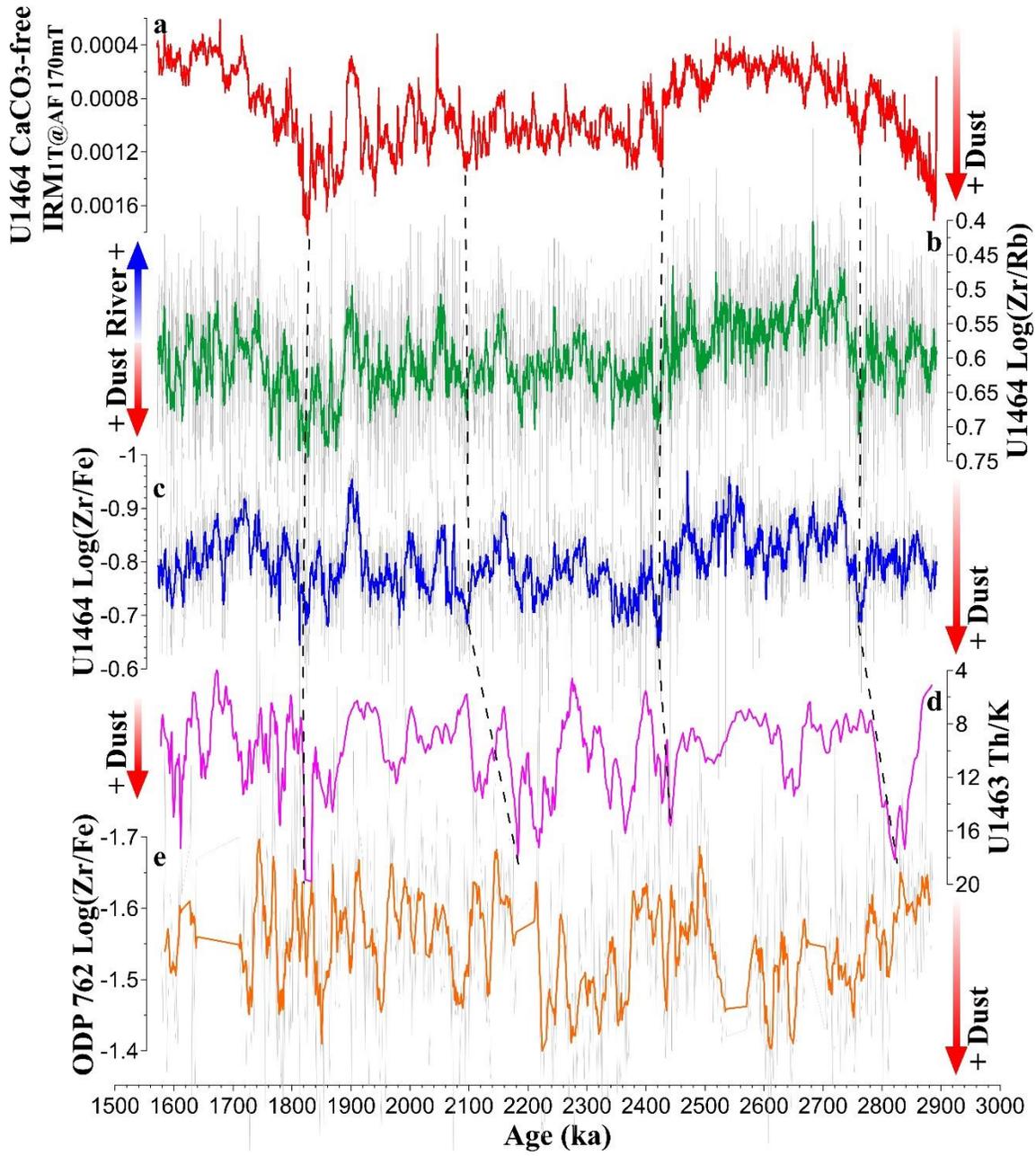


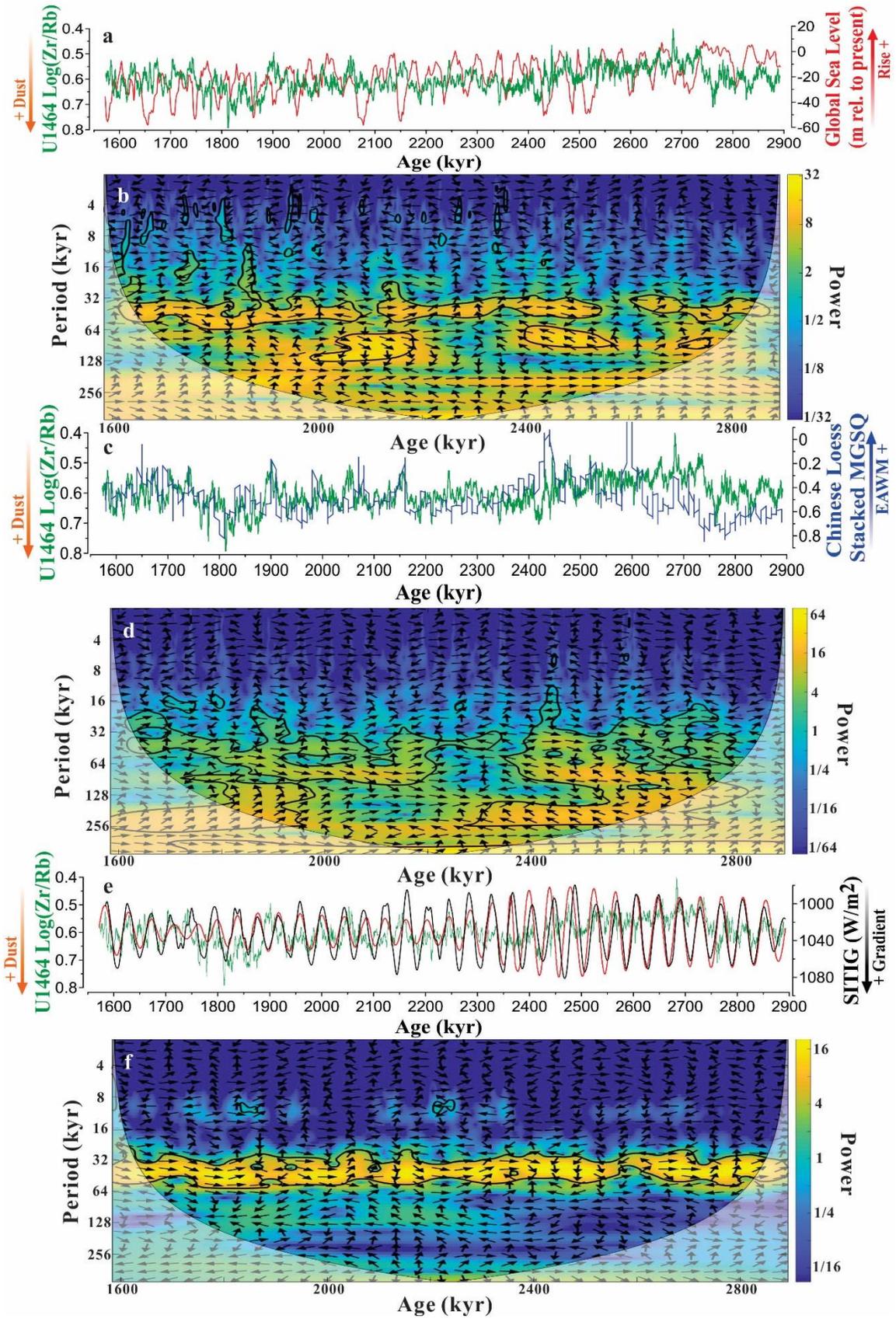












Supporting Information for

Early-Pleistocene orbital variability in Northwest Australian shelf sediments

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Text S1. Alternative eccentricity tunings

In order to fully consider the uncertainty of this eccentricity tuning, twelve scenarios have been found based on different options for each tie-point (Figure S5; Table S4). Pearson and Spearman correlation coefficients are calculated to show the linear and monotonic relationship between eccentricity and our CaCO₃ record (Figure S5). Correlation analyses suggest that 53.62, 71.57, 79.97 and 109.18 mbsf are the best four tie-points for the eccentricity minima at 1682, 2066, 2388 and 2821 kyr, respectively (Figure S5k; Table S4). Additionally, we have calculated the corresponding p value for each of the regressions in twelve scenarios to test if there is a more robust relationship between eccentricity and the U1464 CaCO₃ record.

Following previous studies (Santer et al., 2000), we estimate residuals in CaCO₃ based on the least squares linear regression with eccentricity (i.e., $e(x) = y(x) - \bar{y}(x)$, where e is the regression residual, y is the CaCO₃ and \bar{y} is the regression of CaCO₃ (dependent variable) with eccentricity (independent variable), x is the eccentricity), and assess the autocorrelation in the residuals. We adjust the effective number of samples according to the autocorrelation and then calculate the p value. Basically, we first calculate the lag-1 autocorrelation coefficient of residuals (r_1) to obtain the effective number of samples (n_e) based on $n_e = n_t * (1 - r_1) / (1 + r_1)$, where n_t is the total number of samples. The variance of residuals (S_e^2) is given by $[1 / (n_e - 2)] * \sum_{n=1}^{n_t} e(x)^2$, and the standard error of regression coefficient (b) is defined as $S_b = S_e / [\sum_{n=1}^{n_t} (x - \bar{x})^2]^{1/2}$, where \bar{x} is the eccentricity mean. Accordingly, the ratio between b and S_b can be used as the t value, and then substituted to the cumulative t-distribution with $n_e - 2$ degrees of freedom to get p values. The p value for option (k) is smallest one (Figure S5k), which corresponds with its highest correlation coefficients.

Text S2. Uncertainties in IODP Site U1464 dust proxy records

To quantify the potential influence of measurement uncertainties on the observed variability in our dust-flux proxy records, we calculate measurement uncertainties and their propagation in our $\text{Log}(Zr/Rb)$, $\text{Log}(Zr/Fe)$ and CaCO_3 -free $\text{IRM}_{1T@AF 170mT}$ records. The principal formulas for error propagation in this study are as follows (Taylor, 1997):

- (i) If q is any function of several variables x, \dots, z , then $\delta q = \text{sqrt} \left(\left(\frac{\partial q}{\partial x} * \delta x \right)^2 + \dots + \left(\frac{\partial q}{\partial z} * \delta z \right)^2 \right)$, where variables x, \dots, z are measured with one standard deviation (1δ , expressed as $\delta x, \dots, \delta z$).
- (ii) If q is the sum and difference, $q = x + \dots + z - (u + \dots + w)$, then $\delta q = \text{sqrt} \left((\delta x)^2 + \dots + (\delta z)^2 + (\delta u)^2 + \dots + (\delta w)^2 \right)$.
- (iii) If $q = Bx$, where B is a known constant, then $\delta q = |B|\delta x$, where $|B|$ is absolute value.
- (iv) If q is the product and quotient, $q = (x * \dots * z) / (u * \dots * w)$, then $\frac{\delta q}{|q|} = \text{sqrt} \left(\left(\frac{\delta x}{x} \right)^2 + \dots + \left(\frac{\delta z}{z} \right)^2 + \left(\frac{\delta u}{u} \right)^2 + \dots + \left(\frac{\delta w}{w} \right)^2 \right)$.

Based on equation (i), the 1δ of logarithm function ($\text{Log}(x/y)$) can be derived as follows:

$\delta \text{Log}(x/y) = \text{sqrt} \left(\left(\frac{1}{x} * \delta x \right)^2 + \left(-\frac{1}{y} * \delta y \right)^2 \right)$. In this study, the average fractional uncertainties

(ratios of 1δ values to values of individual measurement) for Zr, Rb and Fe ($\frac{\delta Zr}{Zr}$, $\frac{\delta Rb}{Rb}$ and

$\frac{\delta Fe}{Fe}$) are 0.033, 0.102 and 0.007, respectively. Accordingly, $\delta \text{Log}(Zr/Rb) = \text{sqrt} \left(\left(\frac{1}{Zr} * \right.$

$\delta Zr \right)^2 + \left(-\frac{1}{Rb} * \delta Rb \right)^2 = \text{sqrt} (0.033^2 + 0.102^2) = 0.107$; $\delta \text{Log}(Zr/Fe) = \text{sqrt} \left(\left(\frac{1}{Zr} * \delta Zr \right)^2 + \right.$

$\left. \left(-\frac{1}{Fe} * \delta Fe \right)^2 \right) = \text{sqrt} (0.033^2 + 0.007^2) = 0.034$. To obtain the 1δ of the n-point running

average of $\text{Log}(x/y)$, equations (ii) and (iii) are applied to divide the $\delta \text{Log}(x/y)$ value by

$\text{sqrt}(n)$, where n is the number of data-points in the running average; i.e., $\delta \text{Log}(x/y)_{\text{average}} =$

$(1/n) * \text{sqrt} \left((\delta \text{Log}(x/y)_1)^2 + \dots + (\delta \text{Log}(x/y)_n)^2 \right) = (1/n) * \text{sqrt} (n * (\delta \text{Log}(x/y))^2) =$

$\delta \text{Log}(x/y) / \sqrt{n}$, where each $\delta \text{Log}(x/y)$ is the same. We use a 21-point running average, which yields $\delta \text{Log}(\text{Zr/Rb})_{\text{average}} = 0.107 / \sqrt{21} = 0.023$ and $\delta \text{Log}(\text{Zr/Fe})_{\text{average}} = 0.034 / \sqrt{21} = 0.007$. We further multiply $\pm 1\delta$ values by 1.96 (2.58) to obtain the 95 % (99 %) confidence intervals for $\text{Log}(\text{Zr/Rb})$ and $\text{Log}(\text{Zr/Fe})$ (Figure S10).

For the CaCO_3 -free $\text{IRM}_{1\text{T}@AF 170\text{mT}}$, we only consider the measurement uncertainty of CaCO_3 and its propagation, since the measurement uncertainty of $\text{IRM}_{1\text{T}@AF 170\text{mT}}$ is so small that it is effectively negligible in this study. The predicted CaCO_3 (fractional value) is calculated by $0.096 * \text{Ln}(\text{Ca/Fe}) + 0.474$, where the average fractional uncertainties of Ca and Fe ($\frac{\delta \text{Ca}}{\text{Ca}}$ and $\frac{\delta \text{Fe}}{\text{Fe}}$) are 0.002 and 0.007, respectively. The $\delta \text{Ln}(\text{Ca/Fe}) = \sqrt{(0.002^2 + 0.007^2)} = 0.007$, and the $\delta \text{CaCO}_3 = 0.096 * \delta \text{Ln}(\text{Ca/Fe}) = 0.001$, following equation (iii). The δCaCO_3 can be considered as the $\delta(1 - [\text{CaCO}_3])$ based on equation (iii), and then the fractional uncertainty of $(1 - [\text{CaCO}_3])$ (i.e., $\frac{\delta(1 - [\text{CaCO}_3])}{(1 - [\text{CaCO}_3])}$) is calculated and can be used as the fractional uncertainty of $([\text{IRM}_{1\text{T}@AF 170\text{mT}}] / (1 - [\text{CaCO}_3]))$ (i.e., $\frac{\delta([\text{IRM}_{1\text{T}@AF 170\text{mT}}] / (1 - [\text{CaCO}_3]))}{([\text{IRM}_{1\text{T}@AF 170\text{mT}}] / (1 - [\text{CaCO}_3]))}$) based on equation (iv). Finally, the $\delta(\text{CaCO}_3\text{-free } \text{IRM}_{1\text{T}@AF 170\text{mT}})$, which is $\delta([\text{IRM}_{1\text{T}@AF 170\text{mT}}] / (1 - [\text{CaCO}_3]))$, can be derived to obtain the 95 % (99 %) confidence intervals of $\text{CaCO}_3\text{-free } \text{IRM}_{1\text{T}@AF 170\text{mT}}$ using $\pm 1.96(2.58)\delta$ (Figure S10).

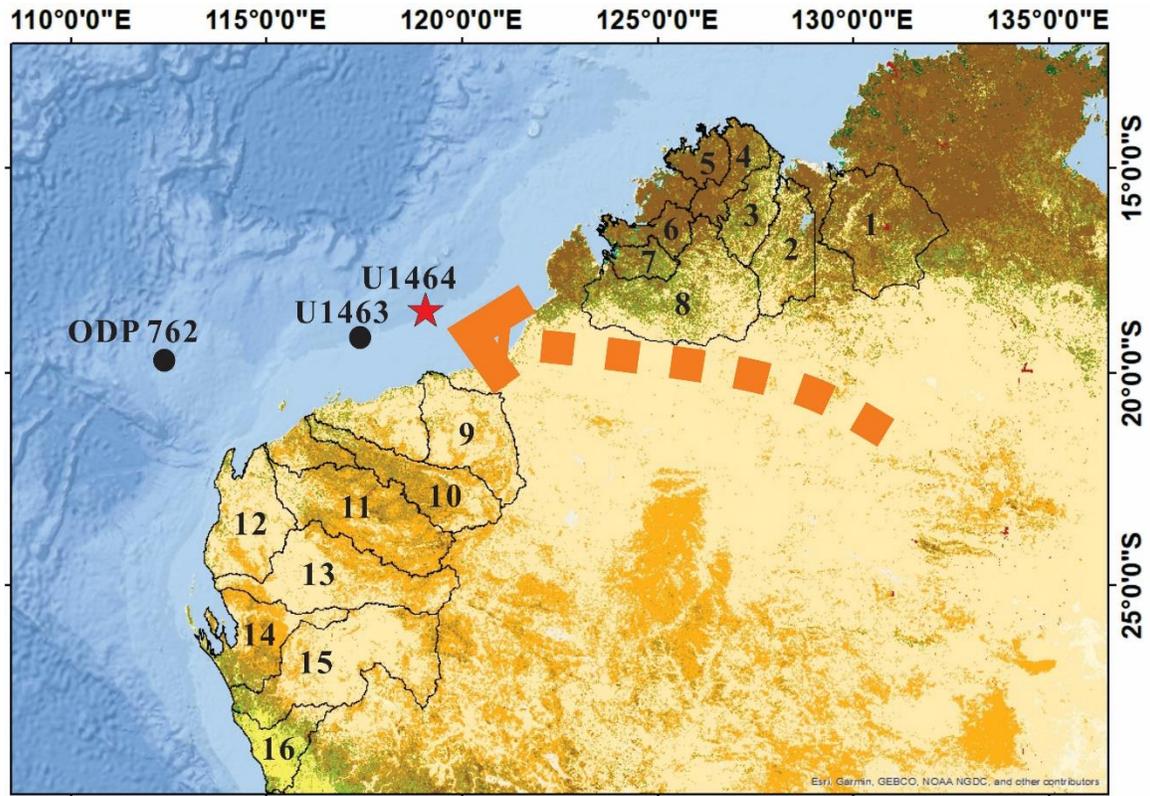


Figure S1. Northwest Australian river basins and locations of three cores (IODP Site U1464, U1463 and ODP site 762). Orange arrow indicates main aeolian dust path (after [Bowler, 1976](#)). Drainage basins of Victoria (1), Ord (2), Pentecost (3), Drysdale (4), King Edward (5), Isdell (6), Lennard (7), Fitzroy (8), De Grey (9), Fortescue (10), Ashburton (11), Lyndon-minilya (12), Gascoyne (13), Wooramel (14), Murchison (15), and Greenough (16) rivers ([Australia's River Basins, 1997](#)) are marked. Base map is from the GlobCover 2009 land cover and World Ocean Base in ArcGIS online.

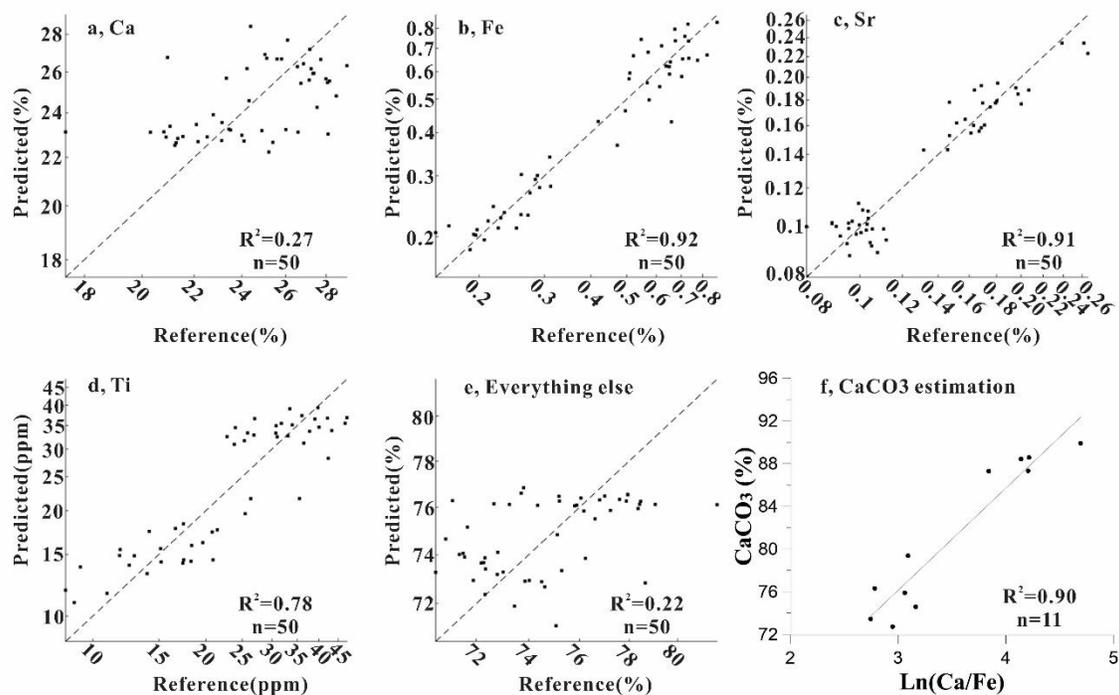


Figure S2. Reference versus predicted element concentrations for IODP Site U1464, based on ICP-OES and scanning XRF (a-e), using a multivariate log-ratio calibration model (Weltje et al., 2015). Shipboard CaCO₃ measurements versus Ln(Ca/Fe) for IODP Site U1464 (f) following the method of Liebrand et al., (2016).

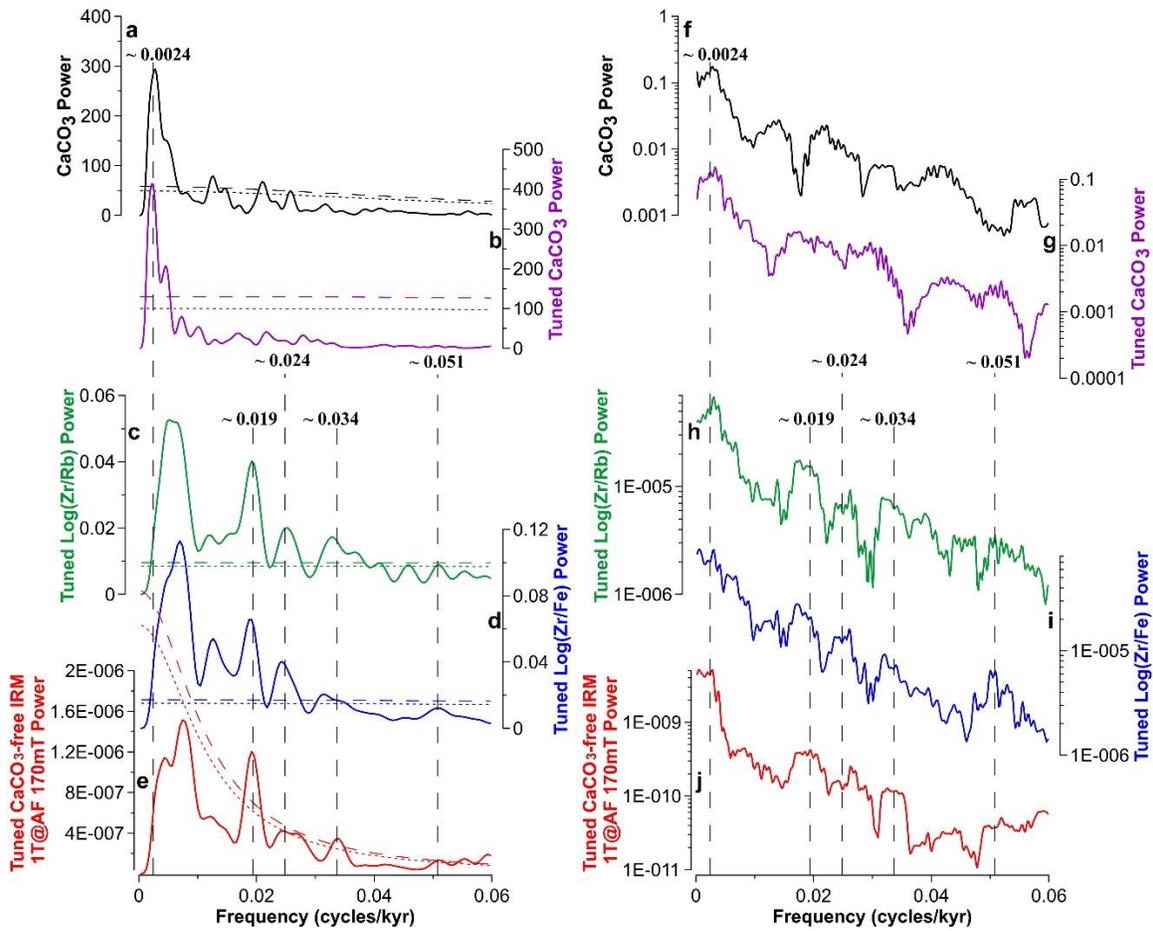


Figure S3. Spectral analyses of Site U1464 proxy records over the 1.6-2.9 Ma interval using the Redfit (a-e) and Multitaper (f-j) methods. Power spectra for CaCO_3 (a, f) on the biostratigraphy-based Bayesian model, and CaCO_3 (b, g), $\text{Log}(\text{Zr}/\text{Rb})$ (c, h), $\text{Log}(\text{Zr}/\text{Fe})$ (d, i) and CaCO_3 -free $\text{IRM}_{1\text{T}@AF 170\text{mT}}$ (e, j) after eccentricity tuning. The a-e represent Redfit spectra with output parameters (oversample = 8, segment = 4 in a-b and oversample = 8, segment = 8 in c-e), and confidence levels (90%, dot; 95%, dashed) are indicated. The f-j represent Multitaper spectra with 5 tapers.

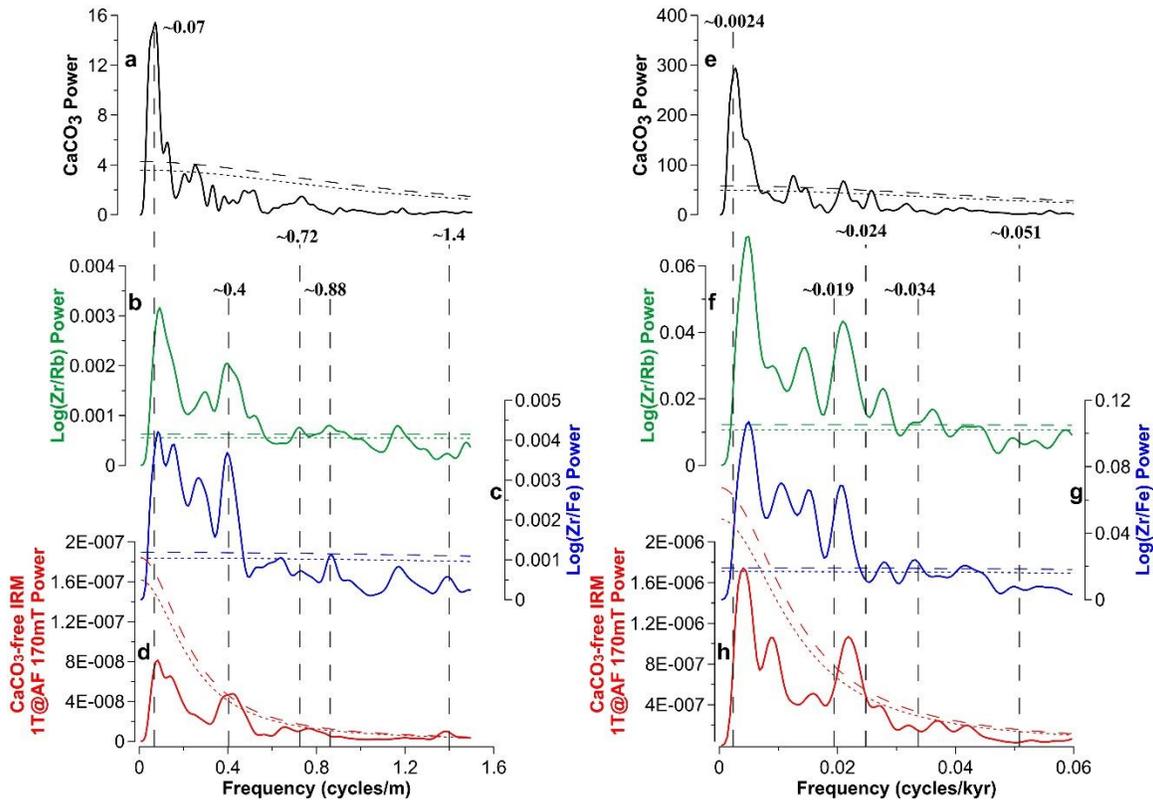


Figure S4. Spectral analyses of Site U1464 proxy records over the interval of ~47-108 mbsf (a-d) or ~1.6-2.9 Ma (e-h) using the Redfit method. Power spectra for CaCO_3 (a), $\text{Log}(\text{Zr}/\text{Rb})$ (b), $\text{Log}(\text{Zr}/\text{Fe})$ (c) and CaCO_3 -free $\text{IRM}_{1\text{T}@AF 170\text{mT}}$ (d) on depth domain. Power spectra for CaCO_3 (e), $\text{Log}(\text{Zr}/\text{Rb})$ (f), $\text{Log}(\text{Zr}/\text{Fe})$ (g) and CaCO_3 -free $\text{IRM}_{1\text{T}@AF 170\text{mT}}$ (h) on the biostratigraphy-based Bayesian model. The a-h represent Redfit spectra with output parameters (oversample = 8 and segment = 4 for CaCO_3 record; oversample = 8 and segment = 8 for dust proxy records), and confidence levels (90%, dot; 95%, dashed) are indicated.

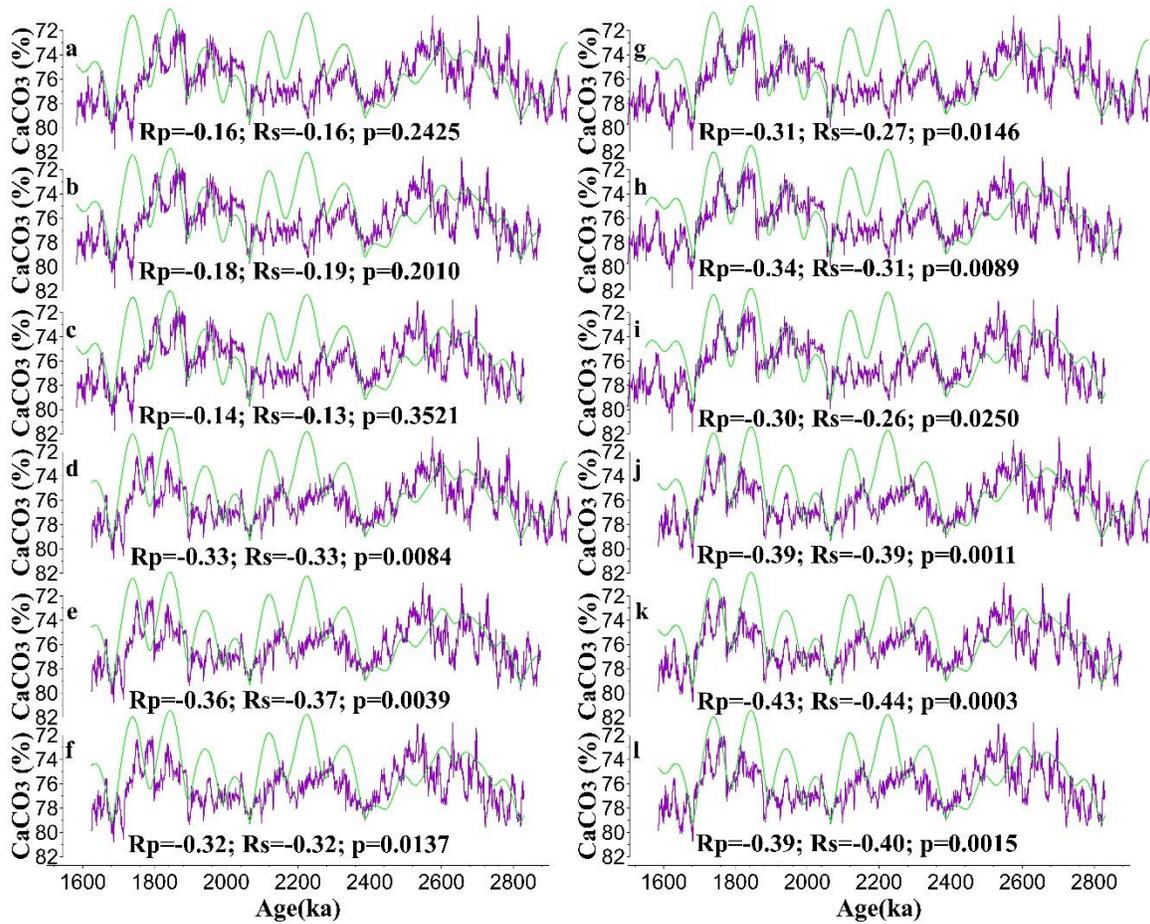


Figure S5. Alternative tunings for Site U1446 to eccentricity. Four eccentricity minima at 1682, 2066, 2388 and 2821 kyr are tuning targets for 12 potential CaCO₃ maxima. See [Table S4](#) for tie-point depths used in (a)-(l). Option (k) is the choice in this study. Purple (green) lines represent CaCO₃ (eccentricity) records. Rp (Rs) represents Pearson (Spearman) correlation coefficient (see [Text S1](#)).

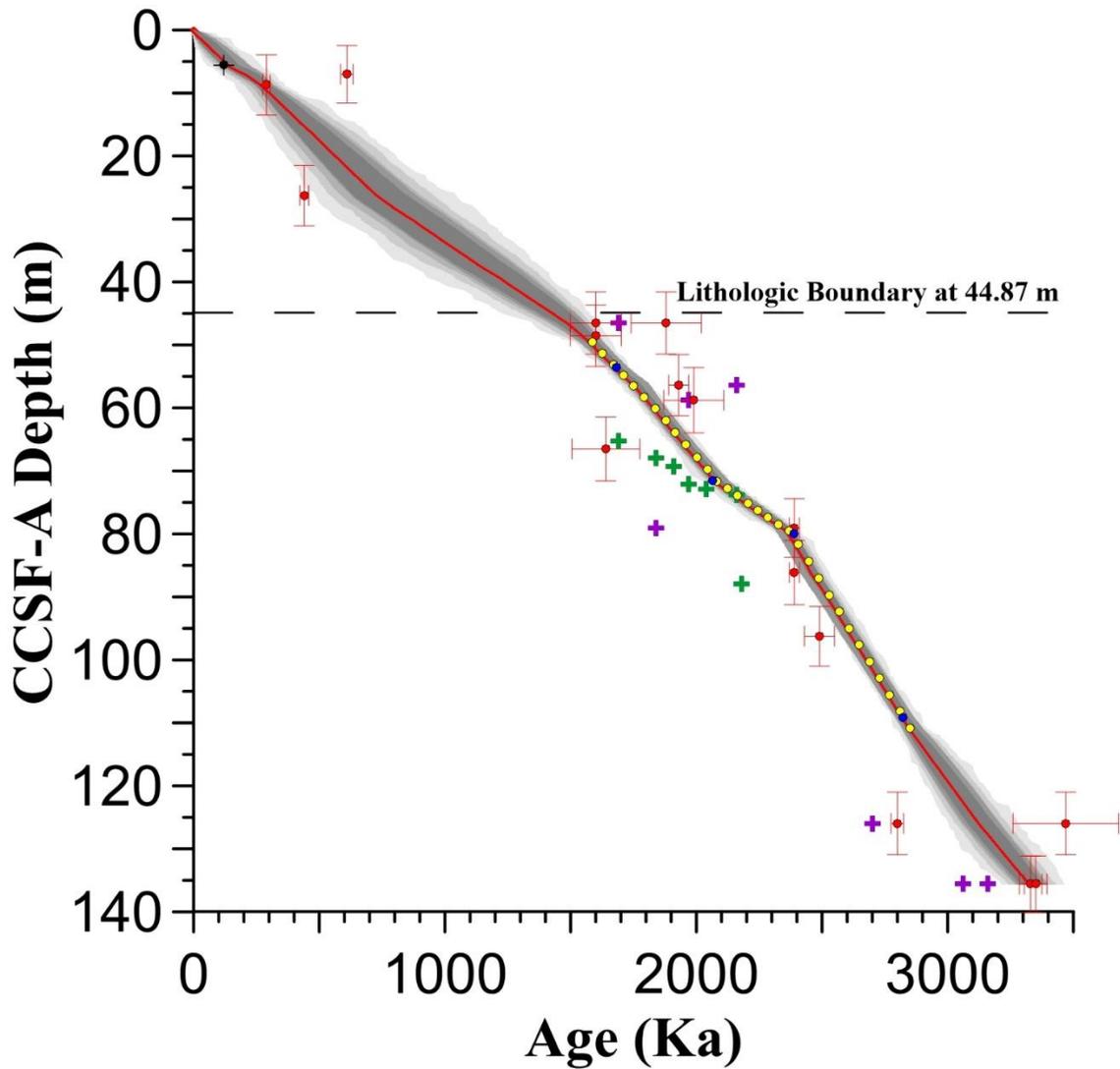


Figure S6. Same as main-text figure 2b, with additional age-depth markers for comparison. Green crosses are inferred chrono-stratigraphy for Site U1464 based on tie-points between U1463 and U1464 using dynamic time warping of natural gamma radiation records (Groeneveld et al., 2021). Purple crosses are revised biostratigraphic datums for neighboring Site U1463 from Groeneveld et al., 2021.

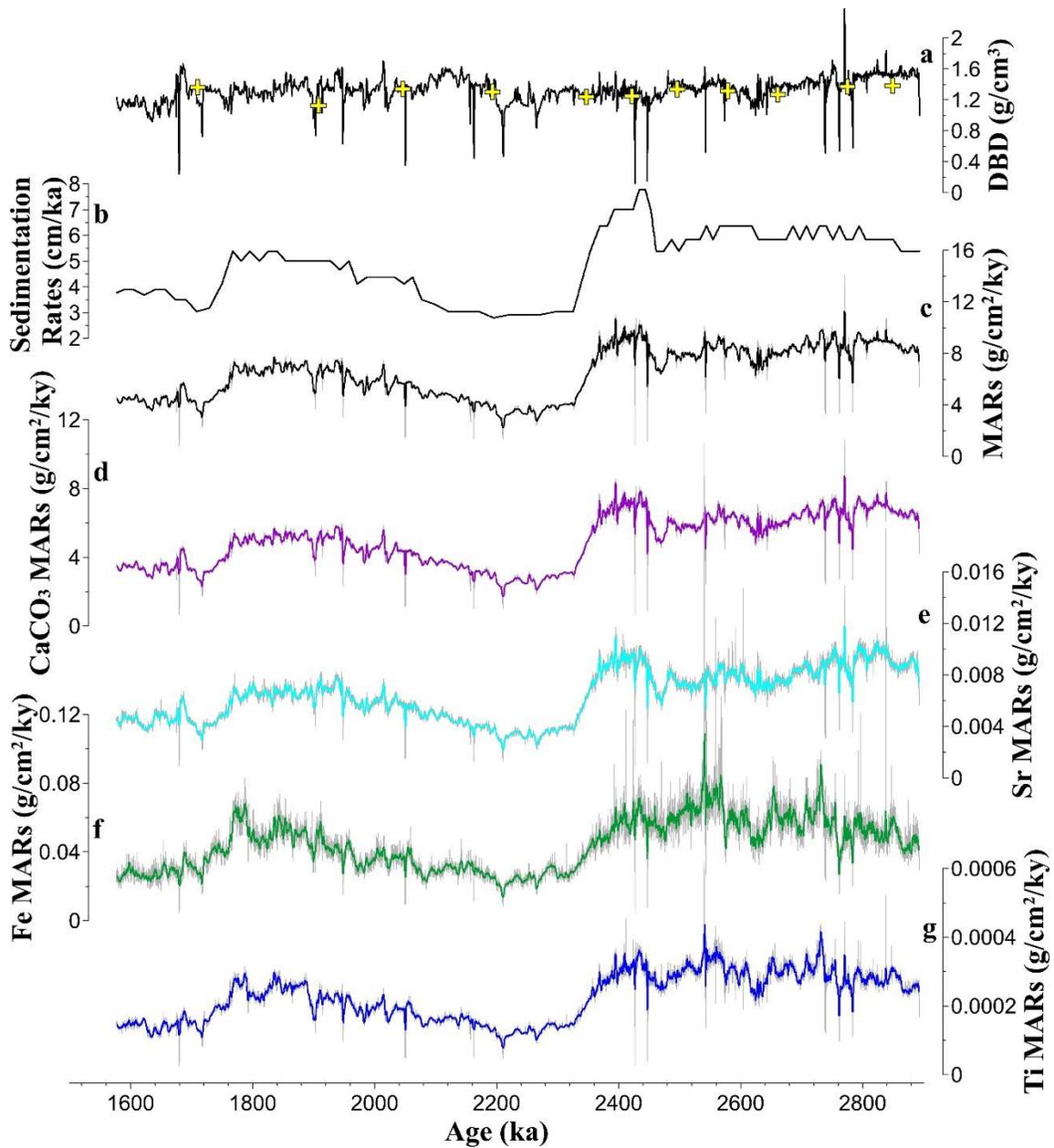


Figure S7. IODP Site U1464 sediment dry bulk density (DBD; black = calculated, yellow = shipboard measurements (Gallagher et al., 2017a), a), sedimentation rates (b) and mass accumulation rates (MARs) over ~1.6-2.9 Ma. Total MARs (c); CaCO₃ MARs (d); Sr MARs (e); Fe MARs (f) and Ti MARs (g). Bold lines (grey lines) in c-g are 21-point running averages (original data).

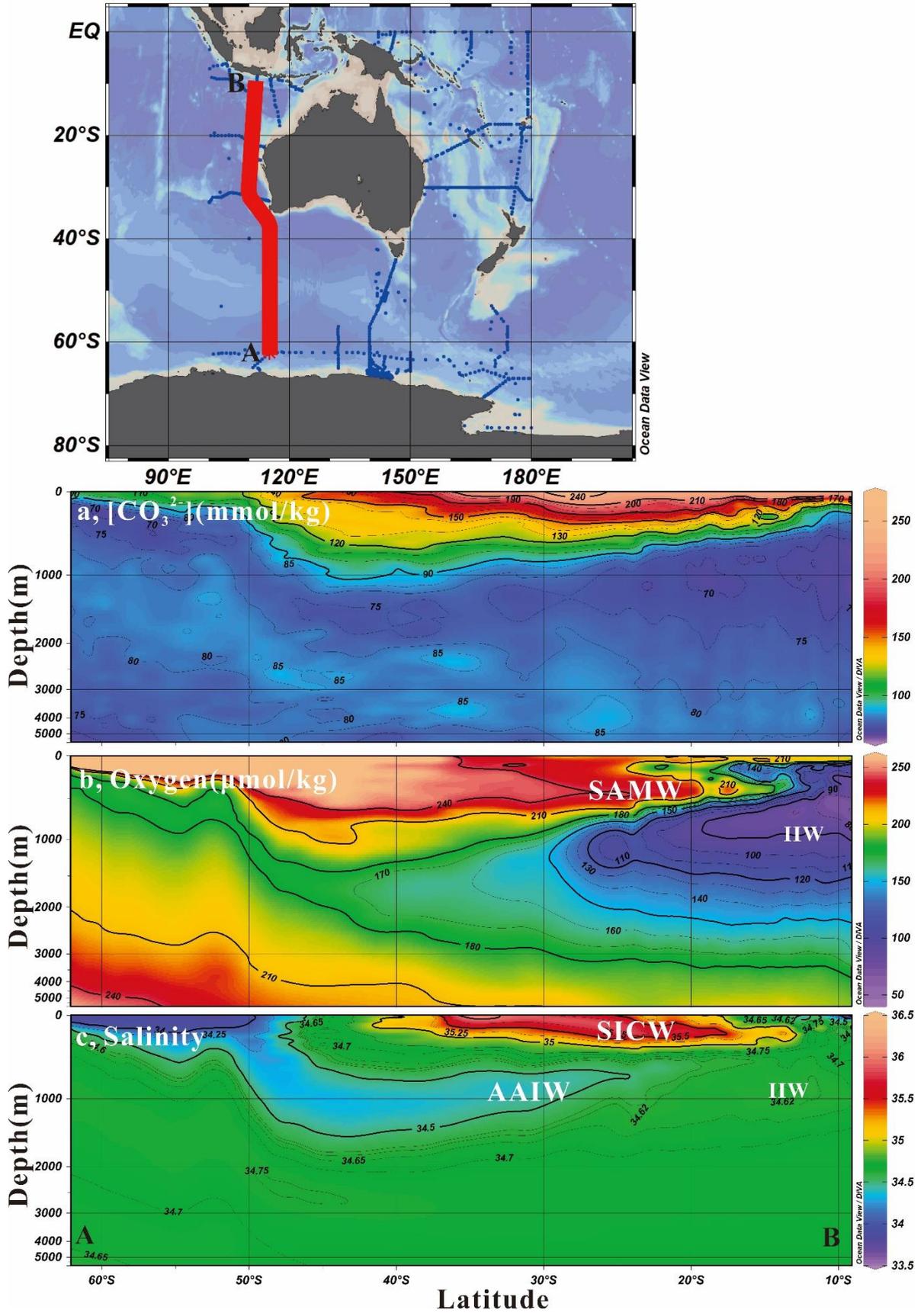


Figure S8. Meridional cross-section profiles of (a) $[\text{CO}_3^{2-}]$, (b) oxygen content and (c) salinity for transect-AB (red line). Data is from the GLODAPv2.2021 dataset (Key et al., 2015). $[\text{CO}_3^{2-}]$ from the GLODAPv2.2021 dataset is calculated using the CO2SYS method (Lewis & Wallace, 1998). Blue dots represent available data-points. SAMW: Sub-Antarctic Mode Water; AAIW: Antarctic Intermediate Water; SICW: South Indian Central Water; IIW: Indonesian Intermediate Water. SAMW is characterized by high-oxygen content (Herraiz-Borreguero & Rintoul, 2011; Woo & Pattiaratchi, 2008); AAIW is characterized by a salinity minimum (Wong, 2005; Woo & Pattiaratchi, 2008); SICW is characterized by a salinity maximum (Woo & Pattiaratchi, 2008); IIW is characterized by a salinity minimum (Talley & Sprintall, 2005) and an oxygen minimum (Fieux et al., 1996).

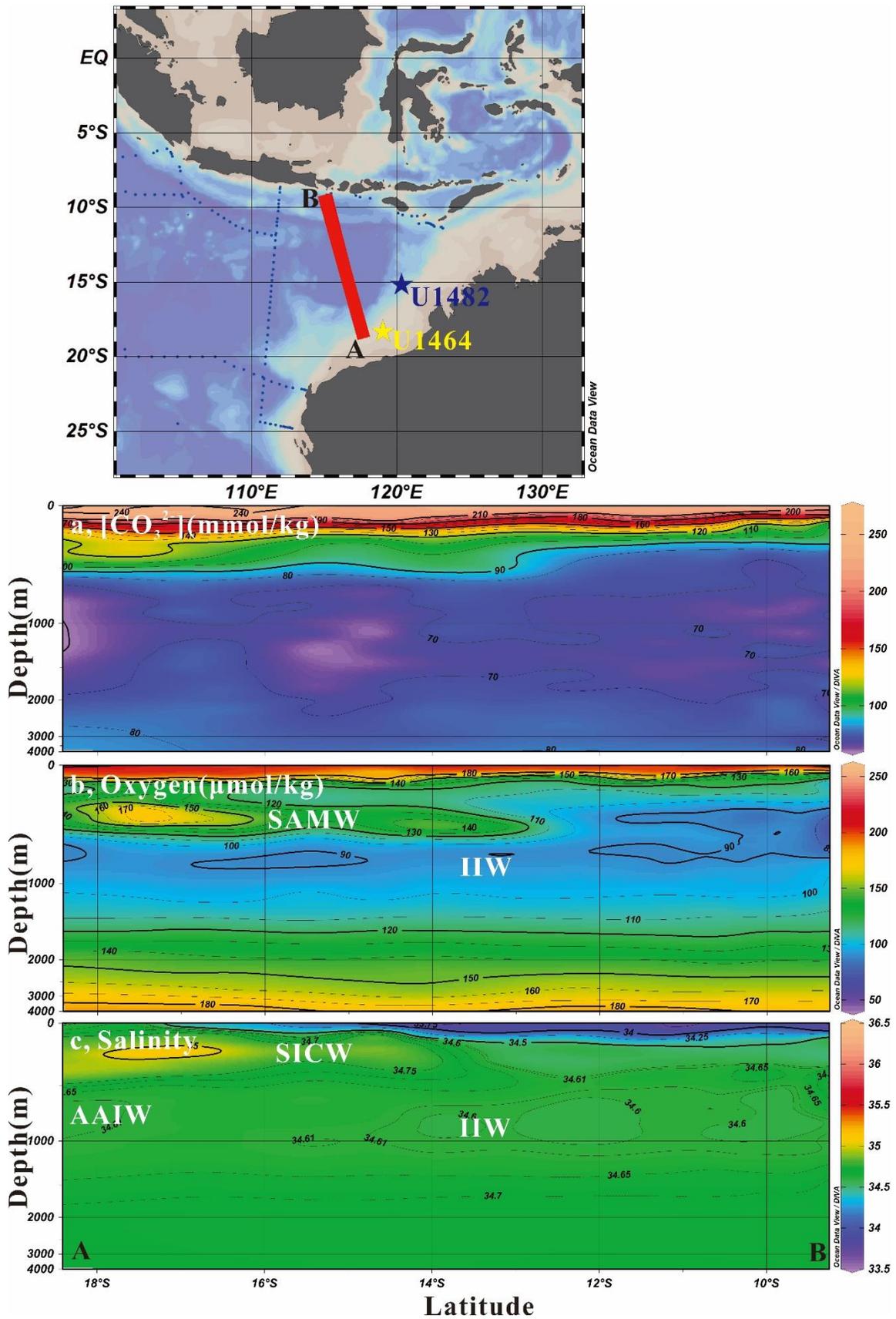


Figure S9. Meridional cross-section profiles of (a) $[\text{CO}_3^{2-}]$, (b) oxygen content and (c) salinity for transect-AB (red line). Yellow (blue) star marks IODP Site U1464 (U1482). This cross-section is the closest GLODAPv2.2021 data to Site U1464 and Site U1482. $[\text{CO}_3^{2-}]$ from the GLODAPv2.2021 dataset is calculated using the CO2SYS method (Lewis & Wallace, 1998). Blue dots represent available data-points. SAMW: Sub-Antarctic Mode Water; AAIW: Antarctic Intermediate Water; SICW: South Indian Central Water; IIW: Indonesian Intermediate Water. SAMW is characterized by high-oxygen content (Herraiz-Borreguero & Rintoul, 2011; Woo & Pattiaratchi, 2008); AAIW is characterized by a salinity minimum (Wong, 2005; Woo & Pattiaratchi, 2008); SICW is characterized by a salinity maximum (Woo & Pattiaratchi, 2008); IIW is characterized by a salinity minimum (Talley & Sprintall, 2005) and an oxygen minimum (Fieux et al., 1996).

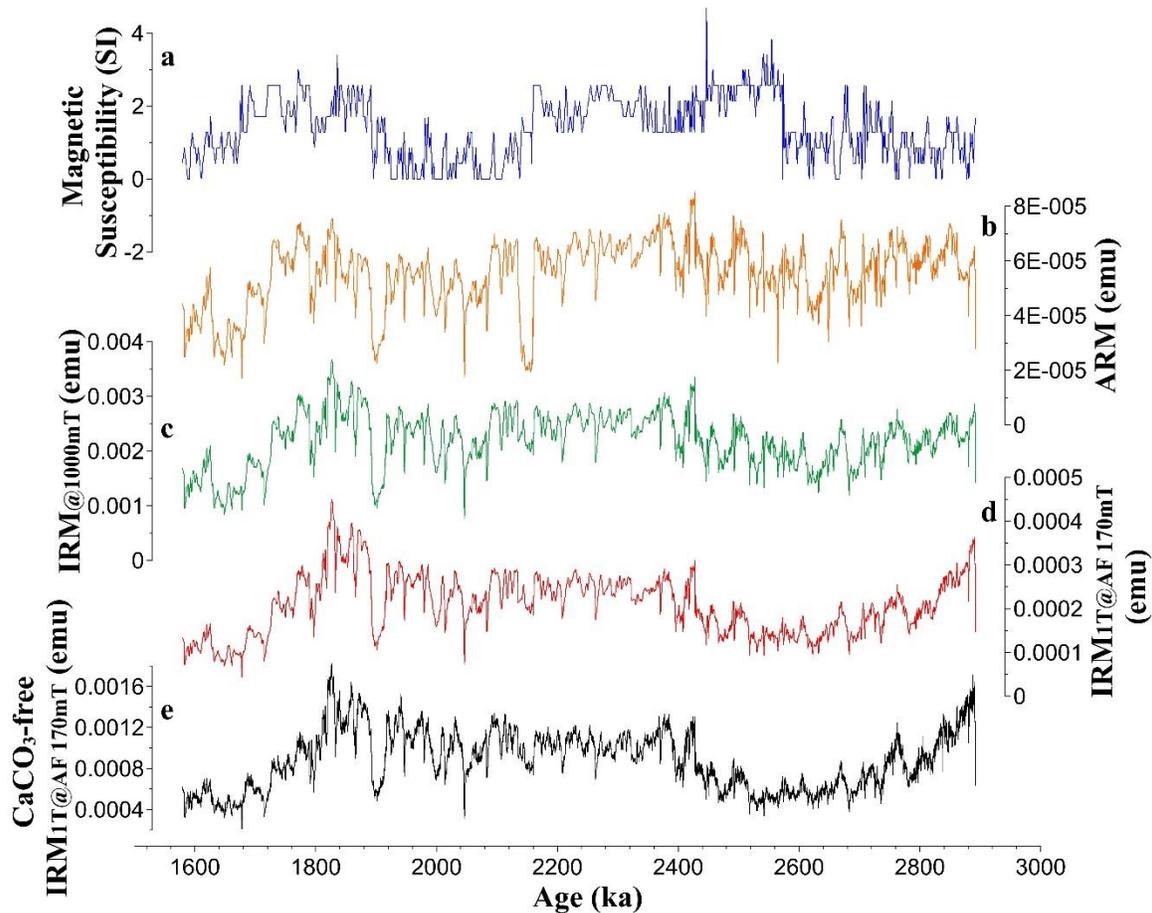


Figure S10. IODP Site U1464 magnetic parameters over the late Pliocene/early Pleistocene. (a) Magnetic susceptibility; (b) ARM; (c) $IRM_{@1000mT}$; (d) $IRM_{1T@AF 170mT}$ and (e) $CaCO_3$ -free $IRM_{1T@AF 170mT}$. Magnetic susceptibility data is from [Gallagher et al., 2017a](#) and other data are measured in this study.

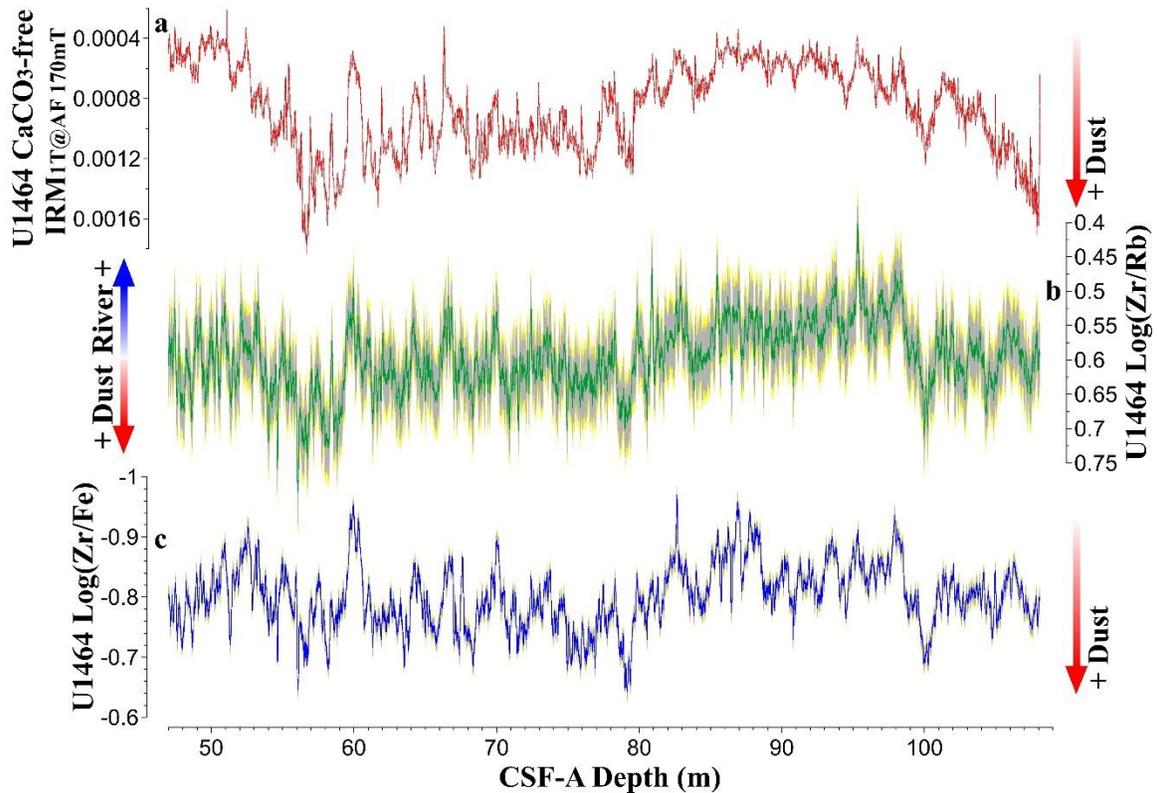


Figure S11. Dust proxy records from the Australian NWS and their uncertainties. $CaCO_3$ -free $IRM_{1T@AF 170mT}$ (a, red), $Log(Zr/Rb)$ (b, green: 21-point running average), and $Log(Zr/Fe)$ (c, blue: 21-point running average) records at Site U1464. The grey and yellow shadings represent the 95 % and 99 % confidence intervals. Uncertainty calculations for these three dust records are shown in [Text S2](#).

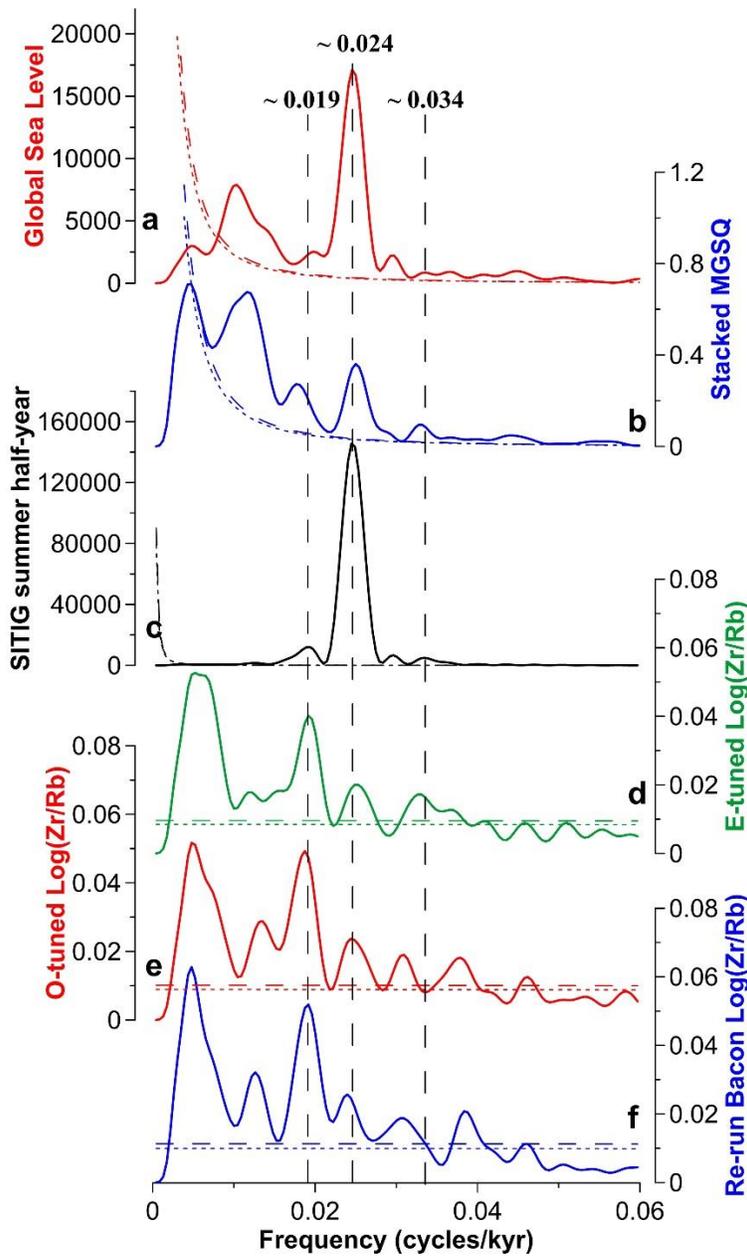


Figure S12. Spectral analyses of Northwest Australian dust fluxes and its three potential origins over the 1.6-2.9 Ma interval using the Redfit method. Power spectra for global sea level (Rohling et al., 2021) (a), stacked mean grain-size of quartz (MGSQ) from the Chinese Loess Plateau (Sun et al., 2006) (b), inter-tropical insolation gradient (SITIG) summer half-year (Laskar et al., 2004) (c), and Site U1464 Log(Zr/Rb) on the eccentricity tuning age model (d), obliquity tuning age model (e) and re-run Bacon age model (f). The Redfit spectra (oversample = 8, segment = 8) and confidence levels (90%, dashed; 95%, dot) are shown.

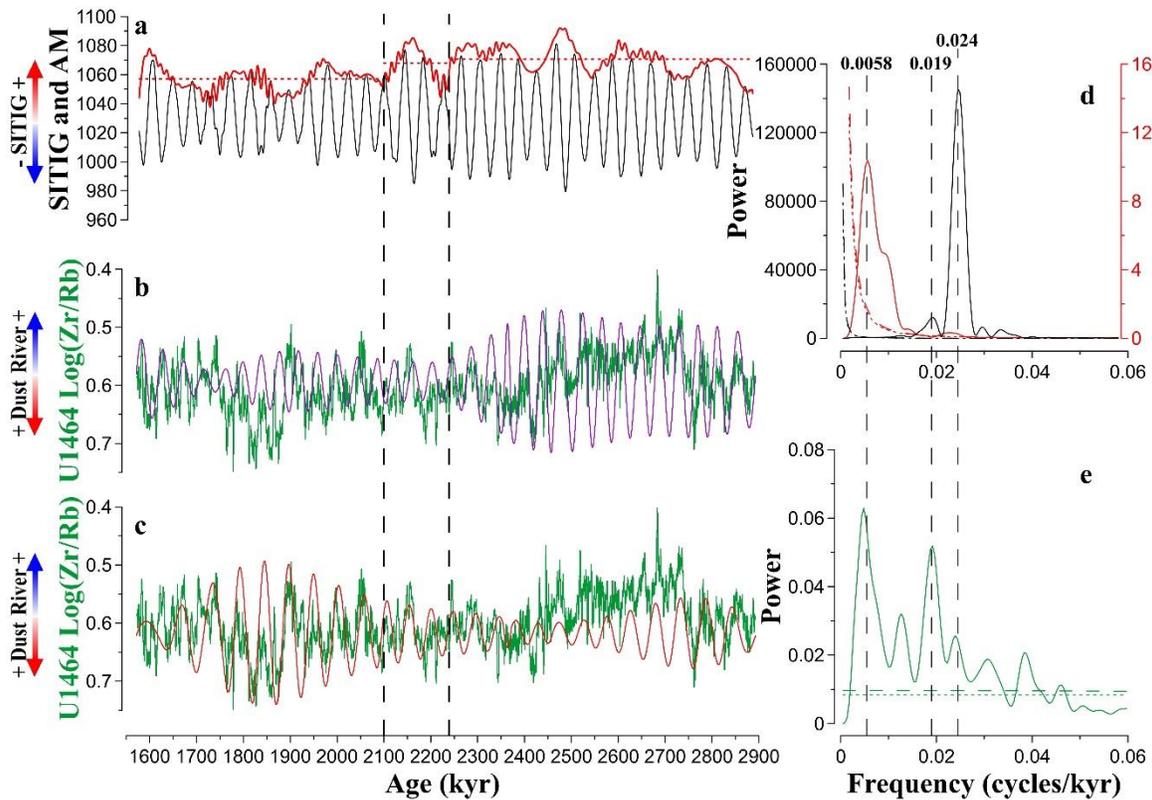


Figure S13. Inter-tropical insolation gradient (SITIG) summer half-year (black) and its amplitude modulation (AM, red) (a); Site U1464 Log(Zr/Rb) (green) and its 0.023-0.027 filter (purple) (b) and 0.017-0.021 filter (red) (c); Redfit power spectra of SITIG summer half-year (black) and its AM (red) (d), and Site U1464 Log(Zr/Rb) (green) (e). The SITIG summer half-year AM envelope is calculated using the MATLAB built-in function “hilbert” (Hilbert transform), and the mean values before 2.24 Ma, during 2.24-2.1 Ma and after 2.1 Ma are shown as red dotted lines in a. The Log(Zr/Rb) filters are calculated using the Fast Fourier transform in the Origin software package. Confidence levels (90%, dot; 95%, dashed) and significant frequencies (dash lines) in d and e are shown.

Table S1. Results of ICP-OES analysis for Site U1464, and standard deviation (SD) for certified reference materials (marine sediments BCSS-1, MESS-1 and estuarine sediments 1646).

Hole, Core, Section, Depth	CSF-A Depth/m	Ca/ppm	Fe/ppm	Sr/ppm	Ti/ppm
D1H1 36-37 cm	0.36	271791.19	2521.55	2385.06	8.46
D1H2 109-110 cm	2.59	260929.47	1891.64	2620.27	8.93
D2H1 78-79 cm	4.28	243912.04	2708.33	2666.67	10.92
D2H2 149-150 cm	6.49	271530.51	3100.39	2066.93	15.15
D2H4 1-2 cm	8.01	242292.39	3117.85	1685.73	20.90
D2H6 17-18 cm	11.17	268626.98	1958.71	1469.04	11.78
D2H7 65-66 cm	12.87	273355.58	2287.89	1572.93	18.33
D3H1 144-145 cm	14.44	273812.92	2592.59	1709.40	19.63
D3H3 142-143 cm	17.42	280691.51	2599.91	1630.00	16.60
D3H4 144-145 cm	18.94	281755.47	2911.51	1669.84	21.47
D3H5 140-141 cm	20.4	279954.03	2742.59	1685.39	14.13
D3H7 53-54 cm	22.05	267369.15	2869.38	1611.86	17.42
D4H2 70-71 cm	24.7	233333.33	2835.28	1516.03	20.79
D4H3 62-63 cm	26.12	277248.93	1934.79	1956.21	9.27
D4H4 149-150 cm	28.49	209504.90	2115.83	1793.55	12.91
D4H5 77-78 cm	29.27	255956.59	2067.14	1696.11	15.20
D4H7 60-61 cm	32	251155.27	1975.51	1802.22	17.37
D5H2 117-118 cm	34.67	290882.92	2252.88	2000.96	18.25
D5H4 18-19 cm	36.68	265655.62	2183.85	1973.87	17.46
D5H6 20-21 cm	39.7	250397.20	1528.04	1808.41	13.95
D5H7 56-57 cm	41.56	258108.11	1657.82	1636.10	12.50
D6H2 1-2 cm	43.01	272419.43	2340.03	1751.52	11.83
D6H3 0-1 cm	44.5	243153.92	4185.55	1458.92	26.35
D6H3 53-54 cm	45.03	285259.99	4712.08	1469.70	25.45
D6H4 56-57 cm	46.56	275391.50	6590.60	1315.44	35.50
D6H4 146-147 cm	47.46	210471.83	5069.81	794.42	25.83
D6H5 87-88 cm	48.37	280966.42	7023.59	952.81	30.72
D6H6 61-62 cm	49.61	265960.04	6374.27	994.15	23.82
D6H6 147-148 cm	50.47	248979.10	6504.62	1035.49	25.33
D7H1 80-81 cm	51.8	231312.17	7162.48	952.16	40.03
D7H2 5-6 cm	52.55	252171.95	8748.87	1004.52	42.15
D7H3 1340 cm	55.34	213450.02	6729.62	967.30	33.98
D7H4 147-148 cm	56.97	212837.54	6752.99	956.18	39.73
D7H6 59-60 cm	59.09	221516.96	7328.14	1044.91	36.04
D8H1 145-146 cm	61.95	260158.80	6130.31	1106.96	30.97
D8H3 2-3 cm	63.52	220878.58	5671.27	1036.53	36.44
D8H3 145-146 cm	64.95	234390.13	6512.58	1030.98	26.85
D8H6 1-2 cm	68.01	231451.21	5742.94	1054.98	22.78
D8H7 27-28 cm	69.77	215530.67	7339.34	1051.61	39.01
D9H2 117-118 cm	72.67	241003.83	6201.62	918.76	26.98
D9H4 17-18 cm	74.67	225276.83	5704.86	983.82	31.78
D9H5 2-3 cm	76.02	235133.89	5090.03	886.43	33.07
D9H6 81-82 cm	78.31	240009.40	7066.29	902.68	23.95
D10H1 100-101 cm	80.5	208100.83	6549.19	1027.90	43.27
D10H3 42-43 cm	82.92	173891.74	8224.45	1119.93	46.95
D10H5 34-35 cm	85.84	212358.44	7304.28	945.35	33.46
D11H2 8-9 cm	90.58	208958.62	5463.85	886.77	30.76
D11H5 28-29 cm	95.28	253995.27	7751.77	1078.01	47.50
D12H1 140-141 cm	99.9	227825.89	4956.70	1011.85	42.33
D12H5 101-102 cm	105.51	203052.39	5207.29	997.72	37.78
Certified Reference Materials	Replicates Count	Ca SD	Fe SD	Sr SD	Ti SD
1646	4	3.422‰	1.064‰	0.008‰	0.004‰
BCSS-1	3	4.981‰	2.607‰	0.009‰	0.005‰
MESS-1	3	2.445‰	2.792‰	0.004‰	0.022‰
Average Value	---	3.616‰	2.154‰	0.007‰	0.010‰

Table S2. IODP Site U1464 shipboard CaCO₃ measurements (Gallagher et al., 2017a), scanning-XRF Ln(Ca/Fe) values, and predicted CaCO₃ content.

Hole, Core, Section	CSF-A Depth/m	CaCO ₃ /%	Ln(Ca/Fe)	Predicted CaCO ₃ /%
B1H1	1.45	89.91	4.693	92.35
B3H3	16.15	88.42	4.140	87.05
B4H3	25.55	87.32	4.207	87.70
B5H4	36.68	88.57	4.213	87.75
B6H3	44.65	87.30	3.839	84.17
B7H3	54.15	73.45	2.746	73.70
B7H3	63.65	75.90	3.062	76.72
B7H3	73.15	72.72	2.949	75.64
B7H3	82.65	76.31	2.784	74.07
B7H3	92.15	74.59	3.164	77.70
B7H3	101.65	79.38	3.093	77.03

Table S3. Age control-points (Gallagher et al., 2017a) in our Site U1464 Bacon age model. CN: calcareous nannofossils; PF: planktonic foraminifers; Base: first appearance depth; Top: last appearance depth. The CCSF-A for each point is calculated from their middle CSF-A depth, based on the established linear relationship between CSF-A and CCSF-A (Gallagher et al., 2017a).

Hole, Core, Section	Depth CSF-A (m)	Depth CCSF-A (m)	Marker Species	Type	Age (Ma)
Core Top	0	0	---	---	0
U1464D-1H-CC	3.55	8.72	Base <i>E. huxleyi</i>	CN	0.29
U1464B-2H-CC	11.51	7.02	Top <i>G. tosaensis</i>	PF	0.61
U1464B-4H-CC	30.82	26.28	Top <i>P. lacunosa</i>	CN	0.44
---	43.00	44.87	Lithologic Boundary	---	---
U1464B-6H-CC	49.82	46.53	Top <i>C. macintyreii</i>	CN	1.6
U1464B-6H-CC	49.82	46.53	Top <i>G. fistulosus</i>	PF	1.88
U1464B-6H-CC	49.82	56.37	Base <i>G. truncatulinoides</i>	PF	1.93
U1464D-6H-CC	51.33	48.54	Top <i>C. macintyreii</i>	CN	1.6
U1464B-7H-CC	59.46	56.37	Top <i>D. brouweri</i>	CN	1.93
U1464D-7H-CC	60.84	58.78	Top <i>G. extremus</i>	PF	1.99
U1464B-8H-CC	69.09	66.51	Top <i>G. apertura</i>	PF	1.64
U1464D-9H-CC	79.83	79.04	Top <i>G. limbata</i>	PF	2.39
U1464B-10H-CC	88.17	86.14	Top <i>D. pentaradiatus</i>	CN	2.39
U1464B-11H-CC	97.42	96.23	Top <i>D. surculus</i>	CN	2.49
U1464B-14H-CC	126.30	125.96	Top <i>D. tamalis</i>	CN	2.8
U1464B-14H-CC	126.30	135.59	Base <i>G. fistulosus</i>	PF	3.33
U1464B-14H-CC	126.30	135.59	Base <i>G. tosaensis</i>	PF	3.35
U1464B-14H-CC	126.30	125.96	Top <i>D. altispira</i>	PF	3.47

Table S4. Different depth options for each Site U1464 CaCO₃-to-eccentricity tie-point (see Figure 4; Figure S5). Twelve possible combinations are shown (Figure S5). Bold numbers represent the best option for the U1464 age model in this study, based on visual alignment and correlation analysis between eccentricity and U1464 CaCO₃.

Eccentricity Tuning-point	Tuning Age (kyr)	Depth (m CCSF-A)	Bacon Age (kyr)
1	1682	51.99	1746
1	1682	53.62	1800
2	2066	63.05	2012
2	2066	71.57	2174
3	2388	79.97	2332
4	2821	104.91	2738
4	2821	109.18	2817
4	2821	112.22	2873

Table S5. IODP Site U1464 tuning tie-points.

Depth (m CCSF-A)	Initial Bacon Age (kyr)	Tuning Age (kyr)	Reference proxy	Orbital Target
49.53	1664	1587	Log(Zr/Rb)	Obliquity
51.33	1725	1627	Log(Zr/Rb)	Obliquity
53.14	1784	1672	Log(Zr/Rb)	Obliquity
53.62	1800	1682	CaCO ₃	Eccentricity
54.85	1841	1709	Log(Zr/Rb)	Obliquity
56.52	1892	1750	Log(Zr/Rb)	Obliquity
58.28	1926	1793	Log(Zr/Rb)	Obliquity
60.10	1958	1837	Log(Zr/Rb)	Obliquity
62.00	1993	1878	Log(Zr/Rb)	Obliquity
63.93	2028	1917	Log(Zr/Rb)	Obliquity
65.88	2063	1959	Log(Zr/Rb)	Obliquity
67.80	2100	2002	Log(Zr/Rb)	Obliquity
69.80	2139	2044	Log(Zr/Rb)	Obliquity
71.57	2174	2066	CaCO ₃	Eccentricity
71.71	2177	2081	Log(Zr/Rb)	Obliquity
72.82	2198	2124	Log(Zr/Rb)	Obliquity
73.94	2220	2164	Log(Zr/Rb)	Obliquity
75.14	2244	2205	Log(Zr/Rb)	Obliquity
76.30	2266	2246	Log(Zr/Rb)	Obliquity
77.41	2288	2284	Log(Zr/Rb)	Obliquity
78.48	2309	2327	Log(Zr/Rb)	Obliquity
79.55	2327	2368	Log(Zr/Rb)	Obliquity
79.97	2332	2388	CaCO ₃	Eccentricity
81.61	2354	2405	Log(Zr/Rb)	Obliquity
84.35	2390	2448	Log(Zr/Rb)	Obliquity
87.06	2429	2487	Log(Zr/Rb)	Obliquity
89.749	2473	2528	Log(Zr/Rb)	Obliquity
92.36	2515	2569	Log(Zr/Rb)	Obliquity
95.05	2559	2607	Log(Zr/Rb)	Obliquity
97.64	2604	2649	Log(Zr/Rb)	Obliquity
100.28	2653	2690	Log(Zr/Rb)	Obliquity
102.90	2701	2728	Log(Zr/Rb)	Obliquity
105.52	2750	2768	Log(Zr/Rb)	Obliquity
108.21	2799	2810	Log(Zr/Rb)	Obliquity
109.18	2817	2821	CaCO ₃	Eccentricity
110.88	2848	2851	Log(Zr/Rb)	Obliquity