

Multi-elemental Statistical Features of Early Paleogene Sediments from the Mid-latitude Eastern Indian Ocean

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Abstract

The early Paleogene is characterized by a “hothouse” environment with repetitive transient warming events known as “hyperthermals.” While these paleoenvironmental changes are well-documented in the Pacific and Atlantic Oceans, records of such changes in the Indian Ocean are limited. Here, we present a new dataset of bulk chemical composition and stable isotopic ratios of the late Paleocene–middle Eocene sediments on the Exmouth Plateau in the mid-latitude eastern Indian Ocean. The bulk $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ suggest a warming period called the Early Eocene Climate Optimum (EECO) and cooling towards the middle Eocene in a long-term perspective. From a short-term perspective, we identified at least five hyperthermals (PETM, H2, I1, J, and ETM3) in the studied sections. We identified six independent components (ICs) corresponding to sediment source materials and post-depositional processes by applying independent component analyses (ICA) to the bulk chemical composition data. The time-series behavior of IC3 indicates an increase in detrital material or a decrease in carbonate rain flux during both long-term (EECO) and short-term (hyperthermal) warming. Additionally, the rise in IC2 implies an increased population of high consumers in the oceanic ecosystem during warming events around the Exmouth Plateau. Other ICs (IC1, IC4, IC5, and IC6), indicators of diagenetic processes and post-depositional remobilization of elements, showed excursions across hyperthermal horizons. These observations indicate that changes in the redox state of pore or bottom water in the Exmouth Plateau are associated with hyperthermals.

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1 **Multi-elemental Statistical Features of Early Paleogene Sediments from the Mid-**
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16
17 **Key Points:**

- 18 • Here, we report bulk $\delta^{13}\text{C}$, $\delta^{18}\text{O}$, and chemical compositions of early Paleogene
19 sediments from Site 762C in the Indian Ocean.
- 20 • We conducted independent component analysis on the bulk chemical composition to
21 evaluate the geochemistry of the depositional area.
- 22 • Environmental factors including terrigenous input, productivity, and redox at the site
23 changed during early Paleogene hyperthermals.
24

25 Abstract

26 The early Paleogene is characterized by a “hothouse” environment with repetitive transient
27 warming events known as “hyperthermals.” While these paleoenvironmental changes are well-
28 documented in the Pacific and Atlantic Oceans, records of such changes in the Indian Ocean are
29 limited. Here, we present a new dataset of bulk chemical composition and stable isotopic ratios
30 of the late Paleocene–middle Eocene sediments on the Exmouth Plateau in the mid-latitude
31 eastern Indian Ocean. The bulk $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ suggest a warming period called the Early Eocene
32 Climate Optimum (EECO) and cooling towards the middle Eocene in a long-term perspective.
33 From a short-term perspective, we identified at least five hyperthermals (PETM, H2, I1, J, and
34 ETM3) in the studied sections. We identified six independent components (ICs) corresponding to
35 sediment source materials and post-depositional processes by applying independent component
36 analyses (ICA) to the bulk chemical composition data. The time-series behavior of IC3 indicates
37 an increase in detrital material or a decrease in carbonate rain flux during both long-term
38 (EECO) and short-term (hyperthermal) warming. Additionally, the rise in IC2 implies an
39 increased population of high consumers in the oceanic ecosystem during warming events around
40 the Exmouth Plateau. Other ICs (IC1, IC4, IC5, and IC6), indicators of diagenetic processes and
41 post-depositional remobilization of elements, showed excursions across hyperthermal horizons.
42 These observations indicate that changes in the redox state of pore or bottom water in the
43 Exmouth Plateau are associated with hyperthermals.

44 1 Introduction

45 Among the long-term climatic trends of the Cenozoic, the early Paleogene is a globally
46 warm period. In the long-term ($\sim 10^6$ yr) climatic trend, the global climate has been warmer since
47 the Paleocene, and the peak of the warming is called the Early Eocene Climate Optimum
48 (EECO, 53.260–49.140 Ma; Westerhold et al., 2020; Zachos et al., 2001). After EECO, the
49 climate trend turned to long-term cooling towards the middle Eocene. Superimposed on the
50 million-years-long warm period during the late Paleocene–early Eocene are well-known transient
51 (10^4 – 10^5 yr) warming events called “hyperthermals” (e.g., Thomas, 1998; Westerhold et al.,
52 2018). In particular, the Paleocene–Eocene Thermal Maximum (PETM, 56 Ma) is the most
53 pronounced and well-studied hyperthermal event, characterized by intense global warming (5–9
54 $^{\circ}\text{C}$ rise in sea surface temperature: SST) and ocean acidification associated with the negative
55 carbon isotope ($\delta^{13}\text{C}$) excursion (CIE, e.g., Kennett & Stott, 1991; McInerney & Wing, 2011;
56 Zachos et al., 2003, 2005). In addition to the PETM, many hyperthermals accompanying
57 negative CIEs, such as the Eocene Thermal Maximum 2 (ETM2), H2, I1, I2, J, and ETM3, have
58 been reported. The background climate, referred to as a “hothouse” climate, and hyperthermals
59 during the early Paleogene have garnered considerable attention as a paleo-analog to the
60 potential fate of contemporary climate changes, including global warming.

61 The paleoceanographic environment during the early Paleogene has been deduced from
62 geochemical records of seafloor sediments. One characteristic paleoceanographic change
63 associated with warming is carbonate compensation depth (CCD) elevation. From a long-term
64 perspective ($\sim 10^6$ yr), the CCD shallowed during the EECO (Pälike et al., 2012); however, from
65 a short-term perspective ($\sim 10^5$ yr), CCD shallowing also occurred in hyperthermals (e.g., Raffi &
66 Bernardi, 2008; Slotnick et al., 2015; Zachos et al., 2005). The CCD shallowing corresponding to
67 the long- and short-term warming has been attributed to the pH lowering of seawater due to high
68 atmospheric $p\text{CO}_2$ (e.g., Anagnostou et al., 2016, 2020; Pälike et al., 2012; Zachos et al., 2005).

69 The ocean stratification, acidification, and deep-water deoxygenation at hyperthermals,
70 particularly in the PETM, have also been inferred by foraminiferal records (e.g., Kaiho et al.,
71 2006; Thomas, 1998) and the enrichment/depletion of redox-sensitive-elements in the marine
72 sediments (Chun et al., 2010; Pälike et al., 2014).

73 The Indian Ocean, the major seawater path between the Atlantic and the Pacific during
74 the early Paleogene (Thomas et al., 2003), is important in elucidating paleoceanographic
75 conditions during the early Paleogene in a global context. Geochemical investigation of Indian
76 Ocean sediments can provide fundamental keys for evaluating global behavior in the early
77 Paleogene hothouse. While geochemical records of hyperthermals, especially of the PETM, from
78 the Atlantic and Pacific Oceans have been well established (e.g., Cramer et al., 2003; Hollis et al.,
79 2015; Lauretano et al., 2015; Leon-Rodriguez & Dickens, 2010; Littler et al., 2014; Lourens et
80 al., 2005; Stap et al., 2010; Westerhold et al., 2018), there are only a few records of
81 hyperthermals from the Indian Ocean (Slotnick et al., 2015; Tanaka et al., 2022; Tremolada &
82 Bralower, 2004; Yasukawa et al., 2017).

83 In this context, we focused on the Ocean Drilling Program (ODP) Site 762 Hole C from
84 offshore of the northwestern margin of the Australian continent (Exmouth Plateau; Figure 1a).
85 The ODP results reported bulk carbonate $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ records from Site 762C (Thomas et al.,
86 1992) and roughly indicate the existence of multiple hyperthermals supported by paleomagnetic
87 and biostratigraphic studies (Galbrun, 1992; Shamrock & Watkins, 2012; Shamrock et al., 2012).
88 However, the temporal resolution of the reported $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ records was low, and
89 hyperthermal horizons were not firmly determined. Here, we report higher resolution records of
90 bulk $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ from the early Paleogene (late Paleocene–middle Eocene) section of Site
91 762C, identify hyperthermal horizons, and show long-term $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ trends at the site.
92 Further, based on the bulk $\delta^{13}\text{C}$ record, we revised the age model of Site 762C core.

93 In addition to the C and O isotopes, we analyzed the bulk chemical composition of the
94 early Paleogene carbonate sediments of Site 762C to discuss the paleoceanographic condition at
95 the site in the Eocene. The bulk chemical composition of seafloor sediments reflects the mixed
96 signals of each source material and post-depositional element redistribution (e.g., Dunlea et al.,
97 2015; Plank & Langumuir, 1998). However, applying conventional approaches based on the
98 bivariate correlations among each element or normalized data is difficult when extracting and
99 interpreting essential information from multi-element datasets. In particular, carbonate sediments
100 are challenging to decipher source materials and post-depositional processes due to the strong
101 dilution effect of biogenic CaCO_3 (e.g., Yasukawa et al., 2017). Thus, most previous studies on
102 geochemical features use only representative elements' abundances, which inevitably exclude
103 signatures underlying the high-dimensional, entire data structure. To overcome these challenges,
104 we applied multivariate statistical analysis to extract geochemical signals hidden in multi-
105 elemental data of the bulk-chemical composition of Site 762C sediments.

106 Multivariate statistical analyses have been utilized for interpreting geochemical data (e.g.,
107 Dunlea et al., 2015; Iwamori & Albarède, 2008; Kato et al., 2011; Yasukawa et al., 2016, 2017,
108 2019, 2022). As the bulk elemental composition of seafloor sediments reflects an admixture of
109 multiple source materials and geochemical processes (e.g., diagenesis, redistribution,
110 precipitation), multivariate statistical analysis is considered a powerful approach for extracting
111 geochemical signals hidden in the high-dimensional data structure. Among various analytic
112 methods, independent component analysis (ICA) extracts features based on non-Gaussianity in a
113 multivariate dataset and is considered an appropriate analytical method for geochemical data

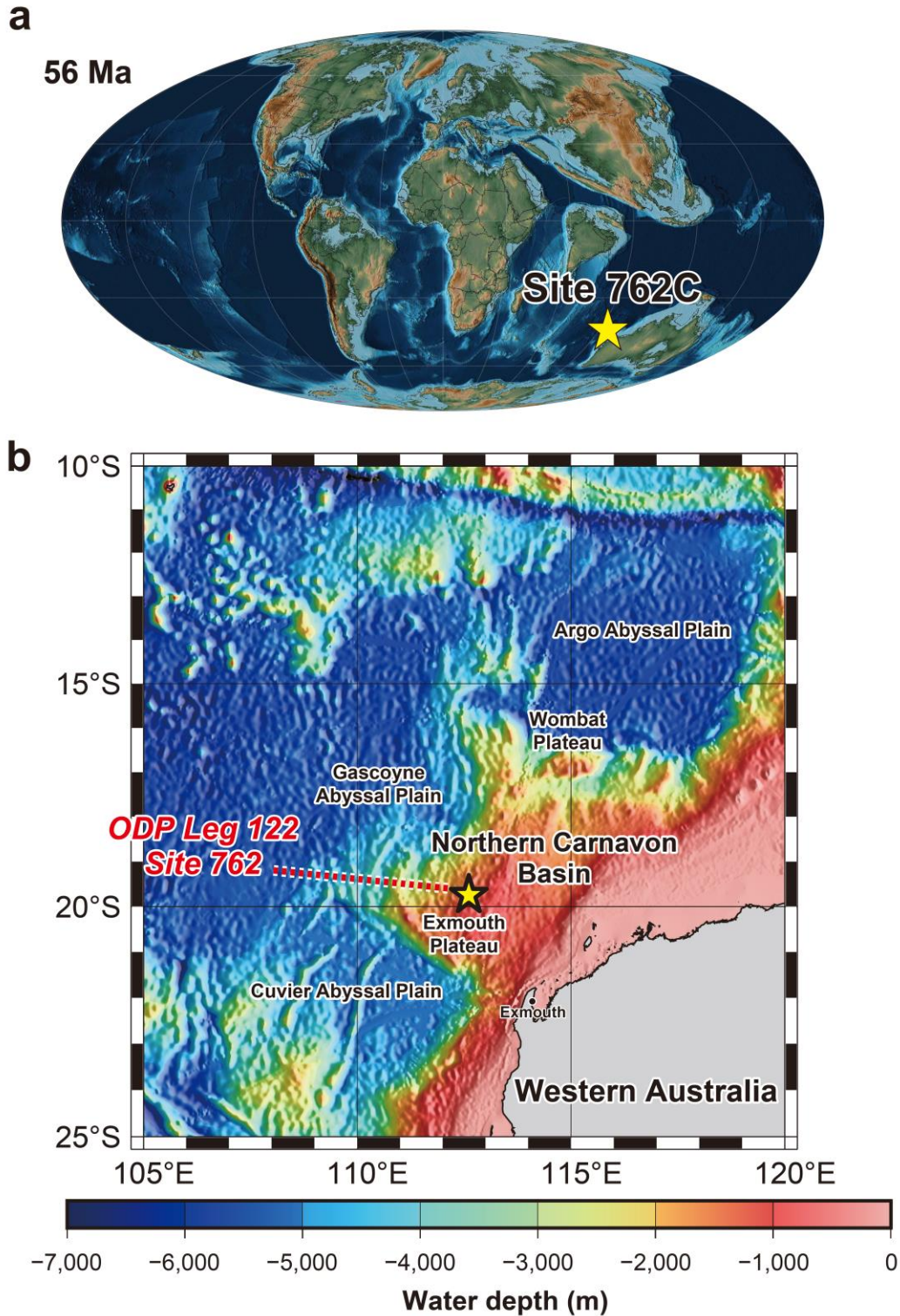
114 with a non-Gaussian data structure (e.g., Iwamori & Albarède, 2008; Yasukawa et al., 2016,
115 2017). In recent years, ICA has been successfully applied to high-dimensional geochemical
116 datasets, including the bulk chemical composition of seafloor sediments (Kato et al., 2011;
117 Yasukawa et al., 2016, 2017, 2019). By applying ICA, the observed multidimensional dataset
118 consisting of the concentration of each element could be transformed into the linear combination
119 of base vectors (independent components: ICs) corresponding to the source materials and
120 geochemical processes of the sediments. For detailed formulation on using ICA in marine
121 sediment analysis, see Yasukawa et al. (2016). In this study, by applying ICA to the geochemical
122 dataset, we evaluated the paleoceanographic changes at Site 762C in the mid-latitude eastern
123 Indian Ocean.

124

125 **2 Geological Settings and Lithology**

126 ODP Leg 122 Site 762 Hole C (19°53.23S, 112°15.24E) is located on the central
127 Exmouth Plateau in the Northern Carnarvon Basin, where ~300 km offshore of the northwestern
128 coast of the Australian continent. According to the paleogeographic reconstruction (Scotese,
129 2001), the Exmouth plateau was located in the mid-latitude of the eastern margin of the Indian
130 Ocean during the early Paleogene (~60–40 Ma; Figure 1a). The Northern Carnarvon Basin was
131 initially formed by the stretching during Carboniferous–Cretaceous, and the Exmouth plateau has
132 been thought to be inactive after mid-Cretaceous. Since then, the Cenozoic sedimentary
133 environment has corresponded to an open-ocean setting (Bilal & McClay, 2022; Haq et al.,
134 1992). The present-day water depth of this site is 1360 m (Haq et al., 1990; Figure 1b), and the
135 paleodepth of the early Paleogene is comparable to that reported today (1000–1200 m; Haq et al.,
136 1992; Jones & Wonders, 1992).

137 The lithology of the early Paleogene section of Site 762C mainly consists of nannofossil
138 chalk that was deposited above the CCD and has been divided into three lithologic subunits (Unit
139 II, Subunit IIIA, and Subunit IIIB; Haq et al., 1990; Shamrock et al., 2012). Unit II (181.5–265.0
140 mbsf: meter(s) below seafloor) consists of white consolidated nannofossil ooze/chalk deposited
141 during the middle Eocene to the early Oligocene. Subunit IIIA (265.0–398.0 mbsf) consists of
142 pale green–light green-grey nannofossil chalk with foraminifers deposited during the early–
143 middle Eocene. Subunit IIIB (398.0–554.8 mbsf) consists of light green-grey–white nannofossil
144 chalk deposited during the early Paleocene–early Eocene. Previous studies based on
145 magnetostratigraphy and biostratigraphy identified four hiatuses in the early Paleogene section of
146 Site 762C (Hiatus A: 289.75 mbsf, Hiatus B: 321.13 mbsf, Hiatus C: 332.18 mbsf, Hiatus D:
147 412.75 mbsf; Shamrock et al., 2012).



148

149 **Figure 1.** Location of studied site. (a) Paleogeographic reconstruction at the latest Paleocene (56

150 Ma) using data by Scotese (2001). Stars indicate the paleo-location of ODP Site 762C. (b)

151 Current location of the Exmouth Plateau and ODP Site 762C. The bathymetry is based on 1 min

152 gridded relief data by NOAA (ETOPO1; <https://www.ngdc.noaa.gov/mgg/global/global.html>).

153

154 3 Materials and Methods

155 3.1 Materials

156 We targeted 248 samples from the late Paleocene (Thanetian)–middle Eocene (Bartonian)
 157 section of Site 762C, corresponding to lithologic Subunits IIIA and IIIB. The sampling
 158 resolution was 5–50 kyr for the latest Paleocene–early Eocene section, which was considered to
 159 record the PETM and early Eocene hyperthermals. We collected samples with >50 kyr intervals
 160 from other sections. All samples were collected from the working half, and the consolidated mud
 161 slurry between “biscuits” of actual sediments was carefully avoided.

162 As a result of the smear slide observations, obtained samples of Site 762C mainly consist
 163 of calcareous nannofossil, which can be considered as fragments of coccolith (70–95%) and
 164 planktonic foraminiferal shells (2–30%). The contribution of detrital components (e.g., quartz,
 165 feldspar, clay minerals) was minor (<5%). In addition, authigenic pyrite grains (100–200 μm)
 166 were observed at Core 28X (412.86–413.36 mbsf).

167 3.2 Carbon and Oxygen Stable Isotope Analyses

168 Before the geochemical analyses, bulk samples were dried in a vacuum freeze dryer
 169 (EYELA FDU-1200) and crushed by an agate mortar and pestle. After sample preparation, we
 170 conducted carbon ($\delta^{13}\text{C}$) and oxygen ($\delta^{18}\text{O}$) stable isotope analyses using GV Instruments
 171 IsoPrime with a dual-inlet system at the Center for Advanced Marine Core Research (present
 172 Marine Core Research Institute), Kochi University. Approximately 100 μg of pulverized bulk
 173 samples were dissolved in phosphoric acid at 90 °C, and released CO_2 was injected into the mass
 174 spectrometer.

175 The measured isotopic ratios were converted from standard delta notation to Vienna Pee
 176 Dee Belemnite by analyzing IAEA-603 standard reference material ($\delta^{13}\text{C} = 2.460 \pm 0.010\text{‰}$,
 177 $\delta^{18}\text{O} = -2.370 \pm 0.040\text{‰}$; Assonov et al., 2020). We also measured $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ of geological
 178 reference carbonate materials (JCp-1) by the Geological Survey of Japan (GSJ) to evaluate
 179 analytical precision. In repeated analyses of IAEA-603 ($n = 60$) and JCp-1 ($n = 7$), the 2SE of
 180 $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ were less than $\pm 0.02\text{‰}$ and $\pm 0.04\text{‰}$, respectively.

181 3.3 Bulk Carbonate Analyses

182 The bulk carbonate content was determined using an elemental analyzer (Thermo
 183 Finnigan EA1112) installed at the Center for Advanced Marine Core Research (present Marine
 184 Core Research Institute), Kochi University. In this analysis, we assumed that the total inorganic
 185 carbon (TIC) of each sediment sample was incorporated as CaCO_3 , and TIC corresponds to the
 186 difference between total carbon (TC) and total organic carbon (TOC) contents. Therefore, we
 187 obtained bulk CaCO_3 content (wt.%) using the following equations:

$$188 \quad [\text{TIC}] = [\text{TC}] - [\text{TOC}] \quad (1)$$

$$189 \quad [\text{CaCO}_3] = [\text{TIC}] \times \frac{W_{\text{CaCO}_3}}{W_c} \quad (2)$$

190 where W_{CaCO_3} and W_{C} in equation (2) are the formula weight of CaCO_3 (100.0869) and the
191 atomic weight of carbon (12.0107), respectively.

192 In the TC measurement, ~2 mg of bulk samples enclosed in tin capsules were introduced
193 to the elemental analyzer and measured by a packed column. In TOC measurement, ~10 mg of
194 bulk samples were digested by 100 μL of organic-free 3N HCl in silver capsules to remove
195 CaCO_3 . After drying, the residues of the samples in silver capsules were overwrapped by tin
196 capsules and introduced to the elemental analyzer. To make calibration curves, we
197 simultaneously analyzed sulfanilamide standard reference materials.

198 3.4 Major- and Trace-element Analyses

199 The major- and trace-element content of bulk samples was determined by Thermo Fisher
200 Scientific iCAP Q, an inductively coupled plasma quadrupole mass spectrometer (ICP-QMS)
201 installed at the Department of Systems Innovation, School of Engineering, the University of
202 Tokyo. Each sample was digested by $\text{HF-HNO}_3\text{-HClO}_4$ and aqua regia, following the
203 established method described by Kato et al. (2005) and Yasukawa et al. (2020). Geological
204 reference materials (JB-2, JB-3, and JMS-2) by GSJ were also prepared simultaneously to
205 evaluate analytical accuracy.

206 In the ICP-QMS analyses, the difference between analyzed and published data of GSJ
207 standards generally falls within 5%, except for Cr, As, and Ta (Supporting Table S1; Imai et al.,
208 1995; Lu et al., 2007; Makishima & Nakamura, 2006; Yasukawa et al., 2020). The reproductivity
209 of replicate analyses ($n = 8$) is basically within 5% relative standard deviation (RSD), except for
210 As, Sb, and Ta, which sometimes result in larger than 20% RSD (Supporting Table S1).

211 3.5 ICA

212 In this study, we applied ICA to a 248×40 data matrix from Site 762C; the rows
213 corresponded to the individual bulk sediment samples, and the columns corresponded to the
214 elements. The variables consist of the abundance of Mg, Al, P, K, Sc, Ti, V, Mn, Fe, Co, Ni, Cu,
215 Zn, Rb, Sr, Y, Zr, Nb, Mo, Cs, Ba, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Hf,
216 Pb, Th, U, and CaCO_3 . Sodium content was not included in the matrix because saline could
217 largely affect Na content (Yasukawa et al., 2016). Contents of Cr, As, Sb, and Ta were not
218 included in the ICA because these elements show large RSDs through the analytical batches and
219 large differences (>20%) from published data for standard materials. Before calculation, the
220 dataset was standardized for each element. The computation of the ICA for our dataset was
221 implemented using the open R package “FastICA” originally released by Marchini et al. (2013),
222 with some modifications by Yasukawa et al. (2016, 2023).

223 In performing ICA, we determined the number of ICs and applied the “Guttman–Kaiser
224 criterion (Guttman, 1954; Kaiser, 1960)” to the eigenvalues obtained from the Principal
225 Component Analysis (PCA) algorithm (Supporting Text S1). Our analysis revealed that the
226 eigenvalues of the first six principal components (PCs 1–6) were greater than unity, and these six
227 components accounted for 87.9% of the total sample variance (Supporting Figure S1; Table S2).

228 After performing ICA, the results were interpreted based on the relative (i.e., variance-
229 normalized) IC loadings and IC scores. The relative IC loadings denote the contributions of each
230 element to the individual ICs. A large loading indicates a strong correlation between the element
231 concentration and the respective IC. As another output of ICA, the IC score represents the
232 coordinate values of sample data projected into the linear space spanned by IC vectors. In
233 simpler terms, IC scores quantify the intensities of each independent source signal in each
234 sample.

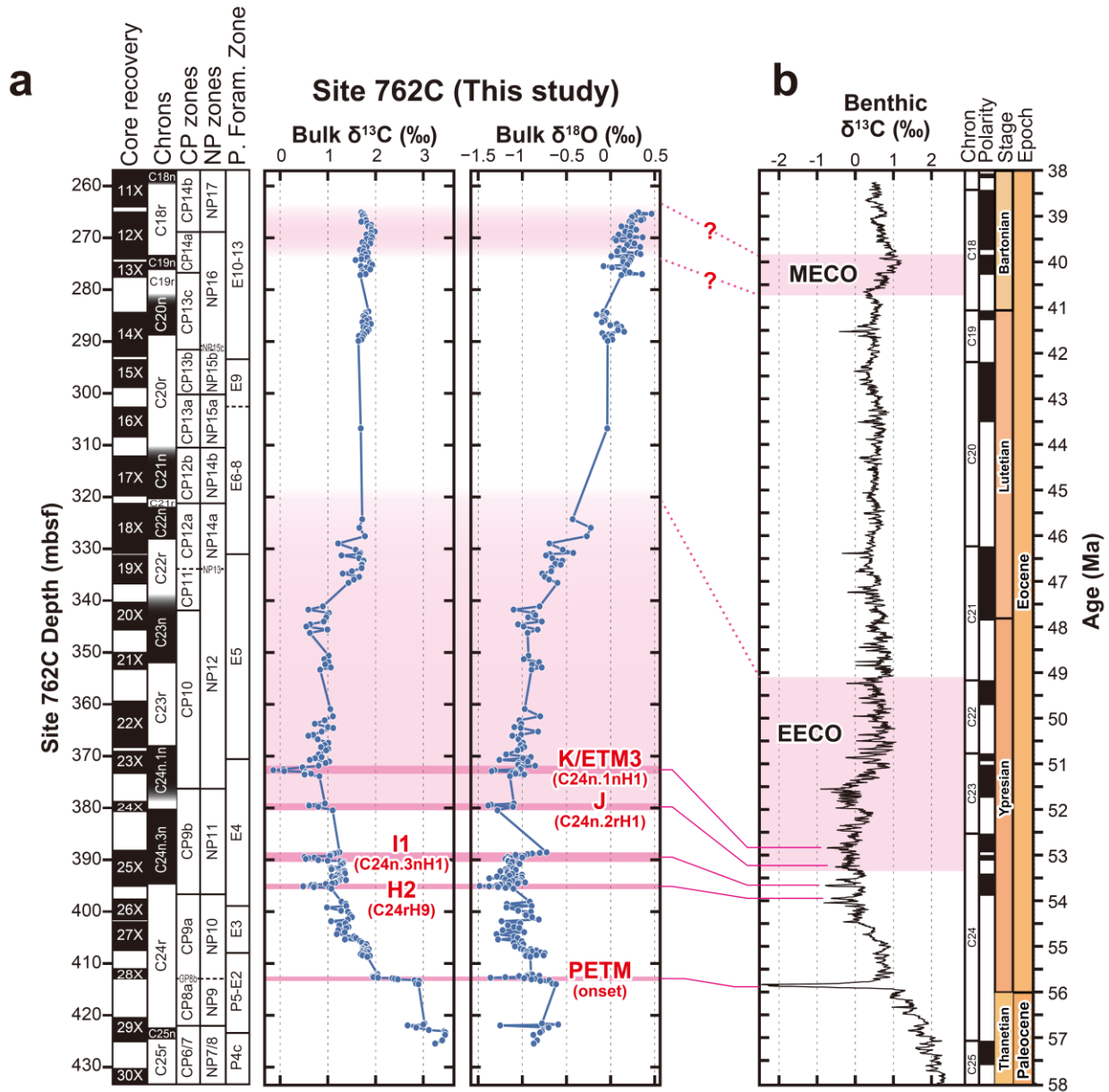
235 **4 Results**

236 4.1 Carbon and Oxygen Stable Isotope Ratios

237 The analytical data of bulk carbonate $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ of Site 762C samples are shown in
238 Supporting Table S3. Depth profiles of $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ at Site 762C are shown in Figure 2. In the
239 overall trend from Core 29X to Core 12X, bulk $\delta^{13}\text{C}$ values were $\sim 3\text{‰}$ during 425–413 mbsf
240 and dropped to $\sim 2\text{‰}$ at ~ 413 mbsf. Thereafter, the $\delta^{13}\text{C}$ gradually decreased to $\delta^{13}\text{C} = \sim 0\text{--}1\text{‰}$
241 from 395 to 340 mbsf. The bulk $\delta^{18}\text{O}$ generally started around -1.0‰ between 425 and 340
242 mbsf and increased to 0.0‰ during 340–307 mbsf. Afterward, bulk $\delta^{18}\text{O}$ rose gradually at
243 depths shallower than 280 mbsf and reached $\sim 0.5\text{‰}$ at 265 mbsf.

244 We found at least four distinct negative excursions of the bulk $\delta^{13}\text{C}$ in Core 25X–23X
245 (Figure 3b). In these cores, bulk $\delta^{18}\text{O}$ also shows negative excursions associated with those of
246 $\delta^{13}\text{C}$, although the $\delta^{18}\text{O}$ fluctuates throughout the section. The magnitude of the four excursions
247 of $\delta^{13}\text{C}$ (difference between excursion peak and onset) at 395, 390, 379, and 373 mbsf are –
248 0.6‰ , -0.7‰ , -0.5‰ , and -0.9‰ , respectively. Conversely, the magnitudes of $\delta^{18}\text{O}$ excursions
249 at each horizon are -0.5‰ , -0.2‰ , -0.3‰ , and -0.4‰ , respectively. In addition, several
250 smaller-scale negative excursions of $\delta^{13}\text{C}$ ($\sim -0.4\text{‰}$) and $\delta^{18}\text{O}$ ($\sim -0.2\text{‰}$) are also observed in
251 Core 22X–18X (Figure 2a). In contrast, no noticeable negative $\delta^{13}\text{C}$ excursion was observed in
252 Core 14X–12X, corresponding to the middle Eocene period.

253



255
 256 **Figure 2.** Correlation between (a) bulk $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ data from ODP Site 762C and (b) the
 257 reference benthic $\delta^{13}\text{C}$ profile (Westerhold et al., 2020). Magnetostratigraphy (Galbrun, 1992;
 258 Shamrock et al., 2012), calcareous nannofossil (CP zone; Okada & Bukry, 1980, NP zone;
 259 Martini, 1971), and planktonic foraminifera biostratigraphy (Hancock et al., 2002; Wade et al.,
 260 2011) are also shown in panel (a). Negative carbon isotope excursions of the hyperthermal
 261 events with red characters are used as age tie-points. The previously inferred Early Eocene
 262 Climate Optimum (EEEO) and Middle Eocene Climate Optimum (MECO) are also shown
 263 (Shamrock et al., 2012), although we could not identify MECO horizon from our bulk $\delta^{13}\text{C}$
 264 record.

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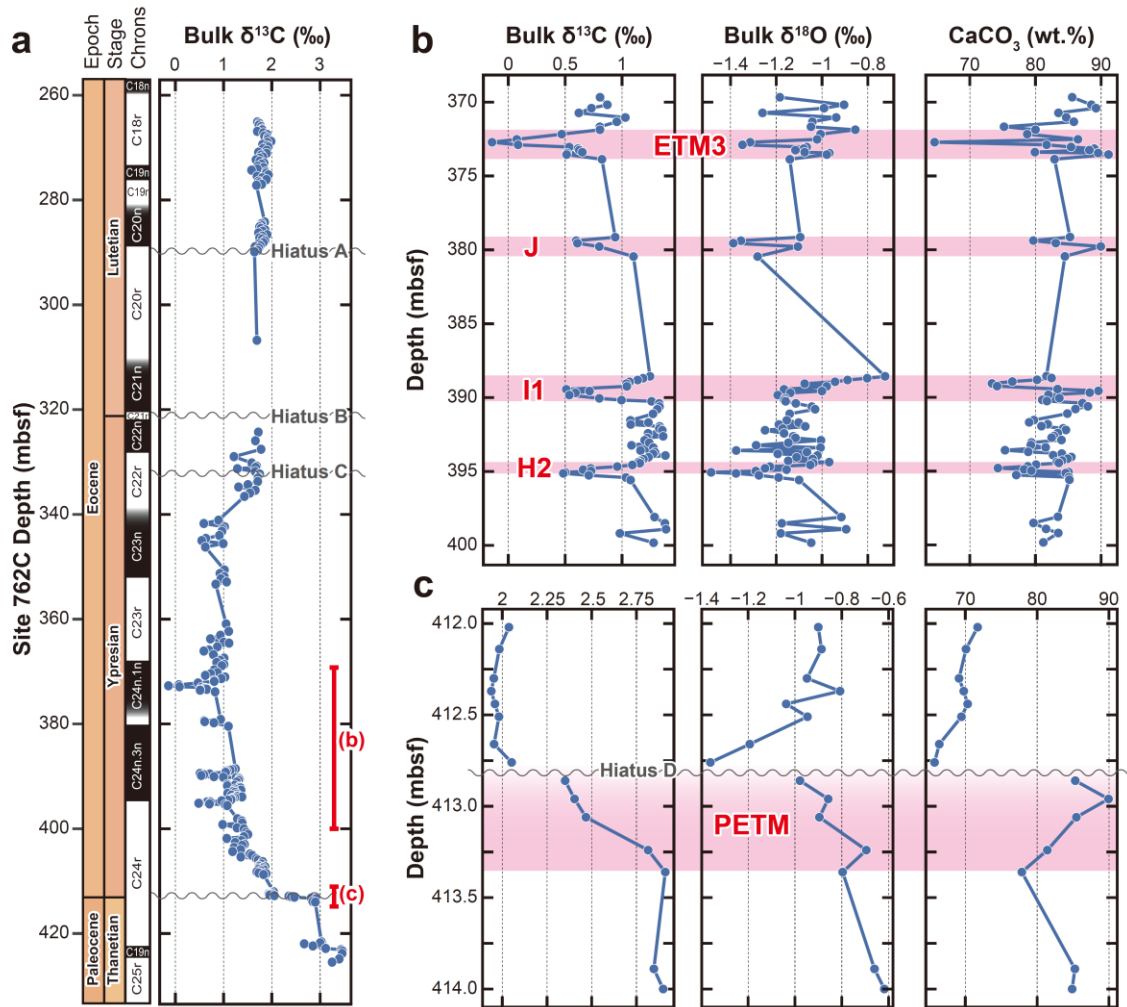
266 4.2 Bulk chemical compositions

267 The bulk chemical composition data of Site 762C, including TC, TOC, and CaCO₃
268 content, are shown in Supporting Table S4. The results show that Ca is the most abundant
269 element in the samples of Site 762C (Ca = 23.7–38.9 wt.%), and CaCO₃ accounts for 60.2–93.3
270 wt.% of bulk samples. Among the other major elements, Fe and Al are contained in percent order
271 (Al = 0.297–2.23 wt.%; Fe = 0.180–2.13 wt.%). In the trace element composition (elements in
272 the order of ppm), Sr and Ba show considerably high concentrations (Sr = 580–1308 ppm; Ba =
273 366–1683 ppm) compared to other trace elements.

274 In the depth profile of each element, the concentration of CaCO₃, the most abundant
275 component in the studied samples, dropped at the horizons of negative $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ excursions
276 (395, 390, 379, and 373 mbsf; Figure 3b, c). In addition, the CaCO₃ abundance also dropped at
277 422, 373, and 288 mbsf (Supporting Figure S2a). Conversely, depth profiles of detrital-origin
278 elements (e.g., Ti, Al) are correspondingly symmetric to that of CaCO₃ (Supporting Figure S3).
279 The depth profiles of P, Y, and total rare earth elements (REEs; here indicating from La to Lu)
280 were similar and showed sharp peaks at 404, 373, and 331 mbsf. Some metal elements (e.g., Cu,
281 Pb) showed several spike-like peaks in the depth profiles (Supporting Figure S3). Except for
282 each peak, these elements generally showed relatively less variable profiles in the long-term
283 trend.

284

285



286

287 **Figure 3.** Depth profiles of bulk $\delta^{13}\text{C}$, $\delta^{18}\text{O}$, and CaCO_3 of Site 762C. (a) All data. (b) Data from
 288 early Eocene (Core 25X–23X). (c) Data around Paleocene–Eocene boundary (Core 28X).
 289 Intervals with red lines indicate ranges of panels (b) and (c). Analytical uncertainties of $\delta^{13}\text{C}$ and
 290 $\delta^{18}\text{O}$ are less than 0.02‰ and 0.04‰, respectively. In panel (a), magnetostratigraphic columns
 291 and identified hiatuses are also shown (Shamrock et al., 2012).

292

293

4.3 Independent Component Analysis

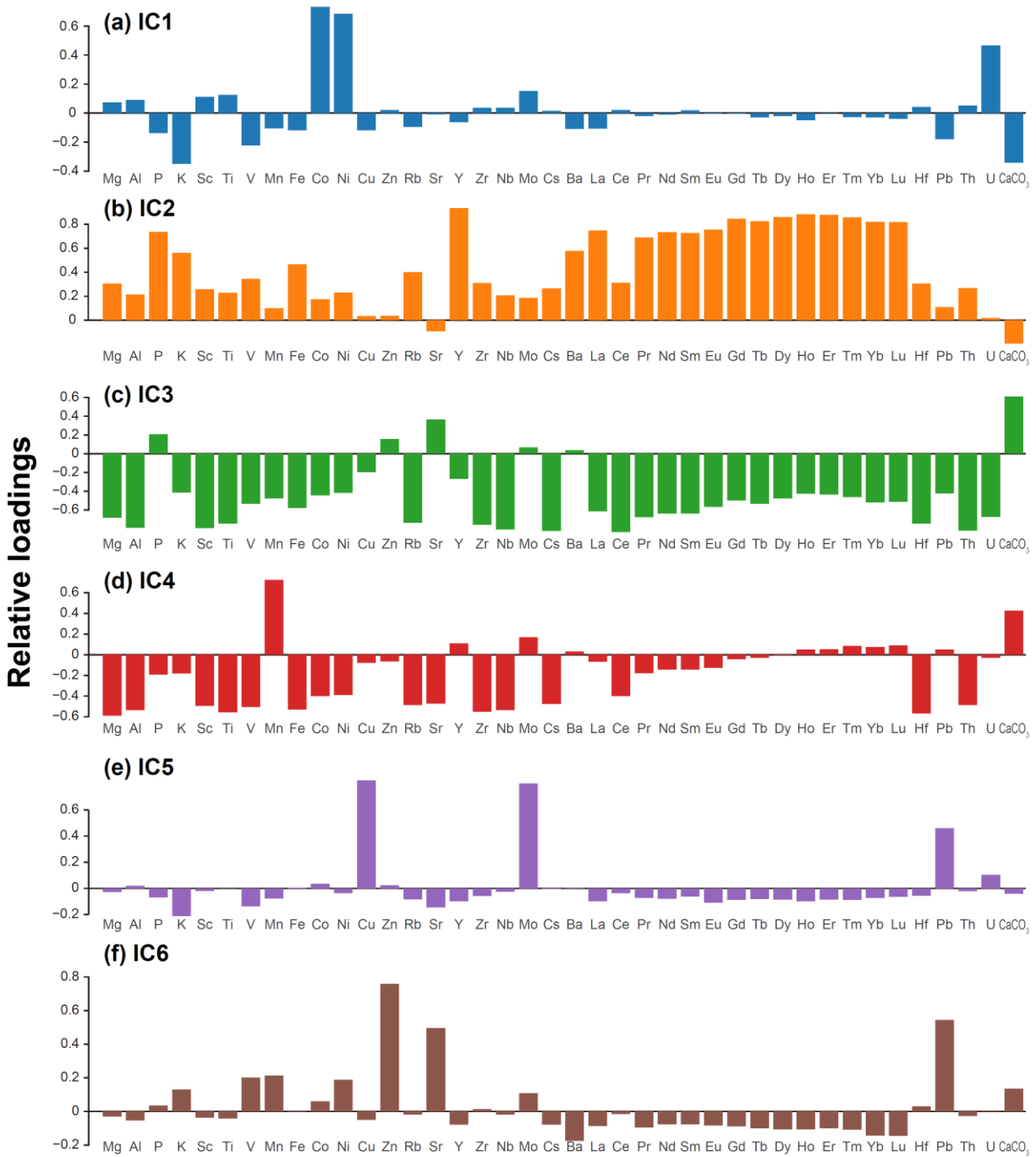
294

295 In this study, we implemented ICA on a dataset consisting of 40 elements and 248
 296 samples from Site 762C. The relative IC loadings are presented in Figure 4. It should be noted
 297 that the numbering of ICs is arbitrary and commutative and does not indicate their mutual
 298 importance. The scatterplots depicting the relationships between ICs indicate clear orthogonality
 299 among the six components, demonstrating their statistical independence from one another
 300 (Supporting Figure S4). The validity of extracting six ICs is discussed on Supporting Text S1 in
 detail. Our subsequent discussion is based on the results corresponding to the six ICs (Figure 4).

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Figure 4. IC loadings of major and trace elements obtained from the Site 762C dataset when the number of ICs = 6.

307

308 **5 Discussion**

309 5.1 Revision of the Age Model

310 Although previous studies have suggested that Site 762C sediments contain multiple
311 hyperthermal horizons (Shamrock et al., 2012; Thomas, 1992; Xu et al., 2021), the precise
312 identification of hyperthermal events has not been achieved owing to the limited temporal
313 resolution of previous bulk $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ records. Thus, the age model of Site 762C must be
314 further considered.

315 In this study, we identified the hyperthermal horizons of Site 762C using the newly
316 obtained bulk carbonate $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ records in combination with the age constraint by
317 magnetostratigraphy and biostratigraphy of Site 762C (Shamrock & Watkins, 2012; Shamrock et
318 al., 2012; Figure 2a) and the well-established compilation of benthic foraminifera $\delta^{13}\text{C}$ records
319 “CENOGRID” (Westerhold et al., 2018, 2020; Figure 2b). Consequently, we identified at least
320 five hyperthermals (PETM, H2, I1, J, and ETM3) in the studied section of Site 762C. We
321 described the base of identifying each hyperthermal horizon on Supporting Text S2. As previous
322 age models of Site 762C did not include the horizons of I1 and J events (Shamrock et al., 2012;
323 Thomas, 1992; Xu et al., 2021) and are based on not up-to-date magnetostratigraphic age, we
324 updated the age model of Site 762C using newly identified hyperthermal horizons and revised
325 magneto- and biostratigraphic ages by “Geologic Time Scale 2020 (GTS2020; Speijer et al.,
326 2020)” and Westerhold et al. (2020). The updated age-tie points are presented in Supporting
327 Table S5.

328 Previous studies inferred that the Middle Miocene Climate Optimum (MECO) horizon
329 exists in Cores 12X and 13X (284.0–265.0 mbsf), the uppermost part of the studied section of
330 Site 762C, based on the nannofossil assemblages (Shamrock et al., 2012; Shamrock & Watkins,
331 2012). The MECO was a one-million-year warming event characterized by a positive anomaly in
332 $\delta^{13}\text{C}$ and a negative anomaly in $\delta^{18}\text{O}$ (e.g., Henehan et al., 2020; Sluijs et al., 2013). However,
333 such excursions were not clearly observed in our bulk $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ records; thus, we cannot
334 identify the chemostratigraphic evidence of the MECO horizon (Figure 2). Therefore, we
335 determined that the age-tie points are based on magneto- and biostratigraphic ages without $\delta^{13}\text{C}$
336 chemostratigraphy during the middle Eocene intervals (Speijer et al., 2020; Westerhold et al.,
337 2020)

338 Based on the age determination in this study, the depositional ages of studied samples
339 from Site 762C were estimated to be from 57.61 Ma (Late Paleocene: Thanetian) to 39.06 Ma
340 (Middle Eocene: Bartonian; Figure 5). The sedimentation rate at Site 762C was calculated to be
341 0.6–44.6 m/Ma (Supporting Figure S8: Table S5). The extremely high sedimentation rate at Core
342 29X (77.9 m/Ma; 439.26–424.45 mbsf) could be due to the presence of voids in the core
343 (424.50–424.15 mbsf; Core 29X-2 115–150 cm) and core fragmentation (Haq et al., 1990).

344

345 5.2 Long-term Variation of Bulk Carbonate $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ Records

346 The bulk carbonate $\delta^{13}\text{C}$ of sediments reflects those of dissolved inorganic carbon in
347 seawater incorporated into the CaCO_3 (e.g., Luciani et al., 2016). As the studied samples from
348 Site 762C are nannofossil chalk consisting of the coccoliths of calcareous phytoplankton (Haq
349 et al., 1990), the bulk carbonate $\delta^{13}\text{C}$ record of Site 762C can be interpreted as the $\delta^{13}\text{C}$ of near-
350 surface seawater.

351 A comparison of the long-term trends of the bulk carbonate $\delta^{13}\text{C}$ records of Site 762C
352 with the global compilation of $\delta^{13}\text{C}$ records of benthic foraminifera (i.e., $\delta^{13}\text{C}$ of deep seawater;
353 Westerhold et al., 2020) revealed that the bulk carbonate $\delta^{13}\text{C}$ records of Site 762C were $\sim 1\text{‰}$
354 higher than those of benthic $\delta^{13}\text{C}$ records throughout the studied period (Late Paleocene–Middle
355 Eocene; Figure 5a). This feature suggests that the ocean surface layer exhibits higher $\delta^{13}\text{C}$ than
356 the deeper layers, which may indicate that deep seawater contains a larger amount of
357 isotopically light carbon derived from the decomposition of organic matter compared to near-
358 surface seawater (e.g., Ravizza & Zachos, 2014).

359 The long-term decrease of bulk $\delta^{13}\text{C}$ records of Site 762C from ~ 57.5 Ma ($\sim 3.3\text{‰}$) to
360 ~ 53.0 Ma ($\sim 0.0\text{‰}$), as seen in CENOGRID benthic $\delta^{13}\text{C}$ records, could be interpreted as the
361 long-term increase in the net release of isotopically light carbon to the deep ocean (e.g., Kirtland
362 Turner et al., 2014; Komar et al., 2013). Among several explanations for the $\delta^{13}\text{C}$ decrease
363 proposed by previous studies, one of the leading hypotheses is the continuous release of carbon
364 due to the decomposition of organic matter (e.g., biogenic CH_4 , permafrost, and peat) triggered
365 by the orbital pacing (e.g., Barnet et al., 2019; Kirtland Turner et al., 2014; Komer et al., 2013).
366 Our data indicate that the $\delta^{13}\text{C}$ decrease toward ~ 53 Ma is also a common phenomenon on the
367 surface of the eastern Indian Ocean.

368 The $\sim 1\text{‰}$ increase of bulk carbonate $\delta^{13}\text{C}$ of Site 762C during the magnetic polarity
369 Chron C23n (52–51 Ma) is also commonly observed in the global benthic $\delta^{13}\text{C}$ data (Figure 5a;
370 Westerhold et al., 2020), although there is ~ 400 kyr difference between them. The timing lag in
371 the rise of $\delta^{13}\text{C}$ of Site 762C during C23n could be attributed to the age uncertainty and poor
372 core recovery in Core 19X, corresponding to C19n (64.8%; Haq et al., 1990). To date, several
373 debatable hypotheses, including increased carbon burial flux, changes in oceanographic
374 circulation patterns, and active volcanism, attempt to explain the rise of $\delta^{13}\text{C}$ during C19n
375 (Anagnostou et al., 2020; Westerhold et al., 2018). A global-scale upheaval of the terrestrial
376 environment (e.g., temperature, precipitation rate; Hyland et al., 2017; Hyland & Sheldon,
377 2013) and tectonic events, including the change of the orientation of plate motion and
378 generation of new subduction zone (e.g., Ishizuka et al., 2011; O'Connor et al., 2013; Whittaker
379 et al., 2007) have been reported during Chron C23. These tectonic or environmental changes
380 could contribute to changes in global carbon cycle involving the rise of seawater $\delta^{13}\text{C}$.

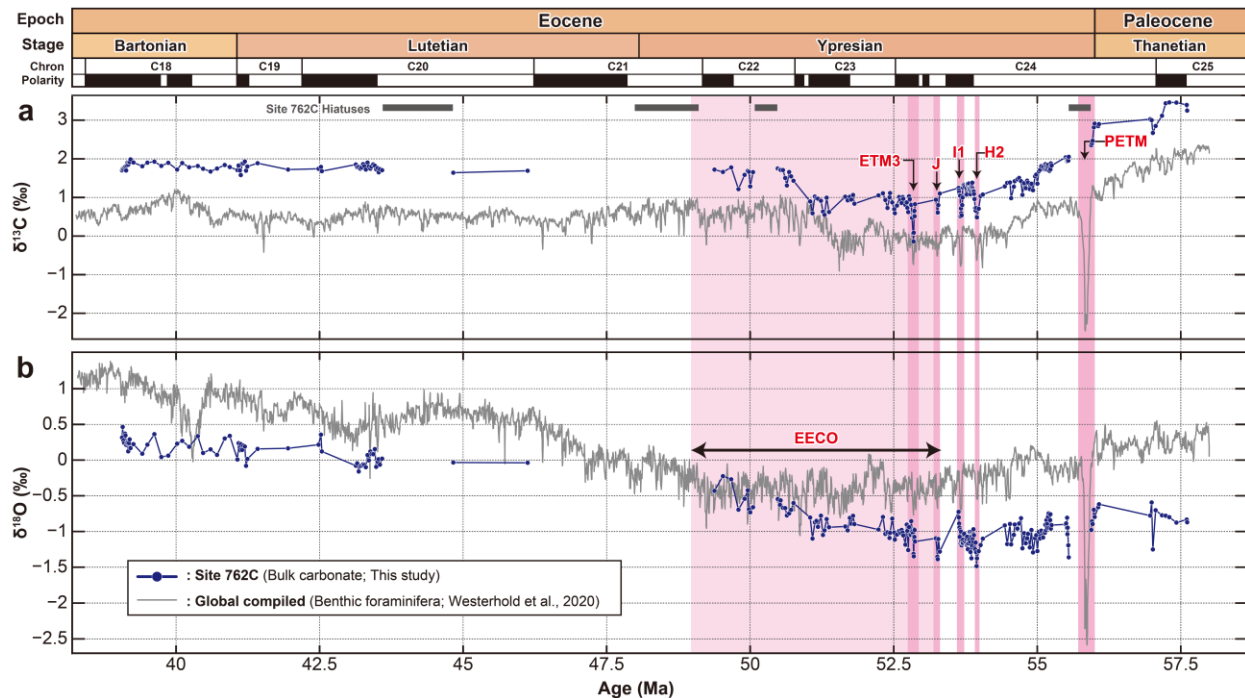
381 During 50–41 Ma, the $\delta^{13}\text{C}$ record did not show long-term changes, indicating the
382 absence of long-term changes in the carbon cycle. While previous studies reported the increase
383 in $\delta^{13}\text{C}$ during 40.5–39.5 Ma known as MECO (Henehan et al., 2020; Sluijs et al., 2013;
384 Westerhold et al., 2020), our Site 762C cores did not show such increase in $\delta^{13}\text{C}$ record. This
385 may be due to the age uncertainty due to the coring gap or hiatuses.

386 The bulk carbonate $\delta^{18}\text{O}$ of Site 762C seems to indicate a general SST trend because the
387 core consists mainly of the calcareous nannofossil, although the $\delta^{18}\text{O}$ value might be affected by
388 diagenetic processes or the contribution of other components. The long-term variation of bulk
389 $\delta^{18}\text{O}$ at Site 762C shows a gradual decrease from the late Paleocene ($\sim -0.7\text{‰}$) to ~ 52.5 Ma ($\sim -$
390 1.3‰), suggesting the long-term rise of SST toward the EECO, as previously reported (Figure
391 5b; Shamrock et al., 2012). After the peak of EECO, the bulk $\delta^{18}\text{O}$ of Site 762C increases until
392 ~ 47 Ma ($\sim 0.0\text{‰}$), which can be interpreted as the cooling of SST, corresponding to the Mid-
393 Eocene Cooling. From 47 Ma to 38 Ma, the bulk $\delta^{18}\text{O}$ of Site 762C fluctuates between 0‰ and
394 0.5‰ , suggesting no significant variation of SST in this period, indicating that the studied
395 samples from Site 762C did not record the warm period of MECO (~ 40.5 Ma) indicated in
396 globally compiled benthic record (Henehan et al., 2020; Sluijs et al., 2013; Westerhold et al.,
397 2020).

398 Contrary to the $\delta^{13}\text{C}$, the trend of bulk $\delta^{18}\text{O}$ of Site 762C is different from those of
399 benthic $\delta^{18}\text{O}$ records (Westerhold et al., 2020). During the EECO period, the gap between bulk
400 $\delta^{18}\text{O}$ of Site 762C (i.e., SST at Site 762C) and benthic $\delta^{18}\text{O}$ (i.e., a global signature of deep
401 seawater temperature) narrows toward ~ 50 Ma (Figure 5b). This feature is also observed in the
402 Southern Ocean (ODP Sites 690 and 738; Auderset et al., 2022), Atlantic Ocean (DSDP 550;
403 Charisi & Schmitz, 1996), and the Pacific Ocean (ODP Site 1209; Dutton et al., 2005).
404 Assuming that benthic $\delta^{18}\text{O}$ is globally homogeneous (Westerhold et al., 2020), the narrowing
405 gap between bulk $\delta^{18}\text{O}$ of Site 762C and globally compiled benthic $\delta^{18}\text{O}$ implies that the
406 vertical gradient of temperature and density in the water column decreased during the EECO.

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410 **Figure 5.** Age profiles of bulk $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ of Site 762C. Grey lines indicate global compiled
 411 $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ of benthic foraminifera (Westerhold et al., 2020). Polarity chron is also shown on
 412 the top. Vertical red bars indicate hyperthermals and EECO.

413

5.3 Variation of Bulk $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ Records during Hyperthermals

414

415 Compared with other marine and terrestrial records of the bulk carbonate $\delta^{13}\text{C}$ record of
 416 hyperthermal events (excluding the PETM), the bulk carbonate $\delta^{13}\text{C}$ at Site 762C exhibited
 417 generally consistent behavior and magnitudes with records from the Atlantic, Pacific, and Indian
 418 Oceans. The magnitude of the CIE during the H2 event at Site 762C ($\sim 0.6\text{‰}$; Figure 3b) is
 419 comparable to the bulk carbonate data from northeastern Atlantic ($\sim 0.4\text{‰}$: DSDP Site 550;
 420 Cramer et al., 2003), northwestern Atlantic ($\sim 0.5\text{‰}$: Blake Nose - ODP Sites 1051; Cramer et al.,
 421 2009; Westerhold et al., 2017), and the southern Indian Ocean ($\sim 0.5\text{‰}$: Kerguelen Plateau - ODP
 422 Site 738C; Yasukawa et al., 2017). However, records from the central Pacific ($\sim 0.3\text{‰}$: DSDP
 423 577; Cramer et al., 2003; Luciani et al., 2016) and the southern Pacific ($\sim 0.2\text{‰}$: Mead Stream,
 424 Dee Stream; Nicolo et al., 2007) exhibit smaller CIEs compared to that of Site 762C.

425

426 Regarding the I1 Events, the CIE at Site 762C ($\sim 0.7\text{‰}$) is comparable to other regions'
 427 data. Similar CIE magnitudes have been reported from the Atlantic ($\sim 0.7\text{‰}$: DSDP 550, ODP
 428 Sites 1051, 1262, 1263; Cramer et al., 2003; Westerhold et al., 2017), Pacific ($0.5\text{--}0.6\text{‰}$: DSDP
 429 577, Mead Stream, Dee Stream; Cramer et al., 2003; Nicolo et al., 2007), and southern Indian
 430 Oceans ($\sim 0.6\text{‰}$: ODP Sites 752 and 738C; Yasukawa et al., 2017). In Site 762C, the CIE
 431 magnitude during the J event ($\sim 0.5\text{‰}$) is similar to that of the Atlantic record ($\sim 0.5\text{‰}$: DSDP
 432 550, ODP Sites 1262, 1263; Cramer et al., 2003; Westerhold et al., 2017) and the Pacific record
 ($\sim 0.4\text{‰}$: DSDP 577; Luciani et al., 2016). The largest CIE ($\sim 0.9\text{‰}$) was observed during the

433 ETM3 horizon in Core 25X of Site 762C sediments. This magnitude is similar to that of the
434 Atlantic (0.6–1.0‰: DSDP 550, ODP Sites 1262, 1263; Cramer et al., 2003; Westerhold et al.,
435 2017), central Pacific (0.7–1.0‰: DSDP 577, ODP Site 1215; Leon-Rodriguez & Dickens, 2010;
436 Luciani et al., 2016), and southern Indian Ocean records (~0.9‰: Site 752; Yasukawa et al.,
437 2017). As the magnitude of CIEs at Site 762C is comparable to that of other regions' data, we
438 can consider that the geochemical records of the modest Eocene hyperthermals at Site 762C are
439 well preserved without hiatus or truncation.

440 5.4 Interpretation of ICs Extracted from Bulk Chemical Composition

441 5.4.1 IC3: Dilution between Biogenic CaCO₃ and Detrital Components

442 The IC3 shows positive CaCO₃ and Sr loadings and negative loadings of most other
443 elements (Figure 4c). The age profile of the IC3 score is similar to that of CaCO₃ abundances
444 (Supporting Figure S9). These features indicate that the positive side of IC3 represents the
445 biogenic CaCO₃ component that constitutes the main component of the Site 762C sediment. As
446 Sr is an alkaline earth metal that can be easily incorporated into biogenic carbonates, Sr is known
447 to coexist with Ca in carbonate sediments (e.g., Plank & Langmuir, 1998; Rudnick & Gao, 2014).
448 Conversely, elements showing distinct negative loadings (e.g., Mg, Al, Ti, Rb, Zr, Nb, Cs, Hf,
449 Th, and light rare earth elements [LREEs]), which are commonly contained in aluminosilicates,
450 indicate the contribution of detrital components. Therefore, IC3 appears to indicate a mixing
451 relationship between biogenic CaCO₃ and detrital component as one of the major structures in
452 our dataset.

453 5.4.2 IC2: Biogenic Calcium Phosphate

454 The IC2 shows positive loadings of P and REY except for Ce (Figure 4b). This feature
455 indicates that the IC2 extracts a biogenic calcium phosphate (BCP) component in the sediment.
456 Biogenic calcium phosphate, a common component in marine sediments, is contained in bones,
457 teeth, and scales of marine vertebrates. As BCP effectively adsorbs REE and Y from ambient
458 seawater, REE and Y are highly concentrated in BCP in marine sediments (Kashiwabara et al.,
459 2014; Takaya et al., 2018; Toyoda & Tokonami, 1990). Indeed, the Al-normalized P and Y
460 abundances of Site 762C show identical age profiles and have a strong positive correlation ($r =$
461 0.91; Supporting Figure S9), suggesting that the excess P and Y (or REE) relative to detrital
462 materials can be explained by BCP contribution. Therefore, the significant positive loadings of P
463 and REE in IC2 may reflect the influence of BCP on the bulk sediment composition. In addition,
464 the small Ce loading and large loadings of Y and heavy rare earth elements (HREEs) on the
465 positive side of IC2 loadings appear to be a similar pattern to the shale-normalized REY
466 composition of seawater (Alibo & Nozaki, 1999). Therefore, the BCP components in Site 762C
467 sediment are considered to contain REE and Y of seawater origin. Notably, in the age profiles of
468 IC scores (Figure 6; Supporting Figure S9), IC2 does not show a mirror profile of the CaCO₃ and
469 IC3. Therefore, the variation of IC2 scores does not simply reflect a simple dilution of BCP
470 abundance by biogenic CaCO₃. Instead, it is controlled by various environmental factors that
471 influence the accumulation rate of BCP at Site 762C.

472 5.4.3 IC4: Diagenetic Signature on Carbonate

473 The IC4 demonstrates positive loadings of Mn and CaCO₃ (Figure 4d). The abundance of
474 Mn has been utilized as an indicator of diagenetic processes because its solubility is affected by
475 the redox conditions in the sediment. In the oxic environment, Mn occurs as MnO₂ in sediments;
476 under reducing conditions caused by the consumption of dissolved oxygen via organic matter
477 decomposition, Mn dissolves in pore water as Mn²⁺ (e.g., Burdige & Gieskies, 1983; Calvert &
478 Pederson, 1993). In carbonate sediments, dissolved Mn²⁺ can be incorporated into or precipitate
479 as (Mn, Ca)CO₃ on the surface of biogenic CaCO₃ shells (e.g., Barras et al., 2018; Boyle, 1983;
480 Thomson et al., 1986, 1993; Pälke et al., 2014). Based on these behaviors of Mn in marine
481 sediments and the positive correlation between Mn and CaCO₃ in the IC4, the positive loadings
482 of IC4 can be interpreted as the incorporation of Mn by CaCO₃ through the diagenetic process at
483 Site 762C. Overall, we interpreted that IC4 represents the diagenetic signature involving redox
484 changes in the Site 762C sediments.

485 5.4.5 IC1, IC5, IC6: Redox-related Behaviors of Metal Elements

486 IC1, IC5, and IC6 are characterized by specific metals, including Co, Ni, U, Cu, Mo, Pb,
487 Zn, and Sr (Figure 4a, e, f). The age profiles of IC1, IC5, and IC6 scores exhibit spike-like
488 excursions at particular horizons (e.g., hyperthermals), indicating that the
489 concentration/depletion of these metal elements occurred at those horizons (Figure 6b, c;
490 Supporting Figure S10). These features suggest that IC1, IC5, and IC6 may indicate redox-
491 related signatures in Site 762C sediments because these metals exhibit specific abundances in
492 sediments corresponding to the ambient redox state. The elements characterizing IC1, IC5, and
493 IC6 (except Sr) are categorized as the “redox-sensitive elements” that exhibit characteristic
494 reconcentration patterns in sediments due to the changes of valences corresponding to the redox
495 state in the sediment pore water. Therefore, redox-sensitive elements are concentrated or
496 depleted at the boundary between oxic and suboxic conditions in the sediments and utilized as
497 the indicator of the redox state in the sediments (e.g., Calvert & Pederson, 1993; Thomson et al.,
498 1993).

499 Various behaviors of such redox-sensitive metals have been proposed to explain their
500 concentration/depletion in marine sediments, including remobilization and precipitation of
501 metals due to the change of valence corresponding to the redox state of sediment pore water (e.g.,
502 Calvert & Pederson, 1993) and co-precipitation with authigenic pyrite (Algeo & Maynard, 2004;
503 Huerta-Diez & Morse, 1992). The separation between IC1, IC5, and IC6 may reflect differences
504 in the concentration/depletion processes described above.

505 IC1 is distinguished by positive Co, Ni, and U loadings, and its score indicates positive
506 excursions at horizons near Hiatus D, I1, and J (Figure 6b, c). The age profile of the IC1 score
507 closely matches that of the Co/Al and Ni/Al profiles, except for the I1 horizon (Supporting
508 Figure S10b, c), strongly suggesting that IC1 reflects the variation of Co and Ni in the sediments.
509 The presence of authigenic pyrite and the positive loading of U (Figure 4a), indicating a reduced
510 porewater environment, suggests that the elevation of Co and Ni abundances, as indicated by the
511 IC1 peaks, may be associated with the precipitation of sulfides (e.g., pyrite) (Huerta-Diez &
512 Morse, 1992; Swanner et al., 2019). As the uptake of Co and Ni into pyrite occurs more slowly
513 compared to other redox-sensitive elements (Algeo & Maynard, 2004), the peaks of IC1,
514 corresponding to those of Co and Ni, may be attributed to the kinetic differences from other
515 redox-sensitive elements (e.g., Cu, Mo, and Pb) (Figure 6c).

516 Positive loadings of Cu, Mo, and Pb characterize IC5 (Figure 4e). Its score exhibits
517 positive excursions at the horizons near Hiatus D, ~55 Ma, H2, and I1 (Figure 6b, c). Conversely,
518 IC6 is characterized by the positive loadings of Zn, Sr, and Pb (Figure 4f), and its scores show
519 positive excursions at horizons near Hiatus D, I1, and ETM3. In addition, the IC6 score also
520 shows two excursions during 41–39 Ma (Figure 6a). Based on the element assemblages of IC5
521 and IC6 and the behavior of their scores, the IC5 and IC6 may reflect the remobilization and
522 reconcentration of redox-sensitive elements due to the change of redox (or pH) conditions in the
523 sediments.

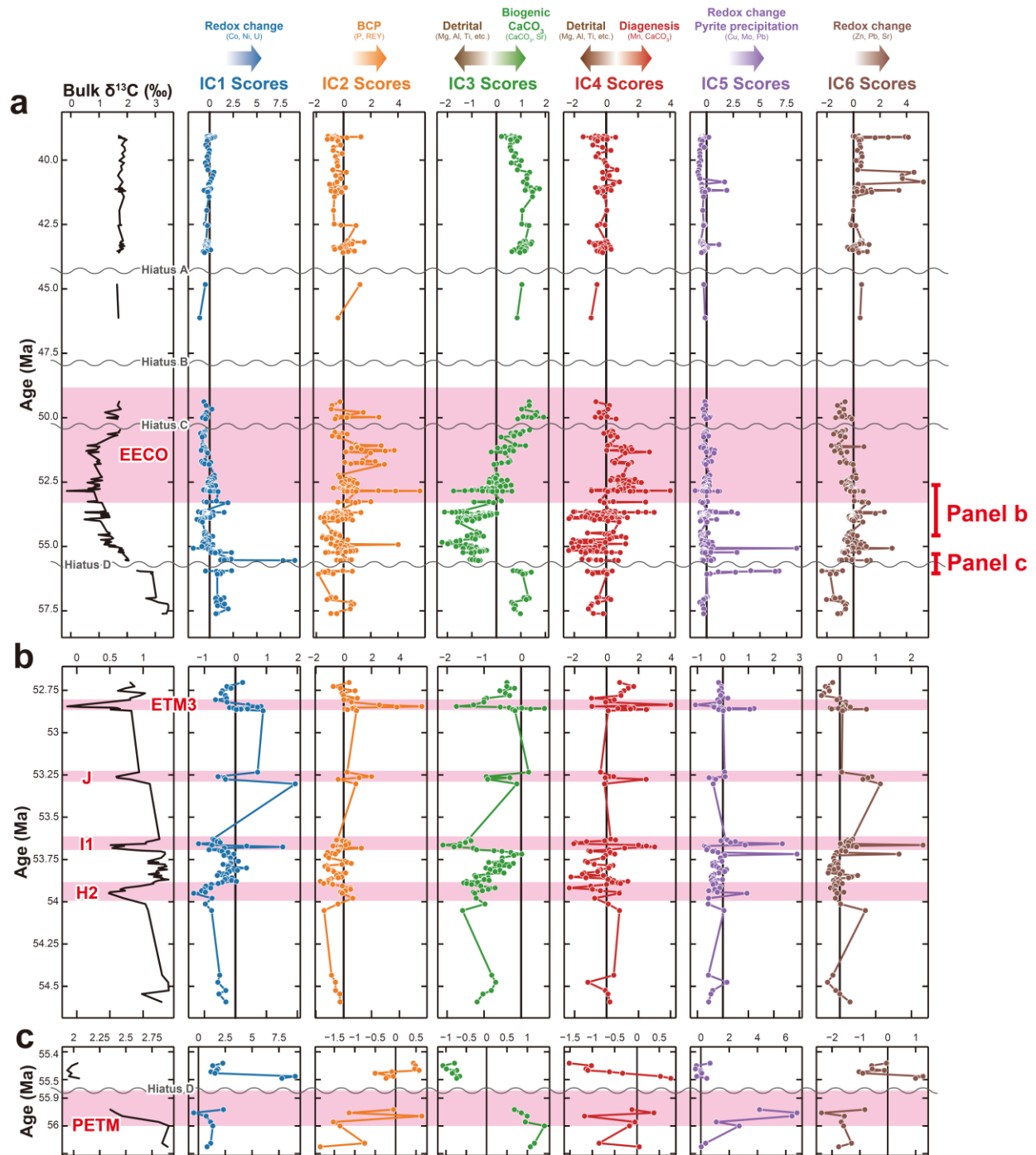
524 The enrichment of Co, Ni, Cu, Mo, Zn, and Pb in marine sediments is also caused by the
525 co-precipitation of authigenic pyrite under reductive conditions, although the uptake speed varies
526 by element (e.g., Algeo & Maynard, 2004; Huerta-Diez & Morse, 1992). Indeed, at the horizons
527 showing positive peaks of IC1, IC5, and IC6, we observed authigenic pyrite grains as suggested
528 by the onboard visual core description of Site 762C sediments (Haq et al., 1990). Therefore, it is
529 also possible that the excursions of these three ICs reflect the occurrence of pyrite precipitation.
530 Although pyrite precipitation or redox-related valence changes of metal elements are difficult to
531 distinguish from the bulk chemical composition, the variation and excursions of IC1, IC5, and
532 IC6 scores can be interpreted as the redox (and/or pH) changes in the pore water.

533 Based on the above discussion, we interpreted that IC1, IC5, and IC6 reflect the post-
534 depositional processes related to the changes in the sediments' redox (or pH) conditions.
535 Therefore, in the following sections, we consider these three ICs as an indicator of the
536 geochemical condition of pore water.

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Figure 6. Age profiles of extracted ICs from the bulk chemical composition and bulk $\delta^{13}\text{C}$ of ODP Site 762C. (a) Whole studied sections. (b) Sections covering multiple hyperthermals (H2, I1, J, and ETM3). (c) A section covering the PETM. Grey lines in each panel indicate bulk carbonate $\delta^{13}\text{C}$ records. Horizontal pink bars in panel (a) indicate EECO, and in panels (b) and (c) indicate hyperthermals. Grey wavy lines are hiatuses indicated by Shamrock et al. (2012).

546

547 5.5 Geochemical Implication of Each IC for Long-Term Trends

548 The IC2 and IC3 show long-term variations throughout the studied section, while
549 the remaining ICs only show transient spike-like excursions. In this section, we discuss the
550 long-term trends of IC2 and IC3.

551 5.5.1 Change of CaCO₃ Contribution Associated with Long-Term Climate Changes

552 IC3, which reflects the mixing relationship between biogenic CaCO₃ and detrital
553 component, exhibited significant variations on the long timescale. Considering that the
554 paleodepth of Site 762C did not show large variations throughout the Cenozoic (1000–1200 m;
555 Haq et al., 1992; Jones & Wonders, 1992), the variation of IC3 scores was not largely affected
556 by changes in paleodepth. In addition, because the location of Site 762C is a passive margin
557 without a large change in the paleogeography (especially the distance to the continent)
558 throughout the studied period (e.g., Scotese, 2001; Figure 1a), the secular changes in tectonic
559 settings can be negligible for changes in the supply of detrital components to Site 762C.
560 Therefore, we considered that the variation in IC3 scores was not mainly caused by tectonic
561 factors but by climatic or paleoceanographic changes.

562 Over the long-term, the IC3 score decreased between 56 and 55 Ma, followed by a
563 gradual increase during 53–50 Ma (Figure 6a). Then, the IC3 scores kept a high value (~ 1)
564 during 50–42 Ma and showed a slight decline towards ~ 39 Ma. In addition, scatter diagrams
565 show that IC3 scores show positive correlations with bulk $\delta^{18}\text{O}$ ($r = 0.70$; Supporting Figure
566 S11d). This indicates that the relative contribution between biogenic CaCO₃ and detrital
567 components at Site 762C fluctuated in accordance with long-term climate changes in the
568 southeastern Indian Ocean, such as the EECO and subsequent cooling.

569 One interpretation of the long-term variation in IC3 scores is the fluctuation in the
570 depositional rate of CaCO₃ associated with the early Paleogene climate. Paleo-CCD estimation
571 from the Pacific and Indian Oceans suggest that shallowing of CCD occurred under high
572 (~1,100–3,000 ppmv) atmospheric $p\text{CO}_2$ and low pH seawater during the EECO (Anagnostou et
573 al., 2016; Pálike et al., 2012; Slotnick et al., 2015). While it is unlikely that the CCD became
574 shallower than the paleodepth of Site 762C during the study period, low IC3 scores in the
575 earliest Eocene could be attributed to a reduced flux of carbonate precipitation to the seafloor
576 because of changes in the paleoceanographic environment, such as vertical mixing, pH levels,
577 and dissolved oxygen concentrations, in the water column, during this period (Luciani et al.,
578 2017). Curiously, the increase in bulk carbonate $\delta^{18}\text{O}$ and IC3 score precedes (by ~2.5 Myr) that
579 of global-compiled benthic $\delta^{18}\text{O}$ record noted as the end of EECO (see Section 5.2., Figure 5b;
580 Westerhold et al., 2018, 2020). Given that previous studies have reported a reduction in the
581 difference between benthic and planktonic $\delta^{18}\text{O}$ during the EECO and have interpreted it as a
582 collapse of oceanic stratification (Auderset et al., 2022; Charisi & Schmitz, 1996; Dutton et al.,
583 2005), increases in bulk carbonate $\delta^{18}\text{O}$ and IC3 score at Site 762C may suggest that the
584 collapse of the vertical structure of the water column (e.g., temperature, pH, and salinity)
585 occurred at the Exmouth Plateau and affected the precipitation flux of biogenic CaCO₃. Indeed,
586 micropaleontological studies have shown that the turnover of the nannofossil and planktonic

587 foraminifera assemblages occurred at the surface ocean associated with major climatic changes,
 588 including the PETM and EECO (Luciani et al., 2016, 2017; Shamrock & Watkins, 2012),
 589 suggesting that climate shift impacted the sea-surface ecosystems, especially carbonate shell-
 590 forming organisms.

591 The increase in IC3 score may also reflect a decrease in the influx of terrigenous
 592 material. Under the warm climate and enhanced hydrological cycle during the early Eocene, it
 593 has been suggested that the supply of terrigenous materials into the ocean was accelerated (e.g.,
 594 Dallanave et al., 2015; Hyland and Sheldon, 2013). Therefore, the decrease in IC3 score during
 595 ~56–55 Ma can be interpreted as an increase in terrigenous input relative to CaCO₃. Indeed, the
 596 upper continental crust (UCC)-normalized REE pattern, which is an indicator of detrital
 597 contribution in marine sediments (e.g., Murray et al., 1991; Plank & Langmuir, 1998), also
 598 indicates an increase in detrital components in Site 762C sediments during the warm climate of
 599 Earliest Eocene. We used Ce/Ce* as an index of Ce negative anomalies (Ce/Ce*) defined as:

$$600 \quad \frac{\text{Ce}}{\text{Ce}^*} = \frac{\text{Ce}_N}{\sqrt{\text{La}_N \text{Pr}_N}} \quad (3)$$

601 where the subscript “N” indicates the UCC-normalized value (Rudnick & Gao, 2014), and
 602 Y_N/Ho_N as an index of Y positive anomalies. Both indices showed clear correlations with IC3
 603 scores (Ce/Ce*: $r = -0.78$, Y_N/Ho_N: $r = 0.81$; Supporting Figure S11e, f). The Ce/Ce* and
 604 Y_N/Ho_N also showed correlations with δ¹⁸O ($r = -0.46$, 0.73 , respectively). The negative Ce and
 605 positive Y anomalies in UCC-normalized REY patterns indicate the signature of seawater (e.g.,
 606 Alibo & Nozaki, 1999; Bolhar et al., 2004; De Baar et al., 1985), while the smaller anomalies or
 607 relatively flat REE patterns indicate a high contribution of the terrigenous material. Therefore,
 608 the correlations between δ¹⁸O and Ce/Ce*, as well as Y_N/Ho_N in sediments from Site 762C,
 609 indicate that there was an increase in terrigenous input during the warm period of the earliest
 610 Eocene (approximately 56–55 Ma) followed by a decrease as the cooling proceeded toward the
 611 middle Eocene.

612 Considering that climatic and hydrological patterns might have significantly changed in
 613 the EECO, the low IC3 score during the earliest Eocene, followed by an increase in the score,
 614 can be interpreted as a decrease in terrigenous (i.e., riverine and aeolian) input to the
 615 depositional area associated with the cooling trend toward the middle Eocene. As the major
 616 source of aeolian dust at the Exmouth Plateau during the Quaternary is considered to be inland
 617 arid areas of the Australian Continent (e.g., Gascoyne plain) (Hasse & McTainsh, 2003), the
 618 aeolian dust supply to the early Eocene sediments at Site 762C could also be derived from
 619 Australia. However, in contrast to the arid environment in modern Australia, the early–middle
 620 Eocene terrestrial environment of the Australian continent was considered to be warmer with a
 621 more active hydrological cycle than that of the present (Reichgelt et al., 2022). If we consider
 622 that the aridification of the Australian continent progressed in relation to the early–middle
 623 Eocene cooling, it would be expected that the dust influx to the Exmouth Plateau would
 624 increase, leading to an increased contribution of detrital material in the sediments. However, the
 625 time-series profile of the IC3 score suggests the opposite behavior (Figure 6a); the dust supply
 626 into the Exmouth Plateau could be diminished by cooling toward the middle Eocene. A possible
 627 explanation is that the influence of the aeolian dust flux on the variation in IC3 was minor. In
 628 contrast, riverine detrital material could have affected the IC3 scores. It has been reported that

629 the contribution of terrigenous fluvial input into the deep-sea sediments around the continent
630 has increased during the earliest Eocene and EECO (Burton et al., 2023). Thus, enhanced
631 denudation, drainage, and chemical weathering in the warm and humid terrestrial environment
632 in the early Eocene may have caused an increase in riverine input of terrigenous materials to the
633 coastal area and, thus, low values in the IC3 score at Site 762C.

634 Overall, the long-term variation in IC3 scores indicates changes in terrigenous
635 (especially fluvial) input or marine ecosystems (especially the abundance of carbonate-bearing
636 organisms) associated with climatic changes, namely the EECO and subsequent cooling.

637 5.5.2 Long-Term Biogenic Calcium Phosphate Abundances

638 The IC2 score, which reflects the contribution of BCP, shows a tendency to increase
639 during the EECO period, particularly in 51–50 Ma (Figure 6a). Although scatter diagrams do
640 not exhibit clear negative correlations between IC2 and $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ ($r = -0.39, -0.21$,
641 respectively; Supporting Figure S11a, b), samples from the EECO (including ETM3 horizon)
642 show high scores. This feature suggests that the high-level consumers, typically vertebrates such
643 as fish, in the marine ecosystems prospered under the warm environment of EECO at Site 762C.
644 This finding is consistent with previous studies demonstrating an increase in fish population
645 during warm environments in the early Eocene based on ichthyolith accumulation rates in
646 pelagic sediments (Britten & Sibert, 2020; Sibert et al., 2016). Our study indicates that such
647 proliferation of marine vertebrates coherently occurred in relatively coastal regions.

648 5.6 Geochemical Implication of Each IC on Hyperthermals

649 5.6.1 Change of CaCO_3 Contribution Associated with Hyperthermals

650 On a shorter ($\sim 10^5$ yr) timescale, IC3 scores exhibited several spike-like decreases,
651 typically at the I1, J, and ETM3 horizons (Figure 6b). These features indicate a decrease in the
652 contribution of biogenic CaCO_3 at multiple hyperthermal horizons at Site 762C. As it is well
653 known that ocean acidification and shallowing of CCD occurred during hyperthermal events
654 (e.g., Slotnick et al., 2015; Zachos et al., 2005, 2010), the decrease in IC3 scores associated with
655 carbon isotope excursions (CIEs) at multiple hyperthermal horizons may reflect the repetitive
656 shallowing of the CCD at Site 762C. Additionally, an increase in detrital input may also
657 contribute to a decrease in the IC3 score during hyperthermals. As it has been argued that the
658 weathering of terrestrial material increased during the hyperthermals (e.g., Ravizza et al., 2001;
659 Tanaka et al., 2022), we cannot exclude the possibility of an increase in detrital input into the
660 depositional area of the Site 762C.

661 In contrast to the record at Site 762C, a previous study that applied ICA to early Eocene
662 carbonate sediments from the southern Indian Ocean (ODP Sites 752 and 738C) did not show
663 clear negative excursions of IC scores reflecting biogenic CaCO_3 content at H2, I1, and ETM3,
664 despite the paleo-depth of both sites being close to that of Site 762C (~ 1000 – 1300 m; Yasukawa
665 et al., 2017). Therefore, the magnitude of the reduction in carbonate rain flux due to CCD
666 shallowing or the increase in detrital materials during hyperthermals could have been regionally
667 variable.

668 The H2 horizon at Site 762C did not exhibit a clear negative excursion of IC3 scores
669 and CaCO₃ abundances (Figure 6b; Supporting Figure S9b). The relatively minor variation at
670 the H2 horizon compared to that at the I1, J, and ETM3 horizons indicates that the magnitude of
671 CCD shallowing and the reduction of carbonate rain flux during the H2 event were smaller than
672 those during other events. This feature is consistent with the Atlantic record (Walvis Ridge;
673 Stap et al., 2009, 2010) and suggests that the magnitude of CCD shallowing could have been
674 variable between each hyperthermal.

675 5.6.2 Biogenic Calcium Phosphate Contribution Associated with Hyperthermals

676 In the sediments of Site 762C, the IC2 score repetitively showed spike-like increases at
677 the PETM (onset), H2, I1, J, and ETM3 horizons (Figure 6b, c). As IC2 primarily reflects the
678 contribution of BCP, the systematic increase in IC2 scores at hyperthermal horizons indicates an
679 increase in the contribution of BCP associated with the warming events. Therefore, the
680 proliferation of fish at Site 762C could be associated with hyperthermals.

681 There have been arguments about whether or not biological productivity was enhanced
682 during hyperthermals (e.g., Bains et al., 2000; Bralower, 2002; Gibbs et al., 2006; Ma et al.,
683 2014; Winguth et al., 2012; Yasukawa et al., 2017; Zhou et al., 2014). The increase in nutrient
684 input due to an increased weathering flux under the warm hyperthermal climate may have
685 triggered the proliferation of marine vertebrates at Site 762C. However, at Sites 752 and 738C,
686 there was no clear signature of BCP increase at H2, I1, or ETM3 (Yasukawa et al., 2017). Our
687 results indicate that the prosperity of marine vertebrates may have sensitively responded to
688 hyperthermals with significant regional variation, even within the Indian Ocean.

689 5.6.3 Post-Depositional Geochemical Processes Associated with Hyperthermals

690 The IC1, IC4, IC5, and IC6, which can be interpreted as post-depositional processes,
691 showed multiple spike-like excursions at hyperthermal horizons (Figure 6b, c). As previously
692 described, IC4 was interpreted as the diagenetic carbonate signature characterized by the
693 positive correlation between Mn and CaCO₃. The depletion and reconcentration of Mn have
694 been interpreted as diagenetic signatures associated with reduction and oxidation in the
695 sediments (e.g., Burdige & Gieskies, 1983), the fluctuations of IC4 score at hyperthermal
696 horizons can be interpreted as changes in the sediment redox condition.

697 As we discuss in Section 5.4.5., IC1, IC5, and IC6 were all characterized by redox-
698 sensitive elements, which suggests changes in the redox condition of sediment pore water
699 and/or bottom seawater. Still, they differed from IC4 because they have no clear correlation
700 with CaCO₃ (Figure 4). IC1, IC5, and IC6 exhibited sharp peaks nearby hyperthermal horizons,
701 and peaks tend to shift slightly between each IC (Figure 6b, c). At the horizon of the PETM-
702 onset, there was a positive excursion of IC5 at the onset of CIE and peaks of IC1 and IC6 above
703 the Hiatus D (Figure 6c). Therefore, we suggest that these ICs capture the enrichment and
704 dissolution of redox-sensitive elements in the PETM layer of Site 762C. At the horizon of I1,
705 IC1 peaks near the CIE level, and IC5 and IC6 peak above and below the CIE (Figure 6b). Such
706 obvious excursions of IC score peaks occurring nearby hyperthermals are also observed in the
707 H2, J, and ETM3 horizons, suggesting the occurrence of elemental enrichment and dissolution

708 systematically occurred at the hyperthermal events. However, there may be variations in the
709 magnitude of redox-change in each event.

710 The reductive horizons in Site 762C identified through ICA may indicate that the redox
711 state of the porewater and bottom water at the Exmouth Plateau in the Eastern Indian Ocean
712 fluctuated during hyperthermals. Regarding the PETM, previous studies have reported that the
713 enrichment and dissolution of redox-sensitive elements indicate that the redox state of the
714 bottom water became reductive (~suboxic) at several sites in the Atlantic and Indian Oceans
715 (Chun et al., 2010; Pälike et al., 2014). Our results are consistent with these previous studies,
716 suggesting that porewater or bottom-water redox changes occurred in the mid-latitude eastern
717 Indian Ocean. Furthermore, our data indicate novel insights into repetitive redox changes
718 associated with the modest, multiple hyperthermals after the PETM. In addition, Zhou et al.
719 (2014) suggested the occurrence of upper ocean deoxygenation during the PETM based on I/Ca
720 record of bulk carbonate sediments from each ocean basin, including Site 762C. Therefore, the
721 redox condition in hyperthermal horizons of Site 762C sediments is potentially linked to the
722 upper ocean dissolved oxygen concentration and primary production rate at the photic zone.

723 5.7. Insights into Decoding the Paleoenvironmental Changes in Hyperthermals with ICA

724 Our study sheds light on the complex interplay between transient global warmings (i.e.,
725 hyperthermals) and paleoceanographic responses. As we noted in Section 3.5., the ICs extracted
726 from the bulk chemical composition were statistically independent. In other words, each IC
727 reflected a signature imprinted in sediments that can be considered a distinct geochemical feature.
728 In reconstructing the paleoceanographic record at Site 762C, each IC can be interpreted as a
729 reflection of a distinct aspect of paleoenvironmental changes due to the hyperthermals. Under the
730 warm hyperthermal climate, increased chemical weathering and riverine drainage could have
731 occurred (e.g., Dickson et al., 2015; Pogge von Strandmann et al., 2021; Ravizza et al., 2001;
732 Tanaka et al., 2022). The increase in the relative contribution of the fluvial detrital component,
733 represented by positive excursions in IC3, can be interpreted as a result of these environmental
734 changes. Additionally, increased continental runoff may have also caused a significant input of
735 nutrients to the continental margin. Given that Site 762C is located on the northwestern margin
736 of offshore Australia, this increase in nutrient supply could have enhanced the marine primary
737 productivity around the site. The positive excursions of IC2, which indicate an increase of BCP
738 accumulation at hyperthermal horizons, suggest another vestige of the warming events from a
739 biological/ecological perspective.

740 The results of ICA also indicate the impact of hyperthermals on seawater chemistry.
741 During hyperthermals, at least locally/regionally, oceans are thought to have stratified due to
742 warming (e.g., Sluijs et al., 2009; Tripathi & Elderfield, 2005; Willard et al., 2019; Zachos et al.,
743 2003). Furthermore, although regional variations may have existed, bottom seawater is
744 considered to have become hypoxic–suboxic (e.g., Kaiho et al., 2006; Pälike et al., 2014;
745 Papadomanolaki et al., 2022; Zhou et al., 2016). Excessive sedimentation of organic matter due
746 to the increased nutrient supply from land and high productivity in the surface ocean may have
747 induced bottom-water hypoxia on the continental shelf owing to enhanced oxygen consumption
748 (Sluijs et al., 2014), as well as decreased dissolved oxygen due to high water temperatures
749 associated with hyperthermals. Such environmental changes during hyperthermals could have
750 resulted in changes in the redox state at the sediment–water interface and the uppermost

751 sediment. This process could have mobilized several redox-sensitive elements after deposition
752 and caused the pyrite precipitation in the reductive zone. Depending on the differences in the
753 kinetics and chemical affinity of each element, redistribution and enrichment of different sets of
754 elements might have occurred in the sediment. These distinct features were subsequently
755 separated as IC1, IC4, IC5, and IC6, which are mutually superimposed as statistically
756 independent data structures in the sediment. Overall, the result of ICA on the bulk chemical
757 composition of Site 762C sediments provides insights into various paleoceanographic processes
758 during hyperthermals; each independent signature of multi-elemental geochemistry reflects
759 different aspects of the effects of global warming on terrestrial and marine environments.

760 Since our study is based on the bulk chemical composition, our discussion remains
761 limited to suggesting the geological phenomena that result in the accumulation and migration of
762 source materials at this site. To further validate these interpretations, future investigations using
763 proxies for continental chemical weathering (e.g., $^{187}\text{Os}/^{188}\text{Os}$ and $\delta^7\text{Li}$; Pogge von Strandmann
764 et al., 2021; Ravizza et al., 2001; Tanaka et al., 2022), biological productivity (e.g., barite
765 accumulation rate and $\delta^{138/134}\text{Ba}$; Bains et al., 2000; Bridgestock et al., 2019; Ma et al., 2014;
766 Miyazaki et al., 2023), paleo-seawater pH (e.g., $\delta^{11}\text{B}$; Anagnostou et al., 2020; Gutjahr et al.,
767 2017; Harper et al., 2020; Henehan et al., 2020), and paleo-redox conditions (e.g., $\delta^{98/95}\text{Mo}$ and
768 $\delta^{15}\text{N}$; Auderset et al., 2022; Dickson, 2017; Siebert et al., 2003) may be necessary. By combining
769 the results of ICA applied to bulk chemistry with a multi-proxy record involving various isotopes,
770 we expect to gain a deeper understanding of the Earth system's responses to transient global
771 warmings during the early Paleogene.

772

773 **6 Conclusions**

774 We newly reported $\delta^{13}\text{C}$, $\delta^{18}\text{O}$, and the chemical composition of bulk carbonate sediments
775 at ODP Site 762C on the Exmouth Plateau in the eastern Indian Ocean and identified the long-
776 term climatic trend (i.e., the EECO followed by the cooling) and five transient hyperthermals
777 (i.e., PETM (onset), H2, I1, J, and ETM3). Through the ICA on the bulk chemical compositions,
778 we extracted six ICs, including ICs representing the mixing relation between biogenic CaCO_3
779 and detrital material (IC3), biogenic calcium phosphate (IC2), diagenetic processes involving
780 Mn-carbonate (IC4), and post-depositional remobilization of redox-sensitive elements (IC1, IC5,
781 IC6) associated with diagenesis and changes in redox conditions. The decrease in IC3 scores
782 during 55–52 Ma suggests a decrease in the contribution of biogenic CaCO_3 or an increase in the
783 riverine terrigenous input relating to significant oceanographic and terrestrial environmental
784 changes in the Early Eocene. Additionally, negative excursions in IC3 scores were observed
785 during hyperthermals, indicating CCD shallowing or an increase of detrital input due to
786 enhanced continental weathering during these events. IC2 scores, which reflect the contribution
787 of BCP, indicate the proliferation of marine vertebrates at hyperthermals. IC1, IC4, IC5, and IC6,
788 characterized by redox-sensitive elements, show peaks around the hyperthermals. This feature
789 suggests that changes in the redox state of porewater or bottom water occurred associated with
790 hyperthermals at Site 762C. Through our study, we shed light on the behaviors of biological and
791 oceanographic responses of mid-latitude eastern Indian Ocean on early Paleogene climate
792 changes.

793

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806 **Open Research**

807 The R script for implementing ICA (fastICA algorithm) used in this study was originally
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 810

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