

28 early Cambrian, accompanied by increasing $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the studied carbonates, likely
29 indicates the enhanced weathering of old continental rock following the assembly of Gondwana.
30 Increased net accumulation of atmospheric oxygen as a result of enhanced organic carbon burial
31 may have benefited from intense continental denudation.

32 **Keywords:** neodymium isotope record; continental denudation; tectonic activity; Neoproterozoic
33 oxygenation event; animal radiation

34 **1. Introduction**

35 The Neoproterozoic Era (1000 Ma to ~ 540 Ma) witnessed the emergence of metazoans and
36 modern ecosystems (Sperling et al., 2013; Knoll and Nowak, 2017), accompanied by a
37 fundamental baseline rise in Earth-surface oxygen levels (Canfield et al., 2007; Och and Shields-
38 Zhou, 2012; Lyons et al., 2014). Nevertheless, oceanic redox conditions underwent dynamic
39 fluctuations with several oxygenation episodes of an overall anoxic ocean are proposed for the late
40 Neoproterozoic (Wood et al., 2015; Sahoo et al., 2016; Wei et al., 2018; Zhang et al., 2018). In
41 addition to uncertainties around the trajectory of redox evolution, the triggers for atmospheric and
42 oceanic oxygenation are also disputed. More effective oxygenic photosynthesis by eukaryotic
43 algae was commonly considered to have produced more oxygen in the late Neoproterozoic (e.g.,
44 Brocks et al., 2017) while the net accumulation of oxygen is also determined by the changing
45 consumption of oxygen by reductants. Tectonic controls on Earth-surface oxygenation have also
46 been invoked in previous research (e.g., Kump and Barley, 2007; Campbell and Allen, 2008; Lee
47 et al., 2016; Planavsky, 2018; Li et al., 2018) although tectonic drivers have not been
48 comprehensively demonstrated by geochemical records. In particular, variations in continental
49 denudation as well as global oceanic circulation following Gondwana amalgamation from the
50 middle Ediacaran to early Cambrian are not well constrained.

51 Initial radiogenic Nd isotope composition of ancient seawater recorded by marine carbonates
52 (defined as $\epsilon\text{Nd}(t)$, relative to Chondritic Uniform Reservoir, CHUR) can be an effective tracer of
53 terrestrial inputs to epeiric seas and ocean basins. Dissolved Nd in the ocean is dominantly derived
54 from the weathering of terrestrial rocks (Piepgras and Wasserburg, 1985; Goldstein and Jacobsen,
55 1987) with additional inputs such as dust, groundwater and seafloor sediment (Frank, 2002; Lacan
56 and Jeandel, 2004; Lacan et al., 2012). Due to its short oceanic residence time (~600 yr) compared

57 with the mixing time scales of the global ocean, the Nd isotopic compositions have shown
58 considerable variability in the modern oceans (Tachikawa et al., 2017). The high spatial variability
59 of present-day ϵNd in the modern ocean is closely linked with various weathered Nd sources to
60 different water-mass and global deep-ocean circulation (Albarède and Goldstein, 1992; Goldstein
61 and Hemming, 2014). Deep North Atlantic water has a significantly lower ϵNd value (~ -13.5)
62 due to riverine input from weathered old continental crust. By contrast, the Pacific ocean, which
63 is surrounded by juvenile magmatic arcs, generally has a comparatively higher ϵNd value (~ -4).
64 ϵNd values of Indian intermediate and deep water are around -8 , due to the advection of seawaters
65 from the Circum-Antarctic Current (Goldstein and Hemming, 2014). The great spatial variability
66 of ϵNd in the global ocean is determined by the heterogeneity of weathered Nd sources to different
67 water-mass and global deep-ocean circulation (Albarède and Goldstein, 1992; Goldstein and
68 Hemming, 2014). Irrespective of spatial heterogeneities in modern global seawater ϵNd values,
69 temporal $\epsilon\text{Nd}(t)$ variations in ancient epeiric water bodies, recorded by marine sediments in a
70 certain area, are also considered to be controlled by fluctuations in sea level (Grandjean et al., 1988;
71 Holmden et al., 1998). Therefore, temporal variations in $\epsilon\text{Nd}(t)$ values of continental shelf
72 seawater generally depend on changes in the weathering source and the degree of water-mass
73 exchange with the more open ocean.

74 In this study, we present new, high-resolution radiogenic Nd isotope data for shallow-marine
75 carbonates from the Jiulongwan–Gaojiayi–Yanjiahe section, South China to reconstruct shallow
76 seawater $\epsilon\text{Nd}(t)$ evolution on the continental shelf margin of the Yangtze block from the Ediacaran
77 to early Cambrian. Neodymium isotopic compositions, combined with radiogenic Sr isotope ratios,
78 are used to directly track the terrestrial inputs and oceanic circulation during this interval. These
79 new data help to constrain the evolution of marine environment on continental shelves and offer

80 valuable insights into the potential correlation of continental denudation and tectonic activity, and
81 their links to atmospheric oxygenation and the emergence of complex metazoans.

82

83 **2. Geological setting**

84 The Doushantuo Formation of the Jiulongwan section in the Yangtze Gorges area is the
85 product of sedimentation in a shallow seawater environment, at or below wave base in an inner
86 shelf lagoon setting (Jiang et al., 2011). At the Jiulongwan section, the Doushantuo Formation has
87 been divided into four members. Member I is an approximately 5-m-thick ‘cap dolostone’
88 featuring a zircon U-Pb age of 635.2 ± 0.6 Ma (Condon et al., 2005), overlying the Nantuo glacial
89 diamictite. Member II is ca. 70 m thick and consists of alternating organic-rich black shale and
90 dolostone bed with pea-sized cherty nodules. Member III comprises about 50-m-thick dolostone
91 variably interbedded with chert in the lower part and limestone in the upper part. Member IV is a
92 ~ 10 m thick black shale, also known as the ‘Miaohe member’ in other localities in the Yangtze
93 Gorges area (e.g., Jiang et al., 2011).

94 The Dengying Formation of the Gaojiayi section is characterized by relatively pure
95 carbonates overlying the Doushantuo Formation, with a zircon U-Pb age 551 ± 0.7 Ma at its base
96 (Condon et al., 2005). The Dengying Formation in the studied area, can be divided into three
97 members: the basal Hamajing Member, the middle Shibantan Member and the uppermost Bamatuo
98 Member. The Hamajing Member is ca. 21 m thick and consists of intraclastic and oolitic dolomitic
99 grainstone. The Shibantan Member consists of dark gray thin- to intermediate-bedded micritic
100 limestone with a total thickness of ca. 50 m and contains Ediacara-type fossils (e.g., Chen et al.,
101 2014). The Bamatuo Member is ca. 40 m thick and consists of micritic and sparitic dolostones.

102 The 54-m-thick Yanjiahe Formation of the Yanjiahe section is characterized by two sub-
103 cycles of shoaling. The lower sub-cycle (~ 29 m) consists of dark grey laminated dolostone
104 interbedded with cherty dolostone, siliceous layers with black shale, and dolostone containing
105 cherty and phosphatic clasts. The upper sub-cycle (~ 25 m) is characterized by black chert with
106 laminated black shale, laminated limestone interbedded with black shale, and limestone with
107 siliceous and phosphate. The Yanjiahe Formation yields early Cambrian Small Shelly Fossil
108 Assemblage Zone 1 (SS1) in the dolostone layer with phosphatic clasts and Small Shelly Fossil
109 Assemblage Zone 3 (SS3) in the upper phosphatic limestone (Jiang et al., 2012). The Ediacaran-
110 Cambrian boundary is considered to be at the bottom of the lower dolostone layer where the SS1
111 first occurs (Chen, 1984).

112

113 **3. Materials and methods**

114

115 We analyzed 75 bulk carbonate samples collected from the Doushantuo Formation, Dengying
116 Formation and Yanjiahe Formation from the Jiulongwan–Gaojiayi–Yanjiahe section, Yangtze
117 Gorges area, Hubei Province, South China (Fig. 1). As reported previously (Ling et al., 2013; Wei
118 et al., 2018, 2019), the best-preserved and least-contaminated carbonate samples primarily
119 composed of micritic dolomite or calcite were selected and drilled for geochemical analyses.

120 3.1 Neodymium isotope analyses

121 For Nd isotope analysis, a two-step leaching method was used to dissolve the marine
122 carbonate component from the bulk samples which may contain other minor components (cf.
123 Bailey et al., 2000; Li et al., 2011; Tissot et al., 2018). Approximately 200 mg powder was firstly
124 rinsed with MQ ultra-pure water, and then leached with 3 ml 0.5 N double-distilled acetic acid
125 (HAc) for 4 hours in order to remove possible contaminants added after marine carbonate

126 deposition. This leachate was discarded after centrifugation, and the insoluble residue was rinsed
127 and centrifuged three times with MQ water. Then 5 ml 1 N double-distilled HAc was added to
128 leach the residue for 4 hours, after which the leachate solution was collected and dried, and then
129 re-dissolved in 5 ml 6 N HCl in preparation for Nd extraction. A two-step resin column method
130 was used for Nd separation and purification (modified after Hirahara et al., 2012). The first elution
131 discards the matrix ion (such as K, Na, Ca, Mg, Sr) from the sample solution, using Bio-rad
132 AG50W-X8 (200-400 mesh) cation resin. The second elution separates Nd from the other rare
133 earth elements (REE), using Eichrom Ln specific resin. The resultant pure Nd solution was dried,
134 and then re-dissolved in 3% HNO₃ for isotopic analyses.

135 Nd isotope analyses were performed on a Thermo Neptune Plus Multicollector, Inductively-
136 Coupled Plasma Mass Spectrometer (MC-ICP-MS) at the State Key Laboratory for Mineral
137 Deposits Research, Nanjing University, and a Nu Plasma II MC-ICP-MS at Nanjing FocuMS
138 Technology Co. Ltd (www.focums.com). ¹⁴³Nd/¹⁴⁴Nd ratios were normalized to a ¹⁴⁶Nd/¹⁴⁴Nd
139 ratio of 0.7219 in order to correct for mass fractionation during analysis. United States Geological
140 Survey standards BRC-2, AGC-2 and BHVO-2 were also digested and analyzed during this study
141 in order to monitor the analytical precision; their ¹⁴³Nd/¹⁴⁴Nd values were measured at 0.512636
142 ±0.000002, 0.512797 ±0.000002 and 0.512986 ±0.000002 (SE), respectively.

143 The studied carbonates are divided into five intervals, with suggested mean depositional ages
144 as follows: ~ 635 Ma for Doushantuo Member I, ~ 580 Ma for Doushantuo Member II, ~ 565 Ma
145 for Doushantuo Member III, ~ 545 Ma for Dengying Formation and ~ 535 Ma for Yanjiahe
146 Formation (age estimates from Condon et al., 2005; Jiang et al., 2012). ¹⁴³Nd/¹⁴⁴Nd ratios were
147 corrected for production of ¹⁴³Nd from ¹⁴⁷Sm decay since the time of deposition of the carbonates,
148 and reported as initial εNd(t) (hereafter termed εNd(t)) relative to ¹⁴³Nd/¹⁴⁴Nd ratios of CHUR at

149 time of deposition (present $^{143}\text{Nd}/^{144}\text{Nd} = 0.512638$ and $^{147}\text{Sm}/^{144}\text{Nd} = 0.1967$, Jacobsen and
150 Wasserburg, 1980). The initial $^{143}\text{Nd}/^{144}\text{Nd}$ ratios of samples are calculated by following equations:

151
$$^{143}\text{Nd}/^{144}\text{Nd}(t) = ^{143}\text{Nd}/^{144}\text{Nd}(0) - ^{147}\text{Sm}/^{144}\text{Nd}(0) \times (e^{\lambda t} - 1) \quad (1)$$

152
$$^{147}\text{Sm}/^{144}\text{Nd}(0) = ([\text{Sm}]/A_{\text{Sm}} \times [^{147}\text{Sm}_{\text{natural}}]) / ([\text{Nd}]/A_{\text{Nd}} \times [^{144}\text{Nd}_{\text{natural}}]) \quad (2)$$

153 whereby $^{143}\text{Nd}/^{144}\text{Nd}(t)$ is the $^{143}\text{Nd}/^{144}\text{Nd}$ ratio of the carbonate when it was deposited;
154 $^{143}\text{Nd}/^{144}\text{Nd}(0)$ is the $^{143}\text{Nd}/^{144}\text{Nd}$ ratio we analyzed in the lab; λ is the decay constant of ^{147}Sm (λ
155 $= 6.54 \times 10^{-12}$, Lugmair and Marti, 1978); t is the age of carbonates in this study; $[\text{Sm}]$ and $[\text{Nd}]$ is
156 Sm and Nd concentrations of the carbonate, measured using a Thermo Element-II ICP-MS at the
157 State Key Laboratory for Mineral Deposits Research, Nanjing University; A_{Sm} and A_{Nd} are atomic
158 weights of Sm (150.36) and Nd (144.242) (Meija et al., 2016); $^{147}\text{Sm}_{\text{natural}}$ and $^{144}\text{Nd}_{\text{natural}}$ are natural
159 abundance of Sm (0.1499) and Nd (0.23798) (Berglund and Wieser, 2011). The initial $^{143}\text{Nd}/^{144}\text{Nd}$
160 of studied carbonate is presented as $\epsilon\text{Nd}(t)$ here, defined by:

161
$$\epsilon\text{Nd}(t) = [^{143}\text{Nd}/^{144}\text{Nd}(t)_{\text{sample}} / ^{143}\text{Nd}/^{144}\text{Nd}(t)_{\text{CHUR}} - 1] \times 10000 \quad (3)$$

162
$$^{143}\text{Nd}/^{144}\text{Nd}(t)_{\text{CHUR}} = ^{143}\text{Nd}/^{144}\text{Nd}(0)_{\text{CHUR}} - ^{147}\text{Sm}/^{144}\text{Nd}(0)_{\text{CHUR}} \times (e^{\lambda t} - 1) \quad (4)$$

163 Whereby t is the same time as deposition of the studied carbonate, $^{143}\text{Nd}/^{144}\text{Nd}(0)_{\text{CHUR}} = 0.512638$,
164 and $^{147}\text{Sm}/^{144}\text{Nd}(0)_{\text{CHUR}} = 0.1967$ (Jacobsen and Wasserburg, 1980).

165 3.2 Strontium isotope analyses

166 For Sr isotope analysis, a two-step leaching method was also used to dissolve the marine
167 carbonate component from the bulk samples (see Bailey et al., 2000; Li et al., 2011 for details). Sr
168 isotope ratios were determined with a Thermo Triton MC-TIMS at the State Key Laboratory for
169 Mineral Deposits Research, Nanjing University. Long-term reproducibility of the analyses was
170 detected by repeated measurement of Sr isotope standard NIST SRM 987 with an average value
171 of 0.710252 (± 0.000016).

172 3.3 Carbon and oxygen isotope analyses

173 Carbon and oxygen isotopes of the carbonate samples in this study were analyzed using a
174 Finnigan Gasbench II online analysis system + Delta Plus XP mass spectrometer at the State Key
175 Laboratory for Mineral Deposits Research, Nanjing University. The results of C and O isotope are
176 presented relative to V-PDB. External precision for C and O isotope analyses is 0.06‰ (2SD) and
177 0.07‰ (2SD), respectively, based on repeated measurements of the Chinese GBW00405 carbonate
178 standard (referred $\delta^{13}\text{C} = 0.57 \pm 0.03\text{‰}$; $\delta^{18}\text{O} = -8.49\text{‰} \pm 0.14\text{‰}$).

179

180 4. Results

181 Results of radiogenic Nd isotope ratio (reported as $\epsilon\text{Nd}(t)$), Nd concentration and radiogenic
182 Sr isotope ratio in this study as well as published C isotope data (Ling et al., 2013) are shown in
183 Figure 2 and Supplementary Table 1.

184 Cap dolostones (i.e. the Doushantuo Member I) in the Jiulongwan section yield relatively
185 high $\epsilon\text{Nd}(t)$ values from -5.86 to -2.91 as well as high Nd concentrations from 1.45 ppm to 5.67
186 ppm. $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in cap dolostones range from 0.7083 to 0.7123 (data from Wei et al., 2019).
187 Carbonates in the Doushantuo Member II have slightly lower $\epsilon\text{Nd}(t)$ values than cap dolostones,
188 ranging from -6.83 to -2.73 . Nd concentrations of the carbonates in this member range from 0.96
189 to 9.28 with some scatter. $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the carbonates in this member are relatively stable,
190 ranging from 0.7079 to 0.7087. Carbonates in the Doushantuo Member III record significantly low
191 $\epsilon\text{Nd}(t)$ values from -10.9 to -7.38 with Nd concentrations between 2.74 and 7.70 ppm (except for
192 a very low value of 0.81). $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the carbonates in the Doushantuo Member III are
193 relatively consistent and overall higher than those in the Member II, ranging from 0.7084 to 0.7095.

194 Carbonates of the Dengying Formation in the Gaojiayi–Yanjiahe section record relatively
195 consistent $\epsilon\text{Nd}(t)$ values and Nd concentrations. Most $\epsilon\text{Nd}(t)$ values of the carbonates in the
196 Dengying Formation rang from -7.35 to -4.92 except for one low point (-8.79 , sample WH-1),
197 which are slightly higher than those in the Doushantuo Member III, but appreciably lower than
198 those in the Doushantuo Member I and slightly lower than those in the Doushantuo Member II.
199 Neodymium concentrations of the carbonates in the Dengying Formation are significantly lower
200 than those in the Doushantuo Formation and range from 0.10 ppm to 1.77 ppm except for four
201 points higher than 3 ppm. $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the carbonates in the Dengying Formation are slightly
202 lower than those in the Doushantuo Member III, ranging from 0.7084 to 0.7092.

203 Carbonates in the Yanjiahe Formation yield lower $\epsilon\text{Nd}(t)$ values and elevated Nd
204 concentrations, relative to carbonates in the Dengying Formation. $\epsilon\text{Nd}(t)$ values of the carbonates
205 in this formation range from -8.90 to -7.16 except for one relatively high point (-3.99 , sample
206 YJH-30) and Nd concentrations generally range from 0.36 to 14.40 ppm with some scatter.
207 $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in the Yanjiahe Formation are higher than those in the Dengying Formation,
208 ranging from 0.7086 to 0.7097.

209

210 **5. Discussion**

211 **5.1 Effects of silicate detritus and diagenetic alteration on carbonate $\epsilon\text{Nd}(t)$**

212 Rare earth elements and yttrium (REY) in shallow marine carbonates are susceptible to
213 detrital contamination due to significantly higher REY contents in silicate detritus than in
214 carbonate minerals and the potentially high clastic sedimentation rates of shallow marine settings.
215 A part of silicate-associated REY could be transferred into the carbonate component during
216 diagenesis or be leached from the silicate fraction during sample preparation. For example, the Nd

217 isotopic compositions of deep marine authigenic sediments have been shown to covary with those
 218 of detrital inputs, resulting from late burial diagenesis (e.g., Du et al., 2016; Jang et al., 2018).
 219 Because aluminum (Al) and thorium (Th) concentrations are high in silicate minerals but
 220 exceedingly low in seawater, covariation between Al and/or Th and other elements or isotope
 221 values is widely used to detect detrital silicate contamination of measured carbonate Nd isotope
 222 values (e.g., Webb and Kamber, 2000; Ling et al., 2013; Zhao and Zheng, 2014, 2017; Wei et al.,
 223 2018, 2019). Leached carbonate components of the samples in this study show significantly low
 224 Al (< 0.2%) and Th concentrations (< 1 ppm) and exhibit no clear covariations between Nd or
 225 $\epsilon\text{Nd}(t)$ and Al or Th concentrations (Fig. 3), which suggests only negligible influence from silicate
 226 detritus on Nd content and Nd isotope data in this study. To further check the impact of detrital
 227 silicate-associated Nd on leachate Nd isotope values, we use the following equations to calculate
 228 Nd concentrations and $\epsilon\text{Nd}(t)$ values of the pure carbonate component by subtracting possible
 229 contributions from silicate detritus, assuming that the Al contents of the bulk carbonates originated
 230 from silicate detritus.

$$231 \quad [Nd]_{det} = \left[\frac{Nd}{Al}\right]_{det} \times [Al]_{bulk} \quad (4)$$

$$232 \quad [Nd]_{carb} = [Nd]_{bulk} - [Nd]_{det} \quad (5)$$

$$233 \quad \epsilon Nd(t)_{carb} = (\epsilon Nd(t)_{bulk} \times [Nd]_{bulk} - \epsilon Nd(t)_{det} \times [Nd]_{det}) / [Nd]_{carb} \quad (6)$$

234 whereby $[Al]_{bulk}$, $[Nd]_{bulk}$, $\epsilon\text{Nd}(t)_{bulk}$ represent the measured concentrations of Al, Nd, and Nd
 235 isotope value, respectively, for each sample using the two-step leaching method (see the result
 236 section and supplementary table for details); $[Nd/Al]_{det}$ and $\epsilon\text{Nd}(t)_{det}$ are Nd/Al ratio and Nd
 237 isotope value of terrestrial detritus contained in carbonate, which are assumed to equal those of
 238 Post-Archean Australian Shale (PAAS) ($Nd/Al = 3.7$, $\epsilon\text{Nd}(t) = -12$) (Taylor and McLennan, 1985,
 239 Allègre, 2008); $[Nd]_{det}$ is the calculated Nd concentration contributed from the detritus to the

240 sample leached solution; $[\text{Nd}]_{\text{carb}}$ and $\epsilon\text{Nd}(t)_{\text{carb}}$ are calculated Nd concentration and $\epsilon\text{Nd}(t)$ value
241 of pure carbonate after detritus correction. The corrected $[\text{Nd}]_{\text{carb}}$ and $\epsilon\text{Nd}(t)_{\text{carb}}$ values of pure
242 carbonates, presented in Fig. 4, typically do not show large differences from the gross values
243 analyzed from the bulk carbonates before correction, except for 4 samples (from 4
244 members/formations) whose $\epsilon\text{Nd}(t)_{\text{carb}}$ values represent outliers, being significantly higher than
245 their gross values (Figs. 4 and 5). This suggests possible overcorrection of these 4 samples and so
246 they are not considered further. Meanwhile, no clear correlations of $\epsilon\text{Nd}(t)_{\text{bulk}}$ vs. $[\text{Nd}]_{\text{bulk}}$ and
247 $\epsilon\text{Nd}(t)_{\text{carb}}$ vs. $[\text{Nd}]_{\text{carb}}$ are observed (Fig. 5A and B). Hence, carbonate samples in this study can
248 reflect the geochemical information of contemporaneous seawater without significant
249 contamination from continental silicate detritus contained in the bulk carbonate samples.

250 In addition to terrestrial silicate detritus, early diagenetic processes may alter the elemental
251 and isotopic compositions of shallow marine carbonates (Oehlert and Swart, 2014; Higgins et al.,
252 2018; Stewart et al., 2015; Chen et al., 2018). However, marine carbonate REY are more likely
253 less sensitive to diagenetic processes, potentially benefiting from high partition coefficients for
254 REY during the carbonate mineral precipitation from seawater, or from usually very low
255 concentrations of REY in low temperature fluids (e.g., Webb and Kamber, 2000; Webb et al., 2009;
256 Zhao and Zheng, 2014, 2017). Even in modern meteoric and marine diagenetic zone (e.g., Great
257 Bahama Bank), marine carbonates were demonstrated to record primary seawater REY signatures
258 (Liu et al., 2019). Additionally, marine dolomitization has been proposed to have a negligible
259 effect on the retention of initial seawater REE signatures in primary carbonates (Nothdurft et al.,
260 2004; Bau and Alexander, 2006). Lack in correlations of $[\text{Nd}]_{\text{carb}}$ vs. Mg/Ca and $\epsilon\text{Nd}(t)_{\text{carb}}$ vs.
261 Mg/Ca (Fig. 5C and D) likewise supports that variations in $[\text{Nd}]_{\text{carb}}$ and $\epsilon\text{Nd}(t)_{\text{carb}}$ of the carbonates
262 in this study are not driven by early diagenetic dolomitization. In conclusion, the Nd isotopic

263 compositions of marine carbonates are proposed to behave conservatively during early diagenesis,
264 which is conducive to the use of Nd isotopes of shallow-marine carbonates as a
265 paleoenvironmental proxy.

266

267 **5.2 Fluctuations of $\epsilon\text{Nd}(t)$ in continental shelf seawater recorded in carbonates from the** 268 **Ediacaran to early Cambrian**

269 Seawater $\epsilon\text{Nd}(t)$ recorded by carbonate rocks from Yangtze Gorges area, South China in this
270 study show three significant excursions to lower values from the early Ediacaran to early Cambrian
271 (N1, N2, N3 in Fig. 4D). These $\epsilon\text{Nd}(t)$ excursions are interestingly coincident with the negative
272 $\delta^{13}\text{C}$ excursions in the Doushantuo Member I (N1), Doushantuo Member III (N2) and Yanjiahe
273 Formation (N3), respectively (Fig. 2A). Generally, abrupt changes in seawater $\epsilon\text{Nd}(t)$ are
274 considered to have been driven by changes in seawater circulation and continental weathering
275 inputs, likely controlled by global climate change and long-term tectonic activity (e.g., Goldstein
276 and Hemming, 2014; Dera et al., 2015). However, studies of carbonates deposited in continental
277 shelf waters suggest that $\epsilon\text{Nd}(t)$ values in those environments may also depend on
278 paleoceanographic position and sea-level fluctuations, reflecting variations in local terrestrial flux
279 to the continental shelf (Fantón et al., 2002; Holmden et al., 2013; Dopieralska et al., 2016). Hence,
280 we suggest that secular $\epsilon\text{Nd}(t)$ variations of the shallow-marine carbonates in this study are
281 primarily caused mainly by changes in Nd source and flux associated with terrestrial input or
282 water-mass exchange. In the context of a marine sedimentary environment for the Yangtze block
283 from the Ediacaran to early Cambrian, Nd isotopic compositions of the Yangtze shelf seawater is
284 dominated by weathered materials from adjacent continental blocks as well as ocean currents with
285 distinct $\epsilon\text{Nd}(t)$ values.

286 To track the potential effect of continental denudation on $\epsilon\text{Nd}(t)$ values of continental shelf
287 seas, $\epsilon\text{Nd}(t)$ data for marine carbonates in this study are compared with those of Neoproterozoic
288 clastic sedimentary rocks sourced from terrestrial silicate detritus (Fig. 6A). Relatively high $\epsilon\text{Nd}(t)$
289 values (> -3) of the clastic rocks suggest extensive weathering of mafic rocks prior to the Ediacaran
290 Period, which is consistent with the emplacement of large igneous provinces (LIP's) (rapid
291 extrusion of huge volume of mafic volcanic rocks in less than several million years) during the
292 early Neoproterozoic (Fig. 6A, Cox et al., 2016 and references therein). Compilations of $\epsilon\text{Nd}(t)$
293 values from Neoproterozoic mudstone, diamictite and iron formation display a decreasing trend
294 from the late Cryogenian to the Ediacaran period (Fig. 6A), suggesting an increasing influence
295 from the weathering of crystalline crustal basement (e.g., Cox et al., 2016). $\epsilon\text{Nd}(t)$ values of
296 shallow-marine carbonates from South China in this study vary in step with the overall trend of
297 $\epsilon\text{Nd}(t)$ in clastic rocks, more likely controlled by secular changes in relative proportions of felsic
298 versus mafic rock weathering (Fig. 6A). However, superimposed on this long-term global trend,
299 the three negative $\epsilon\text{Nd}(t)$ excursions with distinct $\epsilon\text{Nd}(t)$ values from the Ediacaran to early
300 Cambrian (Fig. 4D) may have resulted from relatively short-term changes in the $\epsilon\text{Nd}(t)$
301 compositions of continental shelf seawater, in response to local-to-regional environmental changes
302 (see discussion below).

303

304 **5.2.1 $\epsilon\text{Nd}(t)$ variations in the Marinoan cap dolostone**

305 The Marinoan cap dolostones in this study have relatively high $\epsilon\text{Nd}(t)$ values (average $\epsilon\text{Nd}(t)$
306 $= -3.94$) as well as an appreciable negative $\epsilon\text{Nd}(t)$ excursion in its lower part (lowest to -5.72) (Fig.
307 4D). The average $\epsilon\text{Nd}(t)$ value of cap dolostones is very close to that of the modern Pacific Ocean
308 ($\epsilon\text{Nd}(t) = -4$, Goldstein and Hemming, 2014), and we suggest this is an inherited $\epsilon\text{Nd}(t)$ signature

309 from the syn-glacial Cryogenian ocean (Cox et al., 2016; Gernon et al., 2016). In the context of a
310 short duration for cap dolostone deposition (e.g. Shields, 2005; Hoffman et al., 2017),
311 syngedimentary dolomitization in a brackish mixture of freshwater and seawater has been proposed
312 as a mechanism for cap dolostone deposition, which could explain the dramatic Ca and radiogenic
313 Sr isotope excursions in the lower cap dolostone succession (Wei et al., 2019). The rapid change
314 in $\epsilon\text{Nd}(t)$ could similarly have been influenced by mixing of isotopically distinct water bodies in
315 the aftermath of the end-Cryogenian ‘Marinoan’ glaciation. The abrupt addition of abundant fresh
316 meltwater (from rivers and groundwater) into continental shelf areas could have rapidly decreased
317 carbonate $\epsilon\text{Nd}(t)$ values because freshwater generally has significantly higher Nd concentrations,
318 with less radiogenic Nd, relative to the open oceans (Goldstein and Jacobsen, 1987). Compared
319 with the $^{87}\text{Sr}/^{86}\text{Sr}$ system, which is sensitive to differential weathering of Rb-rich silicate minerals
320 in the post-glaciation (Blum and Erel, 1995; Prestrud Anderson et al., 1997; Li et al., 2007), the
321 $^{143}\text{Nd}/^{144}\text{Nd}$ system is relatively less susceptible to incongruent weathering of different silicate
322 minerals (Bayon et al., 2015). Hence, overall the Doushantuo cap dolostones record high $\epsilon\text{Nd}(t)$
323 values of continental shelf seawater in the aftermath of the Marinoan glaciation, despite a
324 perceptible negative excursion at the base.

325 Relatively high average $\epsilon\text{Nd}(t)$ values (~ -4) and low $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (~ 0.7080) of the
326 carbonates from Doushantuo Member II indicate that the weathering of juvenile magmatic rock
327 dominated riverine inputs into continental shelf seawater (cf. modern Pacific Ocean) throughout
328 deposition of this Member (Fig. 4D). A relatively high proportion of Nd derived from juvenile
329 rocks likely relates to the protracted breakup of the Rodinia supercontinent into the early Ediacaran
330 (Li et al., 2013), suggesting a secular tectonic control on $\epsilon\text{Nd}(t)$ composition of the continental
331 shelf seawater.

332

333 **5.2.2 Significantly negative $\epsilon\text{Nd}(t)$ excursion during the Shuram Excursion**

334 Carbonates of the Doushantuo Member III in this study are characterized by extremely low
335 $\delta^{13}\text{C}$ values (lowest to -10‰), which can be correlated to the Shuram negative $\delta^{13}\text{C}$ excursion (or
336 DOUNCE) (McFadden et al., 2008; Lu et al., 2013). This widespread $\delta^{13}\text{C}$ excursion has been
337 proposed to reflect either a primary marine signal or the product of secondary alteration, both of
338 which suggest global environmental extremes during this period (e.g., Grotzinger et al., 2011).
339 $\epsilon\text{Nd}(t)$ values recorded in these carbonates from South China exhibit a dramatic negative excursion
340 (Fig. 4D) in step with the $\delta^{13}\text{C}$ variation (Fig. 2A). The average $\epsilon\text{Nd}(t)$ value for carbonates in
341 Doushantuo Member III (-9.34) approaches the $\epsilon\text{Nd}(t)$ value of modern circum-Antarctic seawater
342 (-8 to -9) (Piepgras and Wasserburg, 1982), which is much lower than the average values for the
343 cap dolostone (-3.94) and Doushantuo Member II (-3.78).

344 A prolonged and large negative $\epsilon\text{Nd}(t)$ excursion could be caused by a change in continental
345 weathering, exchange between seawater and shelf sediment or a change in ocean circulation (e.g.,
346 Zheng et al., 2013). Exchange between seawater/authigenic carbonate and seafloor sediment
347 during late diagenesis has been suggested to alter Nd isotope signatures of local bottom waters or
348 sediments, controlled by compositions of deeply buried sediments (Lacan and Jeandel, 2004;
349 Abbott et al., 2016; Jang et al., 2018). However, the remobilization of seafloor surface sediments
350 could only affect the $\epsilon\text{Nd}(t)$ composition of deep or intermediate seawater rather than shallow
351 seawater (Lacan and Jeandel, 2004), which seems unlikely to contribute significantly to the
352 negative $\epsilon\text{Nd}(t)$ excursion in Doushantuo Member III carbonates. Moreover, exchange between
353 seawater and seafloor sediment would result in an appreciable correlation between $\epsilon\text{Nd}(t)$ values
354 and Nd concentrations in the carbonate, as detrital sediments on seafloor and sedimentary pore

355 water have significantly higher Nd concentrations, relative to bottom seawater (e.g., Abbott et al.,
356 2016). Carbonates in this study exhibit no clear covariation between $\epsilon\text{Nd}(t)$ values and Nd
357 concentrations (Fig. 5A and B), suggesting little contamination on shallow seawater $\epsilon\text{Nd}(t)$ from
358 dissolution of detrital sediments. The elevated Nd concentrations of the carbonates during this
359 period is more likely caused by the transition from the dolomite to calcite in this stratigraphic
360 interval as calcite generally has much higher Nd concentration relative to dolomite and aragonite
361 (e.g., Webb and Kamber, 2000; Webb et al., 2009). Hence, $\epsilon\text{Nd}(t)$ values of shallow seawater in
362 this study are more likely controlled by the source and flux of Nd imported into the Yangtze margin
363 area, regardless of the nature of early and late diagenetic alteration (e.g., Holmden et al., 1998;
364 Dopieralska et al., 2016; Tachikawa et al., 2017). A previous study suggested that the negative
365 $\epsilon\text{Nd}(t)$ excursion in the middle Ediacaran was induced by extensive weathering of more mature
366 detritus from the Archean Kongling complex in the Yangtze block (Hu et al., 2016). However, this
367 may meet difficulty when considering the subsequent increase of $\epsilon\text{Nd}(t)$ in the late Ediacaran (i.e.,
368 Dengying Formation) since weathering of the Kongling complex would have not been rapidly
369 terminated as the Gondwana assembly was still in the process. Moreover, in view of a submarine
370 sedimentary environment for the Yangtze area in the Ediacaran (Jiang et al., 2011), Nd in the
371 Yangtze marginal seawater is more likely from weathered materials of adjacent exposed
372 continental cratons, rather than only from those of the old terrain in the Yangtze block.

373 Although the age of the Shuram Excursion is not well constrained, the onset of this negative
374 $\delta^{13}\text{C}$ excursion is considered to be later than the Gaskiers glaciation (~ 580 Ma, Pu et al., 2016)
375 and previous studies have favoured a 9–10 Myr duration (Condon et al., 2005; Narbonne et al.,
376 2012; Gong et al., 2017; Minguez et al., 2017; Wang et al., 2017). Therefore, the estimated age of
377 the Shuram Excursion is believed to be broadly coincident with the early Gondwana assembly (cf.

378 Li et al., 2013). Paleo-continental reconstructions suggest that South China was isolated in low-
379 middle latitudes and adjacent to Australia in Gondwana (Li et al., 2008, 2013; Xu et al., 2013;
380 Zhao et al., 2018). $\epsilon\text{Nd}(t)$ values of carbonates from South China approach those of continental
381 detrital sedimentary rocks from Australia (Fig. 6A). Thus, the decreased $\epsilon\text{Nd}(t)$ values of shallow
382 marine carbonates generally may have resulted from enhanced continental weathering of old
383 continental crust exposed during the early assembly process of Gondwana. In this point, the
384 prominently negative $\epsilon\text{Nd}(t)$ excursion during the Shuram Excursion may have been driven by
385 increased export of the weathered old continental felsic rocks to the adjacent seawater
386 accompanying with the initial Gondwana assembly, which alters the preceding local seawater
387 $\epsilon\text{Nd}(t)$ balance.

388 However, $\epsilon\text{Nd}(t)$ values of the carbonates in the Dengying Formation (~ -5) are much higher
389 than those in the Doushantuo Member III and more similar to those in Doushantuo Member II (Fig.
390 4D). The elevated $\epsilon\text{Nd}(t)$ values in the Dengying Formation are considered to reflect an abrupt
391 decrease in terrestrial inputs after the Shuram Excursion, which is unlikely interpreted by tectonic
392 evolution due to the prolonged assembly of Gondwana during this period. This observation
393 proposed that the dramatic $\epsilon\text{Nd}(t)$ anomaly during the Shuram Excursion may have been
394 influenced by other factors in addition to variability in continental denudation. The detrital
395 sediments in NW Laurentia have much lower $\epsilon\text{Nd}(t)$ values (< -15), compared with detrital
396 sediments in other blocks, and show a similar trend in $\epsilon\text{Nd}(t)$ with carbonate $\epsilon\text{Nd}(t)$ record in the
397 middle Ediacaran this study (Fig. 6A). Despite the protracted decrease in $\epsilon\text{Nd}(t)$ from the
398 Neoproterozoic to the early Cambrian, an abrupt $\epsilon\text{Nd}(t)$ fall in detrital sediments of NW Laurentia
399 is also observed in the middle Ediacaran, after which $\epsilon\text{Nd}(t)$ values in NW Laurentia increase
400 slightly and approach to those of Australia (Fig. 6A). Given the observed $\epsilon\text{Nd}(t)$ variation in

401 detrital sediments from NW Laurentia, the dramatically negative $\epsilon\text{Nd}(t)$ anomaly during the
402 Shuram Excursion in this study may have been induced by exchange of seawater between South
403 China and NW Laurentia due to intensified oceanic circulation and/or improved ocean
404 connectivity during the middle Ediacaran. In this regard, water-mass exchange between South
405 China and Laurentia may have been relatively rapid due to the opening gateway between these
406 two cratons via the Mawson ocean (e.g., Li et al., 2013; Zhao et al., 2018). Further, global cooling
407 in the middle Ediacaran (i.e., Gaskiers glaciation typically found on Laurentia, Hoffman and Li,
408 2009; Pu et al., 2016) may have accelerated the export of bottom seawater from high-latitude
409 Laurentia to relatively low-latitude South China (cf. Zheng et al., 2013, 2016).

410 In conclusion, the abrupt low $\epsilon\text{Nd}(t)$ values of shallow-marine carbonates in the Doushantuo
411 Member III, South China suggest an enhanced weathering of old continental rocks in step with
412 more vigorous exchange of different water-masses, potentially driven by both tectonic activity and
413 climate change. In the view of this, the elevated $\epsilon\text{Nd}(t)$ values in the Dengying Formation is more
414 likely induced by recession of water mass mixing between South China and Laurentia, following
415 the subsequent assembly of Gondwana. Additionally, $\epsilon\text{Nd}(t)$ anomaly in the middle Ediacaran
416 Period interestingly coincides with negative C isotope excursion. Incorporation of such bottom
417 water mass containing probably DOC to the shallow water mass in South China could have also
418 contribute to the negative C isotope excursion of Shuram Excursion although other contributions
419 may have also played roles in the light carbon isotope record. More research is needed to unravel
420 the potential links between these two isotope systems during this period.

421

422 **5.2.3 Decreased $\epsilon\text{Nd}(t)$ values of shallow carbonates in the early Cambrian**

423 $\epsilon\text{Nd}(t)$ values of the carbonates in the Dengying Formation are relatively consistent and much
424 higher than those in Doushantuo Member III. Considering that the abrupt negative $\epsilon\text{Nd}(t)$ anomaly
425 in Doushantuo Member III reflects rapid transport of Laurentia-sourced seawater during the
426 middle Ediacaran, we attribute the slight $\epsilon\text{Nd}(t)$ decrease in the Dengying Formation (-5.27 on
427 average), relative to the Doushantuo Member II (-3.78 on average), to successive Gondwana
428 assembly in the terminal Ediacaran (Fig. 4D).

429 Carbonates in the Yanjiahe Formation in this study have a relatively low average $\epsilon\text{Nd}(t)$
430 value (-7.33), compared with carbonates in the Dengying Formation (average $\epsilon\text{Nd}(t) = -5.27$).
431 The decrease in marine carbonate $\epsilon\text{Nd}(t)$ values from the terminal Ediacaran to early Cambrian
432 is coincident with the $\epsilon\text{Nd}(t)$ variation in silicate detritus from Australia and NW Laurentia (Fig.
433 6A), suggesting enhanced denudation of old continental rock. The $\epsilon\text{Nd}(t)$ variation in shallow-
434 marine carbonates in this study is consistent with long-term $\epsilon\text{Nd}(t)$ evolution of the Paleo-Pacific
435 Ocean in the early Cambrian (Keto and Jacobsen, 1988), which is suggested as a global change
436 in the supply of weathered continental materials to the ocean. In the early Cambrian, final
437 assembly of Gondwana with widespread continental collision resulted in a high topographic
438 landscape and a potentially low sea level (e.g., Li et al., 2013). Moreover, the early Cambrian
439 was characterized by a greenhouse climate likely due to extensive subduction outgassing
440 (McKenzie et al., 2016; Hearing et al., 2018). Hence, the decreased $\epsilon\text{Nd}(t)$ values, coincident
441 with elevated $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, from the carbonates in the Yanjiahe section are indicative of an
442 intense continental weathering during the early Cambrian, consistent with previous studies (e.g.,
443 Maloof et al., 2010; Peters and Gaines, 2012).

444

445 **5.3 Long-term evolution of $\epsilon\text{Nd}(t)$ in continental shelf seawater: more records from global**
446 **shallow-marine sediments**

447 Although the residence time of Nd in the ocean is very short (~ 600 yr, Johannesson and
448 Burdige, 2007), temporal changes in $\epsilon\text{Nd}(t)$ values of epeiric seawater can potentially reflect
449 gradual evolution of terrestrial inputs on the continental shelves, which may have been controlled
450 by tectonic activity and climate change (e.g., Scher et al., 2011; Peters and Gaines, 2012). Due to
451 the limited contribution from oceanic hydrothermal sources (Piepgras and Wasserburg, 1985;
452 Goldstein and Jacobsen, 1987), the sedimentary $\epsilon\text{Nd}(t)$ record is more effective for directly
453 tracking regionally terrestrial inputs and water sources, compared with $^{87}\text{Sr}/^{86}\text{Sr}$ or $^{187}\text{Os}/^{188}\text{Os}$
454 ratios. Shallow-marine carbonates in this study exhibit a long-term decreasing trend in $\epsilon\text{Nd}(t)$
455 values from the early Ediacaran to early Cambrian, superimposed by an abrupt negative excursion
456 of $\epsilon\text{Nd}(t)$ values during the Shuram Excursion in the middle Ediacaran. Significantly negative
457 $\epsilon\text{Nd}(t)$ values during the Shuram Excursion likely record incorporation of water-mass from the
458 Laurentia. Whereas, the secular $\epsilon\text{Nd}(t)$ varying trend in shallow carbonates coincides with the
459 evolution of $\epsilon\text{Nd}(t)$ in global terrestrial silicate detritus (Fig. 6A), suggesting a more dominant
460 control of continental weathering materials on gradual change in $\epsilon\text{Nd}(t)$ values of continental shelf
461 seawater over million-year timescale.

462 To better understand holistic $\epsilon\text{Nd}(t)$ variations of shallow seawater, $\epsilon\text{Nd}(t)$ record derived
463 from shallow-marine carbonates in this study is combined with more $\epsilon\text{Nd}(t)$ records from
464 phosphoric rocks (Yang et al., 1997; Felitsyn and Gubanov, 2002) in different regions from the
465 Ediacaran to early Cambrian (Fig. 6B). $\epsilon\text{Nd}(t)$ values of continental shelf seawater derived from
466 these shallow-marine sediments are further compared to $\epsilon\text{Nd}(t)$ record for Paleo-Pacific
467 (Panthalassa) waters (shaded area in Fig. 6B, modified from Keto and Jacobsen, 1988).

468 Compilations of $\epsilon\text{Nd}(t)$ data from shallow-marine biotic and abiotic sediments show a relatively
469 consistent and high $\epsilon\text{Nd}(t)$ composition (around -5) of shallow seawater from the Cryogenian to
470 the middle Ediacaran which coincides with prolonged breakup of Rodinia. Appreciable drop in
471 seawater $\epsilon\text{Nd}(t)$ values initially occurs in the middle Ediacaran and significantly low $\epsilon\text{Nd}(t)$ values
472 (< -15) are observed in the early Cambrian (Fig. 6B). The initial decrease of seawater $\epsilon\text{Nd}(t)$ in
473 the middle Ediacaran is coincident with the beginning of Gondwana assembly (Li et al., 2008,
474 2013). Further, the secular $\epsilon\text{Nd}(t)$ record from the early Ediacaran to early Cambrian is roughly
475 consistent with the $^{87}\text{Sr}/^{86}\text{Sr}$ record in this study (Fig. 7) as well as long-term $^{87}\text{Sr}/^{86}\text{Sr}$ evolution
476 during the late Neoproterozoic (e.g., Maloof et al., 2010), despite the exceedingly high $^{87}\text{Sr}/^{86}\text{Sr}$ in
477 the Doushantuo cap dolostone which is likely deposited in a deglacial meltwater-seawater mixture
478 (Wei et al., 2019). In conjunction with zircon O–Hf isotope record (e.g., Cawood et al., 2013;
479 Spencer et al., 2014), Nd isotopic variations from the Ediacaran to early Cambrian (Fig. 7) are
480 more likely associated with the final Rodinia breakup and subsequent Gondwana assembly.
481 Significantly low $\epsilon\text{Nd}(t)$ values in the early Cambrian (< -15) are corresponding to peak and trough
482 of zircon O isotope and Hf isotope values (Fig. 6, Cawood et al., 2013; Spencer et al., 2014), in
483 response to the timing of extensive continental orogeny driven by final Gondwana assembly. In
484 particular, the initiation of substantial changes in zircon O–Hf isotopes precedes the significant
485 seawater $\epsilon\text{Nd}(t)$ decrease recorded in this study (Fig. 6B), indicating that the formation of mature
486 continent-sourced rocks should be much earlier than weathering of these rocks. In conclusion,
487 secular variation in shallow seawater $\epsilon\text{Nd}(t)$ directly results from changes in weathered materials
488 exported to continental shelf area, driven by long-term tectonic activity.

489

490 **5.4 Potential links between increased continental denudation and oceanic oxygenation in the**
491 **terminal Precambrian**

492 Several isotope systems, such as $^{87}\text{Sr}/^{86}\text{Sr}$, $\delta^7\text{Li}$ and $\delta^{44}\text{Ca}$, have been widely used to track the
493 evolution of continental weathering (e.g., Blättler et al., 2011; Pogge von Strandmann et al., 2013;
494 Chen et al., 2018). $\epsilon\text{Nd}(t)$ values of shallow-marine carbonates can provide a more direct clue for
495 changes in the weathering of regional continental materials (old crust vs. juvenile rock) ultimately
496 controlled by long-term tectonic activity, since differential weathering of silicate minerals in a
497 single rock and submarine hydrothermal fluid have little effects on $\epsilon\text{Nd}(t)$ of continental shelf
498 seawater (e.g., Goldstein and Jacobsen, 1987; Bayon et al., 2015). Although more data are needed
499 from various continental margins for a global chemostratigraphic correlation, gradually decreasing
500 $\epsilon\text{Nd}(t)$ values of shallow-marine carbonates as well as phosphatic rocks in this study support a
501 gradually more extensive and intensive continental orogeny during the assembly of Gondwana
502 from the middle Ediacaran to early Cambrian (Li et al., 2008, 2013; Zhao et al., 2018). Tectonic
503 and magmatic processes are proposed closely linked with Precambrian atmospheric oxygenation
504 (Campbell and Allen, 2008; Planavsky, 2018; Li et al., 2018). The net accumulation of atmospheric
505 O_2 is generally determined by the fluxes of O_2 production and consumption. From the middle
506 Ediacaran to early Cambrian, elevated topography following widespread mountain building
507 related to the amalgamation of Gondwana would have enhanced continental denudation and
508 nutrient export to continental marginal seawater which is supported by increased P contents in
509 fine-grained marine siliciclastic rocks (Reinhard et al., 2017). Thus, O_2 release by marine primary
510 production may have been promoted by this marine fertilization. In addition, the delivery of
511 abundant detrital silicates to continental shelf margin can facilitate organic carbon burial via more
512 rapid burial of marine clastic sediment, as observed in the modern Ganges-Brahmaputra basin (cf.

513 Galy et al., 2011, 2015), yielding a well positive correlation between organic carbon burial rate
514 and sediment burial rate (Bernier and Canfield, 1989). Moreover, a decline in volcanism in the
515 period of intensive intracontinental collision within Gondwana leads to less release of reduced
516 gases from the Earth's interior into the atmosphere (cf. Kump and Barley, 2007). Both enhanced
517 organic carbon burial and decreased release of reduced gases result in less O₂ consumption at the
518 Earth surface. In conclusion, the long-term overall increase in oxygen levels from the middle
519 Ediacaran to early Cambrian is likely a result of secular accumulation of oxygen along with the
520 prolonged assembly of the Gondwana and then extensive continental orogeny. Hence, enhanced
521 continental denudation related to supercontinent assembly may have aroused the Earth-surface
522 environment from a low O₂ steady state, which likely triggers the diversification and radiation of
523 large, complex animals from the late Ediacaran to early Cambrian (Fig. 7).

524

525 **6. Conclusions**

526 Carbonates of the Ediacaran–Cambrian Jiulongwan–Gaojiayi–Yanjiahe section, South China
527 exhibit a gradually decreasing trend in $\epsilon\text{Nd}(t)$ values from around -4 to -8 , which directly indicates
528 a progressively intensifying export of weathered terrestrial materials to continental shelf seawater
529 during the terminal Neoproterozoic and early Cambrian. Additionally, a significant and transient
530 negative $\epsilon\text{Nd}(t)$ anomaly in the middle Ediacaran superimposed on the secular decreasing $\epsilon\text{Nd}(t)$
531 trend, which is indicative of an abrupt and rapid improvement of seawater transport from Laurentia
532 to South China. Together with radiogenic Sr isotopes, the long-term evolution of $\epsilon\text{Nd}(t)$ values on
533 the Yangtze continental margin from the Ediacaran to early Cambrian is considered to be
534 dominantly controlled by extensive continental collision following the assembly of Gondwana.

535 The rise of oxygen level in Earth-surface environment during this key transition is proposed to be
536 linked with enhanced continental weathering, driven by continued continental orogenic processes.

537

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546

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823 **Figure captions**

824 Figure 1. (A) Paleogeographic map of the Yangtze Block, South China (modified after Jiang et al.,
825 2011). The studied area is at the Yangtze Gorges area, representing a sedimentary
826 environment of intrashelf sedimentary basin. (B) Geological map of the Yangtze Gorges area,
827 South China. Samples in this study are collected from 1) the Jiulongwan section; 2) the
828 Gaojiaxi section; 3) the Yanjiahe section.

829 Figure 2. The profiles of (A) carbon, (B) radiogenic neodymium and (D) radiogenic strontium
830 isotopic compositions as well as (C) neodymium concentrations from the carbonates in
831 Jiulongwan-Gaojiaxi-Yanjiahe section, Yangtze Gorges area, South China. $[\text{Nd}]_{\text{bulk}}$ and
832 $\epsilon\text{Nd}(t)_{\text{bulk}}$ represent the pristine data for Nd concentration and Nd isotope without
833 correction of detrital component. $\delta^{13}\text{C}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ data are referred from Ling et al. (2013);
834 Wei et al. (2019) and this study.

835 Figure 3. Cross-plots of (A) $[\text{Nd}]_{\text{bulk}}$ vs. $[\text{Al}]$, (B) $\epsilon\text{Nd}(t)_{\text{bulk}}$ vs. $[\text{Al}]$, (C) $[\text{Nd}]_{\text{bulk}}$ vs. $[\text{Th}]$, (D)
836 $\epsilon\text{Nd}(t)_{\text{bulk}}$ vs. $[\text{Th}]$ in bulk carbonates from Jiulongwan-Gaojiaxi-Yanjiahe section, Yangtze
837 Gorges area, South China.

838 Figure 4. Calculated results of Nd concentration and $\epsilon\text{Nd}(t)$ value of pure carbonate component
839 (presented as $[\text{Nd}]_{\text{carb}}$ and $\epsilon\text{Nd}(t)_{\text{carb}}$, respectively), compared with bulk carbonate $[\text{Nd}]_{\text{bulk}}$ and
840 $\epsilon\text{Nd}(t)_{\text{bulk}}$ in this study.

841 Figure 5. Cross-plots of (A) $\epsilon\text{Nd}(t)_{\text{bulk}}$ vs. $[\text{Nd}]_{\text{bulk}}$, (B) $\epsilon\text{Nd}(t)_{\text{carb}}$ vs. $[\text{Nd}]_{\text{carb}}$, (C) $[\text{Nd}]_{\text{carb}}$ vs. Mg/Ca
842 and (D) $\epsilon\text{Nd}(t)_{\text{carb}}$ vs. Mg/Ca for carbonate samples in this study.

843 Figure 6. (A) $\epsilon\text{Nd}(t)_{\text{carb}}$ variations in shallow carbonates from the early Ediacaran to early
844 Cambrian in this study (filled circles), compared with secular evolution of $\epsilon\text{Nd}(t)$ in
845 continental clastic sediments from NW Laurentia, Australia, South China and Svalbard (data

846 from Cox et al., 2016) (squares). Nd isotopic compositions in marine carbonates, which are
847 considered to record regionally shallow seawater signatures in this study, are overall higher
848 than those in continental detritus, indicating that more dissolved mafic components from
849 weathering of continental crust, than those from felsic components, are exported to shallow
850 seawater. This observation also suggests a more congruent weathering for mafic rock than
851 felsic rock. (B) Compilations of potential seawater $\epsilon\text{Nd}(t)$ records derived from shallow
852 carbonates (this study), sedimentary phosphorites (Yang et al., 1997) and phosphatic Small
853 Shelly Fossils (Felitsyn and Gubanov, 2002). Shaded area is the estimated $\epsilon\text{Nd}(t)$ range for
854 paleo-pacific (Panthalassa) seawater (Keto and Jacobsen, 1988). The dashed curves are zircon
855 Hf and O isotope records, modified from Cawood et al. (2013) and Spencer et al. (2014).

856 Figure 7. Co-varying $\epsilon\text{Nd}(t)_{\text{carb}}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ of shallow carbonates from the early Ediacaran to
857 early Cambrian in this study in step with temporal occurrence ranges for key events of
858 biological evolution (modified after Wood et al., 2019). The black solid curves are results of
859 LOWESS smooth fitting for both $^{87}\text{Sr}/^{86}\text{Sr}$ and $\epsilon\text{Nd}(t)_{\text{carb}}$ data in this study. The dashed curves
860 are analytical errors for the LOWESS fitting (2 SD).

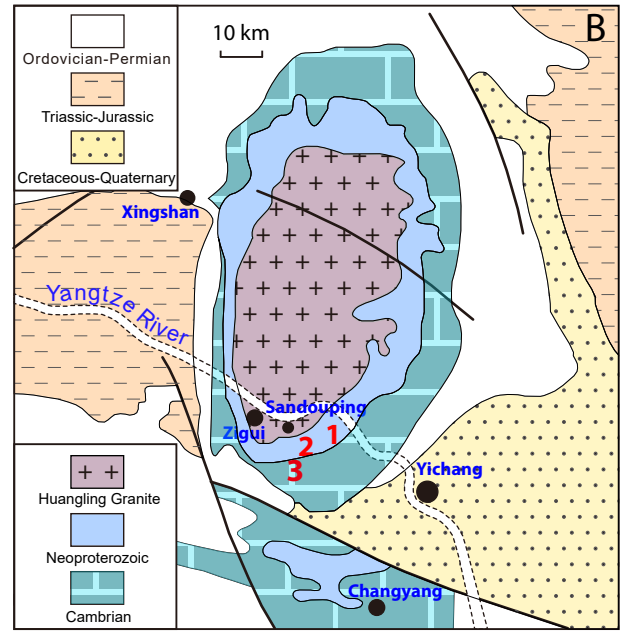
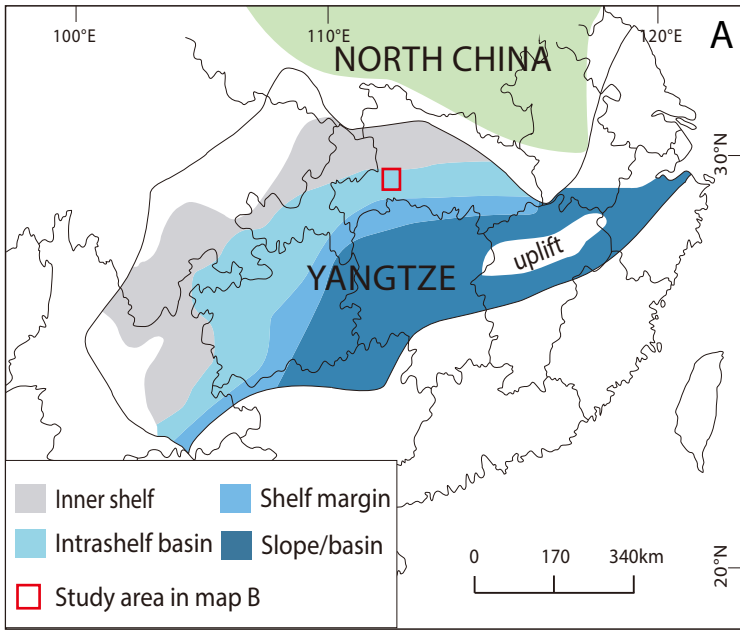


Figure 1

Yangtze Gorges area

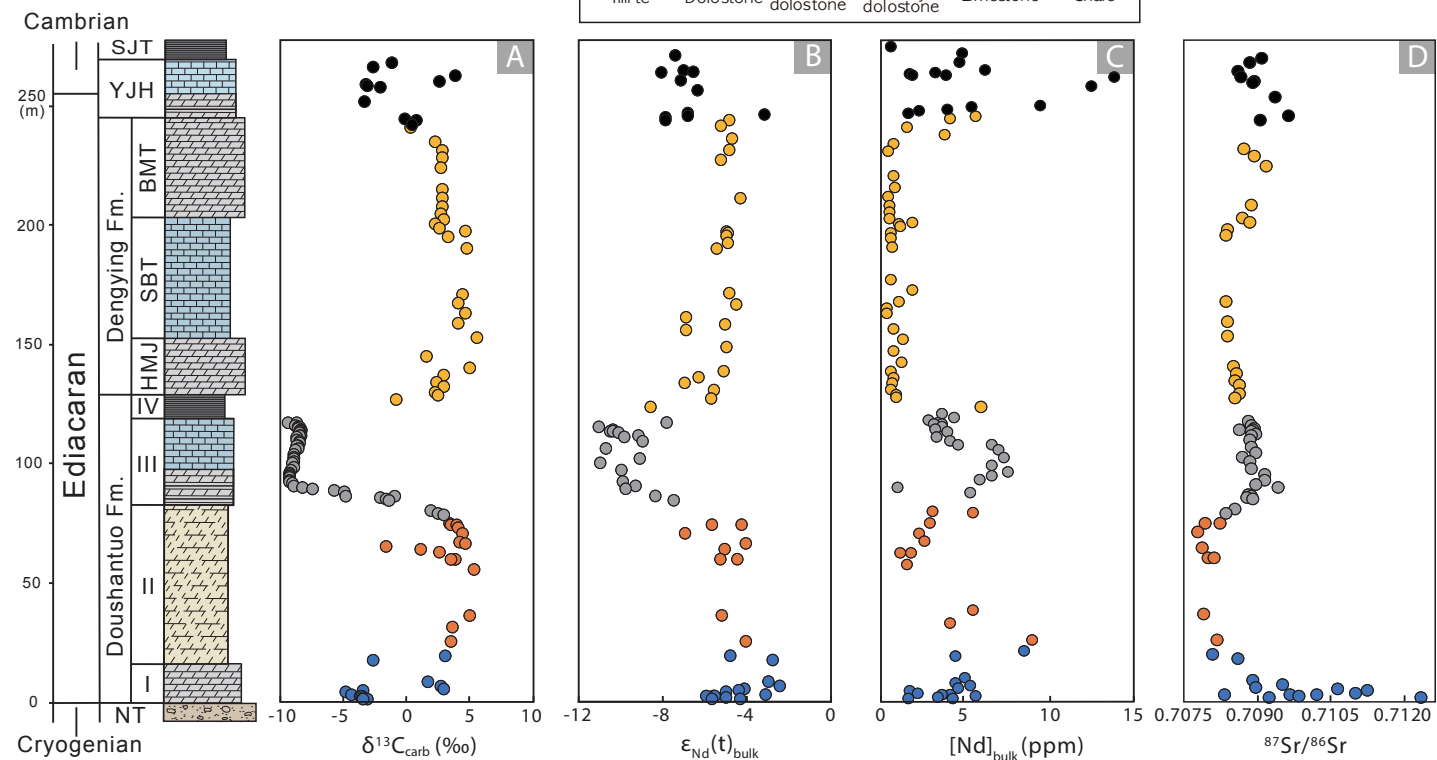


Figure 2

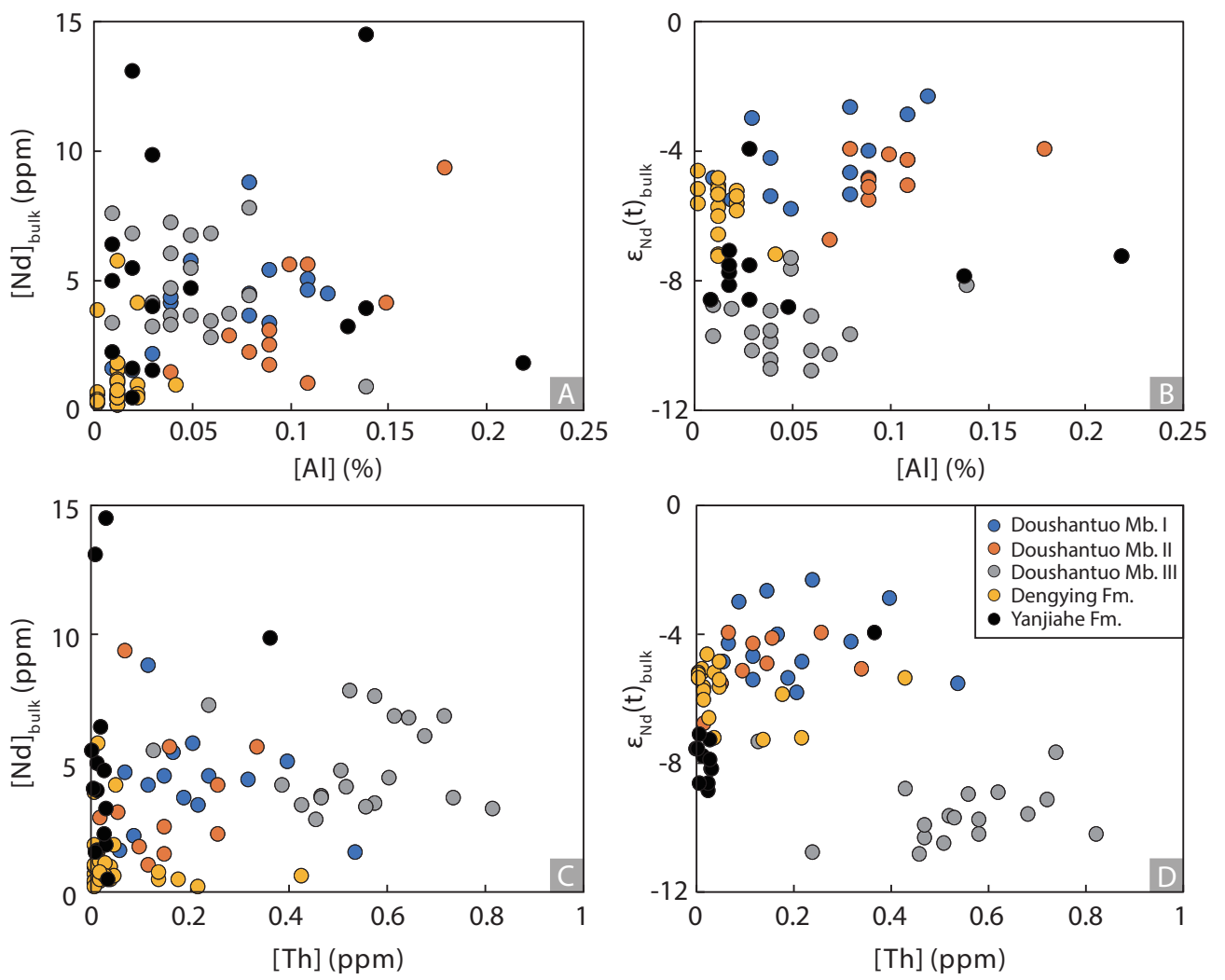


Figure 3

Yangtze Gorges area

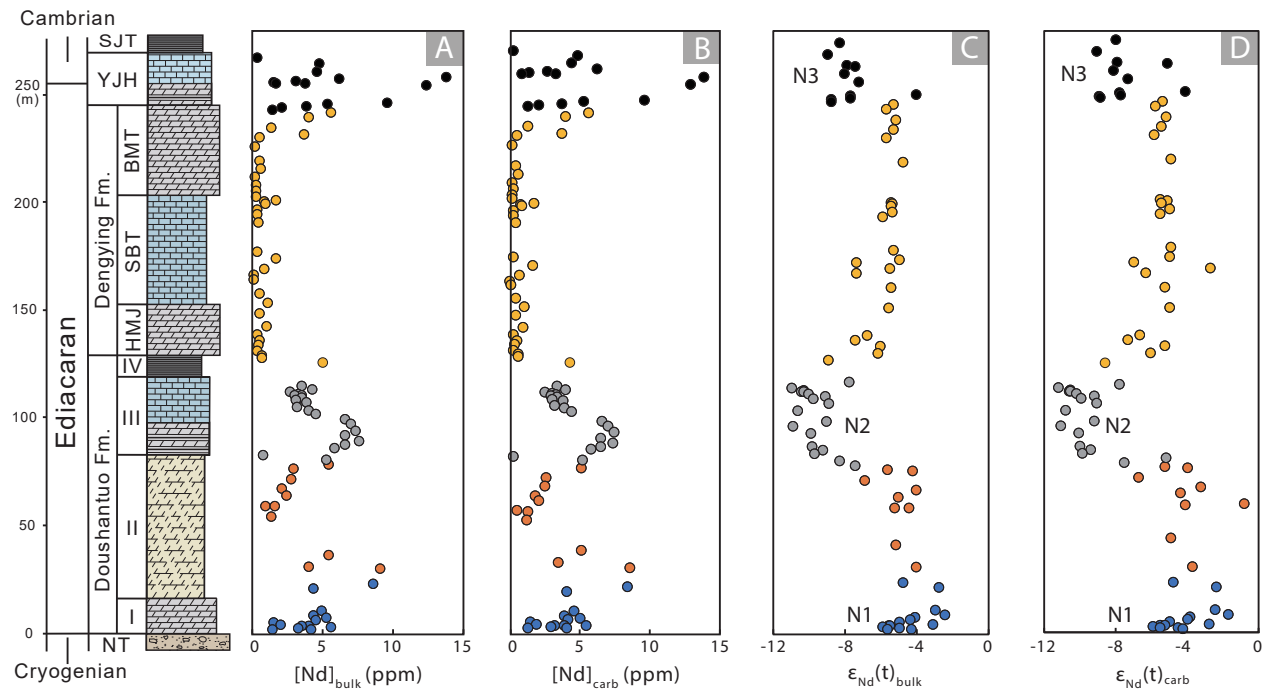


Figure 4

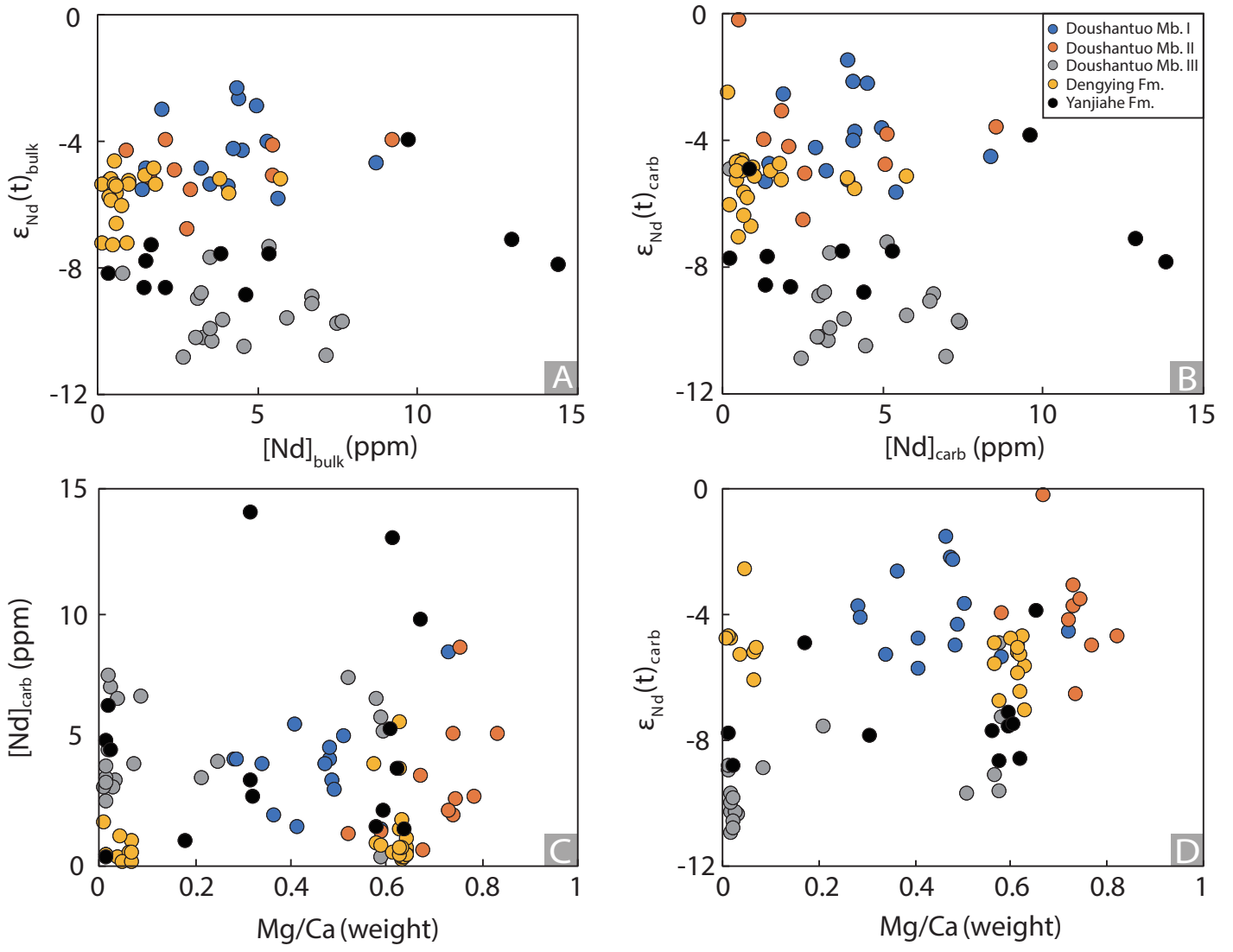


Figure 5

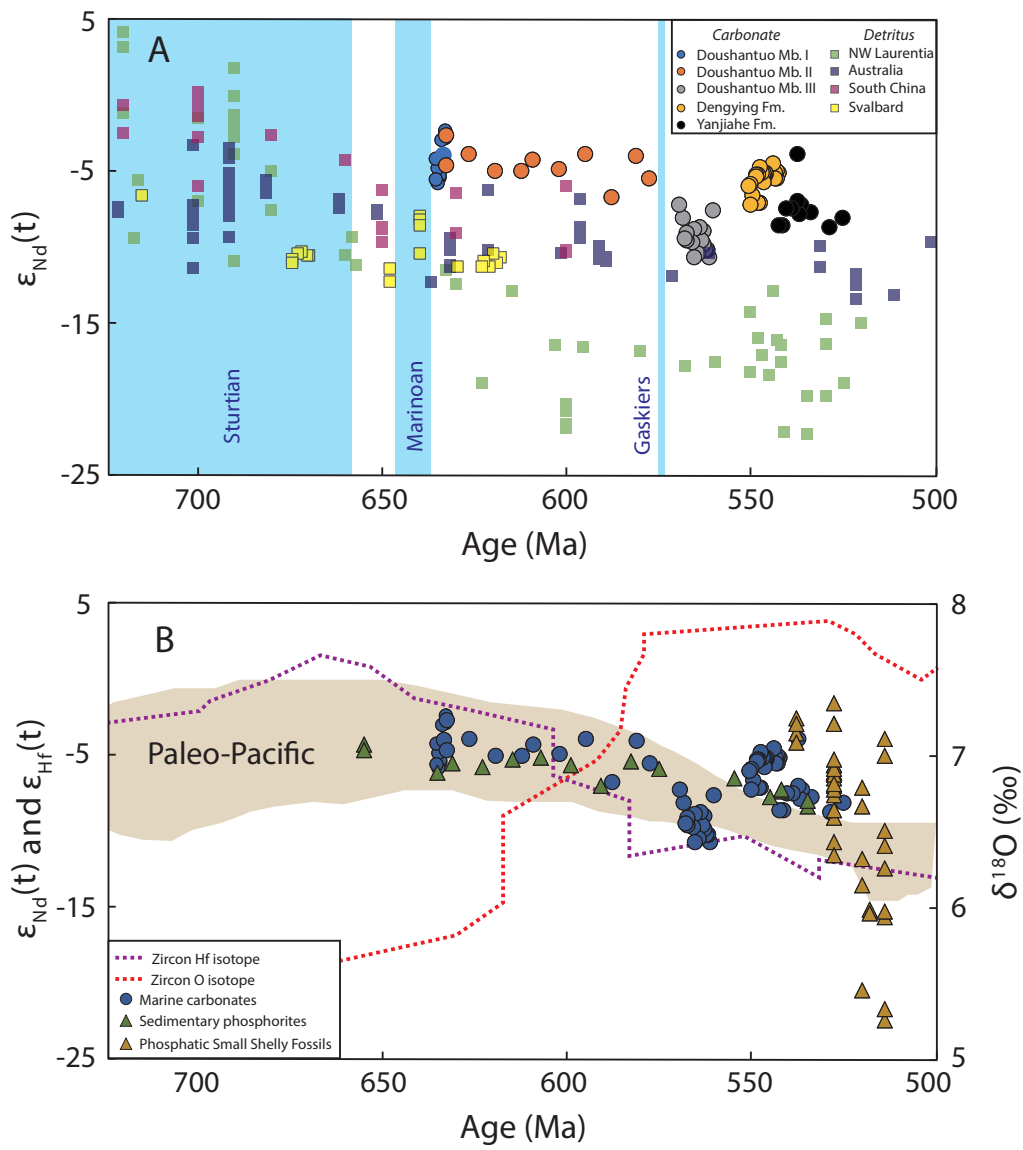


Figure 6

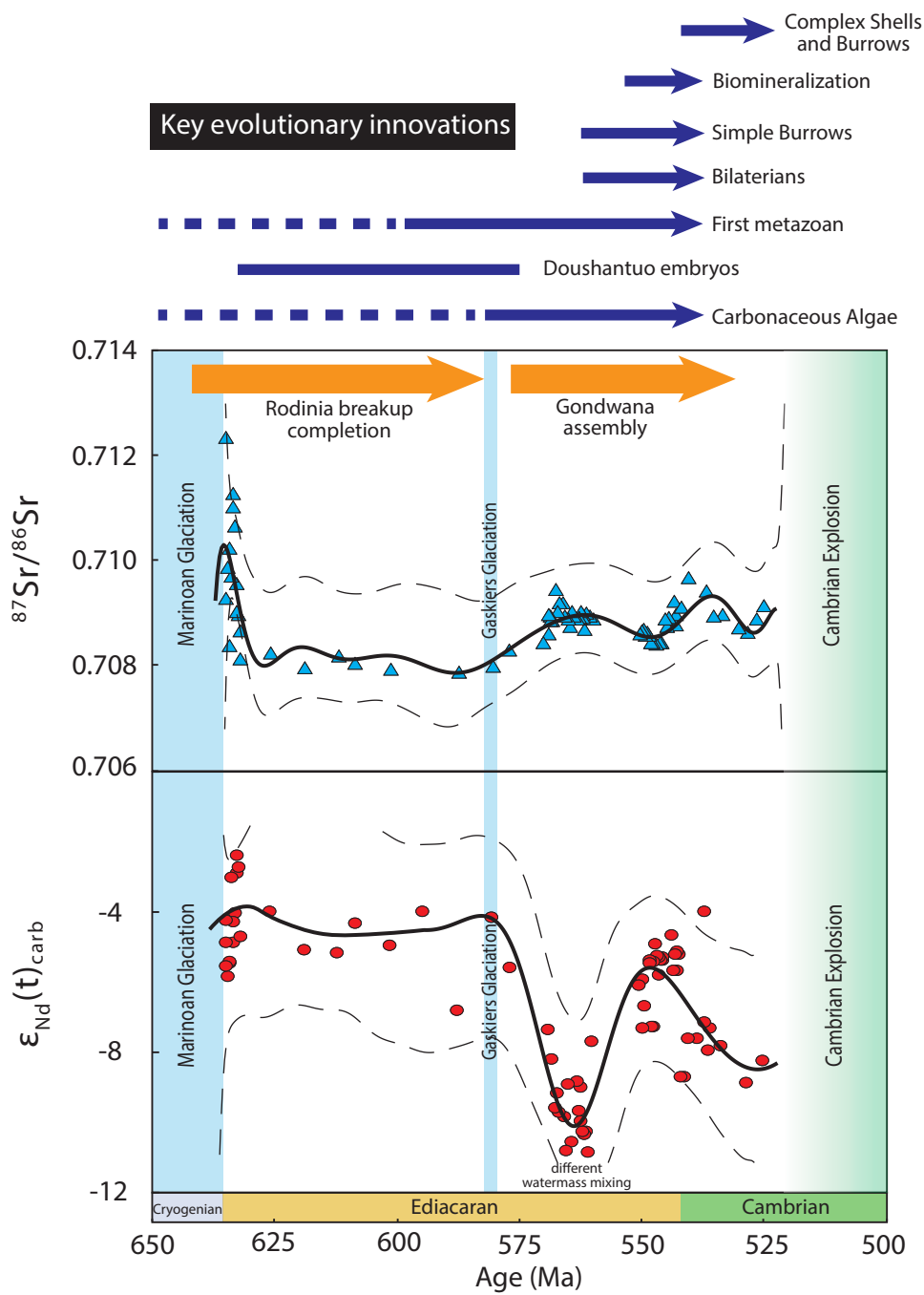


Figure 7