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Integrated carbon, sulfur, and nitrogen isotope chemostratigraphy of the Ediacaran Lantian Formation in South China: Spatial gradient, ocean redox oscillation, and fossil distribution

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Integrated carbon, sulfur, and nitrogen isotope chemostratigraphy of the Ediacaran Lantian Formation in South China: spatial gradient, ocean redox oscillation, and fossil distribution

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Abstract

The Ediacaran Doushantuo Formation in South China is a prime target for geobiological investigation because it offers opportunities to integrate chemostratigraphic and paleobiological data.

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28 Previous studies were mostly focused on successions in shallow-water shelf facies, but data from
29 deep-water successions are needed to fully understand basinal redox structures. Here we report
30 $\delta^{13}\text{C}_{\text{carb}}$, $\delta^{13}\text{C}_{\text{org}}$, $\delta^{34}\text{S}_{\text{pyr}}$, $\delta^{34}\text{S}_{\text{CAS}}$, and $\delta^{15}\text{N}_{\text{sed}}$ data from a drill core of the fossiliferous Lantian
31 Formation, which is a deep-water equivalent of the Doushantuo Formation. Our data confirm a large
32 ($>10\%$) spatial gradient in $\delta^{13}\text{C}_{\text{carb}}$ in the lower Doushantuo/Lantian formations, but this gradient is
33 probably due to the greater sensitivity of carbonate-poor deep-water sediments to isotopic mixing
34 with ^{13}C -depleted carbonate cements. A pronounced negative $\delta^{13}\text{C}_{\text{carb}}$ excursion (EN3) in the upper
35 Doushantuo/Lantian formations, however, is spatially consistent and may be an equivalent of the
36 Shuram excursion. $\delta^{34}\text{S}_{\text{pyr}}$ is more negative in deeper-water than shallow-water facies, particularly in
37 the lower Doushantuo/Lantian formations, and this spatial pattern is interpreted as evidence for
38 ocean redox stratification: pyrite precipitated in euxinic deep waters has lower $\delta^{34}\text{S}_{\text{pyr}}$ than that
39 formed within shallow-water sediments.

40 The Lantian Formation was probably deposited in oscillating oxic and euxinic conditions.
41 Euxinic black shales have higher TOC and TN contents, but lower $\delta^{34}\text{S}_{\text{pyr}}$ and $\delta^{15}\text{N}_{\text{sed}}$ values. In
42 euxinic environments, pyrite was predominantly formed in the water column and organic nitrogen
43 was predominantly derived from nitrogen fixation or NH_4^+ assimilation because of quantitative
44 denitrification, resulting in lower $\delta^{34}\text{S}_{\text{pyr}}$ and $\delta^{15}\text{N}_{\text{sed}}$ values. Benthic macroalgae and putative
45 animals occur exclusively in euxinic black shales. If preserved in-situ, these organisms must have
46 lived in brief oxic episodes punctuating largely euxinic intervals, only to be decimated and preserved
47 when the local environment switched back to euxinia again. Thus, taphonomy and ecology were the
48 primary factors controlling the stratigraphic distribution of microfossils in the Lantian Formation.

49
50 **Key words:** Ediacaran Period, Lantian Formation, South China, carbon isotopes, sulfur isotopes,
51 nitrogen isotopes

52

53

54 INTRODUCTION

55 The co-evolution of life and environment is highlighted in the recent debate on the possible
56 relationship between ocean oxygenation and animal evolution in the Neoproterozoic. Some argue
57 that the progressive oxygenation of the Earth's surface in the late Neoproterozoic removed the final

58 environmental hurdle to animal evolution (Planavsky *et al.*, 2014; Cole *et al.*, 2016), whereas others
59 counter that atmospheric pO₂ levels were sufficiently high to support basal animal metabolism in the
60 Mesoproterozoic, long before the rise of animals themselves (Zhang *et al.*, 2015). In the latter
61 scenario, it is the rise of animals to ecological importance that drove the progressive oxygenation
62 of deep oceans (Lenton *et al.*, 2014). Building upon previous work (Lyons *et al.*, 2009; Och &
63 Shields-Zhou, 2012), recent attempts to test these competing hypotheses are focused on the
64 compilations and meta-analyses of redox proxies including Fe speciation data (e.g., Sperling *et al.*,
65 2015b), redox-sensitive trace elements (e.g., fig. 4 of Sahoo *et al.*, 2016), and stable isotopes (e.g.,
66 Pogge von Strandmann *et al.*, 2015). While these meta-analyses offer a critical view of the
67 big-picture redox trend through the Neoproterozoic, they also omit the more nuanced picture of
68 spatial heterogeneity and often lack the stratigraphic resolution to appreciate the temporal dynamics
69 of Neoproterozoic redox evolution. More importantly, these studies necessarily depend on global
70 stratigraphic correlations to draw conclusions about the co-evolution of life and environment. Such
71 correlations come with uncertainties that are often under appreciated (Xiao *et al.*, 2016). As a
72 complementary approach, one can examine fossiliferous Neoproterozoic successions at high
73 stratigraphic resolution in order to analyze the evolutionary, ecological, and taphonomic factors that
74 control the local and regional distribution of Neoproterozoic organisms. This approach has been
75 applied with success in the investigation of several Ediacaran fossiliferous successions (Fike *et al.*,
76 2006; McFadden *et al.*, 2008; Johnston *et al.*, 2013; Sperling *et al.*, 2015a; Wood *et al.*, 2015; Li *et*
77 *al.*, 2015; Cui *et al.*, 2016a).

78 Ediacaran successions in South China, particularly the Doushantuo Formation, are highly
79 fossiliferous (Xiao *et al.*, 2002; Liu *et al.*, 2014; Xiao *et al.*, 2014). Thus, they are ideal targets for
80 focused and integrated geochemical analysis at high resolution in order to infer the impact of redox
81 conditions on evolution, ecology, and taphonomy. Previous studies of the Doushantuo Formation
82 have been focused on shelf sections in the Yangtze Gorges and surrounding areas, where the
83 Doushantuo Formation is highly fossiliferous and consists of carbonates, shales, and phosphorites
84 (McFadden *et al.*, 2008; Li *et al.*, 2010; Sawaki *et al.*, 2010; Cui *et al.*, 2015). To fully understand the
85 Ediacaran redox structure of South China, it is necessary to gain insights from sections in deep-water
86 slope and basinal facies. However, integrated studies of the Ediacaran successions in deep-water
87 facies are few, and these tend to have limited stratigraphic coverage (Sahoo *et al.*, 2012), have

88 limited stratigraphic resolution (Guo *et al.*, 2007; Och *et al.*, 2016), or come from non-fossiliferous
89 sections (Sahoo *et al.*, 2016; Wang *et al.*, 2016). To address this problem, we carried out an
90 integrated investigation of drill core samples (and supplementary outcrop samples) of the Lantian
91 Formation, which is correlated with the Doushantuo Formation but was deposited in deeper waters
92 and contains exceptionally preserved macroscopic fossils, including putative macrometazoans (Yuan
93 *et al.*, 2011; Wan *et al.*, 2016). In this paper, we present carbonate carbon and oxygen ($\delta^{13}\text{C}_{\text{carb}}$ and
94 $\delta^{18}\text{O}_{\text{carb}}$), organic carbon ($\delta^{13}\text{C}_{\text{org}}$), carbonate-associated sulfate sulfur ($\delta^{34}\text{S}_{\text{CAS}}$), pyrite sulfur
95 ($\delta^{34}\text{S}_{\text{pyr}}$), and sedimentary nitrogen ($\delta^{15}\text{N}_{\text{sed}}$) isotopic compositions of the Lantian Formation. These
96 data provide a first-order chemostratigraphic framework of this unit and shed new light on the redox
97 history and its possible impact on the ecology and taphonomy of the Lantian biota.

98

99 **GEOLOGICAL BACKGROUND**

100 The South China Craton consists of the Yangtze and Cathaysia blocks that amalgamated
101 along the Jiangnan orogen at ~830–820 Ma (Zhao & Cawood, 2012). A rift basin developed to the
102 southeastern margin of the Yangtze block ca. 780 Ma. The rift-to-drift transition occurred during the
103 deposition of Cryogenian sediments. The Ediacaran System on the Yangtze block was deposited on a
104 passive continental margin that deepens to the southeast in present geographic orientation (Jiang *et*
105 *al.*, 2011). Paleogeographic studies show that, during the Ediacaran Period, the Yangtze block
106 consisted of a shelf to the northwest, a deep basin to the southeast, and a narrow slope in between
107 (Fig. 1A) (Jiang *et al.*, 2011).

108 According to various paleogeographic maps (Zhu *et al.*, 2007; Jiang *et al.*, 2011), the Lantian
109 Formation in southern Anhui Province was deposited in basinal facies, but it is also possible that it
110 was deposited in a deep-water but somewhat restricted basin near the enigmatic Poyang Landmass
111 (Fig. 1A). It overlies the terminal Cryogenian diamictite of the Leigongwu Formation, which is
112 equivalent to the Nantuo Formation in the Yangtze Gorges area, and underlies the Piyuancun
113 Formation (“PYC” in Fig. 1C), which contains microfossils characteristic of terminal Ediacaran
114 strata (Dong *et al.*, 2012). A drill core of the Lantian Formation was made near the Lantian village of
115 Xiuning County, Anhui Province (Fig. 1B). In this drill core, the Lantian Formation begins with the
116 cap dolostone or Member I, which is a 4-m-thick unit of light gray siliceous dolostone (Fig. 2A–B).
117 This is overlain by Member II, a 90-m-thick unit consisting of a lower sub-unit of 21 m gray

118 calcareous siltstone interbedded with argillaceous limestone and an upper sub-unit of 69 m black
119 shale with rare argillaceous limestone interbeds (Fig. 2C–D). Carbonaceous compression
120 microfossils and pyritized fossils—including macroalgae (Fig. 2E–F), putative macrometazoans, the
121 enigmatic macrofossil *Orbisiana* (Fig. 2G), and the discoidal fossil *Chuarina* which may be a
122 multicellular eukaryote (Tang *et al.*, 2016)—occur more or less continually in the upper subunit of
123 Member II (Yuan *et al.*, 1999; Yuan *et al.*, 2011; Wan *et al.*, 2014; Wan *et al.*, 2016). Further
124 upsection, Member III is composed of ca. 30 m interbedded gray argillaceous dolostone and black
125 shale, followed by ca. 50 m gray limestone (Fig. 2H–J). Member IV of the uppermost Liantian
126 Formation consists of 10 m black shale (Fig. 2K–L), which is overlain by siliceous rocks of the
127 Piyuancun Formation. Overall, the lithostratigraphic succession intersected by the drill core is similar
128 to those on the outcrop (Wan *et al.*, 2014; Wan *et al.*, 2016), although there are variations in
129 stratigraphic thickness of the four units. The succession is also similar to and can be easily correlated
130 with the four lithostratigraphic members of the Doushantuo Formation in the Yangtze Gorges area
131 (Yuan *et al.*, 2011). Unlike the uncertainty in global correlation of Ediacaran strata (Xiao *et al.*, 2016),
132 the regional correlation between the Liantian and Doushantuo formation is rather straightforward,
133 allowing us to use the radiometric ages from the Yangtze Gorges area to constrain the Liantian
134 Formation between 635 Ma and 551 Ma (Condon *et al.*, 2005).

135

136 METHODS

137 Most samples were collected from the Liantian drill core (29°57.094' N, 118°2.274' E; Fig.
138 1B), but supplementary $\delta^{13}\text{C}_{\text{carb}}-\delta^{18}\text{O}_{\text{carb}}$ samples of Member III were also collected from outcrops
139 near the drill site (29°55.724' N, 118°5.424' E). Sample surfaces were removed to avoid drilling fluid
140 and weathering contaminations. Thin sections were made for petrographic observation. Guided by
141 petrographic observations, powders were taken from polished slabs using a handheld micro-drill,
142 avoiding calcite veins or other visible late diagenetic structures. For comparison and diagenetic
143 evaluation, powders were also micro-drilled from calcite veins. These micro-drilled powders were
144 used for $\delta^{13}\text{C}_{\text{carb}}$ and $\delta^{18}\text{O}_{\text{carb}}$ analyses. In addition, bulk samples were powdered using Retsch RS200
145 for $\delta^{13}\text{C}_{\text{org}}$, $\delta^{15}\text{N}_{\text{sed}}$, $\delta^{34}\text{S}_{\text{CAS}}$, and $\delta^{34}\text{S}_{\text{pyr}}$ analyses.

146 $\delta^{13}\text{C}_{\text{carb}}$ and $\delta^{18}\text{O}_{\text{carb}}$ were measured at the Nanjing Institute of Geology and Palaeontology.
147 An aliquot of 80–100 μg powder was allowed to react with orthophosphoric acid for 150–200

148 seconds at 72°C in a Kiel IV carbonate device connected to a MAT 253 mass spectrometer. CO₂
149 evolved from this reaction was introduced to the mass spectrometer for $\delta^{13}\text{C}_{\text{carb}}$ and $\delta^{18}\text{O}_{\text{carb}}$
150 determination. The standard GBW-04405, with a $\delta^{13}\text{C}$ value of $0.57 \pm 0.03\text{‰}$ and a $\delta^{18}\text{O}$ of $-8.49 \pm$
151 0.13‰ (VPDB), was used for calibration during the analysis. Analytical precision is better than 0.08‰
152 for $\delta^{13}\text{C}_{\text{carb}}$ (VPDB) and 0.1‰ for $\delta^{18}\text{O}$ (VPDB).

153 $\delta^{13}\text{C}_{\text{org}}$, $\delta^{15}\text{N}_{\text{sed}}$, total organic carbon (TOC), and total nitrogen (TN) were analyzed at Indiana
154 University. An aliquot of 1–3 g bulk-sample powder was decarbonated overnight in a Polypropylene
155 centrifuge tube with 3% (V/V) HCl. The process was repeated at least three times to ensure complete
156 removal of carbonate. The carbonate-free residue were then neutralized using Milli-Q water
157 following a stepwise washing procedure, and then dried at 70°C. Samples were loaded into capsules
158 and flash combusted at 1060°C in a Costech Elemental Analyzer (ECS4010) fitted with zero blank
159 autosampler. The resulting CO₂ and N₂ gas were measured by thermal conductivity detector to
160 obtain TOC (wt%) and TN (wt%), then transferred by continuous flow for isotopic ratio
161 determination on a Delta plus XP Isotope Ratio Mass Spectrometer. C/N atomic ratios were
162 calculated from TOC and TN measurements. $^{13}\text{C}/^{12}\text{C}$ and $^{15}\text{N}/^{14}\text{N}$ ratios are reported in the
163 conventional delta notation as per mil deviation from the VPDB and AIR standard, respectively.
164 Calibration of $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values were accomplished using an acetanilide working standard ($\delta^{13}\text{C}$
165 = -27.6‰ and $\delta^{15}\text{N} = 1.61\text{‰}$). Sample $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values were repeatable to a standard error of 0.1‰
166 and 0.32‰ , respectively.

167 $\delta^{34}\text{S}_{\text{CAS}}$ and $\delta^{34}\text{S}_{\text{pyr}}$ were analyzed at Indiana University. To prepare for $\delta^{34}\text{S}_{\text{CAS}}$ analysis,
168 ~200 g powder was rinsed with 10% NaCl and repeated three–five times until no sulfate was present
169 in filtered solutions to remove soluble sulfates (e.g., from sulfide oxidation) and water-soluble sulfate
170 minerals (e.g., anhydrite). CAS was obtained by dissolving the NaCl-rinsed powders in 2N HCl for 2
171 hours at room temperature. Following dissolution, samples were filtered to remove insoluble residues
172 and an excess of 1 M BaCl₂ solution was added to precipitate BaSO₄ overnight for $\delta^{34}\text{S}_{\text{CAS}}$ analysis.
173 To prepare for $\delta^{34}\text{S}_{\text{pyr}}$ analysis, 1–6 g powder was prepared for pyrite extraction using the chromium
174 reduction method (Canfield *et al.*, 1986). Pyrite extraction was performed under N₂ gas by the
175 addition of 6 N HCl and 0.4 M reduced chromium chloride. The reaction was allowed to proceed for
176 at least 3 hours with the sulfide collected as silver sulfide after bubbling through a sodium citrate
177 buffer and into a silver nitrate (0.1 M) trap.

178 Rinsed, filtered, and dried BaSO₄ and Ag₂S precipitates were then combined with an excess
179 of V₂O₅ and analyzed for sulfur isotope composition on a Delta V advantage isotope ratio gas source
180 mass spectrometer fitted with a peripheral Costech Elemental Analyzer (ECS4010) for on-line
181 sample combustion. Sulfur isotope compositions are expressed in standard δ-notation as per mil (‰)
182 deviation from VCDT, with an analytical error of <0.2‰, calculated from replicate analyses of
183 samples and laboratory standards. Analyses were calibrated using four internal standards: silver
184 sulfide (ERE Ag₂S: -4.3‰), chalcopyrite (EMR-CP: +0.9‰), pyrite (PQM2: -14.8‰), and barite
185 (PQB-2: +38.0‰).

186

187 RESULTS

188 Chemostratigraphic data are presented in Table S1 and Fig. 3. The chemostratigraphic profile
189 of δ¹³C_{carb} is similar to those reported previously from outcrop samples at Shiyu (Zhao & Zheng,
190 2009; Wang *et al.*, 2014) and Jinlongshan in the Lantian area (Yuan *et al.*, 2011; see Fig. 1B for
191 locations). Different from the Doushantuo Formation in the Yangtze Gorges area (McFadden *et al.*,
192 2008), δ¹³C_{carb} values of the Lantian Formation range from 0.4‰ to -14.1‰, almost entirely
193 negative values. δ¹³C_{carb} of the cap carbonate (Member I) is around -5‰, decreases to lower values
194 in Member II, and reaches to the lowest value of -14‰ in the fossiliferous upper Member II.
195 Subsequently, δ¹³C_{carb} rises and stabilizes at -3‰ in uppermost Member II and lower Member III,
196 and drops again in dolostone and limestone of upper Member III to values around -10‰; the same
197 pattern is observed at the supplementary outcrop section. δ¹³C_{carb} values of calcite veins are often
198 indistinguishable but are sometimes different from those of host rocks, with δ¹³C_{host rock} - δ¹³C_{vein}
199 ranging from -5.1‰ to 11.0‰ (average = 0.5‰, s.d. = 3.9‰, n = 23).

200 δ¹⁸O_{carb} is mostly lower than -10‰, with the exception of lower Member III. Consistent with
201 previous studies (Zhao & Zheng, 2013; Wang *et al.*, 2014), δ¹⁸O_{carb} of calcite veins is significantly
202 lower than that of host rocks (paired t-test, one-tail $p = 2.8 \times 10^{-6}$; δ¹⁸O_{host rock} - δ¹⁸O_{vein} = 8.1‰ on
203 average, ranging from -1.3‰ to 19.6‰, s.d. = 6.3‰, n = 22).

204 δ¹³C_{org} shows significant variations between -27‰ and -35‰ in the cap carbonate (Member
205 I) and lower Member II. Further upsection, δ¹³C_{org} exhibits more stratigraphic consistency (or less
206 stratigraphic scattering), shifting from a nadir of -35‰ at around 30 m to -30‰ around 70 m and
207 then stabilizing mostly between -29‰ and -30‰ at 70-155 m in upper Member II and much of

208 Member III. There are more variations in uppermost Member III and Member IV, with values
209 ranging from -35‰ to -26‰ . Throughout the Lantian Formation, $\delta^{13}\text{C}_{\text{org}}$ does not seem to co-vary
210 with $\delta^{13}\text{C}_{\text{carb}}$ (Fig. 4D; $R^2 = 0.2$), so that $\Delta\delta^{13}\text{C}_{\text{carb-org}}$ values range from 15.3‰ to 30.0‰ .

211 $\delta^{34}\text{S}_{\text{pyr}}$ profile of the Lantian Formation is broadly similar to but display a more consistent
212 stratigraphic pattern than a previous reconnaissance study (Shen *et al.*, 2008). It can be divided into
213 six intervals, which are highlighted and marked to the right end of Figure 3: (A) mostly positive
214 values in lithostratigraphic Member I and lower Member II (0 to *ca.* 22 m); (B) mostly negative
215 values in the lower part of upper Member II (*ca.* 22–62 m), reaching a minimum of -31.5‰ ; (C) a
216 short interval of positive values in middle part of upper Member II (*ca.* 62–71 m), reaching an apex
217 of 36.5‰ and representing a shift of 68‰ from the minimum in Interval B; (D) mostly negative
218 values in uppermost Member II and lower Member III (*ca.* 71–118 m); (E) mostly positive values in
219 upper Member III (118–174.5 m); and (F) a decreasing trend toward negative values in Member IV
220 (174.5–185 m). Iron speciation data from the same drill core indicate that the redox conditions of the
221 first four intervals oscillate between oxic and euxinic conditions (as marked to the right end of Fig. 3)
222 (Guan, 2014).

223 $\delta^{34}\text{C}_{\text{CAS}}$ data are not available for the lower 60 m of the Lantian Formation because of
224 inappropriate lithology: the cap dolostone is highly siliceous and the lower Member II consist of
225 mostly siltstone and black shale. For the rest of the Lantian Formation, $\delta^{34}\text{C}_{\text{CAS}}$ values were obtained
226 from carbonate interbeds in upper Member II and lower Member III, as well as limestone in upper
227 Member III. These values scatter from 2.2‰ to 32.0‰ (mean = 20.4‰ , standard deviation = 6.8‰)
228 without any clearly defined stratigraphic pattern. There is no significant correlation between $\delta^{34}\text{C}_{\text{CAS}}$
229 and $\delta^{34}\text{S}_{\text{pyr}}$ (Fig. 4E; $R^2 = 0.07$).

230 $\delta^{15}\text{N}_{\text{sed}}$ values range from 2.9‰ to 7.9‰ in the Lantian Formation (average = 5.3‰ , s.d. =
231 1.1‰ , $n = 152$). They vary around 4.5‰ in the first 60 m of the Lantian Formation (or $\delta^{34}\text{S}_{\text{pyr}}$
232 intervals A–B), followed by a positive excursion of over 7.0‰ at *ca.* 70–81 m, a decrease to 2.9‰ at
233 *ca.* 106 m, another minor positive excursion of 6.2‰ at *ca.* 119 m, a broad drop to 3.3‰ at *ca.* 144 m,
234 and a return to values around 6‰ in Member IV.

235 TOC and TN contents show similar variation patterns in the Lantian Formation (positive
236 correlation, $R^2 = 0.5$; Fig. 4A). Atomic ratios of C/N average at 28.0 (s.d. = 28.6, $n = 151$),
237 comparable to those reported from the Doushantuo Formation (Kikumoto *et al.*, 2014). As expected,
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238 black shales in upper Member II, lower Member III, and Member IV have elevated TOC and TN
239 contents. In contrast, the cap dolostone, siltstone in lower Member II, limestone in upper Member III,
240 as well as carbonate interbeds in upper Member II and lower Member III, have lower values. In
241 general, high TOC and TN contents tend to match with low $\delta^{34}\text{S}_{\text{pyr}}$ values ($\delta^{34}\text{S}_{\text{pyr}}$ intervals B, D, F
242 described above) and vice versa.

244 EVALUATION OF POST-DEPOSITIONAL ALTERATION

245 Carbon isotopes

246 $\delta^{13}\text{C}_{\text{carb}}$ can be variably altered during post-depositional interactions with meteoric,
247 diagenetic, and metamorphic fluids (Knauth & Kennedy, 2009; Derry, 2010; Swart, 2015). Indeed,
248 there are reasons to be cautious about the primary origin of the $\delta^{13}\text{C}_{\text{carb}}$ values of the carbonate
249 interbeds in the organic-rich shales of upper Member II and lower Member III. First, the majority of
250 Lantian samples have $\delta^{18}\text{O}_{\text{carb}}$ values less than -10‰ (Fig. 4B). Such low $\delta^{18}\text{O}_{\text{carb}}$ values are
251 indicative of possible diagenetic alteration of $\delta^{13}\text{C}_{\text{carb}}$ values (Kaufman & Knoll, 1995), even though
252 $\delta^{13}\text{C}_{\text{carb}}$ are more rock-buffered than $\delta^{18}\text{O}_{\text{carb}}$ and as a result $\delta^{18}\text{O}_{\text{carb}}$ and $\delta^{13}\text{C}_{\text{carb}}$ are not positively
253 correlated (Fig. 4B). Second, the presence of calcite veins in some intervals of the Lantian Formation
254 suggests the possibility of interaction with diagenetic or hydrothermal fluids (Zhao & Zheng, 2013).
255 Our data show that calcite veins in the Lantian Formation have consistently lower $\delta^{18}\text{O}_{\text{carb}}$ values
256 than the host rock (Fig. 4B–C), suggesting that vein-forming fluids were exogenous. As such,
257 $\delta^{13}\text{C}_{\text{carb}}$ and particularly $\delta^{18}\text{O}_{\text{carb}}$ of the host rock can be affected by interaction with vein-forming
258 fluids, although it can be argued that $\delta^{13}\text{C}_{\text{carb}}$ is less susceptible than $\delta^{18}\text{O}_{\text{carb}}$ to diagenetic or
259 hydrothermal alteration because of the buffering effect of carbonate rocks (Banner & Hanson, 1990).
260 Third, the abundance of black shales in the Lantian Formation raises the possibility that ^{13}C -depleted
261 authigenic or diagenetic carbonate cements, derived from anaerobic oxidation of organic carbon (e.g.,
262 Schrag *et al.*, 2013; Cui *et al.*, 2017), may be a major contributor to the negative $\delta^{13}\text{C}_{\text{carb}}$ values,
263 particularly in upper Member II and lower Member III where TOC content is high. Thus, only
264 $\delta^{13}\text{C}_{\text{carb}}$ values from the organic-lean units (i.e., Member I and upper Member III) are more likely to
265 record primary values.

266 In contrast to $\delta^{13}\text{C}_{\text{carb}}$ data where organic-rich lithologies are more susceptible to
267 post-depositional alteration, primary $\delta^{13}\text{C}_{\text{org}}$ values are likely recorded in organic-rich intervals,

268 including black shales in upper Member II and lower Member III. This interpretation is supported by
269 the observation that the $\delta^{13}\text{C}_{\text{org}}$ profile of the Lantian Formation exhibits the best stratigraphic
270 consistency (or least scattering) in upper Member II and lower Member III where TOC content is
271 generally greater than 0.5 wt%. In contrast, greater stratigraphic variations are observed in Member I,
272 lower Member II, and upper Member III where TOC content is generally <0.5 wt%. When TOC
273 content is low, $\delta^{13}\text{C}_{\text{org}}$ is more susceptible to terrigenous organic carbon contamination and
274 diagenetic alteration (Jiang *et al.*, 2012; Johnston *et al.*, 2012). Thus, upper Member II and lower
275 Member III likely record primary $\delta^{13}\text{C}_{\text{org}}$ values, and the strong $\delta^{13}\text{C}_{\text{org}}$ offset from -35‰ at 30 m to
276 -30‰ at 70 m is a real signal related to primary production. The stratigraphic trends of $\Delta^{13}\text{C}_{\text{carb-org}}$,
277 however, have limited chemostratigraphic value because strata recording the most reliable $\delta^{13}\text{C}_{\text{carb}}$
278 values do not overlap with those recording the most reliable $\delta^{13}\text{C}_{\text{org}}$ values. As a consequence, there
279 is no significant correlation between $\delta^{13}\text{C}_{\text{carb}}$ and $\delta^{13}\text{C}_{\text{org}}$ (Fig. 4D) or between $\delta^{13}\text{C}_{\text{org}}$ and TOC (Fig.
280 4F).

281

282 Sulfur isotopes

283 Sedimentary pyrite in the Lantian Formation is mostly framboidal and cuboidal in
284 morphology (Guan *et al.*, 2014; Guan *et al.*, 2016), but there is no evidence for significant sulfide
285 mineralization related to vein-forming hydrothermal fluids. Additionally, the Lantian Formation
286 contains relatively abundant pyrite (0–9.1%, average = 1.4%, s.d. = 1.6%; Table S1, Fig 3) and
287 shows a stratigraphically consistent $\delta^{34}\text{S}_{\text{pyr}}$ profile. There is no significant correlation between pyrite
288 content and $\delta^{34}\text{S}_{\text{pyr}}$ (Fig. 4G; $R^2 = 0.04$). Thus, $\delta^{34}\text{S}_{\text{pyr}}$ of the Lantian Formation likely records the
289 sulfur isotope signatures of sedimentary pyrite, including both syngenetic pyrite formed in the water
290 column and early authigenic pyrite precipitated in sediments.

291 $\delta^{34}\text{C}_{\text{CAS}}$ values, on the other hand, can be altered by pyrite oxidation (Marenco *et al.*, 2008)
292 and contamination from atmospheric sulfates (Peng *et al.*, 2014), although meteoric diagenesis seems
293 to have a limited impact (Gill *et al.*, 2008). Although $\delta^{34}\text{C}_{\text{CAS}}$ and $\delta^{34}\text{C}_{\text{pyr}}$ do not show significant
294 positive correlation (Fig. 4E), we cannot exclude the possibility of contamination from atmospheric
295 sulfates without ^{17}O data (Peng *et al.*, 2014). Because contamination from both atmospheric sulfates
296 and pyrite oxidation tend to drive $\delta^{34}\text{C}_{\text{CAS}}$ to lower values, $\delta^{34}\text{C}_{\text{CAS}}$ values of the Lantian Formation
297 ($20.4 \pm 6.8\text{‰}$, $n = 40$) likely represent minimum estimates of seawater sulfate sulfur isotopic

298 compositions. Indeed, $\delta^{34}\text{C}_{\text{CAS}}$ values of the Lantian Formation are statistically lower than previously
299 reported $\delta^{34}\text{C}_{\text{CAS}}$ values from the equivalent Doushantuo Formation in South China ($27.9 \pm 8.3\%$,
300 Mann-Whitney test, $p = 2.0 \times 10^{-7}$) (Shields *et al.*, 2004; McFadden *et al.*, 2008; Li *et al.*, 2010; Xiao
301 *et al.*, 2012).

302

303 Nitrogen isotopes

304 Organic matter experiences chemical alteration due to degradation in the water column and
305 sediments, and such alteration is expected to have an impact on $\delta^{15}\text{N}_{\text{sed}}$. In modern marine settings,
306 $\delta^{15}\text{N}$ alteration in the water column and at the water-sediment interface is largely controlled by
307 oxygen exposure time (Robinson *et al.*, 2012), but this factor likely played a weaker role in
308 Ediacaran oceans that were dominated by anoxia. Alteration within marine sediments is largely
309 driven by bacterial decomposition of organic matter, leading to ^{15}N -enrichment in residual organic
310 matter because isotopically light NH_4^+ is released from organic matter to sediment porewater, and
311 this process results in a few ‰ increase in $\delta^{15}\text{N}$ (Lourey, 2003; Thunell *et al.*, 2004; Gaye-Haake *et*
312 *al.*, 2005; Robinson *et al.*, 2012). However, because pore-water NH_4^+ can be effectively scavenged
313 by clay minerals, bulk sediment nitrogen isotopic compositions ($\delta^{15}\text{N}_{\text{sed}}$) can partially compensate
314 for within-sediment alteration and provide a reliable proxy for $\delta^{15}\text{N}$ values of sedimentary organic
315 matter (Higgins, 2012). In the Lantian Formation, TN is relatively abundant in upper Member II and
316 lower Member III, and $\delta^{15}\text{N}_{\text{sed}}$ values in these intervals are better buffered against diagenetic
317 alteration and more likely to preserve primary $\delta^{15}\text{N}$ values. However, because $\delta^{15}\text{N}_{\text{sed}}$ and TN do not
318 show any significant correlation (Fig. 4H), it is possible that $\delta^{15}\text{N}_{\text{sed}}$ values are overall reliable.

319

320 SPATIAL PATTERNS

321 Carbon isotopes

322 $\delta^{13}\text{C}_{\text{carb}}$ of the Lantian Formation is almost entirely in the negative territory, which represents
323 a significant departure from $\delta^{13}\text{C}_{\text{carb}}$ profiles of Ediacaran carbonate successions elsewhere in South
324 China (e.g. Tahata *et al.*, 2013; Wang *et al.*, 2016) and on other continents (Xiao *et al.*, 2016). In
325 particular, $\delta^{13}\text{C}_{\text{carb}}$ values in upper Member II and lower Member III are more than 10‰ lower than
326 those of presumably equivalent units at Jiulongwan and other sections in shelf and upper-slope facies
327 (Jiang *et al.*, 2011; Zhu *et al.*, 2013; Wang *et al.*, 2016). Indeed, all lower-slope and basinal sections

328 of the Doushantuo Formation analyzed so far are lithologically dominated by organic-rich sediments
329 (e.g., black shales) and characterized by almost exclusively negative $\delta^{13}\text{C}_{\text{carb}}$ (Jiang *et al.*, 2007;
330 Wang *et al.*, 2014) (Fig. 5). This facies-dependent spatial pattern was interpreted as evidence for a
331 strong depth-dependent $\delta^{13}\text{C}_{\text{DIC}}$ gradient in excess of $>10\text{‰}$ (Jiang *et al.*, 2007). However, to
332 maintain such a strong gradient for upper Member II and lower Member III (likely representing $>10^7$
333 Myr) would require a strong biological pump to transport isotopically light carbon to the deep ocean
334 and an exceptionally strong supply of phosphorus to sustain the biological pump over millions of
335 years. The depth gradient of $\delta^{13}\text{C}_{\text{DIC}}$ in modern ocean is only $\sim 2\text{‰}$, and modeling results show that
336 to sustain a 10‰ depth gradient requires a combination of biological pump $2\times$ modern value (as
337 measured by the e-folding depth, i.e., the depth by which $1/e$ or $\sim 37\%$ of the organic carbon exported
338 from the surface ocean remains) and a phosphorus supply $>5\times$ modern value (Meyer *et al.*, 2016).
339 Given that Ediacaran primary producers were likely dominated by non-skeletal and slow sinking
340 organisms relative to their modern counterparts (e.g., diatoms, coccolithophorans, fecal pellets, and
341 other large-celled eukaryotes), the biological pump in Ediacaran oceans was unlikely much stronger
342 than its modern counterpart (Lenton *et al.*, 2014). Also, considering the negative feedbacks that
343 stabilize the long-term phosphorus cycles (Van Cappellen & Ingall, 1996), it is difficult to envision a
344 sustained phosphorus supply $>5\times$ modern level. Thus, we regard that a $>10\text{‰}$ depth gradient in
345 $\delta^{13}\text{C}_{\text{DIC}}$ is unlikely to sustain for millions of years and we must seek alternative explanations to
346 account for the spatial pattern of $\delta^{13}\text{C}_{\text{carb}}$ observed in South China.

347 As an alternative explanation, we explore the possibility that post-depositional calcite
348 cements derived from anaerobic oxidation of organic carbon may be a significant contribution to the
349 low $\delta^{13}\text{C}_{\text{carb}}$ values in the organic-rich and carbonate-poor sediments deposited in basinal facies
350 (Schrag *et al.*, 2013; Cui *et al.*, 2017). This hypothesis implies that carbonate-poor sediments such as
351 shales and siltstones are more sensitive to isotopic mixing of ^{13}C -depleted cements, which is
352 expected to be $<50\%$ of rock volume. The plausibility of this hypothesis can be evaluated using a
353 simple mixing model that involves two endmembers (Johnston *et al.*, 2012): (1) a depositional
354 component (i.e., sediments) with carbonate percentage of X_A and carbonate carbon isotopic
355 composition of $\delta^{13}\text{C}_A$, and (2) a post-depositional component (i.e., pure calcite cement) with
356 carbonate percentage of $X_B = 1$ and carbonate carbon isotopic composition of $\delta^{13}\text{C}_B$. When these
357 two endmembers are mixed at a mass ratio of $f_A : f_B$ (where $f_A + f_B = 1$), the mixture would have a

358 carbonate content (in percentage) of $X_M = f_A * X_A + f_B * X_B$. The carbonate carbon isotopic
359 composition of the mixture ($\delta^{13}C_M$) would be defined by both the mixing ratio and the carbonate
360 content of the two components: $\delta^{13}C_M = (\delta^{13}C_A * f_A * X_A + \delta^{13}C_B * f_B * X_B) / X_M$. We assume
361 $\delta^{13}C_A = 0\text{‰}$ (considering the maximum $\delta^{13}C_{carb}$ value of 0.4‰ in upper Member II and lower
362 Member III) and $\delta^{13}C_B = -30\text{‰}$ (considering the likely source of ^{13}C -depleted post-depositional
363 calcite from the oxidation of organic carbon in Member II and lower Member III, which has $\delta^{13}C_{org}$
364 values mostly between -35‰ and -29‰). In this mixing model, calcareous black shales in upper
365 Member II and lower Member III, which typically have TOC content of 1–10 wt%, carbonate
366 content of <50 wt% (and mostly <10 wt%), and $\delta^{13}C_{carb}$ values between 0.4‰ and -13‰ , can be
367 mathematically accounted for when $f_B < 5\%$ and $X_A = 1\text{--}50\%$ (Fig. 6). In other words, only a minor
368 amount of post-depositional cements (i.e., <5% of rock mass) is needed to explain the carbonate
369 content and $\delta^{13}C_{carb}$ data of calcareous black shales in upper Member II and lower Member III. For
370 the carbonate interbeds in upper Member II and lower Member III, a greater amount of cements (but
371 still <20% of rock mass) is implied for the majority of samples (Fig. 6). These predictions make
372 petrographic sense because cements are expected to be <50% of rock mass or volume. This
373 hypothesis should be investigated in the future using petrographically guided microanalysis or SIMS
374 analysis of $\delta^{13}C_{carb}$ (Drake *et al.*, 2015; Cui *et al.*, 2016b).

375 Mathematically, negative $\delta^{13}C_{carb}$ values in Member I (cap carbonate) and upper Member III
376 can also be explained by the mixing model described above. However, $\delta^{13}C_{carb}$ values of Member I
377 and upper Member III—unlike those of upper Member II and lower Member III—seem to show
378 regional and global consistency; in other words, a $>10\text{‰}$ depth gradient is not observed in these units.
379 For example, basal Ediacaran cap carbonates in South China and other continents have consistent
380 $\delta^{13}C_{carb}$ values around -5‰ and this is also true for Member I in the Lantian Formation (Fig. 3).
381 Similarly, the negative $\delta^{13}C_{carb}$ excursion in upper Member III has been known from many sections
382 across the Yangtze block and has been interpreted as equivalent to the Shuram negative $\delta^{13}C_{carb}$
383 excursion (Wang *et al.*, 2014), and potential examples of the Shuram excursion have been reported
384 from numerous Ediacaran successions around the world (Grotzinger *et al.*, 2011). These excursions
385 all have nadir $\delta^{13}C_{carb}$ values around -10‰ . Although it is possible that authigenic/diagenetic calcite
386 may have played a role in the origin of the Shuram excursion (Macouin *et al.*, 2012; Cui *et al.*, 2016b,
387 2017; Furuyama *et al.*, 2016), the possibility that a globally synchronous diagenetic event

388 (Grotzinger *et al.*, 2011; Schrag *et al.*, 2013; Cui *et al.*, 2017) needs to be investigated in order to
389 determine whether the Shuram excursion can be used as a chemostratigraphic marker (Xiao *et al.*,
390 2016).

391 The overall $\delta^{13}\text{C}_{\text{org}}$ profile of the Lantian Formation is broadly similar to that of the
392 Doushantuo Formation at the inner shelf Northern Xiaofenghe section (Xiao *et al.*, 2012), intra-shelf
393 Jiulongwan section (McFadden *et al.*, 2008), outer shelf Zhongling section (Li *et al.*, 2010; Cui *et al.*,
394 2015), and the upper slope Siduping section in South China (Wang *et al.*, 2016). Although the
395 negative excursion of $\delta^{13}\text{C}_{\text{org}}$ in Member III around 160–170 m may be compared with those in
396 Member IV of the Doushantuo Formation in the Yangtze Gorges area (McFadden *et al.*, 2008) or the
397 Shuram Formation in Oman (Fike *et al.*, 2006; Lee *et al.*, 2015), the best preserved $\delta^{13}\text{C}_{\text{org}}$ values of
398 the Lantian Formation are in the organic-rich sediments in upper Member II and lower Member III.
399 There is a gradual negative shift from variable values in lower Member II to -35‰ in middle
400 Member II, followed by a gradual positive shift to about -32‰ that terminates upward with an offset
401 to about -30‰ . Above this offset, there is a plateau of values between -28‰ and -30‰ . A possible
402 equivalent of this excursion to -35‰ is seen in the lower Doushantuo Formation at the upper slope
403 Siduping section and perhaps also at the shelf margin Zhongling section, but not present in the inner
404 shelf and intra-shelf sections at Northern Xiaofenghe and Jiulongwan (Fig. 7). Consistent $\delta^{13}\text{C}_{\text{org}}$
405 values between -28‰ and -30‰ are characteristic of lower-middle Doushantuo Formation at
406 Northern Xiaofenghe and Jiulongwan sections, but slightly more variable and greater values
407 characterize presumably equivalent strata at the Zhongling section (between -25‰ and -27‰) and
408 Siduping section (between -26‰ and -30‰). Other Doushantuo sections that have been analyzed
409 for $\delta^{13}\text{C}_{\text{org}}$ values tend to have relatively low stratigraphic resolution (Guo *et al.*, 2006; Guo *et al.*,
410 2007; Ader *et al.*, 2009) to warrant a reliable comparison with the Lantian Formation for analysis of
411 spatial variations.

412

413 Sulfur isotopes

414 Spatial variations in $\delta^{34}\text{S}_{\text{pyr}}$ and $\delta^{34}\text{S}_{\text{CAS}}$ of Ediacaran successions in South China are shown
415 in Fig. 8. Although $\delta^{34}\text{S}_{\text{pyr}}$ shows significant stratigraphic variations, there is a clear spatial trend that
416 can be discerned from the data. The most important spatial pattern is an overall decrease in $\delta^{34}\text{S}_{\text{pyr}}$
417 values from shallow to deep facies, particularly in the lower Doushantuo and Lantian formations (Fig.

418 8). For example, $\delta^{34}\text{S}_{\text{pyr}}$ values of the Doushantuo Formation at the inner shelf section (Northern
419 Xiaofenghe) and outer shelf sections (Zhongling) are almost entirely positive, mostly 20–40‰ in the
420 lower Doushantuo but slide to 0–20‰ in the upper Doushantuo (Zhongling). In deeper intra-shelf
421 facies (Jiulongwan), $\delta^{34}\text{S}_{\text{pyr}}$ values are mostly 0–40‰ in the lower Doushantuo Formation but
422 decrease to mostly negative values (between 0‰ and –20‰) in the upper Doushantuo Formation. In
423 still deeper sections in slope and basinal facies (e.g., Wuhe and Lantian sections), $\delta^{34}\text{S}_{\text{pyr}}$ values
424 become increasingly negative.

425 There are not sufficient $\delta^{34}\text{S}_{\text{CAS}}$ data from slope and basinal sections of the Doushantuo
426 Formation, because these sections tend to be dominated by black shales. Additionally, the issue of
427 contamination might compromise the reliability of $\delta^{34}\text{S}_{\text{CAS}}$ values as a proxy for seawater sulfate
428 (Marenco *et al.*, 2008; Peng *et al.*, 2014). Thus, the big picture spatial pattern of $\delta^{34}\text{S}_{\text{CAS}}$ is not
429 discernable from currently available data. Nonetheless, $\delta^{34}\text{S}_{\text{CAS}}$ values of the Lantian Formation are
430 broadly comparable to, although slightly lower than, those of the Doushantuo Formation at Northern
431 Xiaofenghe, Jiulongwan, and Zhongling (Fig. 8), all with a large range between 0‰ and 40‰.

432 Following previous authors (Li *et al.*, 2010; Xiao *et al.*, 2012), the observed spatial pattern of
433 $\delta^{34}\text{S}_{\text{pyr}}$ is best interpreted as evidence for oceanic redox stratification with euxinic deep waters and
434 non-euxinic (and probably oxic) shallow waters. When pyrite precipitation occurs in euxinic deep
435 waters, lower $\delta^{34}\text{S}_{\text{pyr}}$ values are expected because sulfate reducing microbes have full access to
436 marine sulfate, which was already a few mM in Ediacaran oceans (Hurtgen *et al.*, 2002), and
437 maximum sulfur isotope fractionation is possible. In contrast, when pyrite precipitation occurs within
438 sediments in shallow-water facies, sulfate availability is limited by diffusion and high $\delta^{34}\text{S}_{\text{pyr}}$ values
439 are expected. Following this simplistic model, it is inferred that Doushantuo sediments in inner and
440 outer shelf facies were mostly deposited in non-euxinic (and probably oxic) conditions, whereas
441 euxinic conditions frequented deeper waters in intra shelf, slope, and basinal facies (Jiang *et al.*, 2011;
442 Muscente *et al.*, 2015). This simple model offers a first-order framework for us to interpret the
443 spatial (and also temporal; see below) variation in $\delta^{34}\text{S}_{\text{pyr}}$ and redox conditions.

444 Of course, the reality is likely more complex because many other factors also control the
445 apparent fractionation of sulfur isotopes between sulfate and pyrite. These factors include, among
446 others, primary production and availability of organic matter (Leavitt *et al.*, 2013), marine sulfate
447 concentration (Gomes and Hurtgen, 2015; Bradley *et al.*, 2016), cell-specific sulfate reduction rate

448 (Sim *et al.*, 2012; Bradley *et al.*, 2016), and oxidative recycling of sulfide (Fike *et al.*, 2015).
449 Importantly, Gomes and Hurtgen (2015) have shown that, in modern euxinic environments, the
450 relationship between $\delta^{34}\text{S}_{\text{sulfide}}$ and sulfate concentration is not a uniform one: the reservoir effect
451 dominates when sulfate concentration is low (<5 mM), whereas sulfate reduction rates and location
452 of pyrite formation in a euxinic water column become more important when sulfate concentration is
453 high. Given that the Ediacaran oceans may have had a sulfate concentration of about 5 mM (Hurtgen
454 *et al.*, 2002) and may have had a euxinic wedge (Li *et al.*, 2010), these factors are relevant and may
455 have been important in affecting the $\delta^{34}\text{S}_{\text{pyr}}$ record of the Lantian Formation. Although we regard
456 pyrite formation in a euxinic water column vs. in sediments as a primary control because the
457 congruence between $\delta^{34}\text{S}_{\text{pyr}}$ values and redox conditions, we do recognize that this is a simple model
458 and other factors must be considered to explain the second-order variation in the $\delta^{34}\text{S}_{\text{pyr}}$ record of the
459 Lantian Formation.

460

461 **Nitrogen isotopes**

462 Biologically available nitrogen in modern oceans has a rather short residence time relative to
463 oceanic circulation (Gruber, 2008). Thus, $\delta^{15}\text{N}$ of nitrate, organic carbon, and seafloor bulk
464 sediments in modern global oceans show significant spatial variations over 10‰ (Gaye-Haake *et al.*,
465 2005; Tesdal *et al.*, 2013). Given these variations, it is remarkable that $\delta^{15}\text{N}_{\text{sed}}$ values of the Lantian
466 Formation are broadly consistent with those of the Doushantuo Formation in South China (Ader *et*
467 *al.*, 2014; Kikumoto *et al.*, 2014) (Fig. 9). Although the Lantian Formation (average $\delta^{15}\text{N}_{\text{sed}} = 5.3\text{‰}$)
468 has statistically higher values than the Doushantuo Formation at Yangjiaping (average $\delta^{15}\text{N}_{\text{sed}} =$
469 4.4‰) and Yangtze Gorges area of South China (average $\delta^{15}\text{N}_{\text{sed}} = 4.8\text{‰}$), the differences in average
470 $\delta^{15}\text{N}_{\text{sed}}$ values are within 1‰. In terms of $\delta^{15}\text{N}_{\text{sed}}$ values, the Doushantuo and Lantian formations are
471 broadly similar to some Ediacaran successions (e.g., $^{15}\text{N}_{\text{sed}} = 5.1 \pm 0.5\text{‰}$ in Svalbard, and $5.1 \pm 2.0\text{‰}$
472 in Brazil) but sharply different from others (e.g., $\delta^{15}\text{N}_{\text{sed}} = 2.9 \pm 0.6\text{‰}$ in northwestern Canada)
473 (Ader *et al.*, 2014; Spangenberg *et al.*, 2014). They are also distinct from Cambrian successions in
474 South China ($\delta^{15}\text{N}_{\text{sed}} = 1.1 \pm 2.1\text{‰}$) (Cremonese *et al.*, 2013; Wang *et al.*, 2013; Cremonese *et al.*,
475 2014; Wang *et al.*, 2015) and Cryogenian successions of South China ($\delta^{15}\text{N}_{\text{sed}} = 6.6 \pm 0.7\text{‰}$) (Wei *et*
476 *al.*, 2016). This overall similarity in $\delta^{15}\text{N}_{\text{sed}}$ values of the Doushantuo and Lantian formations in
477 South China, if confirmed with additional data in the future, suggests that basinal rather than local

478 factors controlled the nitrogen cycle.

479

480 **TEMPORAL REDOX OSCILLATION**

481 On the basis of integrated petrographic and geochemical data, we propose that the Lantian
482 Formation records oscillating redox conditions between likely euxinic and oxic environments.
483 Evidence in support of this hypothesis comes from iron speciation data from the same drill core,
484 which show $\delta^{34}\text{S}_{\text{pyr}}$ intervals A and C in Fig. 3 were likely deposited in oxic conditions, whereas
485 intervals B and D in euxinic conditions (Guan, 2014). This interpretation is supplemented by the
486 congruence of lithological, TOC, TN, and $\delta^{34}\text{S}_{\text{pyr}}$ data. Euxinic intervals with organic-rich shales (B,
487 D, and possibly F in Fig. 3) are expected to have high TOC and TN contents, but low and negative
488 $\delta^{34}\text{S}_{\text{pyr}}$ values. In contrast, oxic intervals with organic-lean carbonates and siltstones (A, C, and
489 possibly E in Fig. 3) are expected to have low TOC and TN contents, but high and positive $\delta^{34}\text{S}_{\text{pyr}}$
490 values. Other independent evidence supportive of this model comes from framboidal pyrite data.
491 Pyrite framboids in black shales of intervals B and D are largely 4–8 μm in size (Guan *et al.*, 2014),
492 consistent with syngenetic pyrite precipitation in euxinic water column (Wilkin *et al.*, 1996),
493 particularly considering that the Lantian data probably represent a maximum estimate of framboid
494 size because of possible pyrite overgrowth (Wacey *et al.*, 2015). Thus, it is possible that oscillating
495 euxinic and oxic conditions are characteristic of the Lantian Formation (Fig. 3), although the redox
496 condition of interval E–F remains to be confirmed with Fe speciation data. In this oscillating redox
497 model, pyrite with high and positive $\delta^{34}\text{S}_{\text{pyr}}$ values was largely precipitated in sediments during oxic
498 intervals A, C, and E, whereas that with low and negative $\delta^{34}\text{S}_{\text{pyr}}$ values was mostly formed in
499 euxinic water column during intervals B, D, and F. The oscillating euxinic and oxic conditions in the
500 Lantian Formation are somewhat similar to those recorded in the time-equivalent Doushantuo
501 Formation in deep-water basinal facies (Sahoo *et al.*, 2016). However, $\delta^{34}\text{S}_{\text{pyr}}$ seems to respond
502 differently in the Doushantuo Formation, where more negative $\delta^{34}\text{S}_{\text{pyr}}$ values occur in oxic events
503 and more positive $\delta^{34}\text{S}_{\text{pyr}}$ values in euxinic intervals—a pattern that was interpreted in terms of
504 sulfate reservoir effect (Sahoo *et al.*, 2016), which would be most prominent when seawater sulfate
505 concentration was less than 5 mM (Gomes and Hurtgen, 2015). Nonetheless, the oscillating redox
506 conditions documented in the Lantian and Doushantuo formations are intriguing, although an exact
507 correlation of redox conditions between these two formations is premature.

508 Redox oscillation in the Lantian Formation can be evaluated using other geochemical data.
509 Due to post-depositional alteration and inappropriate lithologies, reliable $\delta^{13}\text{C}_{\text{carb}}$ data only available
510 in the cap carbonate and upper Member III, and $\delta^{34}\text{S}_{\text{CAS}}$ data in carbonate interbeds in Member II–III.
511 Thus, they do not have sufficient stratigraphic coverage to allow in-depth evaluation of the redox
512 oscillation model. $\delta^{15}\text{N}_{\text{sed}}$ and $\delta^{13}\text{C}_{\text{org}}$ data, on the other hand, do have a relatively continuous record
513 in the Lantian Formation, particularly upper Member II and lower Member III (or roughly $\delta^{34}\text{S}_{\text{pyr}}$
514 intervals B–D). It is interesting to note that, in intervals B–D, statistically lower $\delta^{15}\text{N}_{\text{sed}}$ values tend
515 to occur in the euxinic intervals B and D, whereas higher $\delta^{15}\text{N}_{\text{sed}}$ values in the oxic interval C. A
516 similar correlation may be present in $\delta^{13}\text{C}_{\text{org}}$ variations, although this is more dubious because
517 $\delta^{13}\text{C}_{\text{org}}$ in interval C is not particularly higher than in interval D. Below we explore the possibility
518 that variations in $\delta^{15}\text{N}_{\text{sed}}$ and $\delta^{13}\text{C}_{\text{org}}$ in intervals B–D may reflect redox fluctuation.

519 The most important control on $\delta^{15}\text{N}_{\text{sed}}$ at a global scale is denitrification, which anaerobically
520 reduces NO_3^- to N_2 and involves a significant isotopic fractionation ($\epsilon = 20\text{--}30\%$) (Altabet, 2006).
521 Globally, denitrification needs to be complemented by nitrification, which aerobically oxidizes NH_4^+
522 to NO_3^- . Coupled nitrification-denitrification has been established probably since the Archean
523 (Godfrey & Falkowski, 2009), and we assume that it was also part of the nitrogen cycle in Ediacaran
524 as in modern oceans. In modern oceans, $\delta^{15}\text{N}$ of NO_3^- is elevated in the oxygen minimum zone
525 (OMZ) where denitrification preferentially removes isotopically light NO_3^- to form N_2 . As
526 denitrification proceeds, $\delta^{15}\text{N}$ of NO_3^- (and hence $\delta^{15}\text{N}$ of microorganisms that assimilate NO_3^-)
527 becomes progressively higher as $[\text{NO}_3^-]$ decreases (Altabet & Francois, 1994), and reaches the
528 highest (e.g., 18.7‰) in OMZ under the influence of strong oceanic upwelling and with dissolved
529 oxygen content below 1–2 μM (i.e., sub-oxic to hypoxic conditions) (Voss *et al.*, 2001). However, in
530 euxinic basins where denitrification is quantitative and NO_3^- is exhausted at the chemocline,
531 denitrification does not have an isotopic influence on $\delta^{15}\text{N}$ of organic matter because NH_4^+ and
532 nitrogen fixation, rather than NO_3^- , become the main sources of organic nitrogen. For example, $\delta^{15}\text{N}$
533 of seafloor sediments in the euxinic Cariaco Basin is only 3.5‰ (Thunell *et al.*, 2004). Thus, $\delta^{15}\text{N}_{\text{sed}}$
534 is expected to have a complex relationship with redox conditions (Ader *et al.*, 2014). For euxinic
535 condition to develop, NO_3^- must have been exhausted to allow microbial sulfate reduction to prevail
536 (Canfield, 2006). Thus, $\delta^{15}\text{N}_{\text{sed}}$ of euxinic environments is expected to be low because of NO_3^-
537 exhaustion and hence minimal influenced of denitrification on $\delta^{15}\text{N}_{\text{sed}}$ (Thunell *et al.*, 2004). $\delta^{15}\text{N}_{\text{sed}}$

538 of suboxic environments, on the other hand, would have highly elevated values (>10‰) because
539 of ^{15}N -enrichment related to incomplete denitrification (Quan *et al.*, 2008). However, $\delta^{15}\text{N}_{\text{sed}}$ of oxic
540 environments in OMZ margins and open oceans would approach the modal $\delta^{15}\text{N}_{\text{sed}}$ value of 6‰ of
541 modern marine sediments (Tesdal *et al.*, 2013).

542 Viewed in this light, it is possible to explain the $\delta^{15}\text{N}_{\text{sed}}$ profile of the Lantian Formation
543 under the redox oscillation model. The most reliable $\delta^{15}\text{N}_{\text{sed}}$ values are preserved in upper Member II
544 and lower Member III where TN content is relatively high. Low $\delta^{15}\text{N}_{\text{sed}}$ values (3–5‰) occur in
545 $\delta^{34}\text{S}_{\text{pyr}}$ interval B and the upper part of interval D, which are independently interpreted as
546 representing euxinic environments based on congruent TOC, TN, $\delta^{34}\text{S}_{\text{pyr}}$, and Fe speciation data.
547 Such values are similar to $\delta^{15}\text{N}$ of modern euxinic basins such as the Cariaco Basin (Thunell *et al.*,
548 2004). The highest $\delta^{15}\text{N}_{\text{sed}}$ values occur in $\delta^{34}\text{S}_{\text{pyr}}$ interval C and the lower part of interval D. Interval
549 C is independently interpreted as representing oxic environments, and its $\delta^{15}\text{N}_{\text{sed}}$ values are similar to
550 those of modern open marine sediments (Tesdal *et al.*, 2013). The high $\delta^{15}\text{N}_{\text{sed}}$ values in the lower
551 part of interval D are enigmatic, but they could represent partial denitrification in a suboxic
552 environment at the transition from an oxic condition (interval C) to a euxinic condition (interval D).
553 On the other hand, the Lantian Formation lacks strongly positive $\delta^{15}\text{N}_{\text{sed}}$ values characteristic of
554 suboxic upwelling zones in modern oceans (Voss *et al.*, 2001; Gaye-Haake *et al.*, 2005; Tesdal *et al.*,
555 2013), possibly due to generally low $[\text{NO}_3^-]$ levels in Ediacaran oceans so that nearly quantitative
556 denitrification may have occurred even in suboxic conditions.

557 $\delta^{15}\text{N}_{\text{sed}}$ values of intervals A, E, and F do not seem to be compatible with the simple redox
558 model presented above. However, intervals A and E have relatively low TN contents and interval F
559 has rather low stratigraphic sampling intensity. Thus, it is possible that $\delta^{15}\text{N}_{\text{sed}}$ values of intervals A,
560 E, and F are not reliable, that the inference of local redox conditions based on Fe speciation and
561 $\delta^{34}\text{S}_{\text{pyr}}$ data is not reliable, or that the model about the relationship between $\delta^{15}\text{N}_{\text{sed}}$ and redox
562 conditions is too simplistic. These possibilities need to be investigated in the future.

563 The relationship between $\delta^{13}\text{C}_{\text{org}}$ and redox fluctuation in intervals B–D is less clear.
564 However, it is possible that the lower $\delta^{13}\text{C}_{\text{org}}$ values in euxinic intervals, particularly interval B, were
565 related to recycling of organic carbon in the system. In other words, photosynthetic organic carbon
566 can be anaerobically oxidized (e.g., by sulfate reduction bacteria) and reutilized by chemoautotrophs.
567 Consequently, both photosynthetic and chemosynthetic processes may have made contributions to

568 TOC. This scenario can be tested in the future by comparing $\delta^{13}\text{C}_{\text{org}}$ of TOC and photosynthetic
569 macroalgae in the Lantian Formation.

570 What may have caused the oscillating redox conditions in the Lantian Formation? It is
571 tempting to speculate that global redox changes may have placed an overarching control on the local
572 redox conditions as recorded in the Lantian Formation. Indeed, Sahoo *et al.* (2016) proposed on the
573 basis of redox-sensitive trace metals that global Ediacaran deep oceans were predominantly
574 anoxic/euxinic, with oceanic oxygenation events at 635–630 Ma (e.g., basal Doushantuo Formation),
575 ~580 Ma (e.g., lower Member III of the Doushantuo Formation), and ~560 Ma (Member IV of
576 Doushantuo Formation). The absolute age of these oxygenation events can be a matter of debate, and
577 a correlation of Sahoo *et al.*'s (2016) global oxygenation events with the oxic intervals in the Lantian
578 Formation is also an open question. For example, although oxic interval A in the Lantian Formation
579 is likely correlated with the basal Ediacaran oxygenation event in Member I and lower Member II of
580 the Doushantuo Formation, it is uncertain whether the oxic interval C in upper Member II of the
581 Lantian Formation is correlated with the oxygenation event recorded in lower Member III in the
582 Doushantuo Formation (Sahoo *et al.*, 2016).

583 Regional and local factors may have also played a role in modulating the redox condition in
584 the Lantian Formation. Certainly, regional sea level changes, continental weathering, hydrographic
585 factors, and oceanic upwelling may have influenced local productivity, which is a primary factor
586 controlling redox conditions, chemocline depth, sulfate reduction, and denitrification (and hence
587 $\delta^{13}\text{C}_{\text{org}}$, $\delta^{34}\text{S}_{\text{pyr}}$, and $\delta^{15}\text{N}_{\text{sed}}$). Similarly, varying sedimentation rates may be a factor controlling TOC
588 and TN contents. The observation that oxic conditions largely occur in organic-lean carbonates and
589 siltstones whereas euxinic conditions in organic-rich shales suggests that local and regional factors
590 must have modulated the redox conditions in the Lantian Formation. Because of these local controls,
591 it is not always straightforward to correlate redox history among different basins—for example
592 between South China (Sahoo *et al.*, 2016) and northwestern Canada (Sperling *et al.*, 2015a), or even
593 within the same basin—for example between the Member IV of the Doushantuo Formation at
594 Miaohe and Jiulongwan (Li *et al.*, 2015) or between the Doushantuo Formation and the Lantian
595 Formation (Sahoo *et al.*, 2016; this paper).

596

597 **FOSSIL PRESERVATION**

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598 Macroscopic eukaryote fossils occur mostly in the black shales of upper Member II, or in
599 euxinic intervals B and D (Fig. 3). These fossils include pyrite molds of *Chuarina* (Yuan *et al.*, 2001;
600 Guan *et al.*, 2016) as well as a variety of carbonaceous compressions including the modular fossil
601 *Orbisiana* (Wan *et al.*, 2014), macroalgal fossils such as *Flabellophyton* and *Doushantuophyton*
602 (Yuan *et al.*, 1999), and putative animal fossils such as *Lantianella* and *Xiuningella* (Yuan *et al.*,
603 2011; Wan *et al.*, 2016). So far, no fossils have been discovered from other units of the Lantian
604 Formation, and attempts to recover microfossils using acid digestion techniques have been futile.
605 Thus, fossils appear to be restricted largely, if not exclusively, to euxinic black shales, although not
606 all euxinic black shales contain fossils.

607 What controls the stratigraphic distribution of fossils in the Lantian Formation? In principle,
608 evolutionary, ecological, and taphonomic factors may provide the answer. However, evolutionary
609 factors (i.e., fossil taxa of the Lantian Formation are evolutionarily restricted to intervals B and D)
610 can be ruled out. For example, several Lantian genera (i.e., *Doushantuophyton* and *Enteromorphites*)
611 range into the younger Miaohu biota in Member IV of the Doushantuo Formation (Xiao *et al.*, 2002)
612 but so far have not been discovered in members III-IV of the Lantian Formation. This absence in
613 members III-IV of the Lantian Formation, if confirmed in future investigation, must be related to
614 ecological and taphonomic factors.

615 Both ecological and taphonomic factors probably played an important role in controlling the
616 stratigraphic distribution of Lantian fossils, although it is difficult to tease apart these factors because
617 both are related to depositional environments. The lack of fossils in carbonates and siltstones of the
618 Lantian Formation could be simply due to unfavorable environments or water depths for the
619 colonization or preservation of Lantian fossils. However, the lack of macrofossils in black shales of
620 Member IV is enigmatic. It is possible that black shales in Member II and Member IV represent
621 different redox conditions despite their similar appearance. For example, Li *et al.* (2015) and Och *et*
622 *al.* (2016) have shown, on the basis of Fe speciation and trace metal data, that Member IV black
623 shales of the Doushantuo Formation at sections separated by a few kilometers are characterized by
624 differing redox conditions (euxinic vs. ferruginous). Whether black shales in Member II and Member
625 IV of the Lantian Formation represent different redox conditions need to be tested with Fe speciation
626 and redox-sensitive trace metal data.

627 If the Lantian fossils were preserved in-situ (Yuan *et al.*, 2011; Wan *et al.*, 2014), then the

628 presence of macroalgae and putative animals in upper Member II of the Lantian Formation implies
629 oxic conditions at least at ecological time scales. To reconcile these oxygen-breathing organisms and
630 their preservation in euxinic black shales in intervals B and D, Yuan *et al.* (2011) speculated that
631 these black shales were deposited in largely euxinic environments punctuated by brief oxic episodes,
632 when macroalgae and putative animals thrived, only to be subsequently killed and preserved by
633 frequent switch-back to euxinic conditions. This hypothesis is extremely difficult to test, given that
634 each chemostratigraphic sample likely integrates geochemical signals over several centimeters of
635 strata, thus lacking the stratigraphic resolution to detect redox changes at ecological time scales
636 (Sperling *et al.*, 2015a). To make the task more challenging, it is possible that a euxinic water
637 column would likely overprint the geochemistry of sediments deposited in a preceding oxic episode.
638 These processes result in geochemical time averaging, analogous to taphonomic time averaging in
639 fossil assemblages (Walker & Bambach, 1971). Currently available chemostratigraphic techniques
640 cannot overcome the geochemical time averaging problems in order to resolve such rapidly changing
641 redox conditions.

642 Regardless, the restriction of Lantian fossils in intervals B and D indicates that euxinia may
643 have played an important role in fossil preservation. The taphonomic modes of Lantian fossils
644 include both carbonaceous compression and pyritization (Yuan *et al.*, 2001; Guan *et al.*, 2016; Wan
645 *et al.*, 2016). Carbonaceous compression fossils in the Lantian Formation sometimes show rusty
646 colors (Wan *et al.*, 2016), likely resulting from pyrite weathering and suggesting a continuum
647 between the two taphonomic modes in the Lantian Formation as is also observed in other Ediacaran
648 Lagerstätten (Cai *et al.*, 2012; Hall *et al.*, 2013; Schiffbauer *et al.*, 2014). We hypothesize that anoxic
649 and particularly euxinic waters allowed the precipitation of thin veneer of pyrite on degrading
650 carcasses at or just beneath the water-sediment interface, thus providing a mechanism to replicate
651 soft-bodied organisms in the Lantian Formation. Early authigenic pyrite precipitation may turn out to
652 be a common taphonomic process responsible for various types of fossil preservation in the
653 Ediacaran Period (Liu, 2016).

654

655 CONCLUSIONS

656 High-resolution chemostratigraphic analysis of integrated geochemical data—including
657 $\delta^{13}\text{C}_{\text{carb}}$, $\delta^{13}\text{C}_{\text{org}}$, $\delta^{34}\text{S}_{\text{pyr}}$, $\delta^{34}\text{S}_{\text{CAS}}$, and $\delta^{15}\text{N}_{\text{sed}}$ —from the Lantian Formation deposited in deep-water

658 environments in South China allows a better resolution of the spatial patterns and temporal variations
659 of Ediacaran oceanic redox conditions. Comparison with other geochemical datasets from South
660 China, particularly those from the time-equivalent Doushantuo Formation, makes it possible to
661 determine spatial patterns. A large spatial gradient of $\delta^{13}\text{C}_{\text{carb}}$, with a magnitude of $>10\%$, is
662 confirmed to be present in the lower Doushantuo/Lantian Formation. Unlike previous authors, we
663 argue that this gradient unlikely represents a $>10\%$ depth gradient of $\delta^{13}\text{C}_{\text{DIC}}$; instead, we propose
664 that this gradient is largely due to the influence of ^{13}C -depleted authigenic/diagenetic calcite cements
665 in organic-rich and carbonate-poor sediments deposited in deep water facies. A pronounced negative
666 $\delta^{13}\text{C}_{\text{carb}}$ excursion in the upper Doushantuo/Lantian Formation, however, is spatially consistent and
667 does not display a $>10\%$ depth gradient. This negative excursion has been previously described as
668 EN3 (Ediacaran Negative excursion 3) in South China and regarded as equivalent to the Shuram
669 excursion in Oman. The consistency of this excursion at a basinal scale may have chemostratigraphic
670 significance, but its origin remains to be resolved. Unlike $\delta^{13}\text{C}_{\text{carb}}$, the $\delta^{13}\text{C}_{\text{org}}$ record shows better
671 basinal scale consistency, particularly if only the more reliable $\delta^{13}\text{C}_{\text{org}}$ values of organic-rich
672 sediments are considered.

673 The $\delta^{34}\text{S}_{\text{pyr}}$ record seems to show a spatial pattern, with more negative $\delta^{34}\text{S}_{\text{pyr}}$ values in
674 deeper water facies, but the $\delta^{34}\text{S}_{\text{CAS}}$ record (and hence the $\Delta\delta^{34}\text{S}$ record) is insufficient in both
675 stratigraphic and geographic coverage to allow the determination of spatial patterns. The spatial
676 pattern of $\delta^{34}\text{S}_{\text{pyr}}$ is interpreted as evidence for a chemically stratified basin, with predominantly
677 euxinic deep waters where pyrite precipitated in the water column has lower $\delta^{34}\text{S}_{\text{pyr}}$ values, and
678 predominantly non-euxinic shallow waters where pyrite formed within sediments has higher $\delta^{34}\text{S}_{\text{pyr}}$
679 values. This first-order redox structure is superimposed by local and temporal variations.

680 Relative to the spatial variations of $\delta^{15}\text{N}_{\text{sed}}$ in modern oceans, the $\delta^{15}\text{N}_{\text{sed}}$ record of the
681 Lantian and Doushantuo formations is remarkably consistent. Overall, the $\delta^{15}\text{N}_{\text{sed}}$ data seem to
682 suggest nitrate limitation so that denitrification is often quantitative. The Lantian and Doushantuo
683 formations do not preserve highly positive $\delta^{15}\text{N}_{\text{sed}}$ values characteristic of partial denitrification in
684 suboxic environments of modern oxygen minimum zone, although the $\delta^{15}\text{N}_{\text{sed}}$ database is currently
685 rather limited in spatial and stratigraphic coverage.

686 Integrated TOC, TN, $\delta^{34}\text{S}_{\text{pyr}}$, and Fe speciation data suggest that oscillation between oxic and
687 euxinic conditions is characteristic of the Lantian Formation, at least in Member I to lower Member

688 III. As in the spatial model, pyrite in euxinic intervals was precipitated in the water column and has
689 lower $\delta^{34}\text{S}_{\text{pyr}}$ values, whereas pyrite in oxic intervals was precipitated within sediments and has
690 higher $\delta^{34}\text{S}_{\text{pyr}}$ values due to diffusion-related sulfate limitation. The redox oscillation model can
691 explain some aspects of the $\delta^{15}\text{N}_{\text{sed}}$ profile of the Lantian Formation.

692 Macrofossils in the Lantian Formation include morphologically complex macroalgae and
693 putative animals. Stratigraphic distribution of these fossils is largely restricted to euxinic black shales,
694 although not all black shales preserve macrofossils. In reconciling the preservation of
695 oxygen-breathing organisms in euxinic black shales, it is hypothesized that the euxinic environments
696 were punctuated by brief oxygenation events when macroalgae and putative animals thrived, but
697 were subsequently decimated and preserved when local environment switched to euxinic conditions.
698 If true, then ecological and taphonomic factors were the main controls on fossil distribution in the
699 Lantian Formation (and perhaps in other Ediacaran successions). This hypothesis, however, is
700 difficult to evaluate, owing to the short ecological time scales (compounded by geochemical
701 time-averaging) over which redox variation is presumed to occur.

702

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710

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Figure captions

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Fig. 1. Geological maps and stratigraphic column. (A) Generalized paleogeographic map of the Yangtze Block during the early–middle Ediacaran Period, showing the approximate location of shelf, slope, and basinal facies. Numbered triangles indicate location of Ediacaran sections mentioned in the text, with the Lantian area highlighted in red. (B) Geological map of the Lantian area, showing the location of the Lantian drill core analyzed in this study. Supplementary outcrop samples were collected near the drilling site. The Shiyu and Jinlongshan sections examined in previous studies are also marked in the map. (C) Simplified lithostratigraphic column of the Lantian drill core. PYC: Piyuancun Formation.

Fig. 2. Petrographic photographs of the Lantian drill core (A–D, H–L) and macrofossils from outcrops (E–G). (A) Drill core segment of cap dolostone (Member I). (B) Cross-polarized light photomicrograph of cap dolostone thin section, showing dolomicrite (arrow) and siliceous cement with first-order gray birefringence color. (C) Drill core segment of Member II black shale. (D) Plane-polarized light photomicrograph of Member II black shale, showing silica cements and a compressed *Chuarina* fossil (outlined by red ellipse) surrounded with diagenetic clays. (E–F) The macroalgal fossil *Flabellophyton* from Member II. Specimens illustrated in (E) are preserved with greater three dimensionality, due to pyritization, whereas specimen in (F) is preserved as two-dimensional carbonaceous compression. (G) The problematic fossil *Orbisiana* from Member II. (H) Plane polarized light photomicrograph of argillaceous limestone in upper Member II, showing a mixture of calcite, dolomite, clay, and siliceous cement. (I) Drill core segment of upper Member III limestone. (J) Cross-polarized light photomicrograph of upper Member III limestone, showing calcitic microspar, silty detritus, and siliceous cement. (K) Drill core segment of Member IV black shale. (L) Plane-polarized light photomicrograph of Member IV black shale, showing organic-rich matrix and siliceous cement. Arrows in core segments (A, C, I, K) mark stratigraphic up direction.

1047 **Fig. 3 (in landscape format).** Chemostratigraphic profiles of the Lantian Formation. The $\delta^{18}\text{O}_{\text{carb}}$
1048 and $\delta^{13}\text{C}_{\text{carb}}$ profiles include data from both drill core and outcrop samples, with blue, red, and black
1049 symbols representing measurements of drill core, supplementary outcrop, and calcite vein samples,
1050 respectively. Calcite veins were also sampled from the Lantian drill core. Data in all other profiles
1051 are based on data from drill core samples. Solid symbols denote likely primary signatures based on
1052 evaluation of diagenetic alteration. Redox interpretation for intervals A–D is based on Fe speciation
1053 from the same drill core (Guan, 2014).

1054
1055 **Fig. 4.** Geochemical cross-plots. Note that Fig. 4B plots $\delta^{13}\text{C}_{\text{carb}}$ vs. $\delta^{18}\text{O}_{\text{carb}}$, whereas Fig. 4C plots
1056 $\delta^{13}\text{C}_{\text{carb-vein}}$ vs. $\delta^{13}\text{C}_{\text{carb-host}}$ and $\delta^{18}\text{O}_{\text{carb-vein}}$ vs. $\delta^{18}\text{O}_{\text{carb-host}}$.

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1058 **Fig. 5 (in landscape format).** Spatial variation in $\delta^{13}\text{C}_{\text{carb}}$ of the Doushantuo/Lantian formations.
1059 Dashed lines represent top and bottom boundaries of the Doushantuo/Lantian formations. Gray shade
1060 highlights the negative $\delta^{13}\text{C}_{\text{carb}}$ excursion EN3 that is regionally consistent and is widely regarded as
1061 equivalent to the Shuram excursion. Approximate paleobathymetric location of sections is marked on
1062 a simplified cross-section of the Yangtze Platform (Jiang *et al.*, 2011). See Fig. 1A for exact
1063 geographic location of sections. Data source: Jiulongwan (Jiang *et al.*, 2007), Siduping (Wang *et al.*,
1064 2016), Taoying (Wang *et al.*, 2011), Yuanling (Jiang *et al.*, 2007), Yanwutan (Guo *et al.*, 2007),
1065 Lantian drill core and supplementary outcrop (this study), Shiyu (Wang *et al.*, 2014), and
1066 Jinlongshan (Yuan *et al.*, 2011). DY: Dengying Formation; PYC: Piyuancun Formation. EN:
1067 Ediacaran negative $\delta^{13}\text{C}_{\text{carb}}$ excursion; EP: Ediacaran positive $\delta^{13}\text{C}_{\text{carb}}$ excursion.

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1069 **Fig. 6.** Mixing model showing that variation in $\delta^{13}\text{C}_{\text{carb}}$ could be due to variable mixing of (1) a
1070 depositional component with carbonate mass fraction $X_A = 1$ –100% and carbonate carbon isotopic
1071 composition $\delta^{13}\text{C}_A = 0\text{‰}$, and (2) a post-depositional component of pure calcite cement with
1072 carbonate mass fraction $X_B = 1$ and $\delta^{13}\text{C}_B = -30\text{‰}$. When these two endmembers are mixed at a
1073 mass ratio of $f_A : f_B$ (where $f_A + f_B = 1$), the mixture has a carbonate content of $X_M = f_A * X_A + f_B * X_B$,
1074 and $\delta^{13}\text{C}_M = (\delta^{13}\text{C}_A * f_A * X_A + \delta^{13}\text{C}_B * f_B * X_B) / X_M$. $\delta^{13}\text{C}_{\text{carb}}$ values of carbonate-poor
1075 samples from upper Member II and lower Member III of the Lantian Formation (solid blue dots) can
1076 be accounted for by a relatively small amount of post-depositional cement ($f_B < 5\%$), whereas those

1077 of carbonate interbeds (open circles) require greater amount of cements (but still <20% for most
1078 samples).

1079

1080 **Fig. 7 (in landscape format).** Spatial variation in $\delta^{13}\text{C}_{\text{org}}$ of the Doushantuo/Lantian formations.
1081 Dashed lines represent top and bottom boundaries of the Doushantuo/Lantian formations.
1082 Approximate paleobathymetric location of sections is marked on a simplified cross-section of the
1083 Yangtze Platform (Jiang *et al.*, 2011). See Fig. 1A for exact geographic location of sections. Data
1084 source: Northern Xiaofenghe (NXF) (Xiao *et al.*, 2012), Jiulongwan (JLW) (McFadden *et al.*, 2008),
1085 Zhongling (ZL) (Cui *et al.*, 2015), Siduping (SDP) (Wang *et al.*, 2016), and Lantian (LT) (this study).
1086 DY: Dengying Formation; PYC: Piyuancun Formation.

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1088 **Fig. 8 (in landscape format).** Spatial variation in $\delta^{34}\text{S}_{\text{pyr}}$ and $\delta^{34}\text{S}_{\text{CAS}}$ of the Doushantuo/Lantian
1089 formations. Dashed lines represent top and bottom boundaries of the Doushantuo/Lantian formations.
1090 Approximate paleobathymetric location of sections is marked on a simplified cross-section of the
1091 Yangtze Platform (Jiang *et al.*, 2011). See Fig. 1A for exact geographic location of sections. Data
1092 source: Northern Xiaofenghe (NXF) (Xiao *et al.*, 2012), Jiulongwan (JLW) (McFadden *et al.*, 2008),
1093 Zhongling (ZL) (Cui *et al.*, 2015), Wuhe (WH) (Sahoo *et al.*, 2016), and Lantian (LT) (this study).
1094 DY: Dengying Formation; PYC: Piyuancun Formation.

1095

1096 **Fig. 9.** Spatial variation in $\delta^{15}\text{N}_{\text{sed}}$ of the Doushantuo/Lantian formations. Dashed lines represent top
1097 and bottom boundaries of the Doushantuo/Lantian formations. Approximate paleobathymetric
1098 location of sections is marked on a simplified cross-section of the Yangtze Platform (Jiang *et al.*,
1099 2011). See Fig. 1A for exact geographic location of sections. Data source: Yangtze Gorges
1100 (Kikumoto *et al.*, 2014), Yangjiaping (YJP) (Ader *et al.*, 2014), and Lantian (LT) (this study). DY:
1101 Dengying Formation; PYC: Piyuancun Formation.