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1	Environmental and trilobite diversity changes during the
2	middle-late Cambrian SPICE event
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22	
23	Abstract
24	The Steptoean Positive Carbon Isotope Excursion (SPICE) event at ~497-494 Ma was a
25	major carbon-cycle perturbation of the late Cambrian that coincided with rapid diversity changes
26	among trilobites. Several scenarios (e.g., climatic/oceanic cooling, and seawater anoxia) have been
27	proposed to account for an extinction of trilobites at the onset of SPICE, but the exact mechanism
28	remains unclear. Here, we present a chemostratigraphic study of carbonate carbon and
29	carbonate-associated sulfate sulfur isotopes ( $\delta^{13}C_{carb}$ and $\delta^{34}S_{CAS})$ and elemental redox proxies
30	(U <sub>EF</sub> , Mo <sub>EF</sub> , and C <sub>org/</sub> P), augmented by secular trilobite diversity data, from both upper slope
31	(Wangcun) and lower slope (Duibian) successions from the Jiangnan Slope, South China,
32	spanning the Drumian to lower Jiangshanian. Redox data indicate locally/regionally
33	well-oxygenated conditions throughout the SPICE event in both study sections except for
34	low-oxygen (hypoxic) conditions within the rising limb of the SPICE (early-middle Paibian) at
35	Duibian. As in coeval sections globally, the reported $\delta^{13}C_{carb}$ and $\delta^{34}S_{CAS}$ profiles exhibit
36	first-order coupling throughout the SPICE event, reflecting co-burial of organic matter and pyrite
37	controlled by globally integrated marine productivity, organic preservation rates and shelf hypoxia.

Increasing 834SCAS in the 'Early SPICE' interval (late Guzhangian) suggests that significant 38 environmental change (e.g., global-oceanic hypoxia) was underway before the global carbon cycle 39 40 was markedly affected. Assessment of trilobite range data within a high-resolution biostratigraphic 41 framework for the middle-late Cambrian facilitated re-evaluation of the relationship of the SPICE 42 to contemporaneous biodiversity changes. Trilobite diversity in South China declined during the 43 Early SPICE (corresponding to the End-Marjuman Biomere Extinction, or EMBE, of Laurentia) and at the termination of the SPICE (corresponding to the End-Steptoean Biomere Extinction, or 44 45 ESBE, of Laurentia), consistent with biotic patterns from other cratons. We infer that oxygen 46 minimum zone (OMZ) and/or shelf hypoxia expanded as a result of locally enhanced productivity 47 due to intensified upwelling following climatic cooling, and that expanded hypoxia played a major role in the EMBE at the onset of SPICE. During the SPICE event, global-ocean ventilation 48 49 promoted marine biotic recovery, but termination of SPICE-related cooling in the late Paibian may have reduced global-ocean circulation, triggering further redox changes that precipitated the ESBE. 50 51 Major changes in both marine environmental conditions and trilobite diversity during the late Guzhangian demonstrate that the SPICE event began earlier than the Guzhangian-Paibian 52 53 boundary, as previously proposed.

54

*Keywords:* carbon isotopes; sulfur isotopes; oceanic anoxia; species diversity; Marjuman Biomere;
 Steptoean Biomere

57

### 58 1. Introduction

59 The Cambrian, a key period in Earth history, was characterized by the Cambrian Explosion of 60 marine invertebrate life (Marshall, 2006) as well as unstable marine environments as revealed by fluctuations in carbon isotopes (Saltzman and Thomas, 2012), multiple biotic extinctions, and the 61 62 development of extreme environmental conditions (Servais et al., 2010). The Steptoean Positive 63 Carbon Isotope Excursion (SPICE) was the last major carbon isotope excursion (CIE) of the 64 Cambrian (Saltzman et al., 1998, 2000), recording a shift of ~4 to 6‰ in marine carbonate carbon isotopes ( $\delta^{13}C_{earb}$ ) for an interval of 3.0  $\pm$  0.2 Myr during the Paibian Stage (~497–494 Ma; 65 lowermost Furongian Series; Sørensen et al., 2020; see Supplemental Section 1 for Cambrian 66 timescale). The SPICE event began at the base of the Paibian Stage, as defined by the first 67 appearance datum (FAD) of the trilobite Glyptagnostus reticulatus (Peng and Robison, 2000; Zhu 68 69 et al., 2018), and continued to the base of the Irvingella major Zone in the early Jiangshanian 70 (Peng et al., 2004, 2012; Gerhardt and Gill, 2016), thus spanning from the middle Cambrian 71 (Miaolingian, formerly Series 3) to the late Cambrian (Furongian) series boundary interval. 72 The SPICE event was associated with a pronounced marine biotic turnover, including

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extinctions of trilobites and agnostid arthropods (Palmer, 1984; Saltzman et al., 2000; Gerhardt
and Gill, 2016; Moysiuk and Caron, 2019; Zhang et al., 2021) and brachiopods (Rowell and Brady
1976; Rieboldt, 2005), changes in the composition of reef communities (Lee et al., 2015), and
large increases in plankton diversity (Servais et al., 2008). Middle and upper Cambrian trilobites

77 have been especially well studied, and the SPICE event was accompanied by extinctions of the Marjumiid Biomere at the end of the Guzhangian Stage and the Pterocephaliid Biomere in the 78 79 early Jiangshanian Stage (Palmer, 1979, 1984; Gerhardt and Gill, 2016; Zhang et al., 2021). 80 Several mechanisms have been proposed to explain the extinction of the Marjumiid Biomere, including global temperature changes (Lochman-Balk, 1970; Öpik, 1966), a rise in the 81 82 thermocline and shelf cooling (Stitt, 1975), and/or ecospace changes linked to sea-level fluctuations (Ludvigsen, 1982; Westrop, 1988; Westrop and Ludvigsen, 1987). Geochemical 83 studies have confirmed some of these inferences and proposed other potential causes, for example, 84 85 upwelling of cool deep waters onto shallow shelves (Perfetta et al., 1999; Elrick et al., 2011) as 86 well as widespread oceanic anoxia (Saltzman et al., 1998; Hurtgen et al., 2009; Gill et al., 2011; 87 2021; Dahl et al., 2014). However, these studies are based on geographically limited datasets 88 containing a small number of proxies, rendering the causes of the extinction uncertain.

Middle-Late Cambrian trilobite extinctions were almost certainly linked to marine 89 90 environmental changes, but the nature of such changes during the SPICE event remains poorly 91 known. The SPICE is thought to have coincided with a major phase of global cooling, although 92 this inference is based largely on carbon-cycle changes and physical evidence of sea-level fall 93 rather than direct temperature measurements. In addition to a positive CIE, changes in the global 94 carbon cycle are indicated by evidence of enhanced marine productivity and organic carbon burial 95 from organic carbon-nitrogen isotopes (Hammer and Svensen, 2017), carbon-sulfur isotope modeling of atmospheric O2 content (Saltzman et al., 2011; Krause et al., 2018), N/P ratios 96 indicating P limitation of marine productivity (Saltzman, 2005), and a decrease in seawater 97 87Sr/86Sr values (Zhang et al., 2020). Physical evidence of cooling includes a sea-level lowstand at 98 99 the Sauk-II/III contact (= mid-Paibian) (Saltzman et al., 2000, 2004; Sørensen et al. 2020), ice-erosional features at mid-latitudes of Baltica (Dronov and Popov 2004; Cherns and Wheeley, 100 101 2009), consistent with growth of continental icesheets during the early to middle Paibian 102 (Matthews and Al-Husseini, 2010; Al-Husseini, 2017). Due to the indirect nature of this evidence, 103 climate cooling during the early to middle Paibian remains contentious. The only oxygen isotope (δ18O) study of the Paibian to date documented increased sea-surface temperatures on the western 104 105 margin and cratonic interior of Laurentia (Elrick et al., 2011; Wotte et al., 2019). However, this warming event is likely to have been a local signal related to shallowing on a tropical continental 106 shelf, with glacio-eustatic fall causing the study sections to shallow into the ocean-surface layer, 107 thus recording locally warmer conditions despite the general climatic cooling necessary to induce 108 109 glacial expansion.

In the present study, our goals are to examine chemostratigraphic records of marine paleoenvironmental change during the SPICE, and to link these changes to contemporaneous records of trilobite diversity in order to better understand controls on Middle-Late Cambrian biotic events. We analyzed two middle to upper Cambrian sections in South China, Wangcun and Duibian, representing relatively shallower (upper slope) and deeper (lower slope) depositional settings, applying elemental redox proxies (i.e., U<sub>EF</sub>, Mo<sub>EF</sub>, C<sub>org</sub>*P*) to track local environmental

changes that may have affected biodiversity patterns, and inorganic carbon and sulfur isotopes (i.e.,  $\delta^{13}C_{earb}$  and  $\delta^{34}S_{CAS}$ ; carb = carbonate; CAS = carbonate-associated sulfate) as global seawater proxies. Furthermore, we reprocessed published trilobite data for these two sections, which are among the faunally best studied successions of middle to late Cambrian age globally, having yielded a wealth of trilobite taxonomic data suitable for analysis of biodiversity trends. Our study thus provides an integrated geochemical and paleontological dataset that addresses fundamental relationships between paleoenvironmental changes and biotic evolution during the SPICE event.

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### 124 2. Geological background

125 2.1. Paleogeography

During the middle and late Cambrian, the South China Craton was located on the equatorial margin of Gondwana (Fig. 1A). Three main depositional environments existed along a platform-to-basin transect, with a shallow-platform carbonate facies to the northwest (Yangtze Platform), argillaceous carbonate and shale in the central slope facies (Jiangnan Slope), and fine-grained siliciclastic and chert in the basinal facies (Nanhua Basin) to the southeast (Fig. 1B; Feng et al., 2002; note: all coordinates are modern unless otherwise noted). The present study sections are located on the Jiangnan Slope (Zuo, et al., 2018).

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### 134 2.2. Wangcun section

Wangcun (GPS: 28°43'2.84" N, 109°58'26.10" E) is an outcrop section exposed along a 135 roadcut on the northern bank of the Youshui River in western Hunan Province, South China (Fig. 136 1C). It is a parastratotype of the Drumían-Guzhangian stage boundary, for which the Global 137 138 Stratotype Section and Point (GSSP) is the Luoyixi section, which is located on the southern bank of the same river (Peng et al., 2004, 2005, 2009). The Wangcun section consists, in ascending 139 140 order, of the Aoxi, Huaqiao, and Shenjiawan formations. The Aoxi Formation is mainly composed 141 of dolomite and black shale interbedded with limestone; the Huaqiao Formation is dominated by 142 thin-bedded muddy limestone, with a few oolitic limestone beds in the lower part, and thick-bedded mudstone containing lenticular limestone, conglomeratic limestone, and calcareous 143 144 shale beds in the upper part; and the Shenjiawan Formation consists of limestone and dolomitic limestone (Peng et. al., 2004; Fig. 2). 145

The trilobite biostratigraphy of the Wangcun section is well studied for the Drumian and 146 147 Guzhangian stages, but less so for the Paibian and Jiangshanian stages (Fig. 2A). A total of 9 148 trilobite zones have been established, in ascending order, the Ptychagnostus atavus, Pt. punctuosus, Goniagnostus nathorsti, Lejopyge armata, L. laevigata, Proagnostus bulbus, 149 Linguagnostus reconditum, Glyptagnostus stolidotus, and G. reticulatus zones. The bases of the 150 151 Drumian, Guzhangian, and Paibian stages are defined by the first appearance datums (FADs) of 152 the trilobites P. atavus, L. laevigata, and G. reticulatus, respectively (Peng, 2005; Peng et al., 2009). The placement of the top of the G. reticulatus Zone and the stratigraphic interval of the Ag. 153 orientalis Zone (note: the base of this zone is equivalent to the base of the Jiangshanian) are 154

155 defined based on correlations with Duibian A (Zuo et al., 2018).

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157 2.3. Duibian section

158 The Duibian section consists of outcrops near Duibian village, 10 km north of Jiangshan City, western Zhejiang Province, South China (Fig. 1D). Dubian A (GPS: 28°48'48.38" N, 118°37'19.21" 159 160 E), a parastratotype of the Paibian-Jiangshanian stage boundary, contains, in ascending order, the upper Yangliugang, Huayansi, and lower Siyanshan formations (Lu and Lin, 1989; Peng et al., 161 2012). The Yangliugang Formation consists mainly of dark-gray, thin-bedded dolomitic limestone 162 163 with calcareous mudstone interbeds; the Huayansi Formation comprises dark, thin-bedded limestone with thin shale interbeds and light-colored ribbon limestones; and the Siyangshan 164 Formation consists of pale limestone with breccias in the lower part, and light gray thin-bedded 165 limestone interbedded with calcareous mudstone and muddy limestone in the upper part. Duibian 166 B (28°48'46.14" N, 118°37'17.20" E), the GSSP for the Paibian-Jiangshanian stage boundary, is 167 located ~250 m to the south of Duibian A and exposes only a part of the Huayansi Formation (Fig. 168 1D). Although their chemostratigraphic profiles are shown separately in the figures of this study, 169 170 the geochemical datasets of Duibian A and B were combined for purposes of statistical evaluation owing to their proximity and general similarity. 171

The trilobite biostratigraphy of the Duibian section (Figs. 3A, 4A) is well established (Lu and Lin, 1989; Peng et al., 2012). At Duibian A, the bases of the Paibian and Jiangshanian stages are defined by the FADs of the trilobites *Glyptagnostus reticulatus* and *Agnostotes orientalis*, respectively. Duibian B is the GSSP of the base of the Jiangshanian Stage, based on the FAD of the trilobite *Ag. orientalis* (Peng et al., 2012). The base of the Jiangshanian Stage was placed at 116.6 m and 108.12 m above the base of the Huayansi Formation at Duibian A and B, respectively (Peng et al., 2012).

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### 180 2.4. Comparative global sections

181 Results from the study sections were compared with geochemical data from five widely distributed SPICE sections (Fig. 1A). Four of these auxiliary sections accumulated on continental 182 183 shelves, ranging from subtidal-peritidal to deep-shelf settings: (1) Lawson Cove, Utah (Gill et al., 2011; note: not "Lawsons Cove" as given in that source); (2) Shingle Pass, Nevada (Saltzman et 184 al., 1998; Gill et al., 2007); (3) Mount Whelan core, Australia (Saltzman et al., 2000; Gill et al., 185 2011); and (4) TE-1 core, Texas County, Missouri (Gill et al., 2011). The fifth auxiliary section, 186 187 the Andrarum-3 core (Sweden), was deposited below storm wave base (Gill et al., 2011; Dahl et al., 2013). Paired  $\delta^{13}C_{earb}$  and  $\delta^{34}S_{CAS}$  values for the five auxiliary sections, as well as local/global 188 redox proxy data for two of the five sections (i.e., carbonate uranium isotopes from Mount Whelan 189 190 core, and redox sensitive elements from Andrarum-3 core) were available for comparisons (note: 191 the other three auxiliary sections lack such data). See Supplemental Section 2 and Table S1 for more paleogeographic and stratigraphic background information about the study and auxiliary 192 sections, and Supplemental Figure S1 for correlation of trilobite zones between South China and 193

194 Laurentia.

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#### 196 2.5. Internal stratigraphy of the SPICE event interval

197 Previous studies have divided the SPICE interval into two parts: a 'rising limb' marked by an increasing trend of carbonate carbon isotopes ( $\delta^{13}C_{carb}$ ), and a 'falling limb' marked by a 198 decreasing δ13Ccarb trend (Saltzman et al., 2000; Pulsipher et al., 2021). In the present study, we 199 identify a third interval that we term 'Early SPICE', marking the earliest stage of the SPICE event 200 prior to a major rise in  $\delta^{13}C_{earb}$ . In addition, for the sake of ease of reference, we term the intervals 201 that preceded and followed the SPICE event the 'Pre-SPICE' and 'Post-SPICE', respectively. In 202 203 our study sections, the Pre-SPICE, Early SPICE, Rising SPICE (= 'rising limb'), Falling SPICE (= 'falling limb'), and Post-SPICE intervals correspond to Units I to V (in sequence). For sections 204 having a substantial Pre-SPICE interval, we have designated the somewhat older carbon isotope 205 excursion known as DICE (Drumian Carbon Isotope Excursion) as Unit Ia and the strata between 206 207 DICE and the base of the Early SPICE as Unit Ib. All five units are present at Wangcun (Fig. 2C), but only four units (Ib, II, III and IV) are present at Duibian A (Fig. 3C), and only two units (IV 208 209 and V) at Duibian B (Fig. 4C).

The five intervals described above were defined primarily on the basis of carbonate carbon 210 isotope (813Ccarb) variation (cf. Saltzman et al., 2000; Pulsipher et al., 2021) but with reference to 211 some secondary criteria including the  $\delta^{34}S_{CAS}$  profile and trilobite range data. To facilitate use in 212 other studies, we define the subdivisions primarily in terms of carbon isotope variation and 213 trilobite range data. The Early SPICE interval is marked by a gentle rise of  $\delta^{13}C$  (note: not the 214 steep rise associated with the Rising SPICE), or, in sections lacking such a  $\delta^{13}$ C feature, by a 215 significant rise of 834SCAS, during the late to latest Guzhangian Stage (Linguagnostus reconditus 216 and G. stolidotus zones). The transition from the Early SPICE to Rising SPICE is marked by a 217 sharp acceleration in the  $\delta^{13}$ C profile, or by the onset of rising  $\delta^{13}$ C in sections lacking the slow 218 219  $\delta^{13}$ C rise of the Early SPICE; it spans the earliest Paibian (base of G. reticulatus Zone) to middle 220 Paibian Stage (A. inexpectans Zone). The transition from the Rising SPICE to Falling SPICE is marked by the shift from increasing to decreasing  $\delta^{13}$ C values; in some sections (e.g., Shingle Pass, 221 Lawson Cove, TE-1 Texas County Core, Mount Whelan Core; Saltzman et al., 1998, 2000; Gill et 222 al., 2007, 2011) this transition is rapid, but in other sections (e.g., Deogwoo, Wa'ergang, House 223 Range; Baker, 2010; Chung et al., 2011; Li et al., 2018) there is an extended plateau of nearly 224 uniform high  $\delta^{13}$ C values that makes identification of the exact point of the transition somewhat 225 226 arbitrary. The Falling SPICE corresponds to the middle Paibian to early Jiangshanian Stage (i.e., A. inexpectans Zone to lower part of Ag. orientalis Zone). The Pre-SPICE and Post-SPICE are 227 defined simply as those intervals preceding onset of the Early SPICE and following termination of 228 229 the Falling SPICE, respectively. The Post-SPICE corresponds to middle-late Jiangshanian Stage 230 (i.e., upper part of Ag. orientalis Zone to Eolotagnostus Zone).

231 Our scheme for internal subdivision of the SPICE event redefines the timing of its onset.

Earlier studies placed the base of the SPICE event at the onset of the sharp rise in the  $\delta^{13}C_{carb}$ 6 233 profile (e.g., Saltzman et al., 2000; Zhu et al., 2018; Pulsipher et al., 2021; but note that Schiffbauer et al., 2017 proposed a globally diachronous onset of the SPICE), which is equivalent 234 235 to the base of the Rising SPICE of our study. However, the present study demonstrates that the 236 former definition is inconvenient for three reasons. First, the Wangcun and Duibian sections show positive shifts in  $\delta^{13}C_{carb}$  and  $\delta^{34}S_{CAS}$  heralding the SPICE event well below the 237 Guzhangian-Paibian boundary, which has previously defined the base of the SPICE (Saltzman et 238 239 al., 2000). At Wangcun, obvious positive shifts commence at ~210 m, or ~70 m below the base of the Rising SPICE (which is at ~280 m), and at Duibian A, obvious positive shifts commence at 240 ~-4 m, or ~11 m below the base of the Rising SPICE (which is at ~7 m; Figs. 2-3). Second, the 241 concurrent positive shifts of  $\delta^{13}C_{carb}$  and  $\delta^{34}S_{CAS}$  during the late Guzhangian Stage were a global 242 phenomenon (Gill et al., 2007; Pulsipher et al., 2021), and they coincided with the initiation of 243 shifts in other global proxies (e.g.,  $\delta^{238}$ U values) that continued into the Rising SPICE interval 244 (Dahl et al., 2014). Third, the former definition decouples the SPICE from the EMBE, leading to 245 suggestions that the EMBE had non-SPICE-related causes (Palmer, 1984; Saltzman et al., 2000; 246 247 Elrick et al., 2011), which we regard as almost certainly incorrect. For these reasons, we propose 248 redefinition of the base of the SPICE based on the onset of paleo-environmental disturbances as 249 determined from multiple proxies (i.e.,  $\delta^{13}C_{carb}$ ,  $\delta^{34}S_{CAS}$ ,  $\delta^{98}Mo$  and  $\delta^{238}U$ ), instead of a single proxy (813Ccarb) that exhibits invariant or regionally variable values during the EMBE (e.g., 250 Gerhardt and Gill, 2016; Schiffbauer et al., 2017). The interval of slowly rising 813Ccarb during the 251 earliest part of the redefined SPICE is herein termed the "Early SPICE" interval (Unit II of present 252 study, within the Li. reconditus and G. stolidotus zones) (Figs. 2-3). This redefinition places the 253 254 onset of the SPICE event in the late Guzhangian rather than at the Guzhangian-Paibian stage 255 boundary (cf. Saltzman et al., 2000), and it links middle-late Cambrian trilobite diversity changes more effectively to the trajectory of the SPICE event (cf. Gerhardt and Gill, 2016; Zhang et al., 256 2021). 257

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### 259 3. Methods

### 260 3.1. Isotopic and elemental analyses

261 Weathered surfaces and diagenetic veins were trimmed off, and the remaining bulk-rock carbonate was powdered to <74 µm (200 mesh) using a rock mill. Major elements were measured 262 using wavelength-dispersive XRF and trace elements using ICP-MS, after sample powder 263 digestion by HNO3 and HF, in the State Key Laboratory of Geological Processes and Mineral 264 265 Resources at the China University of Geosciences-Wuhan. Average analytical uncertainty is better than 5% (RSD) for major elements based on repeated analysis of national standards 266 GBW07132, GBW07133 and GBW07407, and better than 2% (RSD) for trace elements based on 267 268 international standards AGV-2, BHVO-2, BCR-2 and GSR-1. In the same laboratory, total organic 269 carbon (TOC) content was measured using an Elementar Vario Micro Cube Analyzer, and inorganic carbon isotopes were measured using a 253 Plus Isotope Ratio Mass Spectrometer 270 (IR-MS) interfaced with a Kiel IV auto-sampler. The analytical precision was better than 0.04‰ 271

for both  $\delta^{13}C_{carb}$  and  $\delta^{18}O_{carb}$  based on duplicate analyses of national standards GBW-04416 and 272 GBW-04417. Multiple NaCl rinses (generally > 30 times) was applied to extract 273 carbonate-associated sulfate (CAS) (Wotte et al., 2012). Sulfur isotopes in CAS were analyzed 274 275 using a Delta V plus IR-MS in the State Key Laboratory of Biogeology and Environmental Geology at the China University of Geosciences-Wuhan. Analytical errors were 0.08 ‰, 0.09 ‰ 276 277 and 0.20 ‰ (1o), respectively, calculated from duplicate analyses of the international standards NBS 127, IAEA SO-5, and IAEA SO-6. Detailed descriptions of all methods are given in 278 Supplemental Section 3. 279

280

### 281 3.2. Trilobite biodiversity

Trilobite biostratigraphic analyses at Wangcun were carried out by Peng and Robison (2000) and Peng et al. (2009), during investigation of the nearby Luoyixi section as the GSSP of the base of the Guzhangian Stage. A total of 66 species (including 2 undefined species) were recognized from ~90 stratal levels at Wangcun, including within the Pre-SPICE and Early SPICE intervals (~150- and ~100-m-thick, respectively), and the lower part of the Rising SPICE (20-m-thick).

Trilobite biostratigraphic work at Duibian was carried out by Lu and Lin (1989) and Peng et al. (2006, 2009, 2012) before designation of this locale as the GSSP of the base of the Jiangshanian Stage. A total of 64 species (including 14 undefined species) were recognized from ~50 stratal levels within a ~200-m-thick interval containing the SPICE event at Duibian A, and 35 species (including 9 undefined species) from ~30 stratal levels within a ~40-m-thick interval at Duibian B.

293 In the present study, we compiled trilobite species range data in order to construct both 294 species diversity and range-through diversity profiles for each study section for the lowermost 295 Drumian to lower Jiangshanian interval. The species diversity curve shows the total number of 296 trilobite species actually identified at a given stratal level in the source studies (Peng and Robison, 2000; Peng et al., 2004, 2005, 2006, 2009, 2012). The range-through diversity curve accounts for 297 298 taxa that are absent at a given stratigraphic level but present in both underlying and overlying horizons, on the assumption that their absence in such cases is due to incompleteness of the fossil 299 300 record.

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### 302 4. Results

### 303 4.1. Variations of isotopic and elemental proxies

The  $\delta^{13}C_{carb}$  profiles of the study sections exhibit a broad first-order positive excursion representing the SPICE, spanning a stratigraphic interval of 229.0 m to 361.0 m at Wangcun and -4.0 m to 29.0 m at Duibian A-B (Figs. 2-4). From background values of ~0-1 ‰ (Unit Ib; Pre-SPICE), the excursion began in the late Guzhangian with a slow initial shift that was slightly larger at Wangcun (~+1 ‰) relative to Duibian A (~+0.5 ‰) (Unit II; Early SPICE). A steeper rise in  $\delta^{13}C_{carb}$  commenced at the Guzhangian-Paibian boundary, marking the onset of the main phase of SPICE (Unit III; Rising SPICE). The positive excursion peaked in the middle Paibian with

values of +3.84 ‰ at Wangcun and +3.15 ‰ at Duibian A.  $\delta^{13}C_{carb}$  values declined during the late Paibian to early Jiangshanian (Unit IV; Falling SPICE), stabilizing at ~1 ‰ in the Post-SPICE interval (Unit V). The full SPICE excursion appears relatively smoother at Wangcun than at Duibian, where some small-scale variability is present (e.g., in Unit III), although this difference may be due to the higher-resolution sampling of the latter section.

The  $\delta^{34}S_{CAS}$  profiles of the study sections also show first-order positive excursions during the 316 SPICE (Figs. 2-4). Following background values of ~25-35 ‰ in the Pre-SPICE, a major rise of 317  $\delta^{34}S_{CAS}$  begins in the Early SPICE, reaching a peak value that is slightly higher at Wangcun 318 (+48.4 ‰) relative to Duibian A (+46.9 ‰). Relatively stable plateau values are observed at 319 320 Wangcun (~+40-50 ‰) and Duibian A (~+35-45 ‰) in the Rising SPICE, but δ34SCAS shows a decreasing trend to a minimum of ~+28 ‰ (Wangcun) and ~+20-25 ‰ (Duibian) by the end of 321 the Falling SPICE.  $\delta^{34}S_{CAS}$  fluctuates within ~5-25 ‰ and finally maintains stable values 322 (~25-30 ‰) in the Post-SPICE at Wangcun, with a comparable range of fluctuations (within 323 ~15-35 ‰) at Duibian B. 324

The TOC profile at Wangcun shows a roughly decreasing trend from ~0.6 to ~0.2 wt.% with 325 326 a few peak values (to ~0.6-1.0 wt.%) in the DICE to Early SPICE intervals, then maintains relatively low values (~0.2 wt.%) punctuated by several sharp peaks (to ~0.6-1.4 wt.%) in the 327 Rising to Post-SPICE intervals (Fig. 5B). The TOC profiles show more regular variations at 328 329 Duibian, stabilizing around 0.10 wt.% in the Pre-SPICE, rising to relatively higher level (~0.2 wt.%) with few peak values (~0.5-2.0 wt.%) in the Rising SPICE, then gradually dropping in the 330 Falling SPICE and reaching a minimum of ~0.02 wt.% by its termination, before sharply rising to 331 a peak value ~0.4 wt.% and fluctuating over ~0.1-0.8 wt.% in the Post-SPICE (Fig. 6B, 7B). 332

333 The Corg/P profile at Wangcun exhibits low values (mostly < 5) in the DICE and Early SPICE, then rises to relatively higher values of ~60-80 in the Early and Rising SPICE, followed by a 334 return to lower values of ~0-50 in the Falling SPICE, before fluctuating in the range of ~0-100 in 335 336 the Post-SPICE (Fig. 5F). At Duibian, relative to background values (~0-20) in the Pre-SPICE, 337 Corg/P rises progressively to ~30 in the Early SPICE, then remains at a plateau (~25-45) punctuated by several peaks (to ~50-200) in the Rising SPICE, before gradually dropping to 338 minimum values (< 5) in the Falling SPICE, and finally rebounding to relatively higher values 339 (~15-45) in the Post-SPICE (Fig. 6F, 7F). 340

Trace-element enrichment factors (EFs) were calculated as XEF = (X/Al)sample / (X/Al)UCC, 341 where UCC is average upper crustal composition (McLennan, 2001). In order to reduce variance 342 343 in EFs related to small denominator values, only samples with Al > 0.5% were used in redox reconstructions (Figs. 5-7; see also Supplemental Fig. S3). At Wangcun, the UEF and MOEF 344 profiles exhibit decreasing trends from ~15 to ~3 and ~64 to ~1, respectively, in the DICE to 345 346 Pre-SPICE intervals, and then both profiles exhibit lower values (mostly < 3) in the Early SPICE 347 to Post-SPICE (Fig. 5D-E). At Duibian A, the U<sub>EF</sub> profile shows a decreasing trend (~7 to ~2) in the Pre-SPICE to Early SPICE, followed by stable values (~3-5) in the Rising SPICE, before 348 increasing (to >10) in the Falling SPICE (Fig. 6D). The Mo<sub>EF</sub> profile mostly exhibits low values 349

350 (~< 3) in the Pre-SPICE to Early SPICE, then rises to slightly higher values (~3 to ~9) in the

351 Rising SPICE, before dropping to a minimum (~1) in the Falling SPICE (Fig. 6E). At Duibian B,

U<sub>EF</sub> fluctuates between ~2 and ~17, and Mo<sub>EF</sub> between ~1 and ~10, in the Falling SPICE to
 Post-SPICE interval (Fig. 7D-E).

354

355 4.2. Trilobite species diversity records

At Wangcun, trilobite range-through species diversity rises from 2 to 9 at ~30-80 m (i.e., within the *Ptychagnostus aculeatus* Zone), remains stable (~5) at ~80-150 m (i.e., lower part of the Pre-SPICE), followed by a slight increase to ~10-15 at ~220 m (i.e., upper part of the Pre-SPICE), before a significant decrease to a minimum of 1 at ~270 m (base of the *G. stolidotus* Zone, i.e., the EMBE), and with the minimum value continuous into the Rising SPICE (base of the *G. reticulatus* Zone) (Fig. 2).

At Duibian A, trilobite range-through species diversity rises significantly from < 10 at the 362 base of the section (lower Guzhangian) to a maximum value of 25 at -5 m prior to the onset of the 363 Early SPICE (base of the Li. reconditus Zone) (Fig. 3). Between -5 m and 0 m (base of the G. 364 365 stolidotus Zone, i.e., EMBE), range-through species diversity drops sharply to < 5, followed by a gradual decline to 0 at 30 m, representing the end of the Rising SPICE (lower part of the A. 366 inexpectans Zone). Upwards, range-through species diversity gradually rises to ~5-10 at 70 m 367 368 (end of Falling SPICE) before a decline to < 5 at the top of the section (within the Ag. orientalis Zone, i.e., ESBE). At Duibian B, trilobite range-through species diversity gradually increases from 369 0-5 at the base of the section to a maximum of 15 at 28 m (end of the Falling SPICE; upper part of 370 the A. inexpectans Zone), followed by a decrease to < 5 at 35-40 m (i.e., ESBE) within the 371 372 Post-SPICE interval at the top of the section (middle part of the Eolotagnostus Zone) (Fig. 4).

### 373

## 374 5. Discussion

375 5.1. Data evaluation

376 5.1.1. CAS extraction methods

377 The extraction method of CAS (i.e., using multiple NaCl rinses) that we applied in the 378 present study is likely to remove a large part of the contaminant secondary sulfate, for example, that from soluble and organically bound sulfur as well as diagenetically oxidized pyrite. Repeated 379 leaching with an NaCl solution is recommended as a standard step in CAS extraction from 380 carbonate rocks, as it can fully remove non-CAS sulfate that was not incorporated into the 381 382 carbonate mineral structure. It is superior to using NaOCl or H2O2 rinses alone, or a combination thereof with NaCl rinses (Wotte et al., 2012). We generally repeated NaCl rinses at least 30 times, 383 384 which is more effective than a small number of NaCl rinses or a single NaCl rinse followed by an 385 NaOCI rinse (Edwards et al., 2019), as the latter two methods are unlikely to fully remove 386 non-CAS sulfate. Although [CAS] is low in Duibian samples (~1-4 ppm), making  $\delta^{34}S_{CAS}$ susceptible to the influence of oxidized pyrite prior to or during laboratory pretreatment, we infer 387 that such effects were probably minor because the  $\delta^{34}S_{CAS}$  profiles of the study sections exhibit 388

relatively high values (mostly > 20 %) relative to those of pyrite sulfur (< -10 %) as well as

- 390 stable stratigraphic trends without anomalous negative outliers.
- 391

392 5.1.2. Effects from local depositional conditions and early marine diagenesis

393 Local depositional conditions and early marine diagenesis determine preservation of marine 394 carbonate and regional/global stratigraphic expressions of carbon-isotopic signals in shallow-water 395 carbonate facies. Large glacio-eustatic fluctuations can result in isotopic shifts unrelated to variations in the global carbon cycle (Swart, 2008, 2015; Swart and Kennedy, 2012). For example, 396 flooding of carbonate platforms can increase the proportion of aragonite in sediments, resulting in 397 398 a globally synchronous positive  $\delta^{13}C$  excursion. Conversely, sea-level fall results in exposure of platform carbonates to freshwater, leading to meteoric diagenetic alteration in which enhanced 399 authigenic carbonate precipitation can generate a negative δ13C excursion (Schrag et al., 2013; 400 Zhao et al., 2016). However, the positive  $\delta^{13}C_{carb}$  excursion of the SPICE event was associated 401 with a major sea-level fall (e.g., Saltzman et al., 2000), arguing against the influence of sea-level 402 variation on marine carbonate carbon-isotopic compositions. Previous studies have inferred that 403 the SPICE was facies-dependent, through intrusion of <sup>13</sup>C-enriched deepwaters onto carbonate 404 platforms during sea-level rise (Schiffbauer et al., 2017). However, the recognition of the Early 405 SPICE interval in the present study suggests that the influence of facies on carbon-isotopic 406 407 compositions was weak. In addition, regional anoxia-euxinia generally corresponds to positive  $\delta^{13}C_{carb}$  excursions during the SPICE (e.g., Gill et al., 2011), indicating that the influence of 408 authigenic carbonates was limited. 409

Local depositional conditions and early marine diagenesis determine the preservation of 410 primary 834SCAS signals (Present et al., 2019; Richardson et al., 2019, 2021). Generally, low and 411 stable 834SCAS values are associated with deep-water facies, whereas higher and more variable 412 413 values characterize shallow-water facies (Richardson et al., 2019). For example, carbonate rocks 414 deposited in slope facies may incorporate sulfate from anoxic marine-phreatic pore fluids that 415 have been isotopically modified from seawater by microbial sulfate reduction (Present et al., 2019). In the present study, Wangcun and Duibian are located in slope areas in which oxic-suboxic 416 conditions prevailed (see Section 5.2.1), suggesting that the influence of facies on  $\delta^{34}S_{CAS}$  signals 417 was limited. However, facies-related influences on 834SCAS signals may have been pronounced in 418 deep-water SPICE successions such as TE-1 Texas County Core (Gill et al., 2011), which is 419 characterized by 834SCAS that is ~10 to 20 ‰ lower than in shallow-water successions such as the 420 Mount Whelan Core (see Supplementary Information). Moreover, 834SCAS profiles exhibit variable 421 values in slope sections globally (see Section 5.3), suggesting potential facies or geographic 422 dependency of primary  $\delta^{34}S_{CAS}$  signals during the SPICE event. 423

424

425 5.1.3. Diagenetic alteration

426 The present  $\delta^{13}C_{carb}$  profiles are interpreted as primary marine signals based on relationships

427 with  $\delta^{18}O_{carb}$  and Mn/Sr ratios (see Supplemental Fig. S2). Generally, Mn/Sr ratios > ~2 and strong

covariation with Mn/Sr or δ<sup>18</sup>O<sub>carb</sub> are taken as evidence of diagenetic alteration in carbonate rocks
(Marshall, 1992; Brand, 2004). At Wangcun and Duibian, Mn/Sr ratios are low (avg. 0.41 ± 0.33,
0.1 ± 0.07 and 0.06 ± 0.06, respectively), consistent with little to no diagenetic alteration of the
samples. Moreover, δ<sup>13</sup>C<sub>carb</sub> shows only weak correlation to Mn/Sr (r = -0.32, n = 37, p(α) < 0.05;</li>
r = -0.39, n = 100, p(α) < 0.01, respectively), and none correlation to δ<sup>18</sup>O<sub>carb</sub> (r = -0.13, n = 48,
p(α) > 0.10; r = -0.11, n = 130, p(α) > 0.10, respectively).
Primary marine δ<sup>34</sup>S<sub>CAS</sub> signals were further evaluated based on relationships to δ<sup>18</sup>O<sub>carb</sub>

Mn/Sr, Mg/Ca, and CAS concentrations (see Supplemental Fig. S2). Generally, diagenetic 435 alteration and dolomitization of carbonate rocks produces strong covariation between  $\delta^{34}S_{CAS}$ , 436 437 δ18Ocarb, Mn/Sr and Mg/Ca (cf. Marenco et al., 2008). However, such effects are not evident at Wangcun or Duibian, because  $\delta^{34}S_{CAS}$  shows weak or no relationship to  $\delta^{18}O_{carb}$  (r = -0.17, n = 34, 438  $p(\alpha) > 0.10; r = -0.24, n = 81, p(\alpha) < 0.05$ , respectively), Mn/Sr ( $r = -0.45, n = 34, p(\alpha) < 0.01; r$ 439 = -0.01, n = 81,  $p(\alpha) > 0.10$ , respectively) and Mg/Ca (r = 0.00, n = 34,  $p(\alpha) > 0.10$ ; r = -0.34, n = -0.34440 81,  $p(\alpha) < 0.01$ , respectively). Although  $\delta^{34}S_{CAS}$  may become coupled to CAS concentrations 441 through diagenetic, dolomitization or chemical extraction processes (Marenco et al., 2008; Wotte 442 et al., 2012), the lack of  $\delta^{34}S_{CAS}$ -CAS relationships at Wangcun and Duibian (r = -0.28, n = 34, 443  $p(\alpha) > 0.10; r = -0.19, n = 81, p(\alpha) \sim 0.05$ , respectively) is consistent with little to no 444 post-depositional alteration. Additionally,  $\delta^{34}S_{CAS}$  variations during the SPICE are similar to those 445 446 reported from multiple middle-upper Cambrian sections globally (see Section 5.3). Therefore, the  $\delta^{34}S_{CAS}$  profile in the present study is inferred to represent a well-preserved primary marine 447 isotopic record. 448

449

### 450 5.2. Oceanic redox conditions during the SPICE

451 5.2.1. Redox conditions in study sections

452 Redox changes on the Jiangnan Slope during the SPICE can be evaluated using elemental 453 proxies (i.e., UEF, MOEF, and Corr/P). Uptake of U commences under suboxic conditions (i.e., 454 around the Fe(III)/Fe(II) redox threshold), whereas uptake of Mo requires euxinic conditions (i.e., presence of aqueous hydrogen sulfide) (Algeo and Li, 2020). Mo-U enrichment can be used to 455 456 roughly assess bottomwater redox conditions, with U<sub>EF</sub> of <3, ~3-10, and >10 and Mo<sub>EF</sub> of <5, ~5-50, and >50 indicative of oxic, suboxic, and euxinic environments, respectively (Algeo and 457 Tribovillard, 2009; Scott and Lyons, 2012). However, the threshold values of MoEF and UEF are 458 likely to be formation-specific and may vary between depositional systems due to differing uptake 459 pathways (Algco and Liu, 2020). Corg/P ratios are especially useful for redox assessments in 460 carbonate facies (in which low organic content can limit trace-metal uptake), with values of <50, 461 462 ~50-100, and >100 indicative of oxic, suboxic, and anoxic environments, respectively (Algeo and 463 Ingall, 2007). The P in the study units was originally deposited in association with organic matter 464 and/or Fe-(oxyhydr)oxides rather than carbonate minerals, and it was thus sensitive to redox changes, as shown by positive correlations with TOC and Fe<sub>2</sub>O<sub>3</sub>, and negative correlations with 465 CaO (see Supplemental Fig. S3). 466

467 In the study sections, U<sub>EF</sub> and Mo<sub>EF</sub> values are mostly low (<10), indicating that oxic to mildly suboxic conditions prevailed throughout the SPICE event (Figs. 5-7). At Wangcun, UEF and 468 469 MoEF peaks are found mainly in samples with low Al content, suggesting that they are artifacts of 470 using a small denominator in the EF calculation. At Duibian A, a shift to moderately reducing (i.e., 471 suboxic) conditions during the Rising SPICE (Unit III) is documented by high U<sub>EF</sub> (to 5-10) 472 combined with low Mo<sub>EF</sub> (~1) for samples with Al<sub>2</sub>O<sub>3</sub> > 1%, obviating the possibility of an artifact 473 associated with low Al content. A UEF vs. MoEF crossplot shows that most samples plot in the oxic 474 field, except for a few close to the suboxic field (Fig. 8A-C). Core/P ratios are mostly <30, which 475 strongly supports oxic conditions, with slightly higher values (to ~50) only within Unit III, 476 consistent with somewhat more reducing (e.g., suboxic) conditions during the Rising SPICE (Figs. 5F, 8D). At Duibian, the Mo<sub>EF</sub> and C<sub>org</sub>/P proxies covary positively (r = +0.62, n = 69,  $p(\alpha) < 0.05$ ) 477 478 but lack a significant relationship to  $U_{EF}$  (r = -0.01, n = 69,  $p(\alpha) > 0.05$ , r = -0.20, n = 31,  $p(\alpha) > 0.05$ 0.05, respectively). We regard MoEF and Corg/P as the more reliable redox proxies given the mutual 479 consistency of their secular patterns and the fact that they indicate more reducing conditions at the 480 481 peak of the SPICE, as expected for an event marked by enhanced marine productivity (Zhou et al., 482 2015). The pattern of declining UEF during the mid-SPICE may have some other cause, e.g., drawdown of global seawater U concentrations due to expansion of oxygen minimum zones 483 (OMZs) (cf. Hetzel et al., 2009). 484

485 The pattern of secular variation in regional seawater redox conditions reconstructed in our study is independently supported by paired CAS and pyrite sulfur isotope (i.e.,  $\delta^{34}S_{CAS}$  and 486  $\delta^{34}S_{pyrite}$ ) analyses. The  $\Delta^{34}S_{CAS-pyrite}$  values of both shallow- and deep-water sections from 487 Laurentia and Gondwana are consistently high (~20-40 ‰) in the late Guzhangian, with a 488 489 decrease to a minimum (~-20 to 0 ‰) in the middle Paibian, followed by a rebound to high values (~20-40 ‰) in the early Jiangshanian (Gill et al., 2011) (Fig. 9D). This pattern is consistent with a 490 strong reduction of the seawater sulfate pool during the Rising SPICE, probably as a result of 491 492 large-scale pyrite burial and increased amounts of free H2S in the water column (Algeo et al., 493 2015), reflecting local expansion of shelf anoxia during the late Guzhangian to the middle Paibian (this study). 494

495

### 496 5.2.2. Redox conditions in global ocean

The SPICE interval was marked by rising oxygen levels in both the atmosphere and oceans 497 (Zhang et al., 2022). Atmospheric oxygen levels (pO2) are variously estimated to have risen from 498 499 ~5 to 10% (Krause et al., 2018) or from ~15 to 25% (Saltzman et al., 2011). This oxygenation event was driven by massive burial of organic matter, as revealed by a global rise in  $\delta^{13}C_{earb}$  (Gill 500 et al., 2011), leading to falling atmospheric pCO2 and climatic cooling. Climatic cooling generally 501 502 steepens the equator-to-pole temperature gradient (Barron et al., 1995) and invigorates oceanic 503 circulation (Cai and Chu, 1998), mainly through intensification of zonal winds rather than oceanic temperature contrasts (Wunsch, 2002; Huybers and Wunsch, 2010). In the middle-late Cambrian, 504 a cooler climate promoted global-ocean circulation and deep-ocean ventilation during the Rising 505

506 SPICE (as revealed by a positive shift in carbonate  $\delta^{238}$ U; Dahl et al., 2014) as well as a 507 concurrent intensification of continent-margin upwelling (Stouffer et al., 2006). Changes in 508 upwelling intensity were focused along specific continental margins, leading to locally elevated 509 productivity and organic carbon sinking fluxes and, thus, expanded oxygen minimum zones 510 (OMZs), despite a general improvement in deep-ocean ventilation. Thus, global-ocean redox 511 changes during the SPICE event were spatially variable, depending on proximity to 512 paleo-upwelling zones.

Comparison of redox proxy data from the study sections in South China with those for the 513 514 globally distributed auxiliary sections (Fig. 1A) demonstrates a systematic pattern of 515 environmental redox variation during the SPICE event. The Pre-SPICE to Early SPICE intervals are marked by weaker organic matter burial (thus lower  $\delta^{13}C_{carb}$  values) and consequently higher 516 atmospheric pCO2 (thus warmer climate), which led to weakened global-ocean circulation and 517 depressed marine productivity (cf. Stouffer et al., 2006). This change resulted in expanded 518 global-ocean hypoxia (e.g., a negative shift in  $\delta^{238}$ U; Dahl et al., 2014), while OMZs on shelf 519 margins contracted (due to low productivity), resulting in mostly oxic conditions on the lower 520 521 slope (this study) and a reduction in euxinia (as revealed by decreased enrichments of Mo, U and V) on the upper slope (Gill et al., 2021). A major redox transition occurred during the Rising 522 SPICE, when massive organic matter burial (thus higher 813Ccarb values) resulted in declining 523 524 atmospheric pCO2 and rising O2 (Saltzman et al., 2011; Krause et al., 2018). Concurrently, climatic cooling due to lower pCO2 led to improved global-ocean ventilation and oxygenation (i.e., 525 first-order positive shifts in  $\delta^{238}$ U; Dahl et al., 2014), while elevated marine productivity led to an 526 527 expansion of OMZs (i.e., locally more hypoxic conditions in lower slope settings; this study). 528 Ocean-redox conditions changed again during the Falling SPICE, marked by a contraction of OMZs in the late Paibian to early Jiangshanian that resulted in a return of oxic conditions to deep 529 slope facies (e.g., Duibian, this study). This development was probably in response to reduced 530 marine productivity, as recorded by declining  $\delta^{13}C-\delta^{34}S$  of the Falling SPICE interval. 531

532 Despite commonalities in temporal patterns of redox variation, the study and auxiliary 533 sections exhibit regionally unique features that may have been due to differences in water depth, 534 watermass restriction, or regional oceanic circulation (see Supplemental file for facies data and water-depth interpretations). Compared to the largely oxic conditions observed in South China, 535 some localities exhibit more intense seawater de-oxygenation. For example, local redox proxy 536 data (e.g., Fe speciation) for the Andrarum no. 3 core (Alum Shale, Sweden) record dominantly 537 538 euxinic conditions during the Pre-SPICE, followed by a shift toward less reducing conditions (ferruginous) close to the peak of the SPICE, and a return to euxinic conditions during the Falling 539 540 and Post-SPICE intervals (Gill et al., 2011, 2021; Dahl et al., 2013). This pattern was punctuated 541 by short-term (millennial-scale) dysoxic episodes, as inferred from sedimentological and 542 ichnological data (Egenhoff et al., 2015). The generally more reducing conditions of the Alum Shale may have been related to stagnation of watermass circulation on the Baltic Craton during the 543 Cambrian eustatic highstand (Thickpenny, 1987; Høyberget and Bruton, 2008). Moreover, OMZ 544

545 expansion during the Rising SPICE did not cause enrichments of trace metals (e.g., Mo, U, and V) in the Alum Shale (Gill et al., 2021), which is consistent with either regional watermass restriction 546 547 (cf. Algeo and Maynard, 2008) or a general drawdown of trace metals in the global ocean linked 548 to expanded anoxia in the Early SPICE interval (cf. Dahl et al., 2014, 2019). A global-ocean redox proxy record (i.e., carbonate  $\delta^{238}$ U) has been generated only for the Mount Whelan core (Australia; 549 Dahl et al., 2014). It shows an intensification of global-oceanic hypoxia during the Pre-SPICE to 550 551 early Rising SPICE, with a shift towards more oxic conditions in the late Rising SPICE, before a probable re-expansion of anoxic facies during the Falling SPICE, although aspects of these 552 interpretations are somewhat uncertain owing to the sparsity and unequal temporal distribution of 553 the  $\delta^{238}$ U data. 554

555

556 5.3. Global carbon-sulfur cycle changes during the SPICE

The marine carbon and sulfur cycles are commonly coupled through biochemical processes 557 such as photosynthesis and microbial sulfate reduction (Jorgensen, 1982; Mazumdar et al., 2012; 558 559 Antler et al., 2013). Major fractionations of carbon and sulfur isotopes are associated with the 560 production of organic matter from dissolved inorganic carbon (DIC) and pyrite from sulfate (Bottrell and Newton, 2006). Owing to the long residence times of DIC and sulfate in the ocean 561 (~100 kyr and ~13 Myr, respectively; Claypool et al., 1980; Zeebe and Wolf-Gladrow, 2001), their 562 isotopic compositions in seawater generally reflect changes in the burial fluxes of organic matter 563 564 and pyrite.

During the Early and Rising SPICE, the carbon and sulfur isotopic profiles show general 565 566 first-order positive excursions. At Wangcun and Duibian A, the late Guzhangian to middle Paibian interval (i.e., Units II to III) is characterized by roughly simultaneous positive excursions of 567  $\delta^{34}S_{CAS}$  (~+35 ‰ to ~+50 ‰ and ~+30 ‰ to ~+50 ‰, respectively), and  $\delta^{13}C_{carb}$  (~0 ‰ to ~+4 ‰ 568 and ~+1 ‰ to ~+4 ‰, respectively). At Wangcun,  $\delta^{34}S_{CAS}$  is positive correlated (r = +0.59, n = 9, 569  $p(\alpha) \sim 0.05$ ) to  $\delta^{13}C_{carb}$  during the Early SPICE, while no relationship (r = -0.43, n = 5,  $p(\alpha) > 0.05$ ) 570 0.05) exists during the Rising SPICE (Fig. 2C-D). At Duibian A, a rise of  $\delta^{34}S_{CAS}$  (~+30 ‰ to 571 ~+45 ‰) corresponds to nearly no changes of δ13Ccarb (~+1 ‰) during the Early SPICE, while a 572 positive correlation is marginally statistically significant (r = +0.67, n = 7,  $p(\alpha) \sim 0.05$ ) during the 573 Rising SPICE (Fig. 3C-D). The first-order positive correlation between  $\delta^{34}S_{CAS}$  and  $\delta^{13}C_{carb}$  during 574 the Early and Rising SPICE suggests coupling of the global marine carbon and sulfur cycles, 575 576 presumably due to co-burial of organic matter and pyrite and a small reservoir of marine sulfate, 577 driven by elevated marine productivity and expanded shelf/slope hypoxia (Dahl et al., 2014; Zhang et al., 2022; this study). 578

579 During the Falling SPICE, the carbon and sulfur isotopic profiles show negative excursions
 and intermittent coupling. The late Paibian-early Jiangshanian interval (i.e., Falling SPICE, Unit
 581 IV) is characterized by simultaneous shifts toward lower δ<sup>13</sup>C<sub>carb</sub> and δ<sup>34</sup>S<sub>CAS</sub> values, declining to
 582 minima of ~+1 ‰ and ~+15 to +20 ‰, respectively. The correlations are significant for δ<sup>34</sup>S<sub>CAS</sub> vs.

583  $\delta^{13}C_{\text{carb}}$  at Wangcun (r = +0.63, n = 7,  $p(\alpha) \sim 0.05$ ) and Duibian A (r = +0.56, n = 14,  $p(\alpha) < 0.05$ ),

but not at Duibian B (r = +0.17, n = 38,  $p(\alpha) > 0.10$ ) (Figs. 2C-D, 3C-D, 4C-D). These differences may exist owing to regional variation in the net burial rates of organic matter and pyrite, but they are not inconsistent with reduced marine productivity in South China during the Falling SPICE.

587 In contrast to the SPICE, the Pre-SPICE and Post-SPICE intervals in the study sections are characterized by non-synchronous variation in  $\delta^{34}S_{CAS}$  and  $\delta^{13}C_{carbs}$  suggesting a general 588 decoupling of the global marine carbon and sulfur cycles. In detail, the middle Drumian to middle 589 Guzhangian interval (i.e., Pre-SPICE, Unit Ib) exhibits fluctuations in  $\delta^{34}S_{CAS}$  (from ~+25 ‰ to 590 ~+35 ‰) that coincided with little change in  $\delta^{13}C_{carb}$  (~0 ‰) at Wangcun (Fig. 2C-D). The early 591 Jiangshanian interval (i.e., Post-SPICE, Unit IV) exhibits a shift toward lower  $\delta^{13}C_{earb}$  values 592 593 (~+1 ‰) that is decoupled from  $\delta^{34}S_{CAS}$  at Wangcun (r = -0.07, n = 8,  $p(\alpha) > 0.10$ ) and Duibian B  $(r = +0.44, n = 11, p(\alpha) > 0.10)$  (Figs. 2C-D, 3C-D). Thus, variations in marine productivity and 594 organic carbon and pyrite burial were not sufficiently large in the pre-SPICE and post-SPICE 595 intervals to override other influences on the global marine carbon and sulfur cycles. 596

All of the auxiliary sections show  $\delta^{13}C_{earb}$ - $\delta^{34}S_{CAS}$  coupling during the late Guzhangian to 597 earliest Jiangshanian (i.e., Early to Falling SPICE) and decoupling during the early-middle 598 599 Guzhangian (i.e., Pre-SPICE) and early Jiangshanian (i.e., Post-SPICE), conforming to the general pattern reported here for Wangcun and Duibian. Covariation of  $\delta^{13}C_{carb}$  and  $\delta^{34}S_{CAS}$  is observed 600 not only in deep-water sections (e.g., TE-1 Texas County Core, Duibian) but also in 601 602 intermediate-depth (e.g., Mount Whelan Core, Shingle Pass) and shallow-water (e.g., Lawson Cove) sections of Laurentia and Gondwana (Fig. 9). This transregional pattern of carbon-sulfur 603 cycling confirms marine productivity as the main control on coupled b13Ccarb-b34ScAS variation 604 throughout the SPICE event (cf. Dahl et al., 2014). In contrast, the Pre-SPICE and Post-SPICE 605 606 intervals were likely associated with a warmer climate, more sluggish oceanic circulation, and lower (and less variable) marine productivity. 607

608

### 609 5.4. Trilobite biodiversity and its relationship to environmental changes during the SPICE event

610 5.4.1. Global comparison of trilobite biodiversity

611 Trilobite biostratigraphic studies from Laurentia resulted in recognition of the EMBE at the 612 end of the Guzhangian Stage and the ESBE in the early Jiangshanian Stage, each of which reportedly exhibits two phases of extinction (Longacre, 1970; Palmer, 1965a, 1965b; Stitt, 1971; 613 Taylor, 2006; Babcock et al., 2017). For the EMBE, the first phase coincided with the base of the 614 Laurentian Coosella perplexa Subzone of the latest Guzhangian (i.e., Early SPICE) and was 615 marked by the disappearance of the majority of shallow-water trilobites with no concurrent change 616 in 813Ccarb values (Palmer, 1979; Gerhardt and Gill, 2016). The second phase of the EMBE was 617 less severe and coincided with the uppermost C. perplexa Subzone of the Early Paibian (i.e., onset 618 619 of the Rising SPICE), marked by the disappearance of surviving members of the C. perplexa 620 Subzone fauna. Generally, the EMBE is characterized not only by a decline in species diversity 621 but also by a shift to biofacies that have broader environmental distributions as well as extensive immigration of taxa from off-shelf and shelf-margin sites to shelf areas (Westrop and Cuggy, 622

623 1999). The ESBE is relatively less studied than the EMBE, but its first and second phases coincided with the lowermost Ir: major Zone and the Taenicephalus Zone of the early 624 625 Jiangshanian (i.e., Post-SPICE), respectively. Collectively, these extinctions resulted in a shift in 626 dominance from the Marjumiid Biomere of the Guzhangian to the Pterocephaliid Biomere of the earliest Paibian, and subsequently to the Ptychaspid Biomere of the early Jiangshanian (Palmer, 627 628 1984; Saltzman et al., 2000) (Fig. 10). Although biomeres were first recognized from patterns seen in Laurentian trilobite faunas, correlative patterns of diversity changes can now be recognized 629 elsewhere, including in South China (Zhou and Zhen, 2008; Zhang et al., 2021). 630

631 The trilobite biodiversity curves generated for the present study are only regionally 632 representative but nonetheless in broad accord with global evolutionary trends during the late Guzhangian. Thus, although the Marjumiid, Pterocephaliid, and Ptychaspid biomeres sensu stricto 633 were endemic to Laurentia (e.g., Saltzman et al., 2000), similar and iterative patterns of trilobite 634 evolutionary diversification can be seen in age-equivalent successions in South China (Zhang et 635 al., 2021; this study), making these biomeres sensu lato of global significance. In the present study, 636 637 the first phase of the EMBE is recognizable as a decrease in trilobite range-through species 638 diversity from ~>10 to 1 at Wangcun (i.e., at ~230-270 m) and from ~25 to ~5 at Duibian (i.e., at -4 to +1 m), during an interval of nearly constant or slightly positive-shifted δ<sup>13</sup>C<sub>carb</sub> values (i.e., 639 Early SPICE) (Fig. 10A, D). A similar decline in trilobite diversity without a major carbon isotope 640 641 excursion has been reported from strata in Laurentia that are age-equivalent to the Early SPICE interval (Palmer, 1979, 1984; Gerhardt and Gill, 2016) (Fig. 10E). This observation suggests that 642 the trilobite diversity curves in the present study are globally representative (cf. Zhou and Zhen, 643 2008; Zhang et al., 2021), and that the EMBE was a widespread event triggered by global 644 645 environmental changes during the Early SPICE interval.

The newly generated trilobite diversity curves for the Rising SPICE interval at both Wangcun 646 647 and Duibian are regionally representative (see Zhang et al., 2021). Although a gradual increase in 648 trilobite diversity occurred immediately after the EMBE in Laurentia (Fig. 10E; Palmer, 1984; 649 Rowell and Brady, 1976), our study reveals a trend toward lower diversity at ~1-30 m in Duibian A, coincident with an increase of  $\delta^{13}C_{carb}$  and stable  $\delta^{34}S_{CAS}$  values during the Rising SPICE, 650 reaching a minimum of one species at the peak of the SPICE (Fig. 10D). A drop in taxonomic 651 diversity during the Rising SPICE was also reported from the Paibi section (which is the GSSP of 652 the base of the Furongian Series and Paibian Stage), ~50 km southwest of Wangcun (Peng et al., 653 2004; Zhang et al., 2021). However, a recent study from South China inferred approximately 654 constant trilobite diversity during the Rising SPICE, before a decline during the Falling SPICE 655 (Zhang et al., 2021). 656

657 The trilobite biodiversity curves from the present study are in accord with documented 658 evolutionary trends for the Jiangshanian of South China (Zhou and Zhen, 2008; Zhang et al., 659 2021), which may be representative of contemporaneous global patterns. Trilobite species 660 diversity remained at a higher level through the end of the SPICE, before declining during the 651 ESBE at the transition to the Post-SPICE interval (Palmer, 1979, 1984; Zhou and Zhen, 2008; this

662 study). Biodiversity changes in South China are comparable to those reported from correlative 663 units in Laurentia (Fig. 10D-E), although, as with the EMBE, the existence of two separate 664 extinction pulses during the ESBE has not been recognized in the present study sections.

Proposed triggers for the EMBE include cooling climate/seawater (e.g., climatic cooling, rise of permanent thermocline, and upwelling of cool nutrient-rich waters) (Öpik, 1966; Lochman-Balk, 1970; Stitt, 1975; Perfetta et al., 1999; Elrick et al., 2011), and seawater anoxia and/or euxinia (Saltzman et al., 1998; Hurtgen et al., 2009; Gill et al., 2011; Dahl et al., 2014). In contrast, the environmental controls on the ESBE have received little consideration to date. Below, we consider possible environmental controls on the EMBE and ESBE, based on a combination of previous studies and our new paleontological and geochemical data.

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#### 673 5.4.2. End-Marjuman Biomere Extinction (EMBE)

The cause of the extinction of the Marjumiid Biomere (sensu lato) and the spread of the 674 Pterocephaliid Biomere (sensu lato) over shelf areas during the Early SPICE has long been 675 debated. Early work focused on differences in the preferred habitats of these two biomeres 676 677 (Palmer, 1984; Pratt, 1992). The fauna of the Marjumiid Biomere mostly occupied shallow-water sandstone and siltstone facies close to paleo-shorelines, resulting in relatively high degrees of 678 endemism. In contrast, the fauna of the Pterocephaliid Biomere was better represented in 679 680 deeper-water, shale-rich facies beneath the oceanic thermocline, allowing it to migrate globally and develop into a eurytopic assemblage (Pratt, 1992). More recent work has focused on the role 681 of temperature change, with climatic cooling, a rise of the permanent thermocline, and/or 682 683 upwelling of deep waters being proposed as the trigger for the EMBE (Palmer, 1984; Saltzman et 684 al., 2000; Elrick et al., 2011). Whereas the Marjumiid Biomere favored warmer waters, the Pterocephaliid Biomere, and especially its agnostoid elements and olenimorphic morphotypes, 685 preferred cooler, deeper waters (Fortey and Owens, 1990), although one of its members, the genus 686 687 Erixanium, had a narrow latitudinal range centered on the paleo-Equator (Lu and Lin, 1989; Stitt 688 et al., 1994; Zhou and Zhen, 2008), suggesting a preference for warmer temperatures (Hughes, 2000). At Duibian, representatives of the Pterocephaliid Biomere comprise a low-diversity fauna 689 690 dominated by proceratopygine and iwayaspine species (Lu and Lin, 1989; Hughes and Rushton, 691 1990; Peng et al. 2012) that may have been especially tolerant of challenging or variable environmental conditions linked to oxygen stress (cf. Zhang et al., 2021). This fauna yielded to the 692 Ptychaspiid Biomere, which was characterized by a decline in endemic species and an increase in 693 694 more cosmopolitan elements (Cook and Taylor, 1975; Żylińska, 2001, 2002; Álvaro et al., 2013). Taxa appearing immediately after the ESBE include both the widespread and arguably pelagic 695 696 genus Irvingella (Fortey, 1985), which was adept at crossing open ocean basins, and the more 697 typical "ptychopariid" Maladioidella, which was restricted to shallow-shelf settings along the 698 margin of Gondwana (Rushton and Hughes, 1996) but spanned an unusually wide range of 699 paleolatitudes (Hughes, 2000). Such widespread occurrence along the Gondwanan margin may 700 attest to a reduced latitudinal temperature gradient following the ESBE, possibly associated with

701 global warming. The biotic succession in the present study sections is thus consistent with cooling

702 in conjunction with the EMBE followed by warming in association with the ESBE.

The biotic extinctions during the Early SPICE may have been analogous to the extinction 703 704 events at the onset and termination of the Hirnantian Glaciation of the Late Ordovician, about 50 705 Myr later (Algeo et al., 2016). The <1-Myr-long Hirnantian Glaciation was marked by a ~5 °C decline of global temperatures (Trotter et al., 2008; Finnegan et al., 2011), a ~70-150 m sea-level 706 fall (Brenchley et al., 2003; Finnegan et al., 2011), and a ~4-6 ‰ positive δ<sup>13</sup>C excursion (i.e., the 707 Hirnantian carbon isotope excursion or HICE, Bergström et al., 2006). An extinction of 708 709 warm-water faunas at the onset of this glacial episode (Barash, 2014) and its replacement by the 710 cool-water-adapted Hirnantia Fauna (Zhan et al., 2010; Rasmussen and Harper, 2011) were possibly analogous to the transition from the warm-water Marjumiid Biomere to the cool-water 711 712 Pterocephaliid Biomere during the Early SPICE (Palmer, 1984; Pratt, 1992).

713 The role of temperature change as a control on trilobite biomeres during the middle-late 714 Cambrian has been inadequately tested to date using oxygen-isotope data. Phosphatic brachiopod  $\delta^{18}$ O from western Laurentia provided evidence of a climate cooling event during the Pre-SPICE, 715 716 followed by climate warming during the Rising SPICE (Elrick et al., 2011). However, this pattern 717 is likely to represent only a local signal linked to shallowing of a tropical shelf as a result of global sea-level fall (Fig. 10F), shifting the local watermass from the cooler thermocline into the warmer 718 719 surface layer of the ocean during the SPICE. The EMBE was followed by the rise of the cool-water Pterocephaliid Biomere fauna in western Laurentia (Stitt, 1975; Rowell and Brady, 720 1976; Palmer, 1984), which is inconsistent with the general climatic warming inferred by Elrick et 721 722 al. (2011). However, global climate cooling is likely to have prevailed during the SPICE, as 723 evidenced by sedimentological, stratigraphic, and geochemical records (e.g., greater burial sequestration of organic matter, and thus a reduced greenhouse effect) (Saltzman, 2005; Cherns 724 725 and Wheeley, 2009; Sørensen et al., 2020) (Fig. 10I).

726 Redox changes are likely to have contributed to trilobite biomere turnovers, although the 727 extinction of the Marjuman Biomere during the latter part of the Early SPICE lagged a major negative shift in carbonate \delta<sup>238</sup>U during its earlier part (Dahl et al., 2014; note: some uncertainty 728 linked to limited fossil data from Whelan core), suggesting that transient expansion of 729 730 global-ocean hypoxia was not the proximate cause of the EMBE. Rather, the EMBE may have been due to the impact of global-ocean circulation changes on regional redox conditions. Global 731 732 climatic cooling is likely to have led to an expansion of seawater hypoxia on shelf margins subject 733 to upwelling, where nutrient-rich deepwaters enhanced regional productivity and organic carbon sinking fluxes, producing locally more reducing conditions (cf. Whitney et al., 2005; Stouffer et al., 734 735 2006). This hypothesis is consistent with a concurrent improvement in global-ocean ventilation 736 during the Early SPICE and Rising SPICE, as evidenced by first-order positive shifts of carbonate  $\delta^{238}$ U (Dahl et al., 2014). The present study provides evidence of OMZ expansion as shown by a 737 transition from oxic to suboxic waters on the Jiangnan Slope of South China during the Rising 738 SPICE (Fig. 10C). This redox change is likely to have placed outer-shelf trilobite communities 739

740 under stress despite generally improved ventilation of the global ocean. The extinction of indigenous cool-water trilobites belonging to the early Paibian Pterocephaliid Biomere in 741 742 deep-slope settings supports our inference of OMZ expansion and increased shelf anoxia as the 743 principal control on the EMBE and the subsequent reduced diversity of the Pterocephaliid Biomere (cf. Pratt, 1992), as does the nature of the trilobites that dominate the Paibian fauna (see 744 above). During the latter part of the Falling SPICE, contraction of OMZs on shelf margins 745 permitted local increases in the abundance and diversity of the Pterocephaliid Biomere fauna 746 sensu lato prior to the ESBE (Fig. 10D). 747

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749 5.4.3. End-Steptoean Biomere Extinction (ESBE)

750 The causes of the ESBE remain uncertain. Given that the ESBE occurred at the end of a 751 period of global carbon cycle instability, this biotic event is likely to have been related to the attenuation of environmental disturbances associated with the carbon cycle. The ESBE was 752 753 probably related to global climate change but evidence for this is presently scant. Bioapatite oxygen isotopes suggest that in Laurentia, the Falling and Post-SPICE episodes corresponded to a 754 755 cooling climate (Elrick et al., 2011), although cooling may have been due to local water-column deepening as a result of global warming and continental ice mass decay. Global climate change 756 during the ESBE has not been studied to date. If the ESBE is analogous to the extinction of the 757 758 Himantia Fauna at the termination of the Himantian Glaciation, then it may also have been associated with global climatic warming (Fig. 101). However, environmental factors controlling 759 the ESBE were probably not simply the opposite of those influencing the EMBE (e.g., climate 760 warming as opposed to earlier climate cooling), because there are cosmopolitan taxa among both 761 762 the latest Paibian and early Jiangshanian biomeres (e.g., Hedinaspis, Irvingella, and Maladioidella at Duibian; Peng et al., 2012). 763

The spatial scale of seawater redox changes during the ESBE requires consideration. As oxic seawater conditions persisted during this time interval throughout the study area, local redox changes cannot explain this extinction event (Fig. 10C). Widespread seawater anoxia may have developed in the global ocean during this interval, as indicated by rapid increases in  $\delta^{34}S_{CAS}$  and  $\Delta^{34}S_{CAS-pyrite}$  in both South China and Laurentia (Fig. 9). However, the geographic extent of oceanic redox changes remains to be tested via a proxy suitable for addressing global trends (e.g., carbonate U isotopes).

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### 772 6. Conclusions

Paired  $\delta^{13}C_{carb}$ - $\delta^{34}S_{CAS}$  profiles and trilobite species diversity curves spanning the lower Drumian to lower Jiangshanian were generated for a shallow-water succession at Wangcun and a deep-water succession at Duibian, located on the Jiangnan Slope in South China. Enrichment factors for U and Mo, along with  $C_{org}/P$  ratios, suggest mostly oxic conditions in the study sections during the SPICE event, except hypoxic (i.e., suboxic) seawater conditions from the earliest Paibian to the middle Paibian (i.e., Rising SPICE) at Duibian.  $\delta^{13}C_{carb}$  was tightly coupled with

 $\delta^{34}S_{CAS}$  during the late Guzhangian to late Paibian, demonstrating first-order control of 779 contemporaneous environmental changes by the marine carbon cycle. Local 813Ccarb trends mirror 780 positive excursions of ~3 ‰ found globally during the Rising SPICE, which were driven by 781 782 elevated global marine productivity and enhanced burial of organic matter. The major positive excursion in  $\delta^{34}S_{CAS}$  started earlier than that of  $\delta^{13}C_{earb}$  during the Early SPICE, and  $\delta^{34}S_{CAS}$ 783 remained stable during the Rising SPICE before declining during the Falling SPICE. The Falling 784 785 SPICE was characterized by diminished global marine productivity, resulting in reduced co-burial of organic matter and pyrite. In order to investigate the global marine carbon-sulfur cycles during 786 the SPICE event, we further compiled 813Cearb-834SCAS variations in four shallow- to deep-water 787 788 successions on the slope/continental margin of Laurentia and Gondwana. The relationship between  $\delta^{13}C_{carb}$  and  $\delta^{34}S_{CAS}$  in these settings is similar to that at Wangcun and Duibian, 789 suggesting globally consistent patterns of carbon-sulfur cycling during the SPICE event. 790

The local trilobite species diversity curves are comparable to those from Laurentia, showing a 791 792 major decline in biodiversity during the End-Marjuman Biomere Extinction (EMBE) in the Early SPICE, as well as the End-Steptoean Biomere Extinction (ESBE) in the Post-SPICE. Therefore, 793 794 the SPICE event should be extended downwards to include the Early SPICE interval of the late 795 Guzhangian Stage, in a manner that more clearly links the EMBE to the SPICE. We further evaluated effects of climate change, global and local seawater redox conditions on these biotic 796 797 extinctions, and propose that expansion of seawater hypoxia on shelf margins as a result of global 798 climate cooling, invigorated global ocean circulation, and intensified continent-margin upwelling may have directly contributed to the extinction of the trilobite fauna of the Marjumiid Biomere 799 800 during the Early SPICE, and the attenuation of those environmental changes during the Falling 801 SPICE set the stage for the subsequent extinction of the Pterocephaliid Biomere.

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1157	Fig. 1. Paleogeographic maps of the Late Cambrian world (A), South China Craton (B), and local
1158	maps of Guzhang County (C) and Jiangshan urban area (D), showing locations of Wangcun,
1159	Duibian A and B, and other globally distributed SPICE sections discussed in the text. Map A is
1160	from https://deeptimemaps.com authorized by Colorado Plateau Geosystems Inc. Map B is
1161	adapted from Li et al. (2018).
1162	
1163	Fig. 2. Trilobite and isotopic data for the Wangcun section: (A) trilobite species ranges, (B)
1164	trilobite species diversity; (C) carbonate (carb) $\delta^{13}$ C (‰ VPDB); and (D) carbonate-associated
1165	sulfate (CAS) $\delta^{34}$ S (‰ CDT). In panel A, trilobite range data are from Peng et al. (2009), with red
1166	and blue lines representing actual range and range-through data, respectively. The base of the
1167	Paibian and the base of the Jiangshanian have been correlated from Duibian A based on
1168	trilobite-carbon isotope biochemostratigraphy from Zuo et al. (2018). The gray fields represent the
1169 1170	stratigraphic extent of SPICE; 'Early SPICE' is newly defined herein, and its base implies an earlier onset of the SPICE than inferred in some earlier studies (see Section 2.5). Abbreviations: A.
1170	inexpectans = Agnostus inexpectans; Ag. orientalis = Agnostotes orientalis; C. plumula =
1171	Corynexochus plumula; E. rectang. = Erixanium rectangularis; G. reticulatus = Glyptagnostus
/-	confinence priming, 2. rectang. 2. manual rectangularis, c. reneuranis dipplognostis

reticulatus; G.s. = Glyptagnostus stolidotus; Go.n. = Goniagnostus nathorsti; L.a. = Lejopyge 1173 armata; Li.r. = Linguagnostus reconditus; L.l. = Lejopyge laevigata; P.a. = Ptychagnostus 1174 1175 aculeatus; P.p. = Ptychagnostus punctuosus; Pr.b. = Proagnostus bulbus; E. = Erixanium; Ir. = Irvingella; T. = Tomagnostella; Jiangshan. = Jiangshanian; Wu. = Wuliuan. 1176 1177 1178 Fig. 3. Trilobite and isotopic data for the Duibian A section: (A) trilobite species ranges, (B) trilobite species diversity; (C) carbonate (carb) δ13C (‰ VPDB); and (D) carbonate-associated 1179 sulfate (CAS) δ34S (‰ CDT). Trilobite data from Peng et al. (2012). See Figure 2 caption for other 1180 1181 details. 1182 Fig. 4. Trilobite and isotopic data for the Duibian B section: (A) trilobite species ranges, (B) 1183 trilobite species diversity; (C) carbonate (carb)  $\delta^{13}$ C (% VPDB); and (D) carbonate-associated 1184 1185 sulfate (CAS) 834S (% CDT). Trilobite data from Peng et al. (2012). See Figure 2 caption for other 1186 details. 1187 1188 Fig. 5. Trilobite and elemental data for the Wangcun section: (A) trilobite species diversity; (B) 1189 total organic carbon (TOC); (C) Al2O3; (D) UEF; (E) MOEF; and (F) Core/P. Red circles represent samples with low detrital content (Al2O3 < 1%) that may result in artificially high UEF and MOEF 1190 1191 values. For abbreviations refer to Figure 2. 1192 1193 Fig. 6. Trilobite and elemental data for the Duibian A section: (A) trilobite species diversity; (B) total organic carbon (TOC); (C) Al<sub>2</sub>O<sub>3</sub>; (D) U<sub>EF</sub>; (E) Mo<sub>EF</sub>; and (F) C<sub>org</sub>/P. For other details see 1194 1195 Figure 5 caption; for abbreviations refer to Figure 2. 1196 Fig. 7. Trilobite and elemental data for the Duibian B section: (A) trilobite species diversity; (B) 1197 1198 total organic carbon (TOC); (C) Al2O3; (D) UEF; (E) MOEF; and (F) Corg/P. For other details see Figure 5 caption; for abbreviations refer to Figure 2. 1199 1200 1201 Fig. 8. Seawater redox conditions in Wangcun, Duibian A and B sections, based on (A-C) MoEF vs. 1202 UEF and (D) TOC vs. P. Panels A-C are after Algeo and Tribovillard (2009): UEF values are mostly 1203 3-10 in suboxic, and Mo<sub>EF</sub> values > 10 in anoxic environments. Redox thresholds in panel D are after Algeo and Ingall (2007): Corg:P ratios are mostly < 50 in oxic, 50 to 100-125 in suboxic and > 1204 1205 100-125 in anoxic environments. In panel D, all data are shown without reference to Al2O3 content, because the Corg:P proxy is independent of Al2O3 content. 1206 1207 Fig. 9. Global comparisons of (A) δ<sup>13</sup>C<sub>carb</sub> (‰ VPDB), (B) δ<sup>34</sup>S<sub>CAS</sub> (‰ CDT), (C) δ<sup>34</sup>S<sub>pyrite</sub> (‰ 1208 CDT), and (D) △34S(CAS-ov) profiles. Data sources: Shingle Pass (Saltzman et al., 1998; Gill et al., 1209 1210 2007); Lawson Cove and TE-1 Texas County Core (Gill et al., 2011); Mount Whelan Core 1211 (Saltzman et al., 2000; Gill et al., 2011); Wangcun, Duibian A and B (this study). All profiles were 1212 replotted and smoothed using the Drumian-Guzhangian (~500.5 Ma), Guzhangian-Paibian (~497 Ma), and Paibian-Jiangshanian (~494 Ma) boundaries as age tie-points, and assuming isochroneity 1213 1214 of the SPICE peak globally. For sections containing incomplete Guzhangian or Jiangshanian stages, age assignments were made assuming a constant sedimentation rate based on the Paibian 1215 stage. Notes: (1) peak  $\delta^{34}S_{CAS}$  values occurred during the Rising SPICE (Gill et al., 2011; this 1216

1217 study); (2) for Shingle Pass, maximum  $\delta^{13}C_{carb}$  is reached slightly below a facies change, so the 1218 peak SPICE interval may have been truncated (Saltzman et al., 2000; Gill et al., 2011). Color 1219 scheme: green = surface layer, blue = intermediate water depth, and black = deep layer. Note 1220 existence of a depth-related gradient for each isotopic system. See the Supplemental file for 1221 descriptions of the sedimentary settings of the sections analyzed here.

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Fig. 10. Summary figure for regional and global marine proxies during the SPICE event: (A) 1223 1224 δ13Ccarb (‰ VPDB), (B) δ34ScAS (‰ CDT), (C) seawater redox, and (D) trilobite diversity of South 1225 China (this study); (E) trilobite diversity of Laurentia, based on House Range (Utah), Highland Range (Nevada), Desert Range (Nevada) and Royer Ranch (Oklahoma) sections (Palmer, 1978, 1226 1227 1984); (F) sea-level changes of Laurentia, based on studies in Upper Mississippi Valley (Runkel et al., 1998); (G) seawater redox conditions of Baltica, based on studies of Mo-U enrichments and Fe 1228 1229 speciation in Andrarum-3 drillcore (Gill et al., 2011); (H) global seawater redox conditions, based 1230 on studies of carbonate U-isotopes in Whelan no. 1 drillcore, Australia (Dahl et al., 2014); and (I) 1231 general global SSTs, based on studies of Saltzman (2005), Matthews and Al-Husseini (2010) and 1232 Al-Husseini (2017). In panels D and E, colors refer to Pre- and Early SPICE vs. Rising and Falling 1233 SPICE vs. Post-SPICE. In panel E, blue horizontal lines are counted diversity from Palmer (1984). 1234 In panels D and E, due to lack of biostratigraphic constraints (Palmer, 1984), correlations of 1235 trilobite diversity curves are based on diversity peaks. For abbreviations see Figures 2-4. 1236













