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Uncovering the spatial heterogeneity of Ediacaran carbon cycling

C. Li¹ | D. S. Hardisty^{2,3} | G. Luo¹ | J. Huang⁴ | T. J. Algeo^{1,4,5} | M. Cheng¹ | W. Shi¹ | Z. An⁶ | J. Tong¹ | S. Xie¹ | N. Jiao⁷ | T. W. Lyons³

¹State Key Laboratory of Biogeology and Environmental Geology, China University of Geosciences, Wuhan, China

²Department of Geology and Geophysics, Woods Hole Oceanographic Institution, Woods Hole, MA, USA

³Department of Earth Sciences, University of California, Riverside, CA, USA

⁴State Key Laboratory of Geological Processes and Mineral Resources, China University of Geosciences, Wuhan, China

⁵Department of Geology, University of Cincinnati, Cincinnati, OH, USA

⁶Faculty of Earth Sciences, China University of Geosciences, Wuhan, China

⁷State Key Laboratory of Marine Environmental Sciences, Xiamen University, Xiamen, China

Correspondence

C. Li, State Key Laboratory of Biogeology and Environmental Geology, China University of Geosciences, Wuhan, China. Email: chaoli@cug.edu.cn

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Abstract

Records of the Ediacaran carbon cycle (635–541 million years ago) include the Shuram excursion (SE), the largest negative carbonate carbon isotope excursion in Earth history (down to -12%). The nature of this excursion remains enigmatic given the difficulties of interpreting a perceived extreme global decrease in the δ^{13} C of seawater dissolved inorganic carbon. Here, we present carbonate and organic carbon isotope $(\delta^{13}C_{carb}$ and $\delta^{13}C_{ore})$ records from the Ediacaran Doushantuo Formation along a proximal-to-distal transect across the Yangtze Platform of South China as a test of the spatial variation of the SE. Contrary to expectations, our results show that the magnitude and morphology of this excursion and its relationship with coexisting $\delta^{13}C_{org}$ are highly heterogeneous across the platform. Integrated geochemical, mineralogical, petrographic, and stratigraphic evidence indicates that the SE is a primary marine signature. Data compilations demonstrate that the SE was also accompanied globally by parallel negative shifts of δ^{34} S of carbonate-associated sulfate (CAS) and increased ⁸⁷Sr/⁸⁶Sr ratio and coastal CAS concentration, suggesting elevated continental weathering and coastal marine sulfate concentration during the SE. In light of these observations, we propose a heterogeneous oxidation model to explain the high spatial heterogeneity of the SE and coexisting $\delta^{13}C_{org}$ records of the Doushantuo, with likely relevance to the SE in other regions. In this model, we infer continued marine redox stratification through the SE but with increased availability of oxidants (e.g., O2 and sulfate) limited to marginal near-surface marine environments. Oxidation of limited spatiotemporal extent provides a mechanism to drive heterogeneous oxidation of subsurface reduced carbon mostly in shelf areas. Regardless of the mechanism driving the SE, future models must consider the evidence for spatial heterogeneity in δ^{13} C presented in this study.

1 | INTRODUCTION

The Ediacaran Period (635-541 million years ago or Ma) is characterized by a lack of global-scale glaciation (unlike the preceding Cryogenian Period), rising but perhaps still variable oxygen levels on the Earth's surface, and biological innovations culminating in innovation among early animals (Droser & Gehling, 2015; Lyons, Reinhard, & Planavsky, 2014; Och & Shields-Zhou, 2012; Pecoits et al., 2012; Sahoo et al., 2016). The Ediacaran is also characterized by a series of major perturbations to the carbon cycle recorded globally in carbonate carbon isotope ($\delta^{13}C_{carb}$) profiles (Grotzinger, Fike, & Fischer, 2011). Carbonates of the mid-Ediacaran Shuram excursion (SE) yield a decrease in $\delta^{13}C$ values to as low as -12‰, placing it among the most negative excursions in Earth history. These values are appreciably more negative than the mantle $\delta^{13}C$ of ca. -5‰, requiring major inputs of isotopically light carbon to the exogenic Earth system during



FIGURE 1 Geological framework of the Ediacaran Doushantuo Formation in South China. (a) Paleogeographic reconstruction of the Ediacaran Yangtze Platform (modified from Jiang et al., 2011) showing locations of the three study sections (Zhangcunping, Jiulongwan, and Siduping). (b) Proximal-to-distal transect across the Yangtze Platform, showing relative paleodepths of the study sections (after Zhu et al., 2013). Legend: 1. thick-bedded grainstone: 2. micritic dolostone: 3. muddy or silty laminated dolostone: 4. limestone: 5. phosphorite: 6. glaciogenic diamictite; 7, black shale; 8, cherty bands and nodules in the carbonate; 9, carbonate interbeds or lenticular beds; 10, cherty nodules in shale; 11, carbonate concretions; 12, sequence boundary; 13, dark or dark gray silty shale or mudstone

the SE (Grotzinger et al., 2011). The SE is also characterized by a general lack of carbonate isotopic covariation with co-occurring organic carbon ($\delta^{13}C_{ore}$), which contrasts with conventional views of the carbon cycle wherein both $\delta^{13}\text{C}_{\text{carb}}$ and $\delta^{13}\text{C}_{\text{org}}$ are derived from the same seawater dissolved inorganic carbon (DIC) reservoir and covary accordingly (Rothman, Hayes, & Summons, 2003). The unusual operation of the global carbon cycle during the Ediacaran may hold a key to understanding the coevolution of life and environments at that time (Grotzinger et al., 2011).

Currently, two leading hypotheses have been proposed to explain the origin of the SE. The first centers on a range of secondary alteration scenarios involving either meteoric waters or burial diagenesis (Derry, 2010a, 2010b; Knauth & Kennedy, 2009; Schrag, Higgins, Macdonald, & Johnston, 2013; Swart & Kennedy, 2012). Indeed, sea-level fluctuation and subsequent periodic subareal carbonate platform exposures can yield widespread meteoric overprints (Swart & Kennedy, 2012). However, models based on secondary alteration, which is inherently a local process, are still generally criticized as inconsistent with the global nature of the event (Grotzinger et al., 2011). The second hypothesis invokes the oxidation of a massive ¹³C-depleted dissolved organic carbon (DOC) reservoir in the Ediacaran oceans during the global oxygenation of a formerly anoxic deep ocean, which drove the isotopic composition of DIC ($\delta^{13}C_{DIC}$) in seawater and, in turn, coeval $\delta^{13}C_{carb}$ to strongly negative values (Fike, Grotzinger, Pratt, & Summons, 2006; Jiang, Kaufman, Christie-Blick, Zhang, & Wu, 2007; McFadden et al., 2008; Rothman et al., 2003). In this model, the size of the hypothesized DOC pool buffered the $\delta^{13} \rm C_{\rm org}$ record against isotopic change. However, the DOC oxidation hypothesis has been challenged because of the extreme oxidant demand that would have been required to drive $\delta^{13}C_{DIC}$ of the entire ocean to -12‰ during marine ventilation

(Bristow & Kennedy, 2008). More generally, the presence and nature of the hypothesized DOC pool and the possible mechanisms behind its generation and maintenance remain poorly known and highly debated.

Here, we shed important new light on interpreting the δ^{13} C records of the SE by investigating basin-scale spatial heterogeneity of the SE as recorded in the Ediacaran Doushantuo Formation along a proximal-to-distal transect across the Yangtze Platform of South China. Our results indicate a strong heterogeneity for the SE across the Yangtze Platform, which challenges the notion that the most extreme negative carbonate δ^{13} C values represent that of global seawater and suggests instead that the mechanism driving the excursion was spatially variable.

GEOLOGICAL SETTING AND 2 STUDY SECTIONS

The Ediacaran Yangtze Platform in South China was a passive continental margin, which formed at ~820 Ma along the rifted southeastern margin of the Yangtze Block during breakup of the Rodinia supercontinent (Wang & Li, 2003). Paleogeographic reconstructions for the Ediacaran Yangtze Platform envisage shallow rimmed platform, slope, and basin environments along a northwest-southeast transect based on lateral variations in lithofacies and stratal thicknesses (Jiang, Shi, Zhang, Wang, & Xiao, 2011; Figure 1a). The Doushantuo Formation was deposited in shallow to deep waters on the Yangtze Platform (Figure 1b) following the last Neoproterozoic global-scale glaciation-the Marinoan (or "Nantuo" in South China) event (Figure 2). Deposition of the Doushantuo Formation continued for ~84 Myr, spanning most of the Ediacaran Period, based on U-Pb



FIGURE 2 C-isotope chemostratigraphy of the Ediacaran Doushantuo Formation at study sections. Data sources include this study (yellowfilled symbols), Li et al. (2010) (red-filled symbols), and McFadden et al. (2008) (Jiulongwan) and Jiang et al. (2010) (Siduping; both blue-field symbols). Green curves represent five-point running averages for high-resolution intervals (note: raw data used for low-resolution intervals). Gray-shaded areas represent negative excursions of $\delta^{13}C_{carb}$. Three age constraints are from Condon et al. (2005) and Liu et al. (2009). See text for more details on stratigraphic correlations and the "IP" and "EN" event designations for study sections. Abbreviations: NT, Nantuo Formation; DY, Dengying Formation; LCP, Liuchapo Formation

ages of 635.2 ± 0.6 and 551.1 ± 0.7 Ma derived from ash beds at its base and top (Condon, Zhu, Bowring, Wang, & Yang, 2005). However, a slightly older age of ~560 Ma was recently suggested for the top of Doushantuo Formation based on stratigraphic correlation of the Miaohe Member of the Doushantuo Formation in the Yangtze Gorges area (An et al., 2015). The Doushantuo Formation is also known for its fossil animal embryos (Yin et al., 2007), macroscopic algae (Yuan, Chen, Xiao, Zhou, & Hua, 2011), and abundant acritarchs (McFadden et al., 2008), marking milestones in eukaryotic evolution and, more generally, the historical march from simple to complex life (Xiao et al., 2014, and references therein).

Our samples of the Doushantuo Formation were collected at several locales: the inner-shelf Zhangcunping site (ZCP; drill core-ZK312), the intra-shelf-basin of Jiulongwan (JLW; outcrop), and the upperslope Siduping sections (SDP: outcrop). These sections contain abundant carbonate and represent a proximal-to-distal transect across the Yangtze Platform (Figure 1), allowing us to investigate spatial variation of the SE. The JLW and SDP sections were previously studied for both $\delta^{13}C_{carb}$ and $\delta^{13}C_{org}$ (Jiang et al., 2010; Li et al., 2010; McFadden et al., 2008; Wang, Jiang, Shi, & Xiao, 2016; see Section "5.1"), and their stratigraphic details were reported in McFadden et al. (2008) and Wang et al. (2016). In light of this previous work, we focus on a description of the new ZCP section in this study and interpret those data within the broader context.

The ZCP ZK312 drill site is located in Duanjiang Village, Baokang County, Hubei Province. In this core, the Ediacaran Doushantuo Formation has a total thickness of ~138 m and can be subdivided into four lithostratigraphic members (Figure 2a). Member I is a 2.91-m-thick cap carbonate that overlies Cryogenian glacial diamictites of the Nantuo Formation; it consists of light gray and thickly bedded microcrystalline dolostone with stromatactis-like cavities. Member II consists of four lithologic subunits: (i) Member IIa is a 12.99-m black shale containing phosphatic layers and nodules, mainly in its upper portion. (ii) Member IIb is a 14.48-m, thick-bedded, gray, microcrystalline dolostone, (iii) Member IIc is a 21.28-m, thin-bedded, gray, microcrystalline dolostone containing phosphorite in its middle third, and (iv) Member 4 WILEY-gebiology

IId is a 26.24-m, thin-bedded, dark gray, microcrystalline dolostone containing large chert nodules. Previous studies of the Zhangcunping outcrops inferred an erosional surface between Members IIb and IIc (Figure 2a; Liu, Yin, Gao, Tang, & Chen, 2009; Zhou, Xie, & Xiao, 2005; Zhu, Zhang, & Yang, 2007; Zhu et al., 2013), but our field observations suggest otherwise because strata above and below this surface show similar lithologies and sedimentary structures. Member III is a 46.93-m, thin-to-medium-bedded, gray, microcrystalline dolostone containing thin intercalations of black shale in its middle portion and chert layers in its upper portion. Member IV is a 12.88-m, thin-bedded, dark grav, microcrystalline dolostone that is distinct from the overlying thick-bedded, light gray dolostone of the Hamajing Member of the **Dengying Formation.**

The microfossil assemblages from the Zhangcunping area were found mostly in chert nodules of the upper Member II (Liu et al., 2009; Zhang, 1986; Zhou et al., 2005; Zhou, Yuan, Xiao, Chen, & Xue, 2004). These assemblages contain cyanobacteria, multicellular algae, and acritarchs that are similar to silicified fossils from Members II and III of the Doushantuo Formation in the Yangtze Gorges area (Liu et al., 2009). A zircon SHRIMP U-Pb age (614.0 ± 7.6 Ma) was obtained from the bottom of the Member IIb at the Wanjiagou section in the Zhangcunping area (Liu et al., 2009).

STRATIGRAPHIC CORRELATION 3

The intra-shelf-basin of JLW and the upper-slope SDP section, as well as other sections across the Yangtze Platform, can be correlated on the basis of distinctive marker beds-the basal cap carbonate of Member I and the black shale of Member IV, in combination with other stratigraphic surfaces of regional extent (Jiang et al., 2011; Zhu et al., 2013; Wang et al., 2016; Figure 2). The new inner-shelf ZCP section of the present study fits readily into this correlation framework (Figure 2). First, the basal cap carbonate overlying the Nantuo glacial diamictites at ZCP is equivalent to cap carbonates in the JLW and SDP sections. Second, Member II of the Doushantuo Formation can be correlated regionally based on lithologic and faunal characteristics. At ZCP, Member II consists of dark-colored black shale and limestone containing acritarchs, multicellular algae-cyanobacteria fossils, and abundant chert nodules. These features correspond to those seen in the JLW section (Liu et al., 2009), although JLW contains a larger proportion of black shale (McFadden et al., 2008). The Member II/III contact is characterized by similar, abrupt lithofacies changes in both sections-from thin-bedded, dark gray, dolostone containing large chert nodules to medium-bedded, light gray dolostone without chert nodules at ZCP, and from thin-bedded dolostone with intercalated black shales to thick-bedded dolostone lacking black shale at JLW. These facies shifts reflect a rapid shoaling at the base of Member III at both sites. Third, the Member III/IV contact can be correlated between ZCP and JLW based on similar lithofacies changes from thick-bedded, light gray dolostone containing chert layers to thin-bedded, dark gray dolostone at ZCP, and from medium-bedded limestone with dolomite caps to black shales at JLW, reflecting a rapid deepening at the base of Member IV

at both sites. Finally, Member IV is unconformably overlain by thickbedded, medium gray dolostone of the Dengying Formation at both sites. Small differences in lithology within correlative intervals of these sections most likely reflect local variation in water depths, water mass circulation, and siliciclastic inputs, but they do not obscure overall similarities in these stratigraphic successions (Jiang et al., 2011; Zhu et al., 2007, 2013). The correlation framework of the study sections offers a unique opportunity to explore the spatial variation of the SE and other Ediacaran excursions.

SAMPLES AND ANALYTICAL METHODS 4

4.1 | Samples

A total of 386 sedimentary rock samples were collected for this study between July 2012 and December 2013 and analyzed for the isotopic composition of carbonate ($\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$) and coexisting organic matter ($\delta^{13}C_{org}$; see Table S1). These samples include 171 (all dolomite) from the ZCP section (ZK312 drill core), 57 (33 dolomite, 24 limestone) from the JLW section (outcrop), and 158 (all dolomite) from the SDP section avoiding the olistostrome breccias (outcrop). The 57 JLW samples were collected with the goal of providing a more complete record of the SE compared to those established in earlier studies of the same section (Li et al., 2010; McFadden et al., 2008). For the outcrop sites, large fresh blocks of rock (>200 g) were collected, and any visibly weathered surfaces and diagenetic veins or cements were removed. Each block was then cut into small pieces (~1 cm³) in the laboratory, and only those pieces with no visible weathering or veins were selected for powdering. Samples were crushed to finer than 200 mesh using a Retsch RS200 vibratory disk mill.

4.2 | Carbonate δ^{13} C and δ^{18} O analyses

About 60 to 300 µg of sample powder was loaded into a vial after drying at 70°C for 24 hr in an argon atmosphere. The samples were then reacted with 100% phosphoric acid under a vacuum at 70°C for 220 s using a Kiel IV device. The resulting CO₂ was subsequently introduced into a MAT 253 isotope ratio mass spectrometer (IRMS) for isotopic measurements. Delta values were calibrated relative to international reference standard NBS-19 ($\delta^{13}C = +1.95\%$; $\delta^{18}O = -2.20\%$) and Chinese national standard GBW04416 ($\delta^{13}C = +1.61 \pm 0.03\%$; δ^{18} O = = 1.59 ± 0.11‰). Carbon and oxygen isotope data for carbonates are reported relative to Vienna Pee Dee Belemnite (VPDB) with a precision of better than ±0.1‰ based on duplicate analyses of GBW04416 and the study samples.

4.3 | Organic δ^{13} C analysis

An aliquot of sample powder (~5-30 g) was reacted with 6 M HCl to completion. The decarbonated residue was rinsed with deionized water to neutral pH, then centrifuged, and freeze-dried for 24 hr. Samples with low TOC contents were treated further with HF to remove silicates before isotopic analysis. Organic δ^{13} C was measured

online using a Flash EA 2000 interfaced with a MAT 253 IRMS and calibrated with the glycine (δ^{13} C = -33.3%) and collagen (δ^{13} C = -9.0%) SIGMA standards. Results are reported relative to the VPDB standard with a precision better than ±0.2‰ based on duplicate analyses of study samples.

4.4 | Analyses of abundance and $\delta^{34} S$ of carbonate-associated sulfate

An aliquot of sample powder (~20-50 g with total inorganic carbon content >3%: 77 samples from JLW section) was washed in a 10% NaCl solution for 24 hr before removing the supernatant. This step was repeated multiple times until there was no barite (BaSO₄) precipitation from the supernatant when adding saturated BaCl₂ solution (~250 g/L). The residual powder was then treated with 4 M HCl until the reaction completed. The filtered solution was treated with 125 ml saturated BaCl₂ to precipitate the target carbonate-associated sulfate (CAS) as barite, which was then filtered, dried, and weighed for calculating CAS concentration in the carbonate fraction of original sample by correcting for the amount of insoluble material (assuming that all of the dissolved material was pure carbonate). The collected barite was mixed with excess V_2O_5 for online combustion, and resulting SO_2 was measured on a Thermo Scientific Delta V Plus IRMS coupled with a Flash elemental analyzer for sulfur isotope composition of the CAS $(\delta^{34}S_{CAS})$. Sulfur isotope compositions are expressed in standard δ notation as permil (‰) deviation from the V-CDT international standard with an analytical error of 0.2% (1 σ) calculated from replicate analyses of IAEA standards (NBS-127, IAEA-SO-5, IAEA-SO-6).

4.5 | Mn and Sr concentration analyses

About 50 mg of dried sample powder was dissolved using a standard HNO_3 -HF digestion as described below. The digestion step included progressive acid treatments at 190°C in a 15-ml Teflon bomb equipped with a screw cap to which HNO_3 -HF (1:1) and HNO_3 were added sequentially until complete digestion was achieved. Distilled HNO_3 and trace metal-grade HF reagents were used for all samples. Following an evaporation procedure to remove concentrated acid, the sample was diluted with 2% nitric acid. Elemental concentrations were measured using either an Agilent 7700x inductively coupled plasma mass spectrometer for Mn and Sr or a Thermo Fisher ICAP 7400 inductively coupled plasma optical emission spectrometry for Mn. Analytical errors are better than $\pm 4.7\%$ for Mn and $\pm 6.3\%$ for Sr based on duplicate analyses of four USGS standards (BCR-2, AGV-2, BHVO-2, and RGM-2) and one Chinese national standard (GSR5).

5 | RESULTS AND DISCUSSION

5.1 | Spatial heterogeneity of $\delta^{13} C$ record across the Yangtze Platform

Our C-isotope data, together with data from earlier investigations from the relevant sections (JLW: Li et al., 2010; McFadden et al.,

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2008; SDP: Jiang et al., 2010), are compiled in Figure 2 and Table S1. A recently published C-isotope profile for the SDP section (Wang et al., 2016) shows stratigraphic trends that are quite similar to those in our dataset, but with some differences in vertical scaling attributable to independent field measurements (note: We show only our dataset in Figure 2).

Our integrated data from the intra-shelf-basin JLW site and upperslope SDP sections reveal three major negative $\delta^{13}C_{carb}$ excursions (EN1, EN2, and EN3; see below) separated by two positive intervals (IP1 and IP2) with $\delta^{13}C_{_{carb}}$ values of ca. +4 to +5‰ at each location (Figure 2; note: We define negative excursion in this study in terms of a decrease in $\delta^{13}\text{C}_{\text{carb}}$ values from the baseline of ca. +4 to +5‰ observed at all our sections). EN1 is associated with the basal cap carbonate (Member I), and EN2 and EN3 are associated with abrupt facies changes at the bases of Members III and IV, respectively (Figure 2b, c; cf. Li et al., 2010; McFadden et al., 2008; Wang et al., 2016). In contrast, our new data from the inner-shelf ZCP section show four negative $\delta^{13}C_{carb}$ excursions (EN1-1, EN1-2, EN2, and EN3) separated by three positive intervals of ca. +5‰ (IP, IP1, and IP2; see Figure 2a). EN1-1, EN2, and EN3 at ZCP can be correlated with the equivalent excursions at JLW and SDP based on independent stratigraphic correlations described above in Section 3, but the EN1-2 excursion has not been previously identified.

Each of the negative C-isotope excursions demonstrates strong spatial heterogeneity among the sections studied. For the cap carbonate-associated EN1 (or EN1-1 at ZCP), the $\delta^{13}C_{carb}$ and $\delta^{13}C_{ore}$ profiles show a decoupling of the stratigraphic trends at ZCP and JLW, but sympathetic trends at SDP (Figure 2). At ZCP, $\delta^{13}C_{carb}$ has a mean of -0.4% (SD = $\pm 1\%$) in the 3 m of cap carbonate and reaches +5.3‰ at ~20 m. The $\delta^{13}\text{C}_{\text{org}}$ values generally remain less variable (-29.4 \pm 0.9‰; mean \pm SD) in EN1-1, although two basal samples have values of >-28‰. At JLW, $\delta^{13}C_{carb}$ values show large variability, but generally increase from -4‰ to +5‰ in the basal 30 m. The $\delta^{13} C_{_{\rm org}}$ values correspondingly decrease up section from –25‰ to -30‰ in the 6 m of cap carbonate and are invariant thereafter. In contrast, at SDP, $\delta^{13}C_{carb}$ values decrease up section from -2.5‰ to -5.4% in the 6 m of cap carbonate and gradually increase to +5%by 40 m. The $\delta^{13}C_{org}$ values correspondingly decrease up section from -25% to -34% in the 6 m of cap carbonate and gradually increase to -25‰ by ~40 m.

The EN1-2 excursion was observed only in the ZCP section and lacks an equivalent within Member II at JLW and SDP. This excursion is marked by a coupling between the δ^{13} C profiles for carbonate and organic carbon. Specifically, between 29 and 43 m, $\delta^{13}C_{carb}$ and $\delta^{13}C_{org}$ decrease up section from +5% to -8% and from -28% to -31%, respectively, and between 43 and 55 m, they recover to +6% and -27%, respectively (Figure 2a).

The EN2 excursion is more distinct at JLW and SDP compared to ZCP (Figure 2). At JLW, $\delta^{13}C_{carb}$ decreases up section from +4 to -10‰ between 68 and 76 m and recovers to +4‰ by 85 m. At SDP, $\delta^{13}C_{carb}$ decreases up section from +4‰ to -2‰ between 60 and 70 m and returns to +5‰ by 75 m. At ZCP, only two small negative shifts from +5‰ to +1‰ are present between 72 and 92 m. No significant

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FIGURE 3 Mn/Sr and $\delta^{18}O_{rath}$ data of the Ediacaran Doushantuo Formation at study sections. The corresponding $\delta^{13}C_{rath}$ data are given for comparison. Stratigraphic details and abbreviations are identical to those in Figure 2

variations were observed in coexisting $\delta^{13}\mathsf{C}_{_{\text{org}}}$ records of EN2 at the three study sites (Figure 2).

The youngest negative C-isotope excursion, EN3 (Figure 2), is thought to be equivalent to the SE as described in Oman (Fike et al., 2006), Australia (Swanson-Hysell et al., 2010), and the western United States (Corsetti & Kaufman, 2003), with a U-Pb age of 551 ± 0.7 Ma for the upper limb of the excursion in China (Condon et al., 2005). The EN3 event at JLW shows three distinct intervals of $\delta^{13}C_{carb}$ variation (from base to top): (i) a negative shift from +5% to -9% over ~ 20 m (EN3a), (ii) a stable interval of roughly -9‰ over ~33 m (EN3b), and (iii) a recovery from –9‰ to –2‰ over >10 m (EN3c). The $\delta^{13}\text{C}_{_{\text{org}}}$ profile is decoupled from $\delta^{13}C_{\text{carb}}$, showing (i) an up section increase from -30‰ to -27‰ in EN3a; (ii) a general decrease to -39‰ from -27‰ in EN3b, although with large sample-to-sample variability; and (iii) a recovery to -34‰ from -39‰ in EN3c. The shallower ZCP and deeper SDP sections show different $\delta^{13}C_{carb}$ features: (i) EN3a corresponds to a negative shift up section from +5‰ to -1‰ over ~20 m at ZCP and from +5% to 0% over ~5 m at SDP, (ii) EN3b is not clearly expressed at ZCP, but is marked by values of ca. +1.5‰ over ~60 m at SDP, and (iii) EN3c shows a recovery from -1‰ to +3‰ at ZCP and a pronounced minimum of -8.3‰ followed by a recovery to +0.1‰ at SDP. The $\delta^{13}C_{\rm org}$ profiles of the ZCP and SDP sections reveal complex variation that is both coupled and decoupled relative to the respective $\delta^{13}C_{carb}$ records (Figure 2).

5.2 | Evaluation of diagenetic influences on $\delta^{13}C_{carb}$

Primary $\delta^{13}C_{carb}$ values can be altered by diagenesis, including recrystallization, which has been suggested previously as a mechanism for local $\delta^{13}\text{C}_{\text{carb}}$ variability during the Shuram (Derry, 2010a; Schrag et al., 2013; Swart & Kennedy, 2012). To avoid potential effects of secondary alteration on $\delta^{13}\text{C}_{\text{carb}}$,we collected microcrystalline dolostones wherever possible and removed any obvious weathered surfaces, veins, and cements prior to powdering. Furthermore, diagenetic alteration of $\delta^{13}C_{carb}$ can be evaluated through multiple geochemical approaches (Brand, 2004). Because strontium is expelled from marine carbonates, particularly aragonite, while manganese is incorporated under the influence of reducing fluids, Mn/Sr ratios can be used to evaluate the degree of alteration (Kaufman & Knoll, 1995). Previous studies suggested that Mn/Sr of <3 is consistent with little to no alteration (Derry, Kaufman, & Jacobsen, 1992; Kaufman, Jacobsen, & Knoll, 1993; Kaufman, Knoll, & Awramik, 1992). However, diagenetic fluids generally contain much less carbon than carbonate rocks, which favors the buffering capacity of carbonate C relative to DIC derived from organic matter remineralization. Therefore, samples with Mn/Sr as high as 10 are likely to preserve primary $\delta^{13}C_{carb}$ values (Kaufman & Knoll, 1995). Most of our samples (79%) have Mn/Sr < 3, and nearly all of them (95%) have Mn/Sr < 10, suggesting that the $\delta^{13}C_{carb}$ profiles of the study sections preserve near-primary marine carbonate signals (Figure 3). A small number of samples (five in EN1-1 and IP at ZCP, four in EN1 at JLW, and four in EN1 and EN3c at SDP) have Mn/ Sr > 10, suggesting possible diagenetic alteration of their δ^{13} C values.

Because post-depositional alteration of marine carbonate rocks commonly produces a decrease in both $\delta^{13}C$ and $\delta^{18}O$ values, extremely low δ^{18} O values (commonly < -10‰) and positive covariation between C and O isotopes have been cited as evidence for a diagenetic origin of negative isotope excursions (Knauth & Kennedy, 2009), including the SE (Derry, 2010a; Grotzinger et al., 2011). Samples with



FIGURE 4 Cross-plots of $\delta^{13}C_{carb}$ versus $\delta^{18}O_{carb}$ for the negative C-isotope excursion intervals in study sections. (a-c) Data from EN1, EN1-1, EN1-2, EN2, and EN3a-c at Zhangcunping, Jiulongwan, and Siduping sections, respectively. The correlation coefficient (r^2) and t-test *p* value (where needed) for each C-isotope excursion are shown in the legend. (d) Separation of EN3 data from the three study sections into dolomitic and calcitic samples

 $\delta^{18}\text{O}$ values of <–10‰ are mostly associated with EN1 at JLW and SDP, EN1-2 at ZCP, and the lower portion of EN3b at JLW (Figure 3). Indeed, the EN1-2 at ZCP is associated with abundant phosphorite deposition, which is susceptible to diagenetic alteration of carbonates (Kaufman & Knoll, 1995). Opposite to the described effects of diagenesis on $\delta^{13}C_{carb}$ - $\delta^{18}O_{carb}$, however, the ZCP section shows no significant $\delta^{13}C_{carb} - \delta^{18}O_{carb}$ covariation for any excursion (Figure 4a), including EN3 (r^2 = 0.0). The SDP section exhibits minor to moderate $\delta^{13} C_{carb}^{} - \delta^{18} O_{carb}^{}$ covariation for each excursion, but the correlations are negative rather than positive for the EN2 and EN3 (Figure 4c)again, the opposite of that expected from diagenesis (Knauth & Kennedy, 2009). These correlation relationships thus do not support significant diagenetic alteration of $\delta^{13}C_{carb}$ values in these sections. In fact, the widespread lack of systematic positive covariation between $\delta^{13} C_{carb}$ and $\delta^{18} O_{carb}$ in other sections of the Yangtze Platform strongly supports a non-diagenetic origin for EN3 found across the Yangtze Platform (Lu et al., 2013).

The JLW section exhibits minor to moderate $\delta^{13}C_{carb} - \delta^{18}O_{carb}$ positive covariation for each excursion (Figure 4b), including EN3 $(r^2 = 0.22)$. It is possible that these positive covariation patterns reflect some degree of diagenetic overprinting of original $\delta^{13}C_{carb}$ values during dolomitization and early burial more generally as suggested by some samples with elevated Mn/Sr and low $\delta^{18}O_{carb}$ values (Figure 3b). However, a few lines of evidence suggest that diagenetic alteration for the EN3 of JLW is insignificant. First, the stratigraphic consistency of EN3 at JLW to other age-equivalent $\delta^{13}C_{carb}$ profiles across the Yangtze Platform (Lu et al., 2013) and its global correlations with other Ediacaran $\delta^{13}C_{carb}$ profiles in India (Kaufman, Jiang, Christie-Blick, Banerjee, & Rai, 2006), Oman (Fike et al., 2006; Osburn, Owens, Bergmann, Lyons, & Grotzinger, 2015), Australia (Calver, 2000; Swanson-Hysell et al., 2010), the southwestern USA (Corsetti & Kaufman, 2003), and northern Mexico (Loyd et al., 2013) suggest that primary secular patterns have been preserved-although local overprints on the global signal are to be expected (as discussed below). Second, petrographic observations reveal that most carbonates from the EN3 in the JLW section are fine grained and uniformly microcrystalline (Lu et al., 2013; McFadden et al., 2008)-inconsistent with a diagenetic origin. Third, the EN3 excursion at JLW is characterized by smooth δ^{13} C variation that is independent of lithofacies changes (dolostone-limestone-black shales) (Lu et al., 2013), which is also consistent with a primary origin of the EN3 signal. Fourth, compared with calcite precipitation, dolomite formation can result in significantly greater oxygen isotope fractionation (up to 5%; Vasconcelos, McKenzie, Warthmann, & Bernasconi, 2005). We note that largely invariant δ^{18} O values are found throughout EN3 in exclusively dolomitic successions (ZCP and SDP sections), but a significant decrease in δ^{18} O values is found in the JLW section within the transition from dolomite beds (<120 m) to overlying limestone beds (120-140 m) without a corresponding change in $\delta^{13}C_{carb}$ (Figure 4d, and Table S1). This relationship is consistent with mineralogical control of carbonate δ^{18} O and a non-diagenetic origin of $\delta^{13}\mathsf{C}_{\mathsf{carb}}$ variation in the EN3 interval of the sections studied. Similar patterns of $\delta^{18}O-\delta^{13}C_{carb}$ for the transition from dolomite to limestone are observed in other sections from the Yangtze Platform (Lu et al., 2013), suggesting that the EN3 signal basin-wide is of primary origin.

Recent studies provide additional evidence that the SE trends recorded in sections on other continents also represent a primary seawater signal. For example, in Australia, Mg- and Ca-isotope data—along with [Mg], [Mn], and [Sr] data from carbonates of the Ediacaran-aged Wonoka Formation—indicate that the most pristine samples carry strongly negative $\delta^{13}C_{carb}$ signals (-7‰ to -8‰; Husson, Higgins, Maloof, & Schoene, 2015), consistent with a primary origin. Furthermore, the carbon isotope compositions of extractable long-chain (>C20) *n*-alkanes and mid-chain monomethyl alkanes from the SE strata in Oman were found to be as low as -40‰ (Lee, Love, Fischer, Grotzinger, & Halverson, 2015), which is rare for marine rocks





of any age and provides evidence for a major carbon cycle perturbation in conjunction with the SE in Oman. Additionally, our CAS concentration data from the Doushantuo and those previously published from global distributed sections-Oman, Mexico, and Death Valley-provide evidence against meteoric diagenesis being responsible for the SE. Specifically, work evaluating CAS concentrations during aragoniteto-calcite neomorphism of Pleistocene coral heads in the presence of meteoric fluids found that CAS concentrations decrease dramatically (Gill, Lyons, & Frank, 2008), often by orders of magnitude, because sulfate concentrations in meteoric fluids are significantly lower than those of seawater. This relationship allows for rock-buffered $\delta^{34}\mathsf{S}_{\text{CAS}}$ during meteoric alterations but with large decreases in CAS concentration (Gill et al., 2008). Opposite to this, the CAS concentrations in the Doushantuo Formation of South China, the Khufai and Shuram formations of Oman (Fike et al., 2006; Osburn et al., 2015), the Johnnie Formation of Death Valley (Kaufman, Corsetti, & Varni, 2007), and the Clemente Formation of Mexico (Loyd et al., 2012) all increase during the SE (see Section 5.3).

Taken together, our integrated geochemical, mineralogical, petrographic, and stratigraphic evidence from this study and previous investigations (Husson et al., 2015; Lee et al., 2015; Lu et al., 2013; Osburn et al., 2015; Tahata et al., 2013) indicates that the EN2 and EN3 (SE) on the Yangtze Platform represent primary seawater signals and that the EN3 is most likely a global signal. Given the possibility of significant post-depositional alteration for the EN1 of our sections (EN1-1 and EN1-2 at ZCP) and the significance of the SE, we limit our discussion below mainly to EN3.

5.3 | Elevated continental weathering and coastal marine sulfate concentration during the SE

To provide a context for interpreting the mechanisms driving the observed spatial heterogeneity in the SE, we evaluated proxies for weathering rates (Sr-isotopes) and marine sulfate availability (CAS concentrations and isotope composition). First, we examined ⁸⁷Sr/⁸⁶Sr ratio-a proxy for continental weathering (Richter, Rowley, & Depaolo, 1992). In South China, the Doushantuo negative C-isotope excursions were accompanied by elevated ⁸⁷Sr/⁸⁶Sr ratios, with the highest ⁸⁷Sr/⁸⁶Sr ratios during the SE (up to 0.708958; Figure 5). Relatively elevated ⁸⁷Sr/⁸⁶Sr ratios provide evidence for enhanced continental weathering during these isotopic events (Cui et al., 2015; Sawaki et al., 2008, 2010). A compilation of data from various continents shows that the SE was globally tied to the highest $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$ ratios for the Ediacaran (Figure 5), consistent with the strongest continental weathering during the SE. An increase in weathering rate might be related to an increase in global tectonic activity associated with global microcontinent collisions that stitched Gondwana during the middle Ediacaran (see reviews in Halverson, Hurtgen, Porter, & Collins, 2010;



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FIGURE 6 C-S chemostratigraphy of successions capturing typical Shuram excursion in South China (a), Mexico (b), southwest USA (c), and Oman (d) where paired carbonate-associated sulfate (CAS) and C-isotope data were reported. Note: All chemostratigraphic data capturing the early onset of the SE (<120 m) in (d) were normalized to a thickness of 120 m based on the base and top surfaces of the Khufai Formation (Osburn, Grotzinger, & Bergmann, 2014), while the >120-m data capturing the rest of the Shuram excursion were normalized to a thickness of 240 m in order to match the normalized thickness of <120 m for the Khufai Formation. Data sources: South China, this study and Li et al. (2010); Mexico, Loyd et al. (2012); southwest USA, Kaufman et al. (2007); Oman, Fike et al. (2006) and Osburn et al. (2015)

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FIGURE 7 Schematic presentation of the proposed heterogeneous oxidation model involving greatly enhanced weathering nutrient and sulfate fluxes and coastal surface-ocean oxygenation for interpreting the high spatial heterogeneity of the Shuram excursion (i.e., the largest negative $\delta^{13}\mathsf{C}_{_{\text{carb}}}$ excursion of the Ediacaran) as recorded in the upper Doushantuo Formation (EN3) across the Yangtze Platform. See text for more details. Abbreviations: DIC_{rc}, dissolved inorganic carbon derived from local oxidation of subsurface reduced carbon; DIC_{bg}, global marine background of dissolved inorganic carbon

and Och & Shields-Zhou, 2012); however, details of the mechanisms behind possible increases in weathering rates remain uncertain.

Second, we compared paired CAS concentration and $\delta^{34}S_{CAS}$ data with $\delta^{13}\text{C}_{\text{carb}}$ results from the JLW section, which is characterized by the most prominent negative $\delta^{13}C_{\text{carb}}$ excursion during the SE among the sections we studied. Furthermore, we compiled the paired C-S data available from other SE successions in Mexico (Loyd et al., 2012), southwest USA (Kaufman et al., 2007), and Oman (Fike et al., 2006; Osburn et al., 2015; Figure 6). The results from JLW demonstrate that the SE was accompanied by a parallel negative shift in $\delta^{34}S_{CAS}$ (up section from >+40‰ to <+10‰), as well as elevated CAS concentration (from <100 to >1,000 ppm; Figure 6a). These coupled trends suggest that marine sulfate concentrations were elevated during the SE relative to before. Importantly, generally similar coupling between the C-S data is observed for all the compiled successions (Figure 6b-d), despite their global distribution, suggesting that the elevated marine sulfate concentration and depleted isotopic composition during the SE were a global phenomenon. Indeed, an increase in seawater sulfate concentration has been inferred for the SE at all previously investigated localities (Fike et al., 2006; Kaufman et al., 2007; Li et al., 2010; Loyd et al., 2013; McFadden et al., 2008; Osburn et al., 2015). The increase in CAS concentration and negative excursion in $\delta^{34}S_{CAS}$ in association with the SE have been previously interpreted to reflect an increase in the global marine sulfate reservoir in response to atmospheric and marine oxygenation during this period (Fike et al., 2006; Kaufman et al., 2007; Loyd et al., 2013; Osburn et al., 2015).

Interestingly, however, different patterns for this C–S coupling are observed among the different continental sites. For example, the magnitudes of the negative shift in $\delta^{34}S_{CAS}$ and increasing CAS concentration in Oman are generally <15‰ and up to >10,000 ppm (Figure 6d), respectively, which are substantially different from those observed in South China and other places (Figure 6a–c). Furthermore, detailed analyses indicate that the negative shift of $\delta^{34}S_{CAS}$ and the elevation of CAS concentration in Oman are associated mainly with shallow waters (Hugf area) rather than with deep waters (Moutain area; Osburn

et al., 2015; Figure 6d). These observations together indicate that the general increase in marine sulfate concentration during the SE was a global phenomenon specific to shallow waters and that local heterogeneities in the magnitude and distribution occurred.

5.4 | Interpreting the spatial heterogeneity of the Shuram Excursion

Previous interpretations of the Shuram $\delta^{13}C_{carb}$ excursion have linked the increase in marine sulfate concentrations to oxidation of a formerly anoxic deep ocean with a large pool of DOC (e.g., Fike et al., 2006). However, recent redox proxy studies have provided evidence against pervasive ventilation of the deep ocean during the SE (Dahl et al., 2010; Johnston et al., 2013; Li et al., 2010; Sahoo et al., 2016; Sperling et al., 2015). Furthermore, oxidant mass balance models for ocean ventilation and DOC oxidation driving the excursion highlight that such a scenario is unlikely, as both marine oxygen and sulfate would be inadequate (Bristow & Kennedy, 2008). Together, these modeling and redox proxy studies suggest that if the SE records an oxidation event, it must have been limited in its areal extent.

Our $\delta^{13}C_{carb}$ dataset provides additional new constraints in this regard. A distinct feature of our proximal-to-distal transect from the Doushantuo Formation is that the magnitude, timing, and $\delta^{13}C_{carb}$ - $\delta^{13}C_{org}$ relationship of the negative $\delta^{13}C_{carb}$ excursions differ among the sites. The high spatial heterogeneity of these primary excursions suggests that the $\delta^{13}C_{carb}$ data for the SE at least partially reflect local controls on DIC in shelf settings. Our claim of local heterogeneity in the $\delta^{13}C_{DIC}$ during the SE is supported by examples from outside of the Doushantuo Formation. For example, the $\delta^{13}C_{carb}$ record for Shuram-aged carbonates from northwest Canada ranges from -2‰ to -8.5‰, similarly pointing to local controls (Macdonald et al., 2013), and the most pristine carbonates from the SE captured in the Wonoka Formation have $\delta^{13}C_{carb}$ of -7‰ to -8‰ (Husson et al., 2015). Additional constraints from the $\delta^{13}C$ of compound-specific biomarkers in Oman suggest that the primary excursion of the SE may have actually

been as small 5‰–7‰ at that locality (Lee et al., 2015). Together, this evidence implies that the global shift in $\delta^{13}C_{DIC}$ during the SE may not have been as extreme as the –17‰ of $\delta^{13}C$ excursion observed in the carbonate record at some localities and that models should consider local processes driving heterogeneity in the local $\delta^{13}C_{DIC}$.

Here, in light of the observations of spatial heterogeneity in δ^{13} C and inferred oxidation, we propose an updated conceptual model for the mechanism behind the negative $\delta^{13}C_{carb}$ of the SE (Figure 7). As shown by previous work, the Ediacaran oceans were redox-stratified with anoxic deep waters (Canfield et al., 2008; Li, Cheng, Algeo, & Xie, 2015: Li et al., 2010: Sahoo et al., 2016) and low in sulfate (<2 mm: Loyd et al., 2012; Osburn et al., 2015). Within this framework, we propose that the strong continental weathering during the SE, as suggested by the highest ⁸⁷Sr/⁸⁶Sr ratios (see Figure 5 and Section 5.3), may have greatly elevated continental nutrient fluxes into coastal oceans, resulting in unusually high coastal productivity. Recent biogeochemical modeling shows that at low atmospheric pO_2 levels (e.g., <2.5% present atmospheric level or PAL), the distribution of O₂ in the surface ocean is controlled principally by marine productivity (i.e., O2 release), with the highest dissolved O2 levels restricted primarily to areas of high productivity (Reinhard, Planavsky, Olson, Lyons, & Erwin, 2016). Thus, any elevation of coastal productivity might have resulted in coastal surface-ocean oxygenation if the atmospheric pO_2 levels prior to and during the SE were low enough. Indeed, recent reviews noted that Ediacaran pO2 levels might have been only a few percent of PAL, although a large range has been suggested (see reviews by Lyons et al., 2014 and Sperling et al., 2015). Coastal surface-ocean oxygenation could in theory have resulted in less oceanic pyrite burial and in turn more negative $\delta^{34}S_{CAS}$ and higher sulfate concentration in the local surface waters (i.e., higher [CAS]). Another possibility is that increased continental weathering may have elevated the riverine sulfate flux into coastal oceans. Ultimately, increased marine oxidant availability led to oxidation in formerly anoxic, relatively shallow settings while maintaining redox stratification (i.e., persistently anoxic deep waters) across the event-resulting in both vertical and lateral gradients in oxidant distribution.

Given an increase in shallow marine oxidant availability (O2 and sulfate), we assume that the source of the isotopically light $\delta^{13}C_{DIC}$ was more widespread oxidation (likely with higher rates and greater depth penetration of those oxidants) of shallow waters adjacent to persistently anoxic subsurface waters containing high concentrations of reduced carbon. We note that elevated reduced carbon concentrations in the Ediacaran ocean have been assumed in previous work, but have yet to be proven, and that this study provides no additional constraints on the possible source of that carbon. Previous studies favored DOC (Fike et al., 2006; McFadden et al., 2008; Rothman et al., 2003; Wang, Li, Hu, & Xie, 2015; Wang et al., 2016) or other forms of organic carbon such as fresh or aged organic matter (Lee et al., 2015). Within this framework, the $\delta^{13} C$ of local carbonate sediments was determined by mixing of global marine background DIC (DIC_{ho}) and isotopically light DIC derived from oxidation of reduced carbon (DIC,,) on a large scale—along with $\mathsf{DIC}_{\mathsf{rc}}$ generated locally through intense oxidation coupled to heterogeneous supplies of O2 and perhaps sulfate (see lower-left insert in Figure 7). As such, both spatial variations in $\delta^{13}C_{DIC}$ and the extremely negative $\delta^{13}C_{carb}$ values observed through the SE are mostly a product of local variation in rates of reduced carbon oxidation in shelf settings rather than shifts in global seawater $\delta^{13}C_{DIC}$. Thus, our proposed local, mostly shelf oxidation of subsurface reduced carbon—as opposed to a large shift in global marine $\delta^{13}C_{DIC}$ and comprehensive ocean ventilation—minimizes the likelihood of the inadequate oxidant supplies imagined in previous studies, which assumed whole-ocean oxygenation and corresponding shifts in $\delta^{13}C$ on the same scale (Bristow & Kennedy, 2008).

Our model can account for the general patterns of spatial heterogeneity of the SE (i.e., EN3) observed in this study. Specifically, relatively low initial reduced carbon availability in nearshore areas (represented here by ZCP) and low oxidant availability to distal areas (represented here by SDP) would have resulted in maximum local DIC production in mid-shelf areas (represented here by JLW) during the EN3a-EN3b (Figure 7). This region would have marked the interface between deeper anoxic waters rich in reduced carbon and shallow regions relatively richer in O2 and with proximal riverine sulfate inputs that rose due to enhanced, tectonically induced weathering. The influence of local DIC_{rc} production is seen in the $\delta^{13}C_{carb}$ profiles of mid-shelf sections, including JLW (this study) and nearby sections (e.g., Tianjiayuanzi; Lu et al., 2013), which exhibit exceptionally large EN3 excursions (Figure 2b). In contrast, nearshore sections, including ZCP (this study) and nearby Hushan-Dayukou (Zhu et al., 2013) as well as those sections, such as Zhongling (Li et al., 2010), that were located on shallow shelf margin rims with low reduced carbon availability (Figure 7), exhibit significantly smaller EN3 excursions (Figure 2a). During the late stage of the SE, reduced local DIC_{rc} availability via persistent consumption of reduced carbon at the nearshore ZCP and midshelf JLW locales is consistent with the observed recovery of $\delta^{13}C_{carb}$ to more positive values in EN3c (Figure 2a, b).

Our conceptual model still requires recognition of additional details of the SE heterogeneity observed in this study (e.g., the large negative $\delta^{13}C_{carb}$ excursion observed in EN3c at the distal SDP), analogous heterogeneous $\delta^{13}C_{\text{carb}}$ at other, globally distributed SE settings, as well as proxy evidence for increased nutrient availability and the source of the isotopically light DIC. However, in principle, our heterogeneous oxidation model can be used to explain existing $\delta^{13}C_{carb}$ records of the SE from the Yangtze Platform as well as those seen in other regions simply by changing the mixing ratio of locally produced DIC and the open seawater. Our model defines a general spatial framework based on the availability of oxidants and reduced carbon in the ocean, which predicts that the SE was recorded most prominently in shelf settings where both oxidants and reduced carbon were in ample supply (e.g., the JLW section of this study). Indeed, the most prominent expressions of the SE worldwide are found in shallow marine carbonate successions (reviewed by Grotzinger et al., 2011). In South China, the SE is particularly well developed in mid-shelf to upper-slope settings and in intra-shelf-basins (Lu et al., 2013; Tahata et al., 2013; Wang et al., 2016), which reflects the optimal balance of local surface-ocean oxidant availability and anoxic, subsurface distributions of reduced carbon.

5.5 | Interpreting the $\delta^{13}C_{org}$ records of the Shuram Excursion in terms of elevated continental weathering

The global record of the SE, including the EN3 in South China, is also notable for the lack of coupling of $\delta^{13}C_{_{carb}}$ and $\delta^{13}C_{_{org}}$ (Figure 2; Grotzinger et al., 2011: Lee et al., 2013). This feature has been explained by varying inputs of detrital, relatively ¹³C-enriched organic matter (OM_{det}) and ¹³C-depleted marine OM (OM_{mar}; i.e., OM in sediment = $OM_{det} + OM_{mar}$; see details in Jiang et al., 2012; and Johnston, Macdonald, Gill, Hoffman, & Schrag, 2012). The OM_{det} may have been relatively ¹³C-enriched, even if originally marine, because it experienced significant thermal maturation during burial prior to uplift and reworking. Thermal processing would have preferentially expelled isotopically light hydrocarbons, thus increasing the δ^{13} C of the remaining organic matter (Des Marais, Strauss, Summons, & Hayes, 1992). High rates of continental weathering are suggested by our compiled Srisotope records (Figure 5). Greatly enhanced continental weathering may have generated large nutrient fluxes while simultaneously stimulating delivery of significant amounts of OM_{det} into the coastal oceans. The collective result of these processes would have been an increase in $\delta^{13}C_{org}$ as observed during the early to middle stage of the SE at all of our locations (EN3a and lower portion of EN3b in Figure 2). Handin-hand with the decline of continental weathering as suggested by decreasing ⁸⁷Sr/⁸⁶Sr ratios (Figure 5) and anticipated decrease in OM_{det} input during the later stage of the SE, the relative contribution of $\mathrm{OM}_{\mathrm{det}}$ versus $\mathrm{OM}_{\mathrm{mar}}$ to sedimentary organic matter gradually decreased. As a result, a decrease in $\delta^{13}\text{C}_{_{\text{org}}}$ occurred, as observed in the upper portion of the EN3b and the entire EN3c at all of our study sites (Figure 2). Minimum $\delta^{13}C_{arg}$ values of <-38% ultimately developed in the more distal JLW and SDP sections, suggesting significant contributions of chemotrophic and/or methanotrophic biomass to OM_{mar} under anoxic bottom waters (Jiang et al., 2012; Wang et al., 2016). Therefore, the $\delta^{13}C_{_{OTP}}$ records of the SE also support our model in which greatly elevated weathering of the continents served as a trigger for these negative $\delta^{13}C_{carb}$ excursions.

6 CONCLUSIONS

Integrated $\delta^{13}\text{C}_{\text{carb}}\text{-}\delta^{13}\text{C}_{\text{org}}$ data from the inner-shelf section of Zhangcunping and the intra-shelf-basin and upper-slope sections of Jiulongwan and Siduping, respectively, reveal large spatial heterogeneity in Ediacaran carbon isotope records, particular for the globally distributed Shuram Excursion (SE). Combined geochemical, mineralogical, petrographic, and stratigraphic data indicate a primary marine signature for at least the SE in South China. Globally elevated ⁸⁷Sr/⁸⁶Sr ratios during the SE suggest that the SE was tied to elevated continental weathering during the mid-Ediacaran. Paired concentration and δ^{34} S data for carbonate-associated sulfate from South China, Mexico, southwest USA, and Oman indicate an increase in marine sulfate during the SE, but with different magnitudes and patterns among the globally distributed sites. In light of these observations, we propose a heterogeneous oxidation model involving greater oxygenation of the surface ocean in coastal regions to explain the SE and the high spatial heterogeneity among the observed records for $\delta^{13}C_{carb}$ and coexisting $\delta^{13}C_{ore}$. Specifically, we argue that elevated continental weathering brought high nutrient fluxes into coastal oceans, which resulted in elevation of coastal primary productivity, greater surfaceocean oxygenation as a consequence, and local elevation of marine sulfate concentration. The latter would reflect the combined effects of greater riverine sulfate inputs and reduced pyrite burial beneath the more widely oxic surface waters along the continental margins. Thus, the spatial $\delta^{13}C_{carb}$ patterns of the SE were controlled primarily by spatially varying extents of oxidation of reduced carbon by oxidants (mainly O₂ and sulfate) in surface waters mostly in shelf areas. The reduced carbon was plentiful in the persistently oxygen-deficient deeper waters of the stratified water column and could be supplied to the shallower waters. The elevated local oxidation of subsurface reduced carbon mainly in shelf settings proposed in our model minimizes the likelihood of the inadequate supply of oxidant suggested in previous research of the SE, which assumed whole-ocean oxygenation. Independent of the mechanistic specifics presented here, however, the evidence for spatial heterogeneity in δ^{13} C presented in this study must be accounted for in future models of the SE.

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AUTHOR CONTRIBUTIONS

C.L. designed this research with input from T.W.L., G.L., J.H., S.X., N.J., and T.J.A. C.L., G.L., J.H., M.C., W.S., Z.A., and J.T. collected samples in field, and G.L., J.H., W.S., and Z.A. performed laboratory analyses. C.L., D.S.H., W.S., M.C., and T.W.L. analyzed the data. C.L., D.S.H., T.J.A., and T.W.L. wrote this article with significant input from all coauthors.

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SUPPORTING INFORMATION

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