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The Pre-Sturtian Negative δ13C Excursion of the Dajiangbian Formation Deposited on the Western Margin of Cathaysia Block in South China

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ABSTRACT: The Dajiangbian Formation in South China is a siliciclastic-dominated sedimentary succession with low-grade metamorphism deposited on the western margin of the Cathaysia Block, and is capped by a glaciogenic diamictite (the Sizhoushan Formation). The Sizhoushan glaciogenic strata can be attributed to the Jiangkou glacial (Sturtian glacial) episode as they share stratigraphic and lithological similarities with Jiangkou strata in South China. Some carbonate, chert and shale units throughout the upper part of the Dajiangbian Formation were sampled for carbonate carbon isotope ($\delta^{13}C_{carb}$) and organic carbon isotope ($\delta^{13}C_{org}$) analyses. A range of geochemical indices including oxygen isotopes ($\delta^{18}O$) and Mn/Sr (Fe/Sr) ratios suggest that primary carbon isotope values were preserved in the upper Dajiangbian Formation. The upper Dajiangbian Formation shows $\delta^{13}C_{carb}$ of -0.1‰, upward decreasing towards to -5.4‰. We suggest that the negative $\delta^{13}C$ excursion beneath the Sizhoushan diamictite is correlative with the Pre-Sturtian Islay $\delta^{13}C_{carb}$ anomaly and allows correlation with the global Neoproterozoic isotope stratigraphy. We find that carbonate and organic carbon isotope data of the upper Dajiangbian Formation are coupled, consistent with the $\delta^{13}C_{carb}$-$\delta^{13}C_{org}$ pattern observed on multiple continents.

KEY WORDS: Neoproterozoic, Cryogenian, carbon isotopes, Islay anomaly, Cathaysia Block.

0 INTRODUCTION

Neoproterozoic carbonate records contain large-amplitude carbon isotope anomalies. The two most extensive Neoproterozoic glaciations were preceded by a distinct excursion of carbon isotope values in carbonate rocks (the Pre-Sturtian Islay anomaly and Pre-Marinoan Trezona anomaly). As negative $\delta^{13}C$ excursions occurred before each of the snowball Earth glaciations, CO_2 drawdown associated with perturbations of the global carbon cycle was considered to be the cause of the first low-latitude glaciation (Tziperman et al., 2011;Schrag et al., 2002). In South China, two glaciations, recorded by Jiangkou Formation and Nantuo Formation, respectively, have been documented (Zhang et al., 2011). The new U-Pb age dating indicates that the older glaciation (Jiangkou glaciation) in South China was initiated at 716 Ma (Lan et al., 2014), implying a synchronous and global Sturtian glaciation.

On the western margin of the Cathaysia Block in South China, pre-Sturtian strata are dominated by siliciclastic rocks, but some carbonate units with low-grade metamorphism are found in units immediately underlying the Jiangkou glaciation (Huang et al., 1994, Tang et al., 1994). These carbonates can be used for carbon isotope stratigraphy. In this study, we present carbonate carbon ($\delta^{13}C_{carb}$) and organic carbon ($\delta^{13}C_{org}$) data from the Pre-Surtian strata (the upper Dajiangbian Formation). The data represent the first carbon isotope data set from Pre-Stutian stratain South China and provide a record of isotope chemostratigraphy for global correlation.

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1 GEOLOGICAL SETTING

In South China, the Yangtze and Cathaysia blocks amalgamated during the Sibao-Jinning orogeny at ca. 1.0–0.9 Ga (Li et al., 2005; Wang and Li, 2003; Li et al., 2002; Li et al., 1995, Fig. 1). Recently more evidence may indicate that the timing of the collision and amalgamation of the Yangtze and Cathaysis blocks is between 860 to 830 Ma (Shu et al., 2011; Zhao et al., 2011; Wang et al., 2007; Zhao and Cawood, 1999). During the break-up of Rodinia, major rift basins formed along the margins of the South China Craton and the Cathaysia Block. Afterwards, thermal subsidence creates significant accommodation space where Middle to Late Neoproterozoic successions was deposited. The Cryogenian succession on the margin of the Yangtze Block includes the Jiangkou glaciogenic strata, the Datangpo interglacial strata and the Nantuo glaciogenic strata (Fig. 2). Litho-, chemo- and chronostratigraphic work of the margins of the Yangtze Craton have been well studied (Lan et al., 2014; Zhang et al., 2011;

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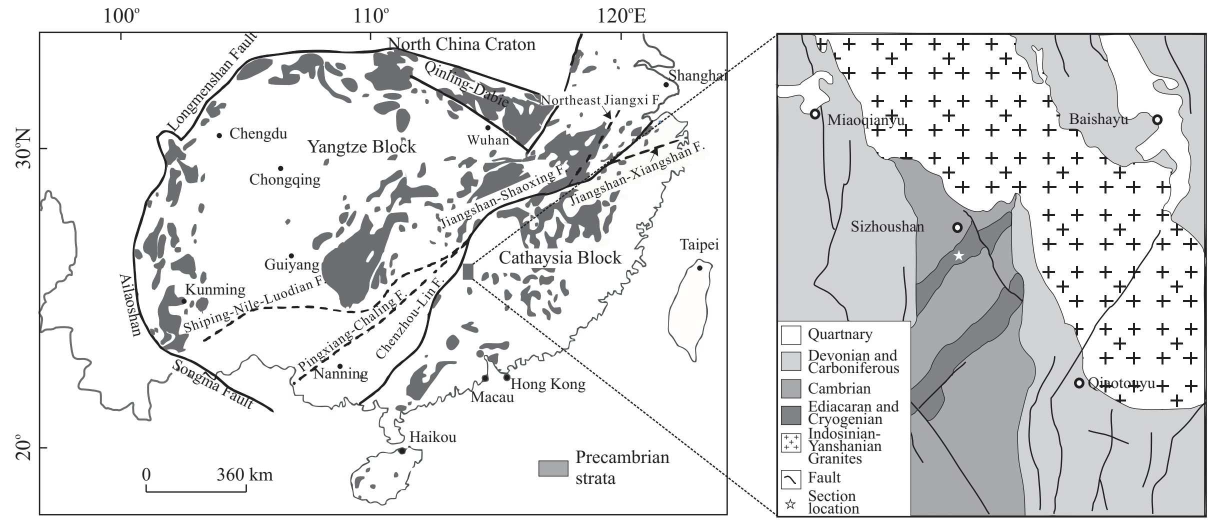

Figure 1. The map of the distribution of the Precambrian stratum in South China (after Zhao and Cawood, 2012) and the simplified geological map of the studied area (after Wu et al., 2013).

Condon et al., 2005; Zhou et al., 2004; Wang and Li, 2003). Compared to those of the Yangtze Block, the sedimentary sequence deposited on the western margin of the Cathaysia Block was poorly studied. Because of adjacent location of geography, the lithostratigraphy of the Neoproterozoic sedimentary successions of southern Hunan (Cathaysia Block) is commonly correlated with that in the central Hunan province (Yangtze Block) (Fig. 2). Huang et al. (1994) and Tang et al. (1994) have shown that the widespread glaciogenic diamictite and hematite can be used for regional correlation (Fig. 2). Huang et al. (1994) suggested that the hematite deposited at the base of the Tianzidi Formation is a Sturtian-equivalent BIF. This establishes a firmer link between southern Hunan and central Hunan, indicating that the Sizhoushan diamictite are broadly correlative with the Jiangkou diamictite (Fig. 2). Some dropstones observed in the Sizhoushan diamictite indicate it is closely associated with the glaciation (Figs. 3a, 3b).

The Sizhoushan syncline is cored by the Dajiangbian Formation, underlying the Sizhoushan diamictite, in the Sizhoushan area, southern Hunan (Fig. 1). Only the upper Dajiangbian Formation is well exposed at the outcrop section. The upper Dajiangbian Formation consists of silty and black shales, cherts and some dolomite interbeds (Fig. 3c), and was probably deposited in a suite of shallow to deeper marine environments (Huang et al., 1994). The lithology and name derivation of this mixed carbonate and siliciclastic formation are as in Tang et al. (1994).

Although no volcanic ash beds have been dated in the Dajiangbian Formation, analysis of detrital zircon populations provide constraints on its depositional age. The youngest concordant U-Pb ages of detrital zircons from the upper part of the Dajiangbian Formations yield a LA-ICP-MS age of 734 Ma (Wu et al., 2013), providing a maximum age constraint for the Dajiangbian Formation.

2 ANALYTICAL METHODS

2.1 Carbonate Carbon Isotope ($\delta^{13}C_{carb}$) Analyses

Carbon and oxygen isotope analyses were conducted at stable isotope geochemistry laboratory, Institute of Geology and Geophysics, Chinese Academy of Sciences. Approximately 50 mg of carbonate powder was reacted for 72 hours at 25 °C with anhydrous H_3PO_4 under vacuum. The purified CO_2 was analyzed on a Finnigan Delta S gas source mass spectrometer. Isotopic results are expressed in the standard notation as per mil (‰) deviations from the VPDB international standard. Carbon and oxygen isotope results are generally reproducible within ±0.2‰.

2.2 Total Organic Carbon (TOC) and Organic Carbon Isotope ($\delta^{13}C_{org}$) Analyses

Rock powders were decarbonated with 4 M HCl for >12 hours for analyzing the total organic carbon (TOC). Total organic carbon (TOC) was determined on acid-washed samples via combustion in a High-frequency Infrared Carbon & Sulfur Analysis instrument (CS-902G/T) (Beijing Wanlianda Inc.). The decalcified samples (30 to 100 mg) +CuO wire (1g) were added to a quartz tube and combusted at 500 °C for 1 hour and 850 °C for another 3 h. Isotopic ratios were analyzed using cryogenically purified CO_2 on a Finnigan Delta S mass spectrometer, and reported in standard δ-notation relative to Vienna PeedeeBelmnite (VPDB) standard. Analytical precision for ^{13}Corg is better than 0.2‰.

2.3 Elemental Analyses

Elemental analyses (Al, Ca, Mg, Sr, Mn, Fe) were performed on an inductively coupled plasma atomic absorption spectrometry (ICP-AES) at Institute of Geology and Geophysics, Chinese Academy of Sciences. All samples were prepared by dissolving ca. 10 mg of carbonate powder in ca.10 ml of 6 M HCl. Replicate analyses were better than 10% for all elements.

3 RESULTS AND DISCUSSIONS

3.1 Preservation of Primary Isotope Signatures

3.1.1 Carbonate carbon isotope

Oxygen isotope is known to be sensitive indicator of

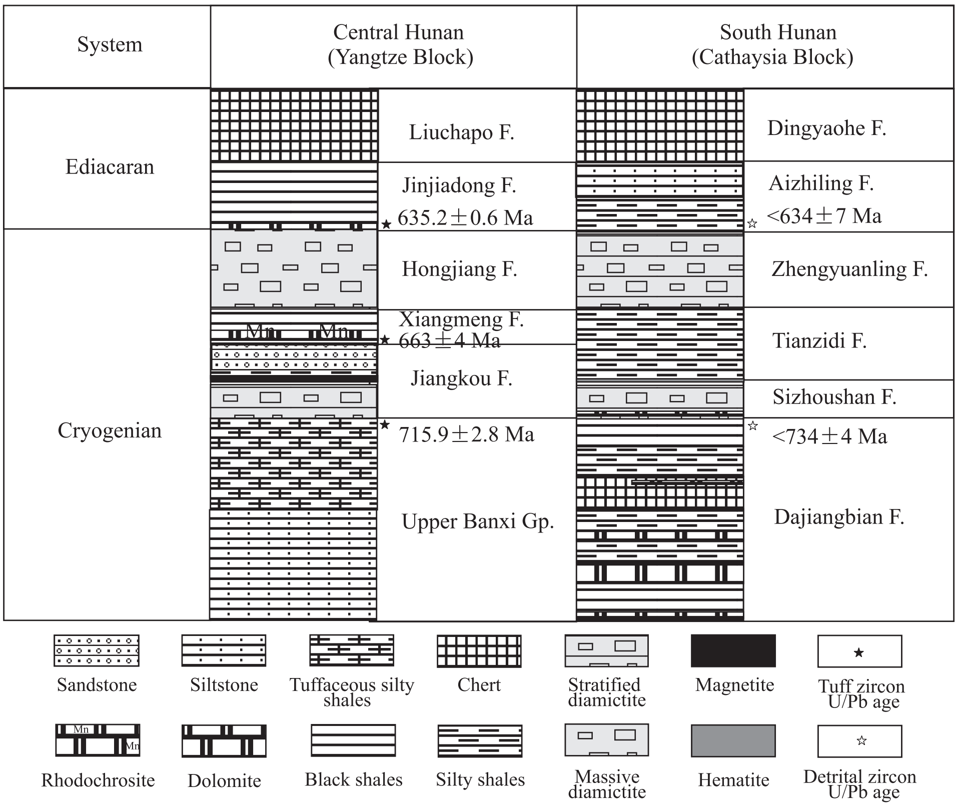

Figure 2. Neoproterozoic stratigraphic correlations between the Central Hunan (left) and South Hunan (right). All age data are from South China (Lan et al., 2014; Wu et al., 2013; Condon et al., 2005; Zhou et al., 2004).

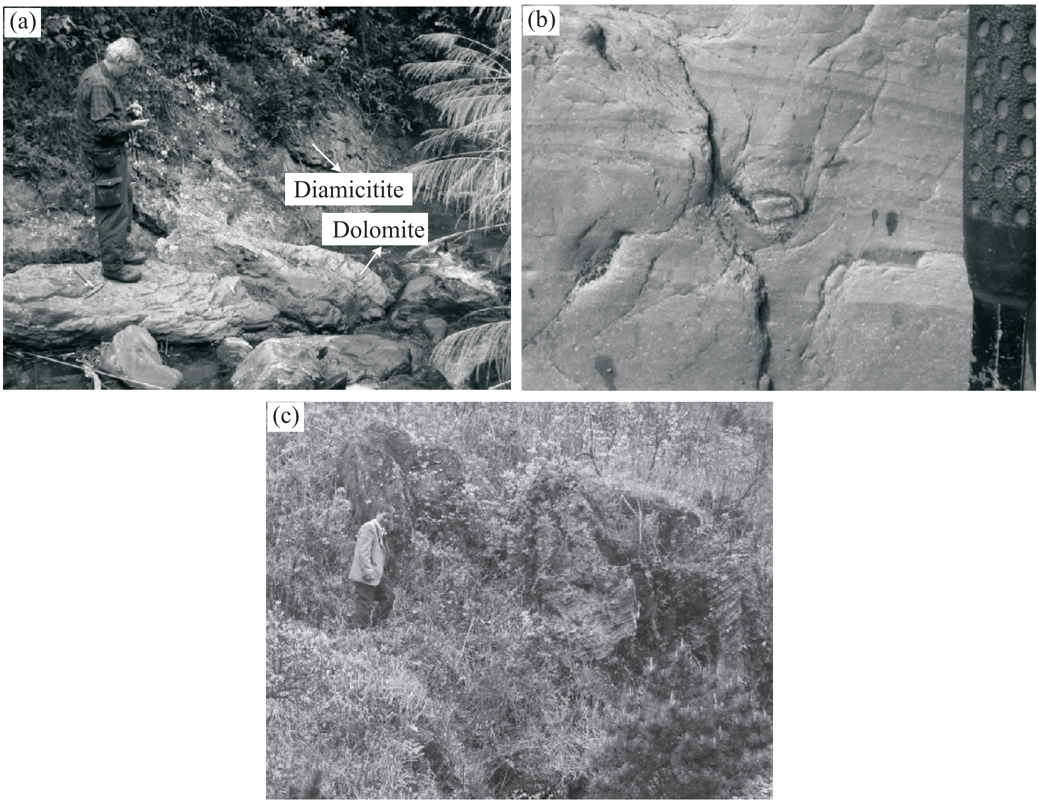

Figure 3. (a) Dolomite beds in the Sizhoushan diamictite; (b) dropstone in the well-stratified Sizhoushan diamictite; (c) dolomite in the Dajiangbian Formation at the subsection DJB2.

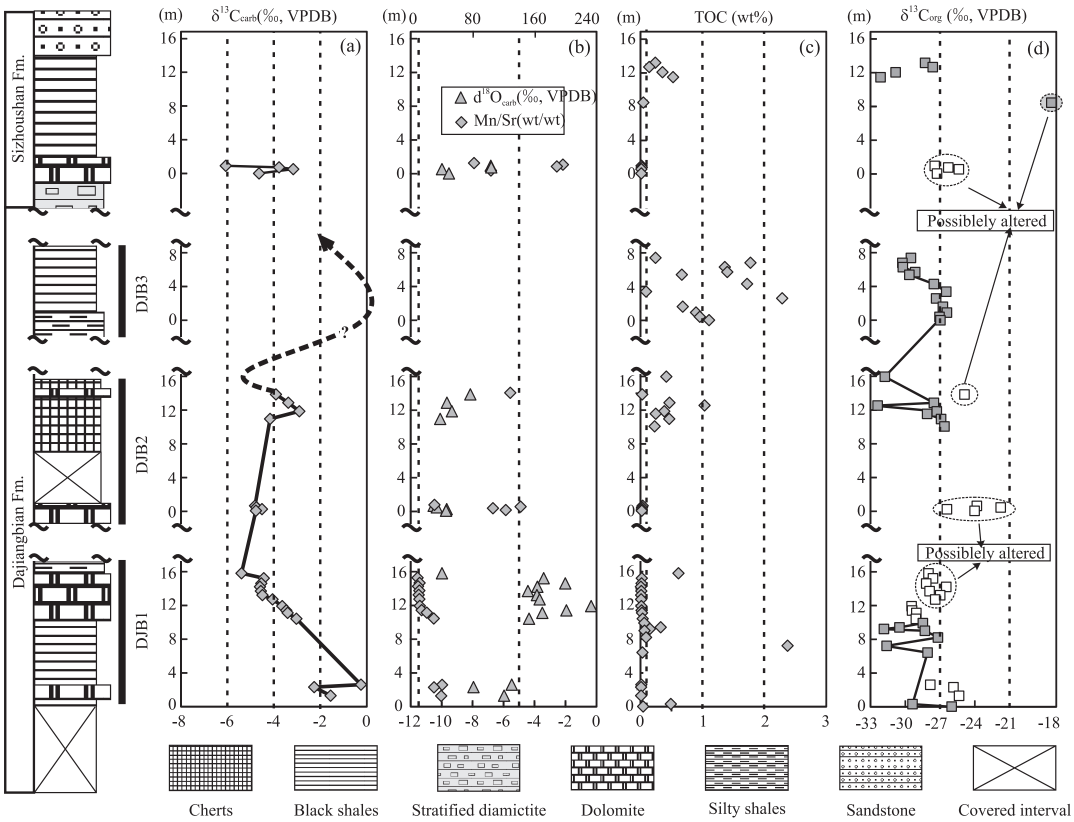

Figure 4. Litho- and cheomostratigraphy of the upper Dajiangbian Formation and the lower Sizhoushan Formation in South Hunan: (a) $\delta^{13}C_{carb}$. Dashed line indicates the trend of carbon isotope of dissolve inorganic carbon (DIC), see the details in the text; (b) $\delta^{18}O_{carb}$, Mn/Sr. (c) TOC. (d) $\delta^{13}C_{org}$. white squares represent the data with paired $\delta^{13}C_{carb}$ and $\delta^{13}C_{org}$. Gray squares represent no paired $\delta^{13}C_{carb}$ and $\delta^{13}C_{org}$ data. DJB1, DJB2, DJB3 are the outcrop subsections at the Sizhoushan Section.

alteration, with marked decrease in $\delta^{18}O$ values resulting from isotope exchange with meteoric or hydrothermal fluids. $\delta^{18}O$ values from carbonates of the Dajiangbian and Sizhoushan formations range from -10.5 to -0.3 (-6.6‰ average, VPDB) (Table 1). About 38% of $\delta^{18}O$ values are >-5‰ and fall into the category of "least altered" according to Kaufman and Knoll (1995). Cross-plots of ^{13}C values versus $\delta^{18}O$ values may show whether carbon isotope values were strongly affected by alteration based on presence or lack of correlation between these two parameters. In comparing $\delta^{18}O$ and $\delta^{13}C$ values, a weak trend suggests that the samples with lowest $\delta^{18}O$ and $\delta^{13}C$ values (D0.9, D-9.4, D-9.6, D-9.8, D-10, Du37.4) might be altered (Figs. 4, 5a; e.g., Kaufman and Knoll, 1995; Fairchild et al., 1990).

Meteoric alteration and digenetic recrystallization of calcite have been shown to increase Mn and Fe contents in carbonates while decreasing their Sr content, resulting in the widespread use of Mn/Sr or Fe/Sr as an index of alteration (Kaufman and Knoll, 1995; Banner and Hanson, 1990; Brand and Veizer, 1981; Lorens, 1981; Brand and Veizer, 1980). However, no particular Mn/Sr or Fe/Sr value defines a threshold between "least-altered' and "more-altered" carbonates. For example, for Proterozoic carbonates, low Mn/Sr ratios (<2, <3, <8, <10, Kaufman and Knoll, 1995; Knoll et al., 1995; Kaufman et al., 1993; Derry et al., 1992, respectively) are interpreted to indicate unaltered or "least-altered" carbonates that retain their primary carbon isotope compositions. In this study, the Dajiangbian dolomites exhibit a wide range of Mn/Sr values from 8 to 141 (43 average, Table 1). Only two analyzed samples (Du38, Du38.6) have Mn/Sr values <10. The Sizhoushan dolomites show high Mn/Sr values (142 average, Table 1). Correlation exists between Mn/Sr (Fe/Sr) values and $\delta^{18}O$ (Figs. 5b, 5d), suggesting that prevalent occurrence of possible alteration for the Dangjiangbian and Sizhoushan dolomite. For the DJB1, the samples with the lower oxygen isotope also have the lower Mn/Sr values, suggesting that the Mn/Sr threshold can be close to 10 for the section, reflecting the contemporaneous sea water could be enriched in manganese during this time. However, the weak correlation between Mn/Sr (Fe/Sr) and $\delta^{13}C$ values indicates that primary carbon isotope trends may be preserved despite potential diagenetic or meteoric alteration (Figs. 5c, 5e). Four extreme high Mn/Sr (Fe/Sr) values (D-9.4, D-9.6, D-9.8, D-10) averaging 100 from the subsection DJB2 might indeed reflect more substantial alteration; However, these samples have $\delta^{13}C$ values that are similar to those of adjacent samples that contain Mn/Sr<10. These relationships indicate that primary carbon isotope may be preserved compared to the oxygen isotope, probably because carbon was not abundant relative to oxygen in diagenetic fluids.

3.1.2 Organic carbon isotope

We report $\delta^{13}C_{org}$ data for the bulk organic carbon of carbonates, shales and cherts from the upper Dajiangbian and

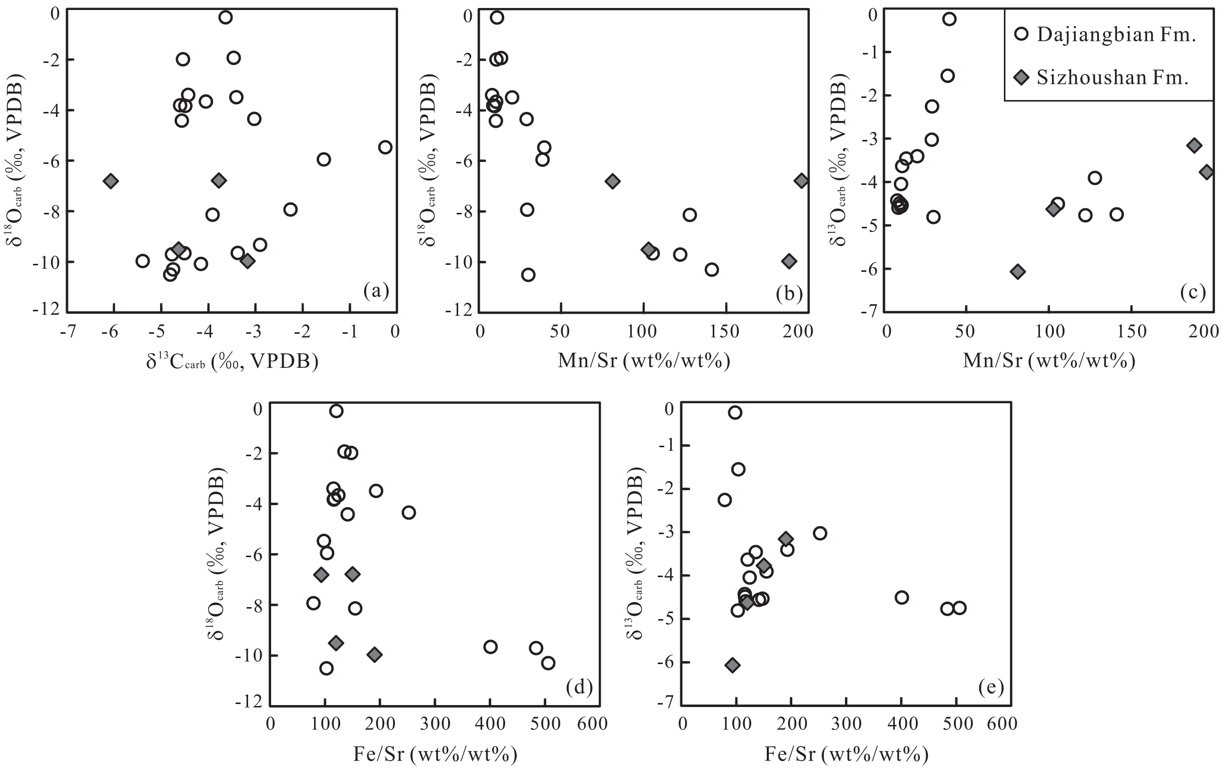

Figure 5. Cross-plots of geochemical measurements for the upper Dajiangbian Formation and the lower Sizhoushan Formations (a–e).Cross-plots of δ13C against δ18O and elemental ratios (Mn/Sr, Fe/Sr) are used to evaluate diagenetic or meteoric alteration.

lower Sizhoushan Formation. It has been argued that thermal degradation relates to the burial history of sedimentary basins; it may change the absolute $\delta^{13}C_{org}$ values but not the temporal $\delta^{13}C_{org}$ trend (Marais et al., 1992). These statements may be true for organic-rich sedimentary rocks, but for organic-poor carbonates, $\delta^{13}C_{org}$ may be more sensitive to diagenesis and thermal maturation. The organic-poor dolomites in the Sizhoushan Formation show more positive $\delta^{13}C$ values ranging from -17.4‰ to -27.4‰ (-24.8‰ average), and the Sizhoushan Formation shows a negative correlation (R^2=0.43) between TOC and $\delta^{13}C_{org}$ (Fig. 6a). High Mn/Sr values (>81) and negative $\delta^{18}O$ (<-6.8‰) values for the Sizhoushan dolomite suggested possible significant contribution from diagenesis. For the Dajiangbian Formation, although little covariation between TOC and $\delta^{13}C_{org}$ were observed (Fig. 6a), the organic-poor carbonate samples at the subsection DJB2 always show more positive $\delta^{13}C_{org}$ values compare to the adjacent organic-rich samples. According to Mn/Sr and $\delta^{18}O$ data, the dolomite of DJB1 seems to undergo a weak diagenetic alteration. However, because of the extreme low TOC contents for the carbonates, we need to further carefully evaluate the $\delta^{13}C_{org}$ data. For the upper DJB1 dolomite samples, there are good negative correlation between TOC and $\delta^{13}C_{org}$ (R^2=0.44), suggesting that diagenetic alteration may affect the $\delta^{13}C_{org}$ record. Throughout the upper DJB1 dolomite, the $\delta^{13}C_{org}$ values show a clear and abrupt increasing trend from -29.4‰ to -27.4‰ along with a decreasing trend of TOC content (Table 1). Therefore, we can divide the upper DJB1 dolomite into two groups. One group includes six carbonate samples (12.7–15.2 m) with a strong negative correlation between TOC and $\delta^{13}C_{org}$ (R^2=0.70),

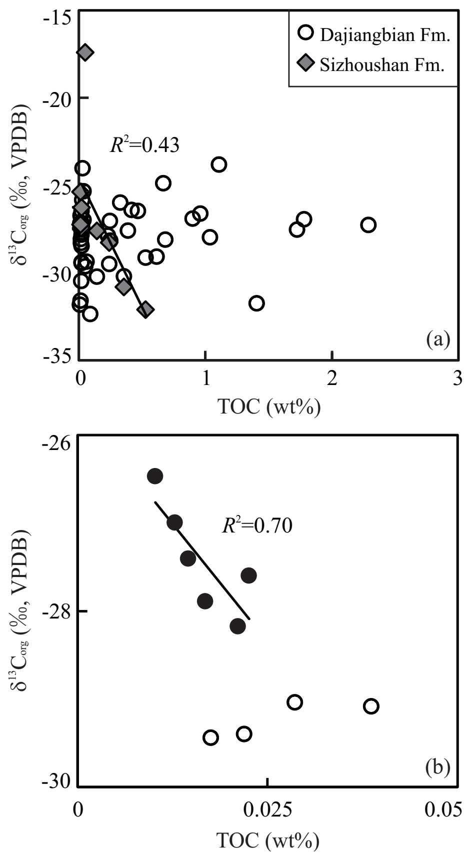

Figure 6. Cross-plots of δ13C against TOC for the upper Dajiangbian and lower Sizhoushan formations. They are used to evaluate the preservation of $\delta^{13}C_{org}$. See the details in the text.

indicating more $\delta^{12}C$ loss as a result of diagenetic alteration (Fig. 6b). Another group (10.4–11.9 m) could preserve primary $\delta^{13}C_{org}$ values which are similar to those of the adjacent shale samples. Other organic-lean dolomite samples from DJB2 and the Sizhoushan Formation could be affected by diagenetic alteration suggested by high Mn/Sr and low oxygen isotope ratios (Figs. 4b, 4c and 4d). One organic-lean shale sample (S4.8) from the Sizhoushan Formation with a higher $\delta^{13}C_{org}$ value (-17.4‰) is largely affected by diagenetic alteration.

3.2 Islay $\delta^{13}C_{carb}$ Excursion

$\delta^{13}C$ values for the carbonates of the upper Dajiangbian Formation vary from -2.3‰ to -0.2‰ with an average value of -1.1‰ in the lower of the subsection DJB1, followed by a decrease to values as low as -5.4‰ at the top of the subsection DJB1. The subsection DJB2 has the negative $\delta^{13}C$ values ranging from -2.9‰ to -4.8‰ (-4.1‰ average). Therefore, it is obvious that the $\delta^{13}C$ data of preglacial carbonates beneath the galciogenic Sizhoushan diamictite show a pronounced negative excursion (Fig. 4a).

In Neoproterozoic carbonates predating the first glaciation of the era, an interval of low carbonate $\delta^{13}C$ values known as the Bitter Springs Stage (BSS) is followed by strata with positive $\delta^{13}C$ values that persist until the ca. 735 Ma Islay anomaly (Rooney et al., 2014; Strauss et al., 2014; Prave et al., 2009). We correlate these $\delta^{13}C$ anomalies with the Pre-Sturtian Islay anomaly because of their stratigraphic position below glacial deposits (Sizhoushan diamictite) (Fig. 2). This $\delta^{13}C_{carb}$ excursion is commonly associated with a distinct recovery to enriched $\delta^{13}C_{carb}$ values prior to the onset of glacial sedimentation (e.g., Hoffman et al., 2012; Prave et al., 2009). It can be clearly seen in the strata of Canada, Ethiopia, and Greenland (Swanson-Hysell et al., 2015; Hoffman et al., 2012). In the Sizhoushan area, the recovery stage for carbonate carbon isotope is missing, which is possibly masked by siliciclastic deposits in the Dajiangbian Formation or could be eroded during glaciation or deglaciation (Figs. 2 and 4).

The Islay anomaly is inferred to be globally synchronous based on correlation of the $\delta^{13}C$ data and loose constraints such as correlation of diamictite. The Islay anomaly now are known to exist beneath Cryogenian glacial horizons in NW Canada (Jones et al., 2010, Macdonald et al., 2010), NW Tasmania (Calver, 1998), the Irish-Scottish Caledonides (Prave et al., 2009; McCay et al., 2006), NE Svalbard and East Greenland (Hoffman et al., 2012) and Arabian-Nubian (Swanson-Hysell et al., 2015). The Re-Os age from NW Canada suggests that the Islay $\delta^{13}C_{carb}$ anomaly predates the onset of the Sturtian glaciation by >15m.y. (Rooney et al., 2014; Strauss et al., 2014).

3.3 $\delta^{13}C_{carb}$-$\delta^{13}C_{org}$ Pattern

The Neoproterozoic carbon isotope excursions were standardly interpreted to be the change of the isotopic composition of DIC in contemporaneous seawater (e.g., Halverson and Shields-Zhou, 2011). If the carbon that is associated with organic matter in carbonate rock is derived from the same dissolved inorganic carbon (DIC) pool as the carbonate carbon, then large-scale changes to the isotope composition of DIC should be reflected both in carbonate ($\delta^{13}C_{carb}$) and organic carbon ($\delta^{13}C_{org}$) isotope records.

In the lower part of DJB1, the dolomite samples show a clearly decreasing trend of $\delta^{13}C_{org}$ (Fig. 4d). Meanwhile, $\delta^{13}C_{org}$ values of the shale samples with relatively high TOC contents also show a decreasing trend. It should be noted that samples with possible significant alteration will be excluded for the argument of the variation of $\delta^{13}C_{org}$ throughout the upper Dajiangbian Formation (Fig. 4d). Therefore, it is obvious that the $\delta^{13}C_{org}$ data covary with $\delta^{13}C_{carb}$ across the negative carbonate carbon isotope excursion, consistent with the observed trend in the NE Svalbard and East Greenland, Tasmania (Hoffman et al., 2012; Calver, 1998). The three independent basins show similar covariation between $\delta^{13}C_{org}$ and $\delta^{13}C_{carb}$ indicate that neither surface oxidation nor meteoric diagenesis have substantially altered the $\delta^{13}C_{carb}$-$\delta^{13}C_{org}$ record. The sympathetic shifts in $\delta^{13}C_{carb}$ and $\delta^{13}C_{org}$ across the Islay stage confirm that the stage reflects a large-scale perturbation to the isotopic composition of the DIC pool and that organic matter in the sediments is representative of coeval biomass that fixed carbon from this $\delta^{13}C$-depleted DIC. The coupled $\delta^{13}C_{carb}$ and $\delta^{13}C_{org}$ should not be taken as evidence for the existence of a large oceanic DOC (dissolved organic carbon) reservoir capable of buffering the $\delta^{13}C$ of marine organic matter, and suggest that the buildup of a larger DOC pool did not occur until Ediacaran time (Johnston et al., 2012; Swanson-Hysell et al., 2010).

For the subsection DJB2, the $\delta^{13}C_{org}$ values show a wide variation ranging from -31.8‰ to -26.6‰. Across the boundary between DJB2 and DJB3, the $\delta^{13}C_{org}$ values seem to show an increasing trend (Fig. 4d). Given that there is the sustainable coupled $\delta^{13}C_{carb}$ and $\delta^{13}C_{org}$ pattern before the Sturtian glaciation, $\delta^{13}C_{carb}$ isotope values of DIC in seawater become positive, and the recovery stage of the Islay anomaly occurred during the deposition of the DJB3 shales at the Sizhoushan Section (Fig. 4a). Meanwhile, the missing of the recovery of $\delta^{13}C_{carb}$ could be related to the erosion of the Pre-Sturtian strata during glaciation and deglaciation.

4 CONCLUSION

Through this study, it was first recognized that a pronounced negative carbonate carbon isotope excursion indeed occurs in the upper Dajiangbian Formation beneath the Jiangkou diamictite in South China. The negative carbonate carbon isotope excursion in the Dajiangbian Formation is possibly linked to the ca. 735 Ma Islay $\delta^{13}C_{carb}$ anomaly, suggesting a perturbation of the global carbon cycling. Meanwhile, the $\delta^{13}C_{org}$ data covary with $\delta^{13}C_{carb}$ in the Dajiangbian Formation, and are consistent with the $\delta^{13}C_{carb}$-$\delta^{13}C_{org}$ pattern observed in the NE Svalbard and East Greenland, Tasmania (Hoffman et al., 2012; Calver, 1998).

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