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Carbon Isotopic Evolution of the Late Ediacaran Gaojiashan Biota on the Northern Yangtze Platform, South China

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Abstract: Metazoan fossils in the Gaojiashan Biota are famous for being well preserved and may provide new insights into the early evolution and skeletonization of Metazoans. We are studying the isotopic compositions of organic and carbonate carbon from a sequence of sedimentary rocks at the Gaojiashan section, northern Yangtze Platform, Shaanxi Province of China. Organic carbon isotope values display a range between –30.8‰ and –24.7‰ with clear stratigraphic variations. Carbonate carbon isotope data vary between 0.1‰ and +6‰. Positive δ^{13}C values from sediments with Gaojiashan biota reflect temporal variations in carbon turnover, i.e. an increasing in photosynthetic carbon fixation followed by an increasing subsequent fractional organic carbon burial, and that related to bio-radiation such as increasing algae, bacteria, and original creatures productivity in biomass. These secular variations are interpreted to reflect perturbations of the regional carbon cycle, specifically changes in the fractional burial of organic carbon, and discuss the relationship between Gaojiashan biota and paleoenvrionmental variation.

Key words: Ediacaran, carbonate and organic carbon isotopic composition, Gaojiashan biota, Ningqiang, Yangtze Platform

1 Introduction

The Precambrian–Cambrian transition is one of the critical time intervals in Earth's history. Profound geotectonic, climatic and biological changes occur during the Late Ediacaran and its transition into the Early Cambrian. China's Yangtze Platform is characterized by a well developed succession of Late Ediacaran sedimentary rocks, and offers the opportunity to study the interaction between atmosphere, hydrosphere, biosphere and lithosphere during this critical interval in Earth's history. The Gaojiashan biota (551–541 Ma) is hosted in calcareous siltstones or calcisiltites that intercalate with calcareous mudstone or calcilutite (Lin et al., 1986; Zhang, 1986; Cai et al., 2010). Metazoan fossils in the Gaojiashan Biota are famous for being well preserved and include both trace and tubularbody fossils (Zhang, 1986; Hua et al., 2007), and may provide new insights into the early evolution and skeletonization of Metazoans (Xiao et al., 1998; Yin et al., 2011; Wang et al., 2011). It includes various soft-bodied or lightly biomineralized tubular fossils, calcareous protolagenid microfossils (Zhang et al., 1992; Hua et al., 2007; Cai et al., 2010). The paleobiological significance of the Gaojiashan biota has been discussed extensively, mostly in some literatures (Lin et al., 1986; Zhang, 1986; Zhang et al., 1992; Hua et al., 2000; Chen and Sun, 2001; Hua et al., 2007; Cai et al., 2010). Few researches (Zhu et al., 2007) have reported geochemical results from the Gaojiashan section; however, the geochemistry of the Gaojiashan biota and the relationship among paleonenvironments, bio-evolution, and organic matter burial have not been studied in detail.

This study focuses on the sediments those are from fossiliferous, pyrite-rich, soft-bodied or lightly skeletonized tubular fossils in the Gaojiashan Member at the Gaojiashan section and provides temporal records for the isotopic compositions of organic and carbonate carbon throughout this time Interval and to investigate the relationship between δ^{13}C variations, bio-evolution, geological and paleoenvironmental changes of regional and global importance.

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2 Geological Setting, Section and Samples

The Gaojiashan section is located about 45 km northeast of Ningqiang County town, the northern of the Yangtze Platform, southern Shaanxi Province (Fig. 1). The Ediacaran sedimentary sequence in Gaojiashan area comprises of the Liantuo, Nantuo, Doushantuo and Dengying formations. The Dengying Formation, however, has a wide geographic distribution and consists predominantly of dolomitic rocks interbedded with limestone and sandy shale, which is conveniently classed into three members, designated as the Algal Dolomite, Gaojiashan and Beiwan members(Hua et al., 2007; Cai et al., 2010). These three members, in turn, can be roughly correlated with the Baimatuo, Shibantan and Hamajing members of the Dengying Formation in the Yangtze Gorges area (Zhu et al., 2003; 2007).

The succeeding Gaojiashan Member is 55 m thick and can be divided into three units (Cai et al., 2010): The lower Gaojiashan Member is characterized by 19 m greenish and brownish siltstone, greenish silicified tuffaceous siltstone, and silty shale; *shaanxilithes ningqiangensis* (the first Gaojiashan level) occurs in silty shale about 18 m from the base (Xing et al., 1984; Lin et al., 1986; Zhang, 1986; Zhang et al., 1992; Hua et al., 2001; Hua et al., 2007; Cai et al., 2010). The middle Gaojiashan Member consists of 8 m of non-fossiliferous interbedded calcisiltite-siltstone and calcilutite-mudstone contains abundant pyritized fossils (*Conotubus* and *Gaojiashania*), calcareous microfossils (*Protolagena*), and horizontal trace fossils toward the upper part of this unit (the second Gaojiashan biota) (Cai et al., 2010). In the succeeding 14 m of strata, limestone becomes increasingly dominant over siltstone, fossils become increasingly scarce and are dominated by *Cloudina*, but wrinkled microbial sedimentary structures and rip-up clasts are common(Cai et al., 2010). The upper Gaojiashan Member is characterized by 3 m coarse sandstone and conglomerate. The Gaojiashan Member is succeeded by light gray, thick-bedded dolostone of the Beiwan Member, which, like the Algal Dolomite Member, is also characterizedby peritidal dissolution structures (Cai et al., 2010).

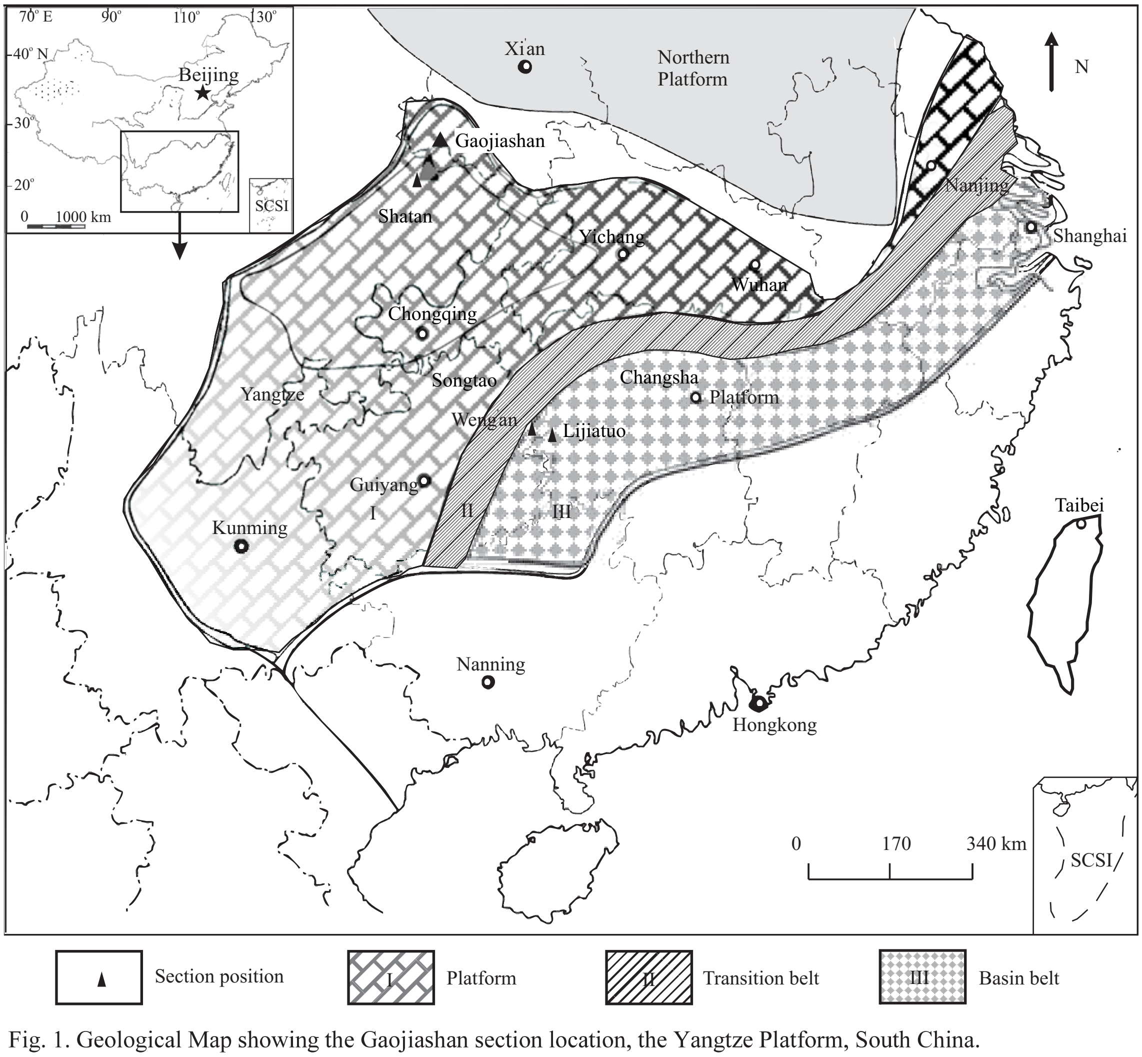

Fig. 1. Geological Map showing the Gaojiashan section location, the Yangtze Platform, South China.

Nineteen samples of carbonate were collected for geochemical studies at Gaojiashan, Ningqiang County, Shaan'xi Province. The stratigraphic positions for the samples are provided in Figs. 1 and 2 and Table 1.

3 Analytical Methods

Prior to geochemical analyses, all samples were chipped and pulverized (200 mesh). Subsequently, they were studied for their total, inorganic and organic carbon abundances and their organic and in part also for their carbonate carbon isotopic compositions. For carbonate samples, elemental abundances of Mn and Sr were measured (Table 1).

3.1 Organic matter

Total organic carbon (TOC) concentrations were determined gravimetrically, following the removal of carbonate with 15% HCl. Organic carbon isotopic compositions were measured for the kerogen fraction. Kerogen extraction has been performed according to a procedure modified after Lewan (1986).

Kerogen preservation was assessed by its H/C atomic ratio, following the determination of C, H, N elemental abundances.

Isotopic composition ($\delta^{13}C_{org}$) were measured via sealed quartz tube combustion (e.g. Strauss et al., 1992) and subsequent mass spectrometric analysis (Finnigan EA-ConF-IRMS) at the Geologisch - Paläontologisches Institut, Westfälische Wilhelms - Universität Münster, Münster, Germany.

Organic carbon abundances (TOC) were further determined as the difference between the total carbon (TC) and the total inorganic carbon (TIC) measured with a CS-MAT 5500 at the Geologisch - Paläontologisches Institut, Westfälische Wilhelms - Universität Münster, Germany.

3.2 Carbonate

CO_2 was liberated from whole rock samples via phosphorylation (McCrea, 1950) with enriched H_3PO_4 at 25°C for 24 h (limestone), and 50°C for 24 h (dolostone) (Wachter and Hayes, 1985; Guo et al., 2007). All carbonate carbon and oxygen isotopic compositions were measured in the Institute of Geochemistry, the Chinese Academy of Sciences, Guiyang, China, using at Finnigan MAT 252 mass spectrometer. The analytical procedure was

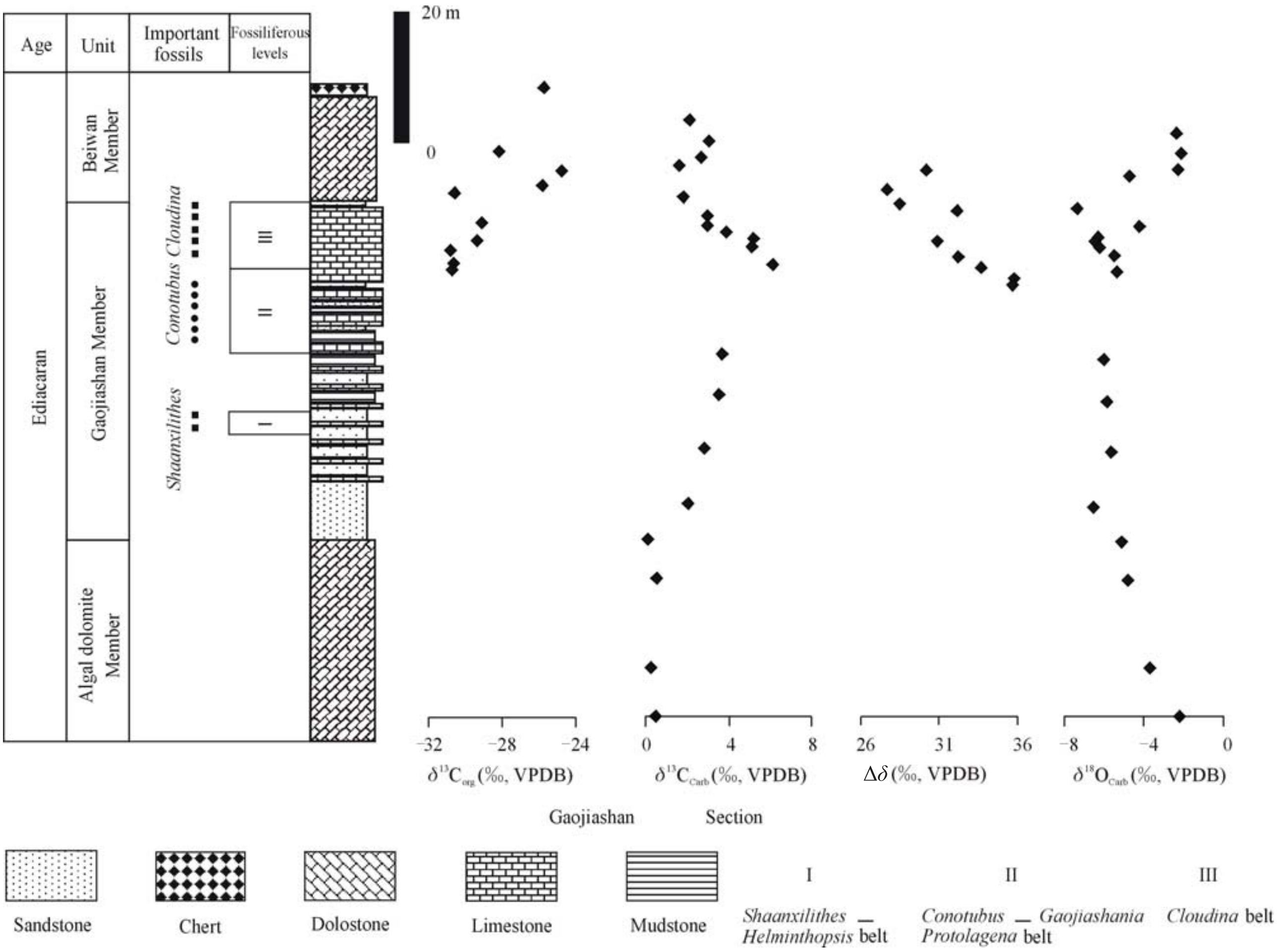

Fig. 2. Profiles of $\delta^{13}C_{carb}$, $\delta^{13}C_{org}$ and $\Delta\delta$ at the Gaojiashan section of Ningqiang, Shaanxi Province, China.

Table 1 Analytical results for samples from the Gaojiashan section, Shaanxi Province, South China

Samples	Unit	Lithology	Depth (m)	$\delta^{13}C_{org}$ (‰, V_{PDB})	$\delta^{13}C_{carb}$ (‰, V_{PDB})	$\triangle\delta$	$\delta^{18}O_{carb}$ (‰, V_{PDB})	TOC %	H/C	Mn (ppm)	Sr (ppm)	Mn/Sr
1	Beiwan Mem.	Chert	97.65	−25.7					1.74	310	30	10.3
2	Beiwan Mem.	Dolomite	87.75	−28.1	2.1	30.2	−2.4	0.3	1.47	150	240	0.6
3	Beiwan Mem.	Dolomite	84.75	−24.7	3.0	27.7	−2.1	0.2	1.53	100	70	1.4
4	Beiwan Mem.	Dolomite	82.35	−25.8	2.7	28.5	−2.2	0.2	0.78	120	70	1.7
5	Beiwan Mem.	Dolomite	81.35	−30.6	1.5	32.1	−4.7	0.2	0.71	160	80	2.0
6	Gaojiashan Mem.	Limestone	76.6	−29.1	1.8	30.9	−7.4		1.99	40	120	0.3
7	Gaojiashan Mem.	Dolomite	73.8	−29.3	2.9	32.3	−4.2	0.6		170	70	2.4
8	Gaojiashan Mem.	Limestone	72.3	−30.8	2.9	33.7	−6.3			140	370	0.4
9	Gaojiashan Mem.	Limestone	71.45		3.8		−6.4			110	740	0.1
10	Gaojiashan Mem.	Limestone	70.4	−30.7	5.2	35.8	−6.2	0.1		180	680	0.3
11	Gaojiashan Mem.	Limestone	69.3	−30.7	5.0	35.8	−5.5			110	820	0.1
12	Gaojiashan Mem.	Limestone	66.8		6.0		−5.3			120	960	0.1
13	Gaojiashan Mem.	Limestone	53.6		3.7		−6.0	0.1		310	420	0.7
14	Gaojiashan Mem.	Limestone	43.5		3.5		−5.8	0.05				
15	Gaojiashan Mem.	Limestone	34.8		2.8		−5.6	0.04				
16	Algal dolomite Mem.	Dolomite	26.35		0.1		−5.1	0.08		80	130	0.6
17	Algal dolomite Mem.	Dolomite	17.63		0.5		−4.8	0.1				
19	Algal dolomite Mem.	Dolomite	9.4		0.2		−3.7	0.06				
20	Algal dolomite Mem.	Dolomite	0		0.4		−2.2	0.1		130	80	1.6

controlled by measuring the Guiyang laboratory standard GBW 04406 for its $\delta^{13}C_{carb}$ ($\delta^{13}C_{carb\text{-}standard}$: −10.85‰; standard deviation: 0.05‰) and $\delta^{18}O_{carb}$ ($\delta^{18}O_{carb\text{-}standard}$: −12.40‰; standard deviation: 0.015‰) values. Results are reported as $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ relative to the Vienna Peedee Belemnite Standard (VPDB). Standard deviation was usually better than ±0.1 ‰.

In order to constrain carbonate diagenesis, samples were further studied for their elemental abundances of Mn and Sr (Veizer, 1983; Popp et al., 1986; Kaufman et al., 1993; Veizer et al., 1999). Samples were digested in 3N HCl and elemental concentrations were measured with atomic absorption spectroscopy. Results were corrected for the amount of insoluble residue (soluble(%)=(total weight - weight of insoluble residue)/(total weight)).

4 Results

Carbonate carbon isotope data lie between 0.1 and 6‰, averaging 2.7±1.7‰ (n=18), and respective $\delta^{18}O_{carb}$ values range from –7.4‰ to –2.1‰, averaging –4.8±1.6‰ (n=18). The organic carbon isotopic composition for kerogen samples ($\delta^{13}C_{org}$) displays values between –30.8‰ and –20.7‰, averaging –28.5±2.4‰ (n=10). The difference between the organic and carbonate carbon isotopic compositions ($\Delta\delta_{carb\text{-}org}=\delta^{13}C_{carb}-\delta^{13}C_{org}$) varies between 27.7‰ and 35.8‰, averaging 31.9±2.9‰ (n=9). The total organic carbon content (TOC) ranges from < 1 to 26 mg/g. Elemental abundances of Mn and Sr are highly variable (Mn: 40–310 ppm; Sr: 30–960 ppm). Mn/Sr ratio varies from 2.4 to 0.1. Geochemical analysis as following:

Algal dolomite Member: $\delta^{13}C_{carb}$ varies between 0.1‰ and 0.5‰; Lower part of Member: $\delta^{13}C_{carb}$ varies between 2.8‰ and 3.7‰; including I *Shaanxilithes-Helminthopsis* belt; Middle part of Member: $\delta^{13}C_{carb}$ varies from 3.7‰ via 6‰ to 5‰; there is the highest $\delta^{13}C_{carb}$ value in the level; including II *Conotubus – Gaojiashania – Protolagena* belt; Upper part of Member: $\delta^{13}C_{carb}$ varies from 5.2‰ to 1.8‰; $\delta^{13}C_{org}$ varies between –30.8‰ and –29.1‰; including III *Cloudina* belt; Beiwan Member: $\delta^{13}C_{carb}$ varies from 1.5‰ to 3.0‰; $\delta^{13}C_{org}$ varies between –30.6‰ and –24.7‰. Analytical results are given in Table 1.

5 Preservation and Diagenesis

5.1 Preservation of organic matter

Evaluating the preservation of the organic matter is crucial. The H/C ratio is of major importance for understanding the state of preservation of kerogen. Strauss et al. (1992a) suggested a minimum H/C ratio of 0.2 in order to exclude poorly preserved kerogens, which would have experienced significant shifts in ^{13}C kerogen.

The H/C ratios obtained for the Gaojiashan samples are very high (0.71 to 1.99) (Table 1), with none close to 0.2. Note that instead of yielding heavier ^{13}C kerogen values with lower H/C ratios are unusually ^{13}C depleted.

5.2 Carbonate diagenesis

It is necessary to evaluate carbonate diagenesis, which can obliterate primary depositional signatures, which reflect seawater chemistry. Generally, an increase in the elemental abundances of Fe and Mn and a decrease in Sr concentrations can be observed during diagenesis (e.g. Veizer, 1983; Marshall, 1992). Furthermore, diagenesis would cause decreases in $\delta^{13}C$ and $\delta^{18}O$. That reflects the incorporation of CO_2 derived from the oxidation of organic matter during carbonate precipitation and the consequence of meteoric water alteration. In order to quantify the degree of carbonate diagenesis, certain thresholds have been proposed. Only samples with Mn/Sr<10, $\delta^{18}O$>–10‰ and

$\Delta\delta$ values around 30±2‰ (with values up to 33±2‰ for organic-rich sediments) are thought to have likely retained their primary carbon isotope signals (Kaufman et al., 1993, 1995).

No correlation exists between commonly applied geochemical indicators for carbonate diagenesis, such as Mn/Sr and the respective δ^{13}C and/or δ^{18}O values (Fig. 3). Elemental abundances and ratios clearly indicate that carbonates have been not altered during diagenesis.

6 Discussion

The organic and carbonate carbon isotope record is based on 21 samples (Fig. 2; Table 1), starting with Algal dolomite Member of Dengying Formation dolostones at the base and followed by sandstone, limestone, shale of the Gaojiashan Member with Gaojiashan Biota, and followed by dolostone of the Beiwan Member.

δ^{13}C values of organic matter vary between –30.8‰ and –24.7‰. The carbon isotopic compositions for carbonate carbon display variations between 0.1‰ and +6‰.

There are positive $\delta^{13}C_{carb}$ (0.1‰ to 6‰) values from 0.4‰ at the base of the section via the first Gaojiashan biota level (I: *Shaanxilithes–Helminthopsis*) to a positive 6‰ value through the middle of the second Gaojiashan biota level (II: *Conotubus – Gaojiashania – Protolagena*) (Fig. 2), the carbon isotopic composition from shallow water sediments becomes significantly ^{13}C enriched, which reflects temporal variations in carbon turnover , i. e. an increasing in photosynthetic carbon fixation followed by Gaojiashan Biota (*Shaanxilithes*, *Helminthopsis*, *Conotubus*, *Gaojiashania*, *Protolagena*) radiation and an increasing subsequent fractional organic carbon burial, and that related to bio-radiation such as increasing original creatures productivity in biomass, which arrives at the most biological production, and here lithology changes from dolomite to limestone which discloses that the paleonenvrionment changes to shallow, warm, nutritious, and oxygen-rich environment (Fig. 4), and there is an obvious relationship among bio-radiation, positive carbon isotope excursion and increasing organic carbon burial.

An evolution towards a negative $\delta^{13}C_{carb}$ shift at the middle of Gaojiashan Member from 6‰ to 1.8‰ through dark and organic-rich limestone sediments with high TOC abundance (TOC: 0.6% to 0.1%) into the boundary between the Gaojiashan Member and Beiwan Member with minimum values around 1.8‰ which is still positive, and the negative $\delta^{13}C_{carb}$ shift from the top of *Conotubus–Gaojiashania-Protolagena* belt (II) to the third Gaojiashan Biota level (*Cloudina* belt) suggest that gradually changing environmental conditions have different bio-zones, discloses that temporal variations in environmental conditions and carbon turnover are the main reason for the second Gaojiashan biota bed extinction and the appearing of the *Cloudina* belt. A lowering in photosynthetic carbon fixation followed by a decrease in photosynthetic carbon fixation and demand for carbon dioxide during primary

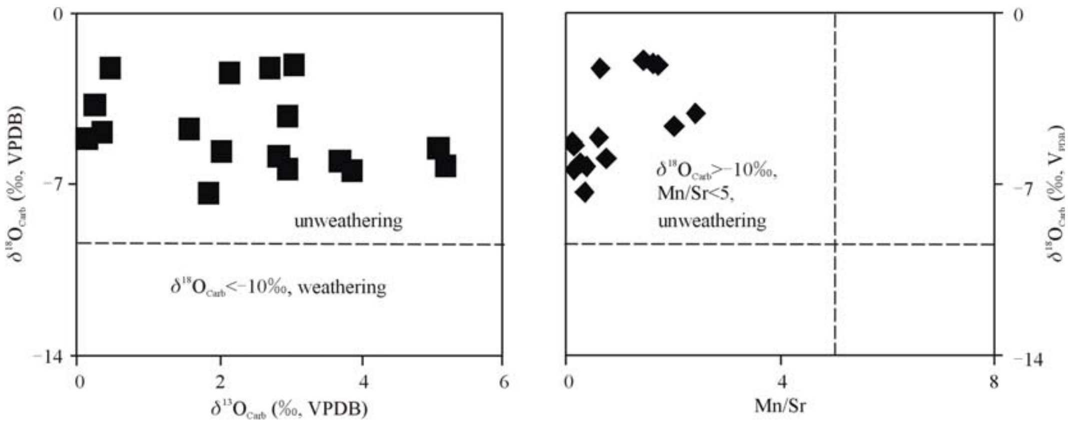

Fig. 3. Cross-plot of $\delta^{18}O_{carb}$ and $\delta^{13}C_{carb}$ (A) and $\delta^{18}O_{carb}$ and Mn/Sr (weight ratio) (B).

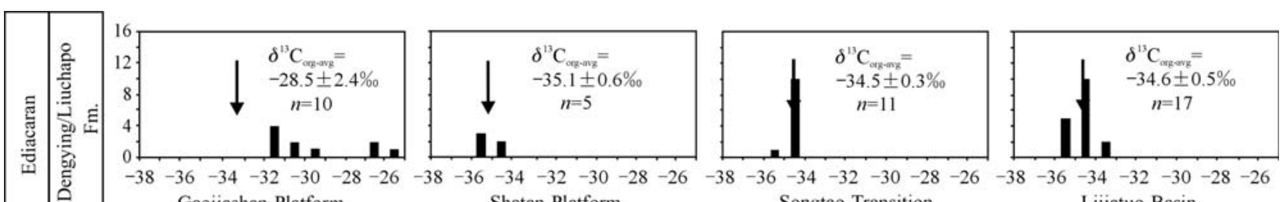

Fig. 4. Distribution of organic carbon isotopic composition of this study in the Dengying period, the Ediacaran on the Yangtze Platform.

production and subsequent fractional organic carbon burial (Fig. 5), likely reflects some contribution from bacterial biomass in addition to organic matter derived from primary production in which much abundant biology still lives in is a little bit deeper water environment which is oxic/sub-oxic which can compare with that of other sections in the Yangtze Platform (Goldberg et al., 2007; Guo et al., 2007) and leads to increasing organic carbon burial in the Gaojiashan and Beiwan time interval (Fig. 5).

An evolution towards a light positive $\delta^{13}C_{carb}$ excursion (from 1.5‰ to 3‰) and a less negative $\delta^{13}C_{carb}$ values continues into the lower part of the Beiwan member with maximum values around −24.7‰. This suggests a clear change either in environmental conditions (such as a change in oxygenation of the water column) or in the type of organic material deposited in the sediment. While the carbon isotopic composition defines a clear stratigraphic trend, no distinct difference in TOC abundance across stratigraphy is discernible, and there is no obvious correlation between TOC and $\delta^{13}C_{carb}$, the positive $\delta^{13}C$ values in the Beiwan Member likely reflect some contribution from bio-radiation in addition to ^{13}C-rich matter derived from primary production.

Usually, it is now widely accepted that the isotopic compositions of sedimentary organic and carbonate carbon are dependent on the relative rates of their burial. Isotopic mass-balance calculations, therefore, can be used to identify and quantify episodes of enhanced organic carbon burial. Isotopic fluctuations to ^{13}C-enriched (heavy) carbon generally reflects high rates of burial of organic carbon while movement to ^{13}C-depleted (light) values reflects weathering and re-oxidation of previously buried organic carbon (Strauss et al., 1992). In this study, the implications of a $\delta^{13}C_{org}$ signal that is not parallel to the $\delta^{13}C_{carb}$ fluctuations. And $\Delta\delta_{carb-org}=\delta^{13}C_{carb}-\delta^{13}C_{org}$ varies between 27.7‰ and 35.8‰, and which probably imply that the organic matter and carbonate should be not digenetic or alteration. The carbonate carbon isotopic composition varies from 1.8‰ to 6‰ (n=10) in Gaojiashan member, yielding an average εTOC ($\delta^{13}C_{carb}-\delta^{13}C_{org}$) of 33.7±2.2‰ ($n$=5).

Comparably high εTOC values > 30‰ have been reported in Gaojiashan member, and these high values are interpreted to reflect the incorporation of ^{13}C-depleted chemoautotrophic biomass to the total organic carbon.

The carbonate carbon isotopic composition varies from 1.5‰ to 3‰ (n=4) in Beiwan member, yielding an average εTOC ($\delta^{13}C_{carb}-\delta^{13}C_{org}$) of 28.8±1.3‰ ($n$=3).

Comparably low εTOC values< 30‰ have been reported in Beiwan member, and these low values are interpreted to reflect the incorporation of ^{13}C-enriched chemoautotrophic bio-extinction to the total organic carbon.

The research results can also compare with the results of a similar time interval with the other researches (Guo et al., 2007; Ling et al., 2007; Zhu et al., 2007) (Fig. 4) in the world: the carbonate and organic carbon isotopic results of the Dengying Formation. from Gaojiashan area ($\delta^{13}C_{carb}$=2.5±1.7‰ (n=20); $\delta^{13}C_{org}$=−28.5±2.4‰ (n=10)), $\delta^{13}C$ values from platform are more positive comparison with those such as Shatan ($\delta^{13}C_{carb}$=−0.6±0.8‰ (n=14); $\delta^{13}C_{org}$=−35.1±0.6‰ (n=5)), Songtao (=−34.5±0.3‰ (n=11)) , Lijiatuo ($\delta^{13}C_{org}$=−34.6±0.5‰ (n=17)) (Guo et al.,

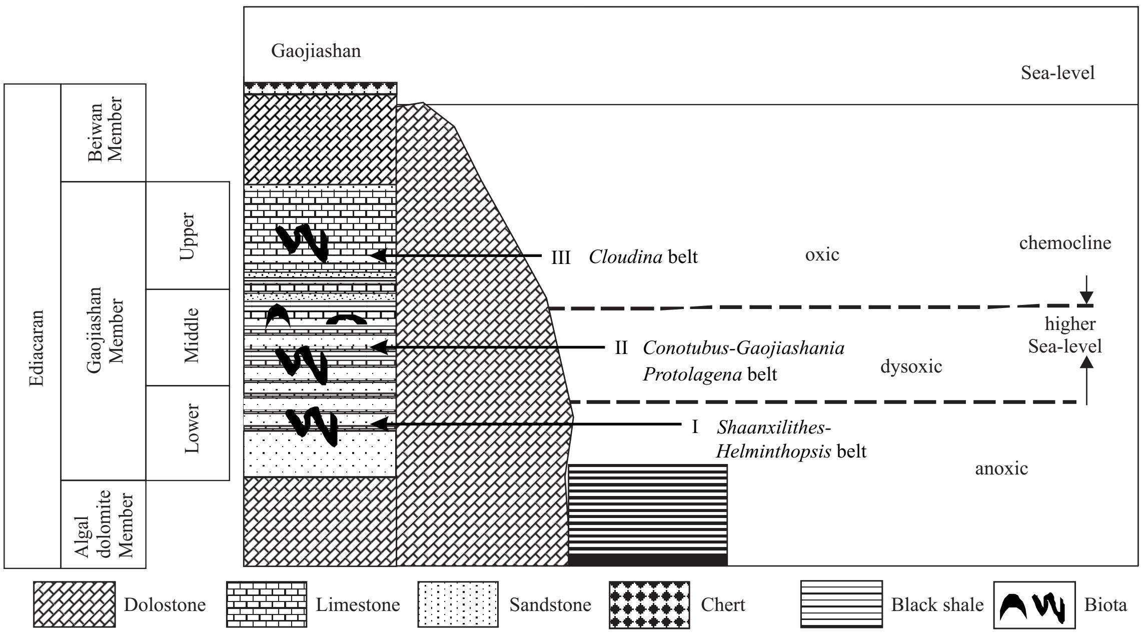

Fig. 5. Environmental evolution across the transition from Algal dolomite Member via Gaojiashan Member to Beiwan Member in Dengying Formation.

2007; Fig. 1; Fig. 4) from shelf, and from shallow water sediments becomes significantly ^{13}C enriched (Guo et al., 2007; Zhu et al., 2007), which reflects temporal variations in carbon turnover, relating to an increasing subsequent fractional organic carbon burial; and the negative carbonate carbon isotopic results show that some contribution from bacterial biomass in addition to organic matter derived from primary production.

7 Conclusions

Biological generals (three fossils assemblages) difference and different environments have affected on the organic and carbonate carbon isotopic compositions:

(1) The carbon isotopic composition from shallow water sediments becomes significantly ^{13}C enriched, which reflects temporal variations in carbon turnover, i.e. an increase in photosynthetic carbon fixation followed by Gaojiashan Biota radiation and an increasing subsequent fractional organic carbon burial, related to bio-radiation such as increasing original creatures productivity in biomass, and there is an obvious relationship among bio-radiation, positive carbon isotope excursion and increasing organic carbon burial.

(2) The negative $\delta^{13}C_{carb}$ shift from the top of *Conotubus – Gaojiashania – Protolagena* belt (II) to the third Gaojiashan Biota level (*Cloudina* belt) suggest gradually changing environmental conditions, leading to the second Gaojiashan biota bed extinction and the appearing of the *Cloudina* belt. A lowering in photosynthetic carbon fixation followed by a decrease in photosynthetic carbon fixation and demand for carbon dioxide during primary production and subsequent fractional organic carbon burial. Lower carbonate carbon isotopic results show that some contribution from bacterial biomass in addition to organic matter derived from primary production, much abundant biology lives in a little bit deeper environment which is still oxic/sub-oxic and leads to increasing organic carbon burial in the Gaojiashan and Beiwan time interval.

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