Front. Earth Sci. China 2007, 1(4): 425–430
DOI 10.1007/s11707-007-0052-z

RESEARCH ARTICLE

Organic fraction of the total carbon burial flux deduced from carbon isotopes across the Permo-Triassic boundary at Meishan, Zhejiang Province, China

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Abstract By combining the carbon cycle model with the records of carbonate and organic (kerogen) carbon isotope, this paper presents the calculation of the fraction of organic carbon burial (f_{org}) of beds 23–40 at the global boundary stratotype section and point (GSSP) of the Permian–Triassic boundary at Meishan, Zhejiang Province. The resulting calculation produces two episodes of f_{org} maxima observed to occur at beds 23–24 and 27–29, which respectively corresponds to the two episodic anoxic events indicated by the flourish of green sulfur bacteria. Two episodic f_{org} minima occurred at beds 25–26 and 32–34, generally coincident with the flourish of cyanobacteria (bed 26 and upper part of beds 29 to 34) as shown by the high value of 2-melthyhopnoanes. It appears that the f_{org} is related to the redox conditions, with greater f_{org} values observed under the reductive condition. The relationship between f_{org} and the total organic carbon (TOC) content was complex. The f_{org} value was low at some beds with a high TOC content (such as bed 26), while high observed at some beds with a low TOC content (e.g. bed 27). This association infers the important contribution of primary productivity to the TOC content. The original organic burial could be thus calculated through the configuration of the function of the primary productivity and f_{org}, which can be used to correct the residual TOC measured today. This investigation indicates that compiling the organic-inorganic carbon isotopes with the carbon cycle model favors to understand the fraction of organic carbon burial, providing information for the reconstruction of the coupling among biota, environments and organic burial.

Keywords carbon isotope, carbon cycle, organic burial, Permian–Triassic boundary

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Journal of China University of Geosciences, 2007, 32(6): 767–773
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1 Introduction

Carbon isotope records of oceanic carbonates play an important role in the research on the formation and origins of oil and gas (Xu et al., 2001; Guo et al., 2004; Teng et al., 2004, 2006; Dai et al., 2005; Gao et al., 2005; Zhang et al., 2006a, 2006b). The carbon isotope includes inorganic carbon isotope ($\delta^{13}C_{\mathrm{carb}}$) and organic carbon isotope ($\delta^{13}C_{\mathrm{org}}$), both of which are widely used to evaluate and explore hydrocarbon source rocks, and took effects in recognizing origins and environmental conditions of hydrocarbon source rocks. However, both the $\delta^{13}C_{\mathrm{carb}}$ and the $\delta^{13}C_{\mathrm{org}}$ are impacted by several factors, such as productivity, sedimentary environments and preservation of organic matter. Even each factor might take different actions in different environments. This prevents from evaluating which is the main factor and which is the second one.

Intensive investigations on the global change result in important breakthroughs on the basis of the quantitative simulation of the global carbon cycle. Under the guide of the carbon cycle theory (especial the oceanic carbon cycle model), the paleooceanic carbon cycle model was put forward, and the geologists try to explain its significance in the environmental study (Kump, 1991; Kump and Arthur, 1999; Hayes et al., 1999; Voigt et al., 2006). For example, the difference of the two carbon isotope values, ΔC ($\delta C_{\mathrm{carb\text{-}org}}$), can be used to decode the variation of carbon cycling and the flux change between the two carbon reservoirs (organic carbon reservoir and inorganic carbon reservoir). Riccardi et al. (2007) analyzed the variation of ΔC during the Permian–Triassic transition at Meishan and Shangsi Sections, and showed that the ΔC reduced distinctly at the “event bed”. They proposed that this variation was caused by the microbial variation from algae/cyanobacteria to green sulfur bacteria. Hayes et al. (1999) and Kump and Arthur (1999) constructed the formula to calculate the fraction of organic carbon burial, the fractionation between inorganic and organic carbon, and the isotope fractionation during the photosynthesis on the basis of carbon cycle models.

In a global scale, the variation of the productivity can impact the values of $\delta^{13}C_{carb}$ and $\delta^{13}C_{org}$, while the ΔC, which was controlled by the degradation and the burial efficient of organic matter, kept constant in some geological time. Voigt et al. (2006) studied the carbon isotope of Late Cenomanian in Europe, and calculated the variation of the fraction of organic carbon burial (f_{org}) in different sea-levels based on the carbon cycle model proposed by Kump and Arthur (1999). The parallel and paired study on carbonate and organic carbon isotope can provide information on the coupling relationship among biota, environments and organic carbon burial and on the fraction of organic carbon burial.

This paper studies the paired carbonate and organic carbon isotope records during the Permian–Triassic transition at Meishan Section, where the biostratigraphy and geochemistry have been studied intensively. Based on the carbon cycle model of Kump and Arthur (1999), the authors try to calculate the fraction of organic carbon burial during the important geo-turn, and discuss the relationship among the fraction of organic carbon burial, the primary production and the redox condition. The final purpose of this paper is to provide new methods for the geobiological evaluation of hydrocarbon source rocks and their comparison with the conventional methods.

2 Materials and methods

The Meishan Section is located in northwest of Changxing County, Zhejiang Province. The sedimentary facies of the Changhsing Formation of Late Permian is a carbonate platform, while the Yinkeng Formation of Early Triassic is developed at the deep water slope of the shallow sea. The Meishan Section outcrops at six sites, named as A, B, C, D, E and Z from west to east, respectively. The Section D is the global boundary stratotype section and point (GSSP) of Permian–Triassic boundary.

All the samples were collected from the Section B, from bed 22 of the Changhsing Formation to bed 40 of the lower member of Yinkeng Formation. In this paper 384 samples have been collected from this 20.6 m-thick interval, of which 26 samples were analyzed for the paired carbonate and organic carbon and oxygen isotopes. The analytic method of organic carbon isotope is as follows. The kerogen was extracted out of the whole rock and put into the quartz tube with oxidizer and the platinum used as the catalyzer. The quartz tube was combusted at 850°C and then the MAT-251 was used to detect and measure the carbon and oxygen isotope of the pured carbon dioxide. Carbon and oxygen isotope compositions of the bulk carbonate were determined by using the conventional H_3PO_4 digestion method (McCrea, 1950). The carbonate samples were crushed to less than 100 meshes and the powders were reacted with 100% phosphoric acid for 24 h at 25°C. $^{13}C/^{12}C$ and $^{18}O/^{16}O$ ratios of CO_2 generated by the acid reaction were measured on the MAT-251. All the isotopic data are reported as per mil (‰) relative to Vienna Pee Dee belemnite (V-PDB) standard, and the analytical precision is better than ± 0.1‰. The analysis of TOC content and the extraction of kerogen were conducted at the Laboratory of Jianghan Oil Field Academy, and the others were analyzed at the State Key Laboratory of Geological Processes and Mineral Resources, China University of Geosciences (Wuhan). The data are shown in Table 1.

3 Results and discussion

3.1 Organic and the carbonate carbon isotope composition

Bulk carbonate $\delta^{13}C$ values range from −1.5‰ to 4.0‰ (PDB) in the interval investigated (Fig. 1), showing two episodes of negative excursions. The first negative excursion (4‰) is characterized by a gradual start at beds 23–24 and a sharp end at beds 25–26. The second negative excursion of carbon isotope started from the middle part of bed 27, and got the nadir at bed 34, and thereafter the carbon isotope composition turned to a positive excursion. The associated kerogen carbon isotope values varied in the range of (−33‰)–(−24‰) (PDB), with a mean value of −27.71‰. The kerogen carbon isotope composition falls into two regions (Fig. 2), one was lower than −30‰ indicative of a mixed type, and the other was greater than −28‰ indicative of a humic type. Coincident with the carbonate carbon isotope record, the organic carbon isotope values also showed two episodes of negative excursions, which, respectively occurred at bed 26 in Changhsing Formation and beds 30–34 in Yinkeng Formation. As a whole, the values of $\delta^{13}C_{org}$ in Yinkeng Formation were larger than those in Changhsing Formation. The TOC content during the Permian–Triassic transition was very low, and shows no relationship with the $\delta^{13}C_{org}$. In beds 23–24 and 29–34, the TOC content was relatively high and fluctuated frequently, which formed two distinct intervals. The TOC content in bed 26 was also relatively high. However, in beds 25, 27 and 28, the concentration of TOC was low, less than 0.1% in general.

3.2 Construction of the carbon cycle model and the calculation of the fraction of organic carbon burial (f_{org})

The carbon isotope during the Permian–Triassic transition has been studied intensively (Magaritz et al., 1988, 1992; Xu and Yan, 1993; Smith and MacLeod, 1994; Li and Zhou, 2002; Krull et al., 2004; Wang et al., 2005; Payne et al., 2005; Zhang et al., 2006; Haas et al., 2007), and played an important role in understanding the variation of the biota and the associated environments during this critical time. Until now, lots of the scholars have accepted the fact that the carbon isotope underwent a distinct negative excursion, though the causes and the variation patterns are still controversial. The variation range of the carbon isotope composition is also debated

Table 1 $\delta^{13}C_{org}$ and $\delta^{13}C_{carb}$ data of the samples analyzed from Meishan Section B

Bed	Sample No.	Depth/cm	Lithology	$\delta^{13}C_{org}$/(‰, PDB)	$\delta^{13}C_{carb}$/(‰, PDB)
38	B-38-8	1 797	Calcareous mudstone	–25.7	2.4
37	B-34-188	1 334	Calcareous mudstone	–24.4	0.9
35	B-34-148	975	Calcareous mudstone	–25.0	–0.0
34	B-34-108	692	Calcareous mudstone	–25.7	–1.1
	B-34-79	549	Black mudstone	–25.7	–1.1
	B-34-55	439	Calcareous mudstone	–25.3	–1.1
	B-34-26	307	Calcareous mudstone	–28.9	–1.5
	B-34-8	237	Calcareous mudstone	–25.4	–0.7
32	B-32-16	134	Calcareous mudstone	–26.1	–0.2
30	B-30-12	75	Limestone	–30.6	–0.3
	B-30-16	59	Marl	–28.8	–0.2
29	B-29-8	41	Limestone	–25.5	1.2
28	B-28-4	27	Clay	–25.2	–0.2
27	B-27-12	19	Limestone	–25.4	0.7
	B-27-24	11	Limestone	–30.6	0.2
26	B-26-2	9	Calcareous mudstone	–32.7	–0.4
	B-26-5	6	Calcareous mudstone	–31.5	–0.9
25	B-25-2	3	Clay	–28.8	–0.6
24	B-24e-2	–3.5	Limestone	–29.6	1.3
	B-24e-3	–6.5	Limestone	–28.7	1.3
	B-24d-4	–18.5	Limestone	–27.8	0.8
	B-24c-8	–48.5	Limestone	–28.7	2.5
	B-24b-6	–66	Limestone	–28.3	2.8
	B-24a-6	–75	Limestone	–28.1	2.1
23	B-23-3	–86	Limestone	–29.8	3.6
	B-23-42	–190.5	Limestone	–28.0	3.6

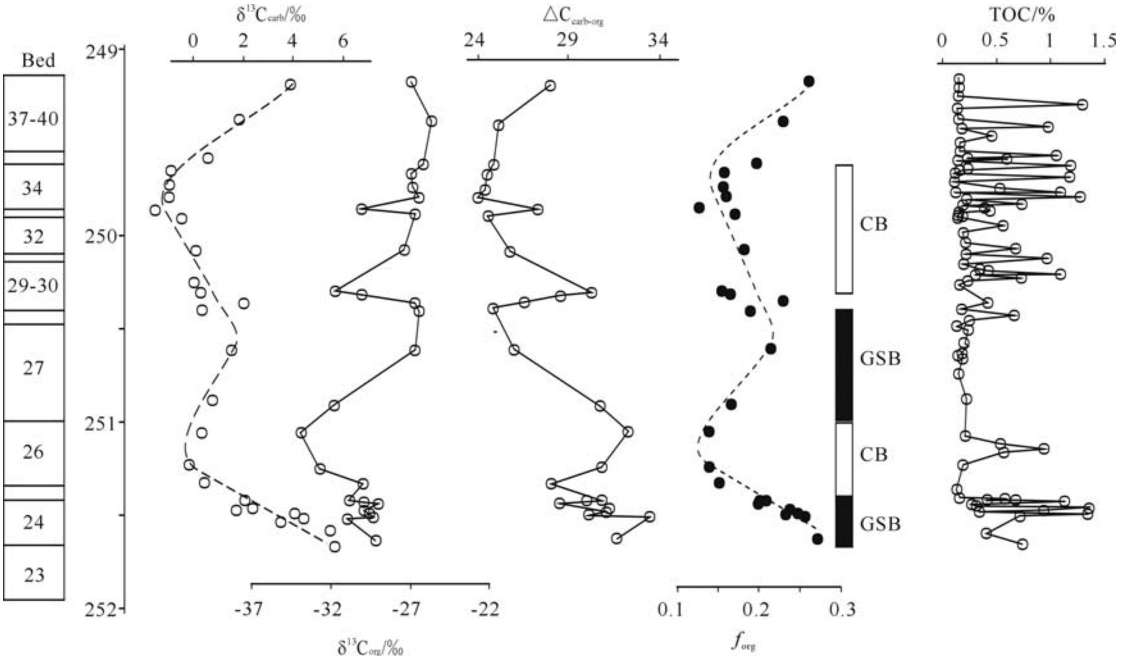

Fig. 1 Variation trends of $\delta^{13}C_{org}$, $\delta^{13}C_{carb}$ and TOC at Meishan Section B (dating data from Bowering et al., 1998)
CB and the solid columns denote the intervals of cyanobacterial expansions; GSB and the open columns show the intervals of the flourish of green sulfur bacteria

to date. Xu and Yan (1993) proposed a variation range as large as 8‰ during the Permian–Triassic transition, with the minima of carbon isotope values occurred at the Permian–Triassic boundary. However, some scholars supposed a less variation range, i.e. only 3‰, and the minima of the carbon isotope values not at the PTB (Cao et al., 2002; Payne et al., 2004)

In order to quantitively investigate the variation of carbonate and organic carbon isotope composition during the Permian–Triassic transition, it is necessary to understand the

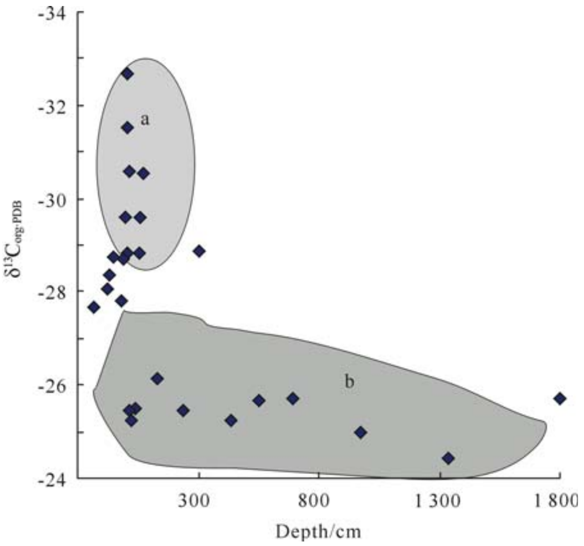

Fig. 2 $\delta^{13}C_{org}$ variation with depth at Meishan Section B

evolution mechanisms. Based on the research on the modern ocean carbon cycle model, lots of scholars proposed some paleo-oceanic carbon cycle models to recognize the shifts between the organic and the inorganic carbon reservoirs (Kump and Garrels, 1986; Kump, 1991; Kump and Arthur, 1999; Hayes, 1999; Voigt et al., 2006). Based on the achievements of previous scholars, Kump and Arthur (1999) added the contributing vectors of the volcanism and the weathering occurring at the earth surface into the carbon cycle model, and constructed a formula to calculate the fraction of organic carbon burial, which provided us with the mathematic basis for the semi-quantitative or quantitative research on the carbon isotope and the fraction of organic carbon burial. The following formula is proposed by Kump and Arthur (1999).

$$f_{\text{org}} = \frac{(\delta_w' - \delta_{\text{carb}})}{\Delta_{\text{B}}}$$

where δ_{w}' is the isotopic composition of the carbon input into the surface ocean from the volcanisms and weathering, which was proposed to be the same as that of the mantle, about −5‰. The Δ_{B} represents the difference between the organic and the carbonate carbon isotope, i.e. $\Delta_{\text{B}} = \delta^{13}\text{C}_{\text{org}} - \delta^{13}\text{C}_{\text{carb}}$.

Based on the above formula, the fraction of organic carbon burial was calculated (Fig. 2). The results indicated that the variation of the fraction of organic carbon burial showed comparable trends with that of the carbonate carbon isotope composition. In the studied interval, there were two episodes of high and low values of f_{org}. The two intervals of high f_{org} occurred at beds 23–24 and bed 27 to lower part of bed 29, while the two low f_{org} occurred at beds 25–26 and 32–34. It was interesting that the two high f_{org} intervals, such as beds 24 and 27, corresponded to the two episodes of the extreme anoxia as indicated by the flourish of green sulfur bacteria (Grice et al., 2005). It appeared that the increased fraction of the organic carbon burial was related to the anoxic sedimentary environment. The two low f_{org} intervals corresponded to the two episodes of cyanobacteria expansion, such as bed 26 and upper part of bed 29 to 34 (Xie et al., 2005, 2007). The molecular fossil studies indicated that the whole water column during the deposition of beds 26 and 29 was oxygenic (Huang et al., 2007; Wang et al., 2005). The oxygenic water might be caused by the oxygenic photosynthesis of cyanobacteria, which in turn reduced the fraction of the

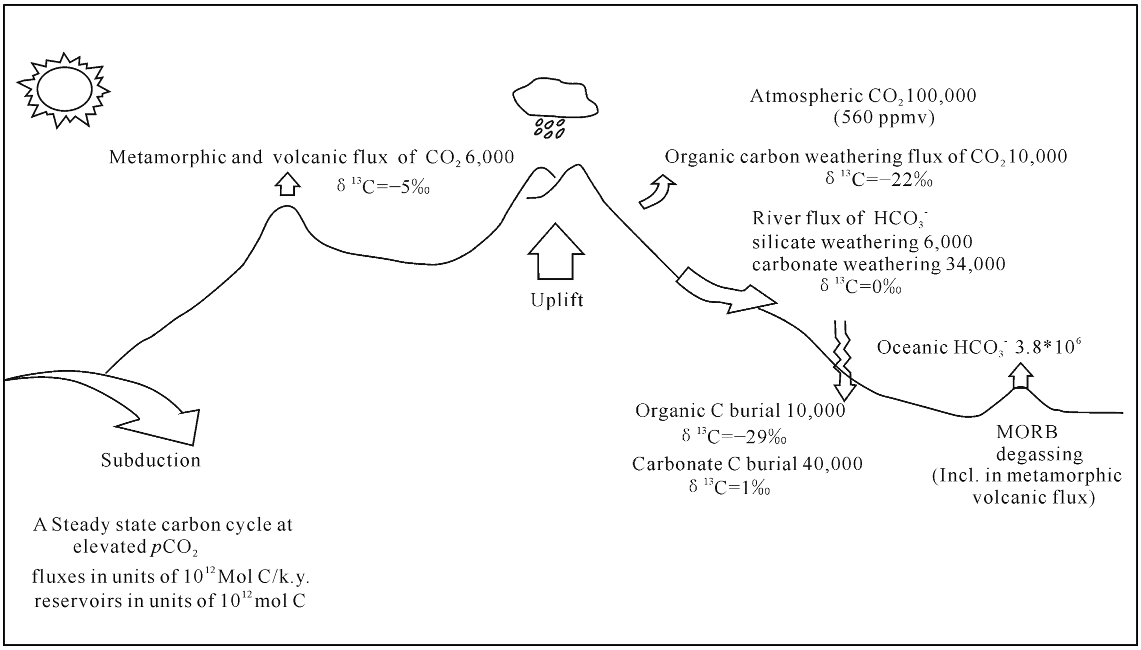

Fig. 3 Carbon cycle model proposed by Kump and Arthur (1999)

organic carbon burial. On these points, the fraction of the organic carbon burial correlated to the redox condition. And in some cases, the f_{org} could be used as the proxy of the coefficient of organic matters preserved.

3.3 Relationship between the fraction of the organic carbon burial and the TOC content

The variations of the fraction of the organic carbon burial and the TOC content showed that the high fraction of the organic carbon burial in Late Changxingian (bed 24) corresponded to the high TOC content at that time. This indicated that both the high primary productivity and the low oxygen concentration caused lots of organic carbon preserved. However, the positive correlation between the f_{org} and the TOC content was not observed in other intervals except at bed 24. At some beds, such as beds 26, 27 and 34, the fraction of the organic carbon burial was opposite to that of the TOC content. At bed 26, the f_{org} was low but the TOC content was relatively high. At bed 27, contradictory to the high f_{org} was the lower TOC content.

Such a kind complex relationship between the fraction of the organic carbon burial and the TOC content (Table 2) might be reasonable in considering that the TOC is not only related to the f_{org}, but also contributed by the primary productivity. Specifically, although the f_{org} was low at bed 26, the primary production was high due to the cyanobacterial expansion, which will also lead to a high TOC content. In contrast, at bed 27, although the f_{org} was high owing to the anoxic condition, the TOC content was low due to the low primary productivity. The complex relationship between the fraction of the organic carbon burial and the TOC content inferred that both the primary productivity and the redox condition were important contributors to the organic matter preserved in hydrocarbon source rocks. In some cases, the primary production was proposed to be more important. Taking bed 26 as an example, although the oxic condition was not favorable for organic preservation because it increased the degradation of organic carbon, and thus decreased the fraction of the organic carbon burial, but the TOC content was high due to the high primary productivity as a consequence of cyanobacterial expansions. It appeared that, in some case such as the slope environments investigated here, the high primary productivity in association with the poor preservation condition would also result in a high TOC content. In addition, the enhanced input from terrestrial organics might also contribute to the TOC in these beds (beds 26 and 32−34) as the moretane molecular fossils suggested (Xie et al., 2007b).

Table 2 Fraction of the organic carbon burial (f_{org}), the primary production (P) and the TOC content in some beds in Meishan Section B

Bed No.	f_{org}	P	TOC content
32−34	Low	High	High
27	High	Low	Low
26	Low	High	High
24	High	High	High

According to the fraction of the organic carbon burial and the primary production, a function can be configured to address their relationship with the original organic burial (TOC_{burial}), i.e. $TOC_{burial} = F(f_{org}, P)$. The difference between the TOC_{burial} and the measured TOC content is caused by the lost organic carbon during the diagenesis. On this basis, the geobiological data gained through the carbon isotope records can be used to correct the conventional evaluation methods for hydrocarbon source rocks.

4 Conclusions

There are at least two episodic negative excursions of both carbonate and organic carbon isotope composition, occurred mainly at beds 25–26 and 32–34, respectively, during the Permian–Triassic transition at Meishan Section. The kerogen carbon isotope composition indicated that the kerogen extracted in this section has two origins, one was a mixed type featured by a $\delta^{13}C$ value less than −30‰, and the other was a humic type indicated by a $\delta^{13}C$ value larger than −28‰. The difference between carbonate and organic carbon isotope composition ($\Delta C_{carb\text{-}org}$) varied distinctly during this time. The values of $\Delta C_{carb\text{-}org}$ in Yinkeng Formation were generally lower than those in Changhsing Formation.

Based on the carbon cycle model proposed by Kump and Arthur (1999), the calculated values of the fraction of the organic carbon burial f_{org} showed two maxima and minima occurred at beds 23–24 and 27–29, and beds 25–26 and 32–34, respectively. The two high f_{org} intervals corresponded to the two episodes of extreme anoxia, while the two low f_{org} intervals corresponded to the two episodes of cyanobacteria expansions. The relationship between f_{org} and TOC content appeared complex. At some beds, the low f_{org} was associated with the high TOC content; the latter was supposed to be contributed by high primary productivity. The measured residual TOC content can be corrected on the basis of the original organic burial deduced from the primary productivity and the fraction of organic carbon burial.

The preliminary results of the paper indicate that based on the carbon cycle model, the parallel and paired analysis of the carbonate and organic carbon isotope composition can be used to calculate the fraction of the organic carbon burial. The resulting data can further provide information for reconstructing the model to decipher the coupling relationship among biota, environments and the organic carbon burial.

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