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Research paper

Stable isotope ($\delta^{13}C_{ker}$, $\delta^{13}C_{carb}$, $\delta^{18}O_{carb}$) distribution along a Cambrian outcrop section in the eastern Tarim Basin, NW China and its geochemical significance

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Hu Liu [a,b], Zewen Liao [a,*], Haizu Zhang [c], Yankuan Tian [a], Bin Cheng [a], Shan Yang [a]

[a] State Key Laboratory of Organic Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou 510640, China
[b] Sichuan Key Laboratory of Shale Gas Evaluation and Exploitation, Chengdu 610091, China
[c] Research Institute of Petroleum Exploration and Development, PetroChina Tarim Oilfield Company, Korla 841000, Xinjiang, China

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ABSTRACT

This study investigated the geochemical features of the lower Paleozoic strata of Yaerdang Mountain outcrop along with the core samples from well TD2∈ in the eastern Tarim Basin, NW China. The total organic carbon abundance, hydrocarbon-generating precursor biospecies, and stable isotope ratios of organics and carbonate ($\delta^{13}C_{ker}$, $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$) were comprehensively studied for their possible correlative constraints during sedimentary evolution. The results revealed that the $\delta^{13}C_{ker}$ (VPDB) of Cambrian kerogens along the outcrop section varied from −34.6‰ to −28.4‰, indicating an increasing tendency from the lower Cambrian to the upper Cambrian. This was on the whole accompanied by the variation in the $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ along the profile, which might be associated with the changes in the sea level and also in the compositional variation of benthic and planktonic biomass. The large variation in the stable carbon isotope ratios up to 6‰ along the outcrop section reflected the heterogeneity of the Cambrian source rocks from the eastern Tarim Basin. Hence, the ^{13}C-enriched crude oils from well TD2∈ might have been derived from a localized stratum of Cambrian source rocks. The results from this study showed the possibility of multiple source kitchens in the Cambrian–lower Ordovician portion of Tarim Basin.

1. Introduction

The Tarim Basin is one of the most important petroleum basins in China (Li et al., 1996), a paleozoic cratonic basin, overlaid by Mesozoic–Cenozoic foreland depressions. The two main marine oil source units identified in the basin are the Cambrian–lower Ordovician and middle–upper Ordovician strata (Liang et al., 2000; Zhang et al., 2000). Recently, it has been reported that oils typically from the Cambrian–lower Ordovician source rocks in the Tarim Basin are ^{13}C-enriched, such as crude oils of TD2∈ and TZ62S with bulk stable carbon isotopic ratios around −28‰ (Zhang et al., 2004a; Xiao et al., 2005; Tian et al., 2012), which are much heavier (usually by 3–6‰) than the oils from middle–upper Ordovician source rocks. Researchers have previously attributed the ^{13}C-enrichment in the crude oils to the post-depositional evolution due to the geochemical transformation of organic material after sedimentation, and the origin-controlling mechanisms resulting from contribution from primary biomass (Liu et al., 2013).

In this work, outcrop samples from the Yaerdang Mountain profile and core samples from well TD2∈ were investigated for the basic geochemical features of the lower Paleozoic strata in the eastern Tarim Basin, NW China. Primarily, the correlations between total organic carbon (TOC) abundance, stable isotope ratios and their depositional conditions were probed. And then TOC abundance, primary biomass, $\delta^{13}C_{ker}$ and their constraints between lower Cambrian and upper Cambrian strata were discussed. Finally, the diagnostic explanations to the origins of ^{13}C-enriched oils in the Tarim Basin such as TD2∈ and TZ62S were supposed.

* Corresponding author. Tel.: +86 20 85290190.
E-mail address: liaozw@gig.ac.cn (Z. Liao).
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2. Samples and experimental analysis

2.1. Geological background and sample collecting

As an important part of the Rodinia supercontinent during the Jinningian (1000–800 Ma), the Tarim Basin had undergone two evolution phases — the initial Sinian formation phase of passive continent and the Cambrian–early Ordovician maturation phase. The primitive basin experienced a series of evolutions of Sinian craton central uplifting, craton marginal depression and Cambrian–early Ordovician intracratonic depression, craton marginal depression (Jia, 1997; Jia et al., 2007; Zhang et al., 2007).

During the Cambrian period, the partial pressure of carbon dioxide in the atmosphere ($p(CO_2)$) was quickly declining (Berner, 1991, 1994, 1998; Berner and Kothavala, 2001), accompanying the climate alternation between warm humid and hot arid. The Manjiaer depression in the eastern Tarim Basin (Fig. 1) had experienced a sea level change cycle from early Cambrian sea level rising to middle–late Cambrian falling (Zhang et al., 2006). In the early Cambrian, along with the sea level rising, the upturning of ocean currents could reach the inner shelf, even to the onshore region. On one hand, the cool upwelling currents which were oxygen-deficient, phosphorus and silicon-rich are beneficial for the thriving of planktonic algae and benthic algae. On the other hand, the enhanced basinal accommodation could favor the marine sedimentation enriched in phosphorus, silicon and organic matter (Zhang et al., 2004b). During the middle–late Cambrian period, along with the sea level declining, the basinal accommodation controlled by the preservation and upwelling complex pattern gradually decreased. As a result, light colored carbonate and clastic rocks were deposited from the platform to the shallow shelf region.

In order to investigate the basic geochemical features of the lower Paleozoic strata, which would hopefully explain the origins of ^{13}C-enriched oils in the Tarim Basin, a series of outcrop samples were collected from the Yaerdang Mountain profile (YI and YII, at the basin edge) and a few core samples from the well TD2∈ in the eastern Tarim Basin, NW China (Fig. 1, Tables 1 and 2). Among which, 14 rock samples were from profile YI covering the strata of

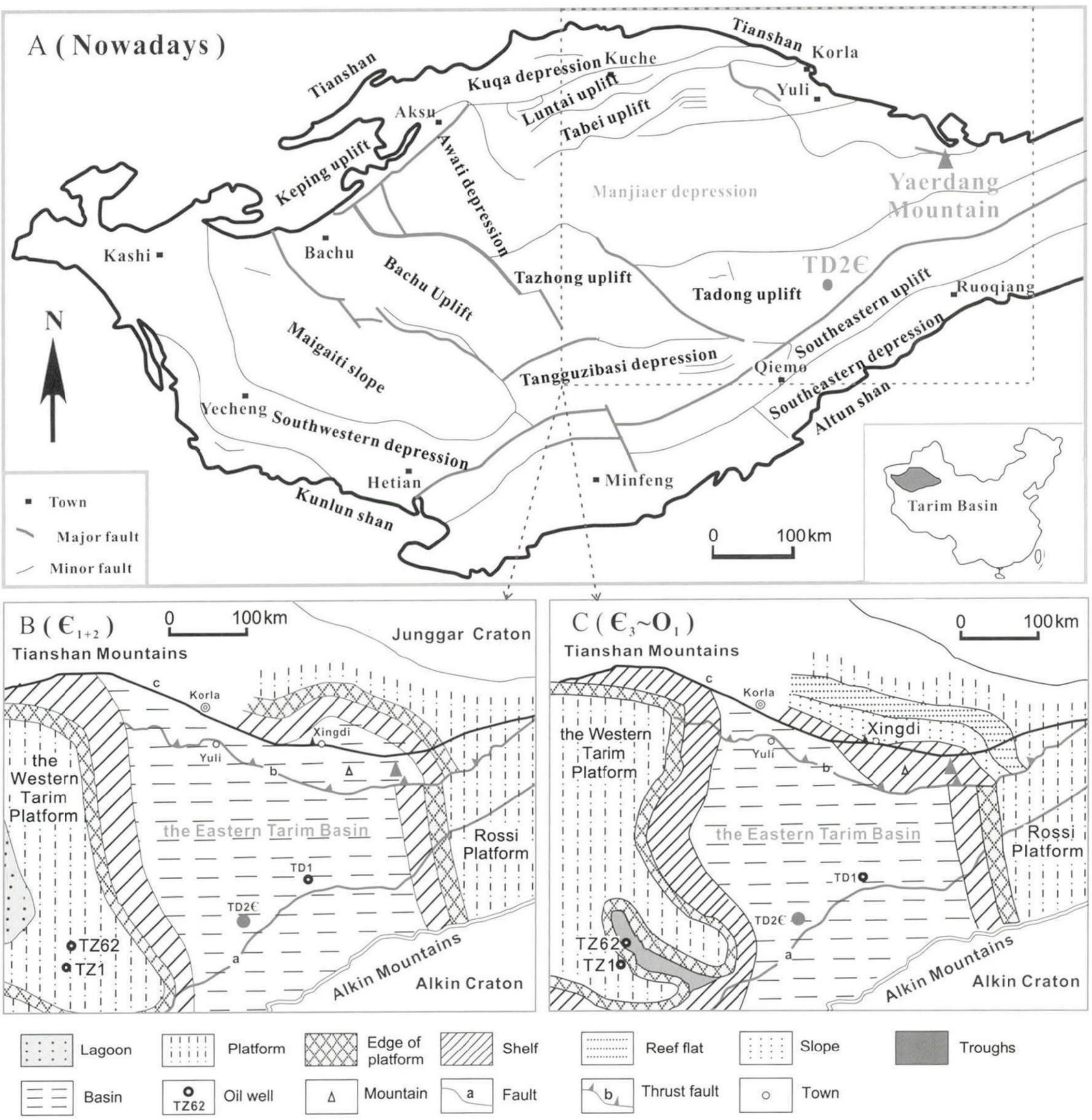

Figure 1. (A) Structure map of the present Tarim Basin (modified after Zhang et al., 2000). (B, C) Cambrian sedimentary facies of the eastern Tarim Basin (modified after Huang et al., 2009). (a) Che'erchen fracture. (b) Kongquehe fracture. (c) Xingdi fracture.

Table 1
The Cambrian strata of the southern Kuruktag region (Yaerdang Mountain) in comparison with the international standard (modified after Jia et al., 2004; Huang et al., 2009).

Erathem	System	International standard (2009)		Chinese standards (2009)	Southern Kuruktag region	Age(Ma)
		Series	Formation	Series	Formation	
			Himantian		Yinpinshan	444
					Yuanbaoshan	
		O_3	Katian	O_3	Zatupo	
			Sandbian			
	Ordovician				Queerqueke	461
		O_2	Darriwilian	O_2		
			Dapingian		Heituao	472
		O_1	Floian	O_1		
			Tremadocian			488
			Stage 10			
		Furongian	Stage 9	$Є_3$	Tuershaketagequn	
			Paibian			501
Paleozoic			Guzhangian			
		Series 3	Drumian			
			Stage 5	$Є_2$	Moheershan	
	Cambrian					510
			Stage 4			
		Series 2			Xidashan	
			Stage 3			521
			Stage 2	$Є_1$	Xishanbulake	
		Terreneuvian	Fortunian			
						542
		Ediacaran		Z_2	Hangeerqiaoke	
	Sinian				Shuiquan	630
				Z_1	Yukengou	
Neoproterozoic					Zhamoketi	680
		Cryogenian		Nh_2	Teruiaiken	
	Nanhua				Aletonggou	
				Nh_1	–	
					Beiyixi	800

Nanhua (800–680 Ma), Aletonggou Formation (Nh_{1a}) and Sinian Shuiquan Formation (Z_{2s}), and 64 rock samples from profile YII covering lower Cambrian Xishanbulake Formation ($Є_{1xs}$), Xidashan Formation ($Є_{1xd}$), middle Cambrian Moheershan Formation ($Є_{2m}$) and the upper Cambrian Tuershakequn Formation ($Є_{3t}$). There were 4 core rock samples and one crude oil from well TD2Є for comparison.

Table 2
Cambrian sedimentary facies of Kuruktag region (Zhang et al., 2006; Huang et al., 2009).

Geologic time	The southern Kuruktag region	Well TD2Є
$Є_3$–O_1	Inner Shelf	Outer Shelf
$Є_2$	Outer Shelf	Bathyal
$Є_1$	Bathyal	Bathyal

2.2. Sample treatment and kerogen preparation

Rock samples without obvious weathering were surface washed by water, and then dried for more than 4 h in a vacuum oven at 55 °C. The dry rock samples were cracked and ground to fine powders (about 80 meshes). Powdered rock sample was decarbonated with hydrochloric acid (HCl), and demineralized with hydrofluoric acid (HF). Kerogens were prepared from the rock samples according to the procedures described by Fu and Qin (1995). The resulting kerogens were dried in a vacuum oven around 60 °C for more than 4 h, and then ground to fine powder.

2.3. Instrumental analysis

2.3.1. Total organic carbon (TOC) abundance analysis

Total organic carbon abundance of the rock samples were measured by a CS-400 analyzer from American LECO Corporation. The powder sample was firstly decarbonated by hydrochloric acid (HCl), and then combusted in oxygen flow and the generated CO_2 were detected by an infrared detector to acquire the TOC values.

2.3.2. Stable carbon and oxygen isotope analysis of rock samples

To measure the $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ of the rock samples, anhydrous phosphoric acid (100%) was added drop-wisely to the powered rock samples in order to digest the carbonate. Evolving CO_2 was separated, purified and detected by GV IsoPrime II Stable Isotope Ratio Mass Spectrometer. The analysis error was less than 0.06‰ for $\delta^{13}C_{carb}$ and 0.10‰ for $\delta^{18}O_{carb}$, respectively.

2.3.3. Stable carbon isotope analysis of kerogens

$\delta^{13}C_{ker}$ values were obtained using a Delta XL plus IRMS (isotope ratio mass spectrometer). The isotopic compositions were calculated by integration of the masses 44, 45 and 46 ion currents of the CO_2 peak. The $^{13}C/^{12}C$ composition is reported relative to that of a reference gas pulse produced by allowing carbon dioxide of known $^{13}C/^{12}C$ content into the mass spectrometer (Grice et al., 2007). The accuracy is better than 0.5‰ (VPDB). Each sample was analyzed at least twice, and the average values reported.

3. Results and discussion

3.1. Stable carbon/oxygen isotope results of rock samples from the Yaerdang Mountain profile

During the Cambrian period, although the Yaerdang Mountain profile and the location of TD2∈ represented the shallow shelf carbonate facies and starved basin sedimentary facies, respectively (Fig. 1), they were both controlled by synchronous sea level variations and climate change. Therefore, the TOC distribution trends with geologic age and the stable carbon isotope distribution curves for these two profiles should be comparable. As shown in Fig. 2, there is a descending trend for the TOC abundance distribution from lower Cambrian to upper Cambrian strata along with the profile. High TOC abundance is generally associated with high organic carbon buried rate and high biomass productivity. The relationships between TOC abundance, stable carbon/oxygen isotope distributions and the depositional background of well TD2∈ have

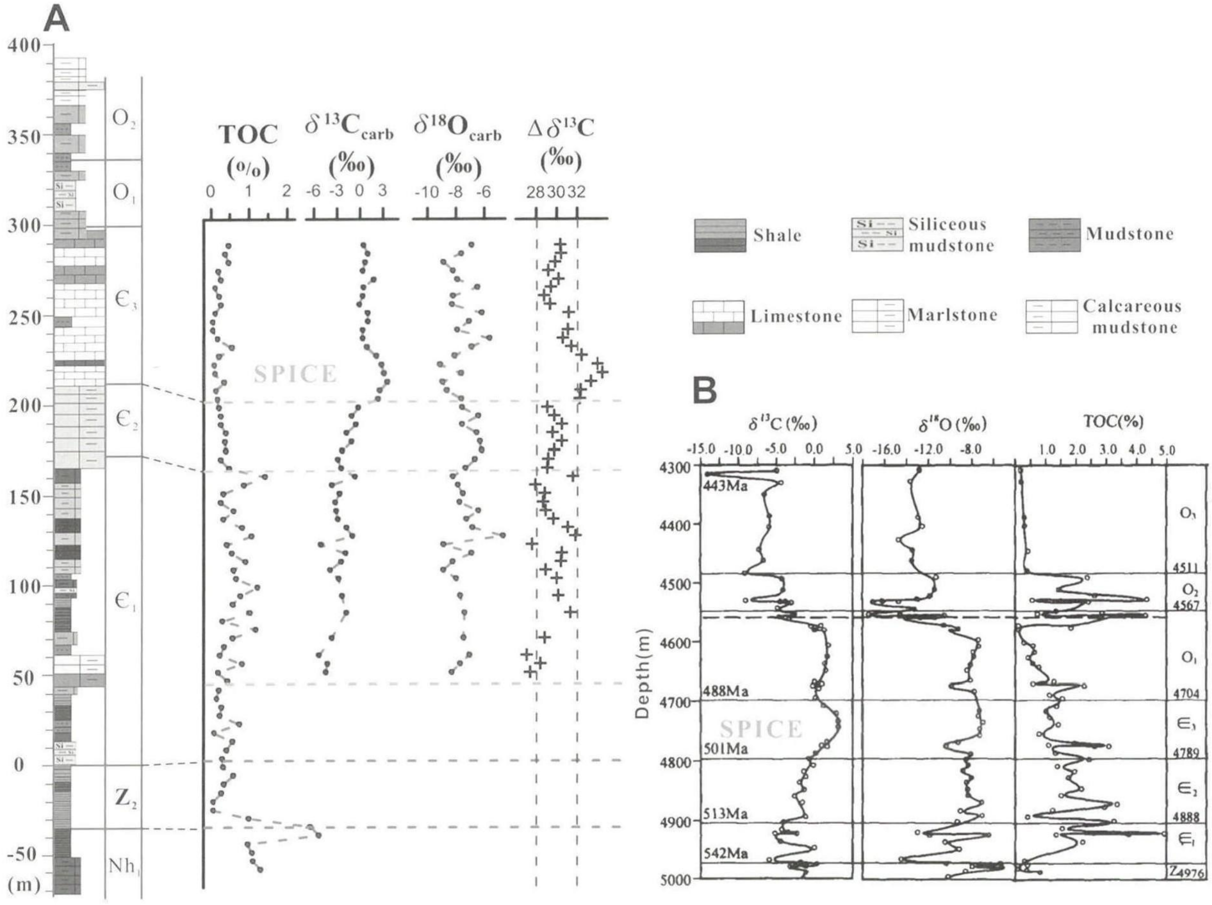

Figure 2. (A) Stable isotope strata curves from the Yaerdang Mountain profile and (B) well TD2∈ (Zhang et al., 2006) in eastern Tarim Basin. Arrow to the right corresponds to the SPICE (the Steptoean positive carbon isotope excursion). Nh_{1a} – Aletonggou Formation, Z_{2S} – Shuiquan Formation, $∈_{1xs}$ – Xishanbulake Formation, $∈_{1xd}$ – Xidashan Formation, $∈_{2m}$ – Moheershan Formation, $∈_{3t}$ – Tuershakequn Formation.

been discussed by Zhang et al. (2006). The positive excursion of carbon and oxygen isotopes at the boundary of upper Sinian to lower Cambrian strata implies a cold climate and declining sea level (Fig. 2). Subsequently, the negative excursion of stable carbon and oxygen isotopes indicate a warming climate and rising sea level, which corresponds to the highest TOC abundance of the core column.

It's worth noting that the SPICE (the Steptoean positive carbon isotope excursion) event corresponding to transgression event as well as extinction of trilobites (Glumac and Walker, 1998; Buggisch et al., 2003; Zhu et al., 2004; Lindsay et al., 2005; Kouchinsky et al., 2008; Fan et al., 2011) was recorded by the stratigraphic curves of carbonate $\delta^{13}C$ from the Yaerdang profile. And from Fig. 2B we can also see that the SPICE event has been recorded in the TD2 core samples, which implies the two profiles are comparable.

3.2. $\delta^{13}C_{ker}$ distributions along the Yaerdang Mountain profile and their constraint factors

3.2.1. $\delta^{13}C_{ker}$ distributions along the profile

The correlations among TOC, carbonate stable carbon/oxygen isotope strata curves and the depositional background from well TD2∊ have been studied by Zhang et al. (2006). Nevertheless, response relationship from the organic carbon stable isotope and the constraints of the isotopic fractionation between organics and inorganics was not included.

The $\delta^{13}C_{ker}$ distributions along the Yaerdang Mountain profile obtained from this study showed a fairly wide range from −34.6‰ to −28.4‰ within the Cambrian strata (Fig. 3). As shown in Fig. 3, $\delta^{13}C_{ker}$ values from well TD2∊ were generally heavier than those from the Yaerdang Mountain profile by value around 3‰. TD2∊ might have experienced thermal alteration under high temperature more than 200 °C. It is also possible that samples from TD2∊ have been formed from the residual solid bitumens instead of the original kerogens (Zhang et al., 2006). However, from early Cambrian to late Cambrian strata, the $\delta^{13}C_{ker}$ from both sedimentary facies shows consistent trend of ^{13}C enrichment (Fig. 3).

3.2.2. Correlations between the stable isotope results of organics/inorganics and their constrained factors

The correlation analysis between the stable carbon/oxygen isotope distributions and organics/inorganics is important to probe their constrained factors (Knauth and Kennedy, 2009; Jiang et al., 2012). According to Rothman et al. (2003), there is a good linear relationship between $\delta^{13}C_{carb}/\delta^{13}C_{org}$ and $\Delta\delta^{13}C$ ($\delta^{13}C_{carb}-\delta^{13}C_{org}$) in geological history, and the slope and y-axis intercept in $\delta^{13}C_{carb}-\Delta\delta^{13}C$ plot are geologic time dependent. Data points from the Xidashan Formation (∊$_{1xd}$) have a slope of 0.84 and a y-intercept of −27.65‰ (Fig. 4A), while those from the Tuershakequn Formation (∊$_{3t}$) show a slope of 0.59 and a y-intercept of −16.95‰ (Fig. 4C). The former are typical of late Neoproterozoic carbonates (Rothman et al., 2003), which are

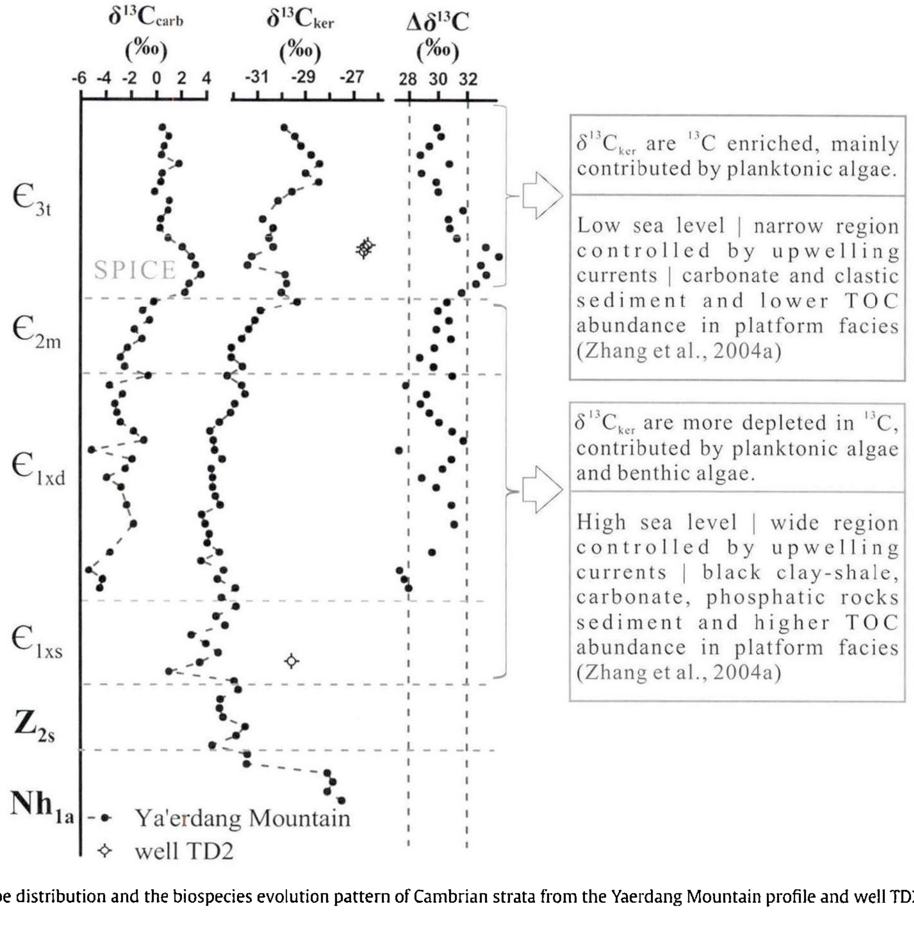

Figure 3. Stable carbon isotope distribution and the biospecies evolution pattern of Cambrian strata from the Yaerdang Mountain profile and well TD2∊ ($\Delta\delta^{13}C = \delta^{13}C_{carb} - \delta^{13}C_{org}$).

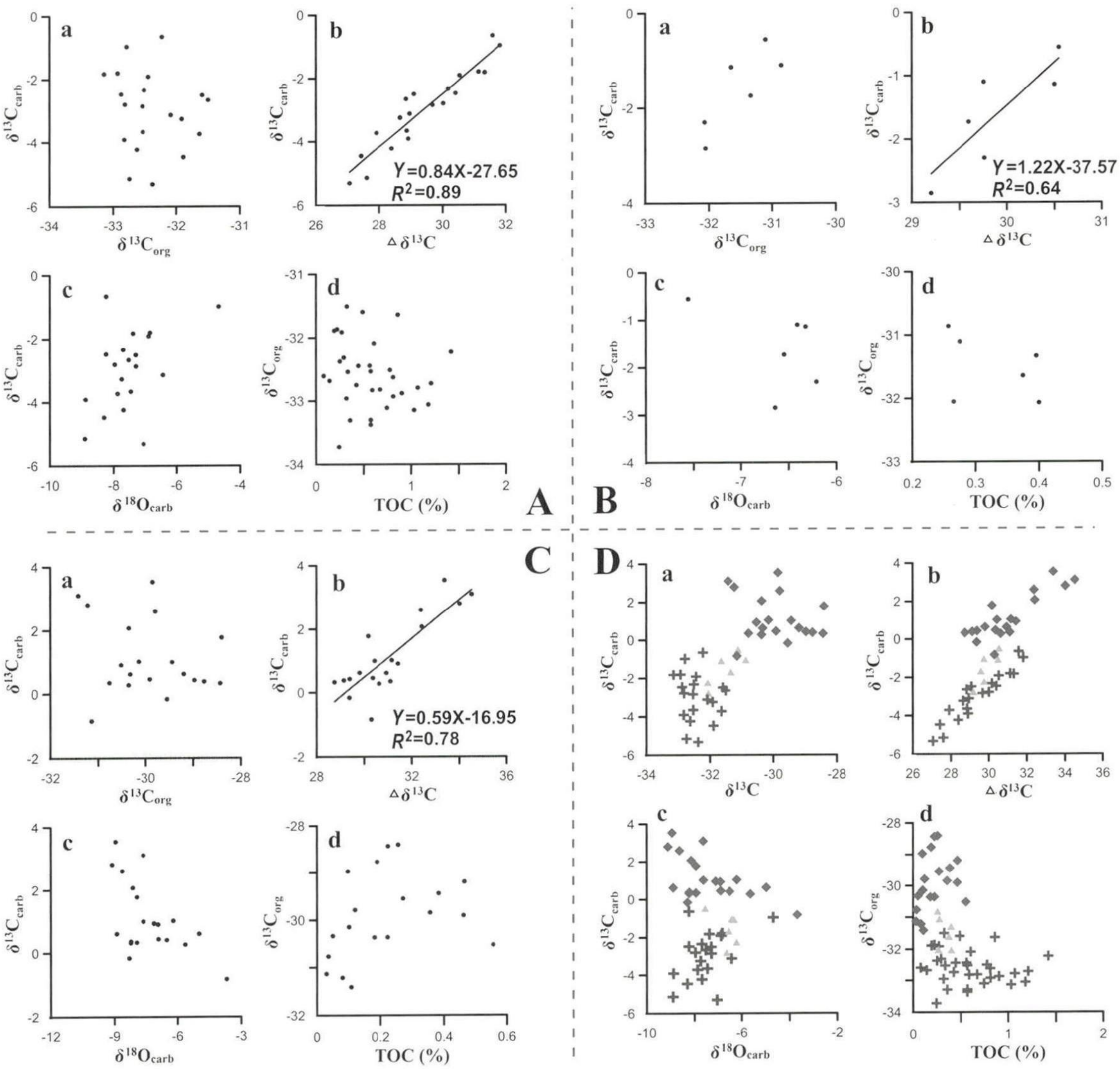

Figure 4. Cross plots of stable carbon and oxygen isotopes and TOC results from the Yaerdang Mountain profile in Tarim Basin ($\Delta\delta^{13}C = \delta^{13}C_{carb} - \delta^{13}C_{org}$). (A) Xidashan Formation ($\in_{1xd}$), (B) Moheershan Formation ($\in_{2m}$), (C) Tuershakequn Formation ($\in_{3t}$), (D) comprehensive cross plots of isotope data. (a) $\delta^{13}C_{carb}$ versus $\delta^{13}C_{org}$, (b) $\delta^{13}C_{carb}$ versus $\Delta\delta^{13}C$, (c) $\delta^{13}C_{carb}$ versus $\delta^{18}O_{carb}$, (d) $\delta^{13}C_{org}$ versus TOC.

similar to the result from lower Cambrian Yanjiahe to Shuijingtuo formation in the Yangtze Gorges area from the southern China (Jiang et al., 2012). Meanwhile, in the comprehensive cross plots of isotope results as shown in Fig. 4D, the data points of Xidashan Formation ($\in_{1xd}$) and Tuershakequn Formation ($\in_{3t}$) can be grouped into different groups, which suggested that they were constrained by different factors.

During the past 800 Ma, the averaged values of TOC (the average isotopic fractionation between total organic carbon and sedimentary carbonates) can be generally divided into three grades: higher than 32‰, between 28‰ and 32‰, and less than 28‰, and according to Hayes et al. (1999) each grade had different denotative significance. As shown in Fig. 3, $\Delta\delta^{13}C$ (geochemically similar to the parameter TOC) data points from the Xidashan Formation ($\in_{1xd}$) are almost distributed between 28‰ and 32‰, which may be from the maximal fractionation of carbon isotopes by phytoplanktonic producers, while some $\Delta\delta^{13}C$ data points from the Tuershakequn Formation ($\in_{3t}$) are higher than 32‰, which are probably due to the influence from ^{13}C-depleted methane clathrate or input of chemoautotrophic organisms in the ocean (Dickens et al., 1995; Hayes et al., 1999).

3.3. Heterogeneous occurrence of Cambrian kerogens in the Tarim Basin and its geochemical significance

3.3.1. Possible explanations to the varied distribution of Cambrian $\delta^{13}C_{ker}$

There exists a similar distribution feature for the $\delta^{13}C_{ker}$ from Cambrian strata worldwide (Table 3). The primary constraints for

Table 3
$\delta^{13}C_{ker}$ distribution scopes of global Cambrian source rocks.

Area	Strata	Formation	$\delta^{13}C_{ker}$ (‰)	References
Siberian platform	$\in_{1+2}$	Kuonamka	−33 to −27	Parfenova et al., 2010
Oman Basin	$\in_1$	Ara	−37 to −34	Grosjean et al., 2009
Australian	$\in_1$	Ouldburra and Observatory Hill	−32.5 to −26.7	Logan et al., 1997
Sweden	$\in_{2+3}$	Alum	−31 to −27	Buchardt et al., 1986
South China	$\in_1$	Niutitang	−34 to −29	Guo et al., 2011
Tarim Basin	$\in$	All	−36 to −26	Wang, 2012

$\delta^{13}C_{ker}$ of Cambrian source rocks are thermal maturity, weathering fractionation, precursor biospecies and sedimentary lithofacies (Hayes, 1993). These can be classified into the post-depositional evolution due to geochemical transformation after deposition, and the origin-controlling mechanisms resulting from contributions from initial biomass and geological settings. In this study, the $\delta^{13}C_{ker}$ of core drilling samples from well TD2∈ are more ^{13}C-enriched than those of contemporaneous outcrops by around 3‰, which cannot be properly ascribed to their thermal differences (Tang et al., 2000). It is expected that weathering fractionation should result in $\delta^{13}C_{ker}$ from outcrops been heavier than those from core samples. This however, is not true for the present study and therefore suggesting that weathering fractionation might not be an important constraint factor. However, the prepared residual kerogens from TD2 samples might have commingled with the thermally pyrolyzed solid bitumens (Zhang et al., 2004a), which might have resulted in the ^{13}C enrichment of the samples.

$\delta^{13}C_{carb}$ of rock carbonate is more affected by diagenetic evolution than primary constraining factors, while $\delta^{13}C_{ker}$ is controlled more by the latter. According to the report from Zhao and Huang (1995), $\delta^{13}C_{oil}$ derived from humus varied from −27‰ to −25‰, while the $\delta^{13}C_{oil}$ from Sapropel fall in the scope of −35‰ to −29‰. Meanwhile, it has been reported that the oils originated from blue-green algae, diatom or sulfur green algae were enriched in ^{13}C to various degrees (Stanley, 2010; Close et al., 2011). Furthermore, the $\delta^{13}C$ of organic matter was found to be constrained by lithology and depositional environment, as $\delta^{13}C_{org}$ from shallow carbonate sedimentary facies is heavier than −30‰ which was probably controlled by photosynthesis, while those from deep water black shale and dark siliceous sedimentary facies is lighter than −32‰ which was mainly contributed from chemoautotrophic organisms and methanotrophs under anaerobic environment (Wang et al., 2014).

In this work, $\delta^{13}C_{ker}$ along the outcrop profile varied continuously and this is accompanied by a contemporaneous ocean regression and cold event. It seems that the Cambrian $\delta^{13}C_{ker}$ of the eastern Tarim Basin was predominantly constrained by sedimentary background, lithofacies and precursor biomass.

3.3.2. Geochemical implication to the oil-source correlation

Heterogeneous $\delta^{13}C_{ker}$ distribution of Cambrian source rocks of the Tarim Basin has been reported by Wang (2012). The simplified evolution pattern of hydrocarbon-generating alga from the early Cambrian to late Cambrian strata in the eastern Tarim Basin is presented in Fig. 3. During the Cambrian period, there was a drastic change of partial pressure of carbon dioxide in atmosphere ($p(CO_2)$) and world sea level fluctuated widely. It brought about the regional shrinkage of starved basin facies and reduced the living space for planktonic algae, especially for the benthic algae in the eastern Tarim Basin. And thus the TOC abundance, hydrocarbon-generating alga, $\delta^{13}C_{ker}$ and their constraints were heterogeneously developed among the Cambrian source rocks.

Currently, the Cambrian–lower Ordovician strata and the middle–upper Ordovician strata are generally believed to be the main locations of high quality marine hydrocarbon source rocks in Tarim Basin (Liang et al., 2000; Zhang et al., 2000). The ^{13}C-enriched crude oils of TD2∈ and TZ62S have been used as the end member from the Cambrian–lower Ordovician source rocks and have been used to quantitatively assess the mixed marine oil reservoirs that occur extensively in the Tarim Basin (Li et al., 2010; Tian et al., 2012). However, the Cambrian source rocks in the eastern Tarim Basin were found to have been heterogeneously developed. Therefore, the use of most ^{13}C-enriched marine crude oils as the representatives of oils generated from the Cambrian–lower Ordovician source rocks to evaluate the marine oil reservoirs throughout the whole Tarim Basin could be misleading. This is because the Cambrian source rocks were formed from mixed origin. Also, the contribution from the middle–lower Cambrian source rocks with much higher TOC abundance cannot be neglected. The results from the present study showed that the different organic facies of the source rocks should be taken into consideration in the oil-source rock correlation study of the basin.

3.3.3. Enlightenment to the diagnostic explanations concerning the ^{13}C-enriched crude oils such as TD2∈ and TZ62S in Tarim Basin

It has been recently found that crude oils typically from the Cambrian–lower Ordovician source rocks are ^{13}C-enriched, such as crude oils of TD2∈ and TZ62S with their bulk stable carbon isotopic ratios approaching −28‰ (Zhang et al., 2004a; Xiao et al., 2005; Tian et al., 2012), which were much heavier than the corresponding kerogens and the crude oils from the middle–upper Ordovician source rocks. Zhang et al. (2006) considered that the thermal cracking was the dominant diagnostic factor for the ^{13}C-enriched in the heavy crude oil of TD2∈, as the reservoir oil had experienced serious thermal alteration under high temperature over 200 °C. Nevertheless, thermal alteration cannot explain the stable carbon isotope characteristics of ^{13}C-enriched crude oil of TZ62S or gas condensate of YN2, as they were not subjected to serious thermal alteration as TD2∈. As aforementioned, there is a correlation between limestone/mudstone, total organic carbon abundance, hydrocarbon-generating precursor biospecies, and stable isotope ratios of organics and carbonate ($\delta^{13}C_{ker}$, $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$) and fluctuations of $p(CO_2)$ and sea level. This is to say that the transformation of hydrocarbon-generating precursor biospecies was mainly controlled by depositional environment, such as sea level and oxygen content in the water. As a consequence and from the present study, we cannot rule out the possibility that the ^{13}C-enriched crude oils were derived from some localized Cambrian source rocks.

4. Conclusions

TOC and stable isotope distributions of the organics/inorganics of the isochronous deposition in the Yaerdang Mountain profile and well TD2∈ core rocks in the eastern Tarim Basin have been compared in the present study. Shallow shelf carbonate facies was the dominant feature of the Yaerdang Mountain profile, while the starved basin facies characterized well TD2∈. The results revealed that the $\delta^{13}C_{ker}$ (VPDB) of Cambrian kerogens along the outcrop section studied in this work varied from −34.6‰ to −28.4‰, indicating an increasing tendency from the lower Cambrian to the upper Cambrian. This was on the whole accompanied by the variation in $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ along the profile, which might be due to the changing in the sea level and also in the compositional variation in benthic and planktonic biomass. The huge variation in the stable carbon isotope ratios up to 6‰ along the outcrop section in the present work suggests remarkable heterogeneity of the Cambrian source rocks from eastern Tarim Basin.

Therefore, the ^{13}C-enriched in the crude oils from TD2∈ and TZ62S suggest oils derived from localized Cambrian source rocks. It is therefore questionable to take the ^{13}C-enriched crude oils of TD2∈ and TZ62S as representatives of all the marine crude oils from the Cambrian–lower Ordovician source rocks in the Tarim Basin.

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