

Comprehensive diagnostic review of the ^{13}C -enriched crude oils exemplified by TD2C and TZ62S in Tarim Basin, NW China

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Abstract The crude oils typically from the Cambrian-Lower Ordovician source rocks of Tarim Basin, NW China, such as TD2C and TZ62S, are ^{13}C -enriched with the stable carbon isotopic ratios (VPDB) approaching -28‰ . In this paper, the main research viewpoints on this issue are summarized, and combined with results from organic and inorganic carbon isotope stratum curves of the outcrop at the Ya'erdang Mountain in Tarim Basin. In addition, more alternative interpretations are discussed. On one hand, the inverse fractionation features of stable carbon and hydrogen isotopes of these crude oils may imply their protogenous nature. On the other hand, the anisotropy of source rocks and contribution from older stratum source rocks need verifying as well. For the sake of the final resolution of this issue, some further study topics are recommended.

Keywords Carbon isotope · Coevolution · Tarim Basin · Organic geochemistry

1 Introduction

Hayes et al. (1999) summarized the factors influencing the stable carbon isotopic fractionation and coevolution features among the crude oil, kerogen and carbonate in the petroleum system. There have been abundant achievements made on the unusual distributions of stable carbon isotope ratios over geologic time, and their geochemical significances have been explored (Zhang et al. 1987, 2006a, b, 2008; Close et al. 2011). However, controversies still existed over the diagnostic explanations of ^{13}C -enriched crude oils in Tarim Basin, NW China. The question remained whether the fact that crude oils from Cambrian-Lower Ordovician source rocks enriched in ^{13}C was representative in Tarim Basin needed clarification, or whether they were just sourced from some specific geologic background or some particular precursors.

Tarim Basin is a large-scale overlapped and composite basin that has experienced a complex deposition history from marine carbonate and clastic rocks to paralic clastic rocks and carbonate, finally to continental clastic rocks (Zhang et al. 2011). The Tarim craton was active in the early paleozoic, but became relatively stable later on. This character has controlled the development of two sets of high quality marine hydrocarbon source rocks, the Cambrian-Lower Ordovician and the Mid-Upper Ordovician source rocks.

In recent years, it has been found that oils typically from the Cambrian-Lower Ordovician source rocks are ^{13}C -enriched, such as crude oils of TD2C and TZ62S which have bulk stable carbon isotopic ratios approaching -28‰ (Table 1). These are much heavier than the corresponding kerogen and oils from the mid-upper Ordovician source rocks. This isotope ratio has been used as an end member value to quantitatively assess the mixed

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Table 1 $\delta^{13}\text{C}$ values of crude oils and their kerogens typically from Cambrian-Lower Ordovician source rocks in Tarim Basin

Well	Reservoir	$\delta^{13}\text{C}_{\text{Ker}}$ (‰)	$\delta^{13}\text{C}_{\text{Crude oil}}$ (‰)
TD2	Є-O ₁	~−31	−28.2 ^a
TZ62	S	~−32	−28.6 ^b
YN2	J	~−32	−27.7 ~ −27.1 ^a
T904	O	–	−29.6 ^c
TS1	Є	–	~−28 ^c

^a Zhang et al. (2004)^b Tian et al. (2012)^c Zhai et al. (2007)

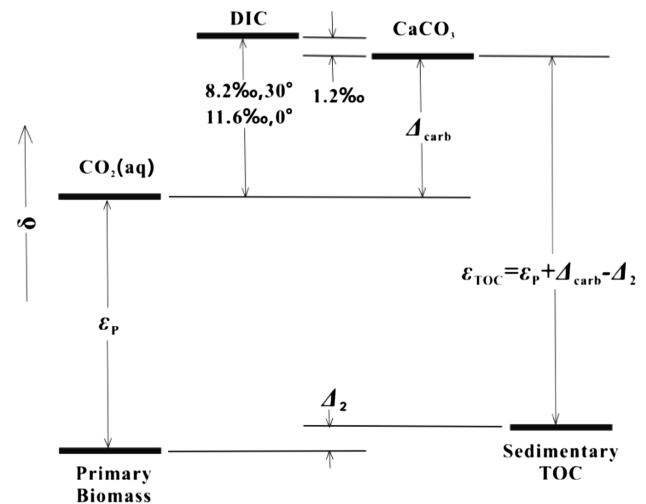
marine oil reservoirs in Tarim Basin (Li et al. 2010) (Table 1).

However, it is important to verify the representative of the ^{13}C -enriched isotope ratio of TD2Є and TZ62S for the general crude oils sourced from the Cambrian-Lower Ordovician strata. In addition, the mechanisms for the reverse stable carbon isotope distribution patterns between oils and kerogens should be explored. This kind study is especially important for the further exploration of the marine oil reservoirs in the Tarim Basin, where the source rocks are still not clearly constrained.

2 Research background

2.1 Scientific constraint factors

The stable carbon isotope compositions of kerogens ($\delta^{13}\text{C}_{\text{Ker}}$) over geologic time are generally consistent, and they document a trend of ^{13}C enrichment with decreasing age. The $\delta^{13}\text{C}$ values of crude oils have a similar trend (Andrusevich et al. 1998). Yet the carbon isotope compositions of marine carbonate and organics vary throughout a certain specific period. This process is usually accompanied by the coevolution of organics and inorganics (mainly sedimentary carbonate) in carbon pools (Hayes et al. 1999; Kump and Arthur 1999). Therefore the ^{13}C enrichment of crude oils is attributed to the coevolution of local or global organic and inorganic carbon at the macro level, while at the micro level it is restrained by organic facies, post-deposition thermodynamic activity, bio-degradation, etc. In order to probe the stable carbon isotopic excursions, Hayes et al. (1999) systematically analyzed more than 4000 stable carbon isotope ratio data (including $\delta^{13}\text{C}_{\text{Carb}}$ and $\delta^{13}\text{C}_{\text{Org}}$) from different parts in the world and the past 800 Ma. The relationships between stable carbon isotopes and all important constraint factors are shown in Fig. 1. The inorganic carbon isotope fractionations between CO_2 (aq) and DIC (dissolved inorganic carbon) and DIC and

**Fig. 1** Schematic summary of isotopic relationships between dissolved CO_2 and carbonate minerals, and between primary products and sedimentary TOC (Hayes et al. 1999)

carbonate are included. The organic carbon isotope fractionation between CO_2 (aq) and primary biomass and sedimentary TOC and primary biomass are also considered. Hence, the CO_2 content in the atmosphere and the marine, temperature, primary biomass type and alteration of post-deposition are all closely related to the stable carbon isotope distributions in organic and inorganic carbon.

The reasons for the global carbon isotope excursions are complex, as there are different factors co-controlling the stable carbon isotope distribution in different regions. However, the diagnostic explanations of the unusual stable carbon isotope distribution can be summarized into the protogenetic and the post-depositional evolution mechanisms (Knauth and Kennedy 2009; Derry 2010; Swanson-Hysell et al. 2010; Grotzinger et al. 2011; Sansjofre et al. 2011). The former is known as the main contribution from the biogeochemistry characteristics of primary biomass, and the latter is ascribed to the alteration of various geochemical processes after the deposition stage.

2.2 Unusual organic stable carbon isotope distributions from other areas

The reversed carbon isotope patterns between the soluble components and kerogens were discovered in the lower Paleozoic black shale of South China by Zhang et al. (Fig. 2A). The authors considered this to be primarily connected with the original source material, but could not exclude the thermal evolution effect. The entire or partly reversed stable carbon isotope distribution patterns among different groups of crude oils or soluble organics of source rocks are also found in the Tarim Basin (Fig. 2B, C). The authors proposed that the reversed carbon isotope patterns

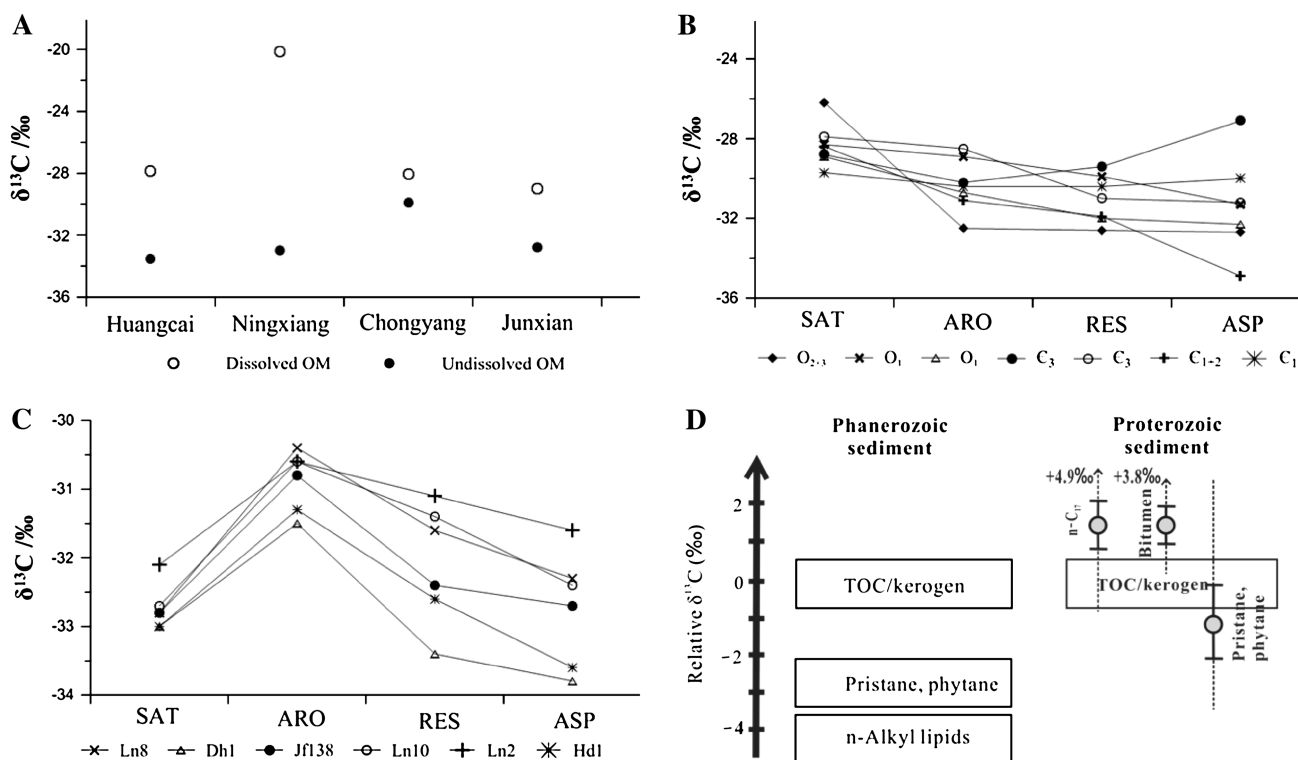


Fig. 2 Unusual organic stable carbon isotope distributions from other areas

among groups of crude oil or soluble organics indicated the post-depositional alteration of deep source rocks (Zhang et al. 2006a, b, 2008), while the partly reversed pattern between asphaltene and other crude oil groups may have suggested micro-biological degradation or oil mixing (Wang et al. 2005; Mi et al. 2006). In addition, the inverse carbon isotope distribution patterns of lipids and kerogen were widely discovered in Proterozoic sedimentary organics (Fig. 2D), which were proposed to have resulted from the heterogeneous primary biomass and the selective preservation of ^{13}C -enriched lipids from prokaryotes (Close et al. 2011).

A—inverse carbon isotope pattern between dissolved organic matter (OM) and undissolved OM from South China, modified after Zhang et al. (1987). B—Carbon isotopic composition of soluble organic components (SAT, ARO, RES and ASP) of the Cambrian and Ordovician deep source rocks in Tarim Basin (Zhang et al. 2006a, b). C—Carbon isotopic compositions of crude oils and its group components of the deep marine oil reservoirs in Tabei Uplift (Zhang et al. 2008). D—Comparison on stable carbon isotope patterns from Phanerozoic sediment and Proterozoic sedimentary (Close et al. 2011). Values show average and standard deviation (1σ), and the total range of individual values is shown with dotted lines.

The inverse carbon isotope distribution patterns between the soluble components and kerogen from South China lower Paleozoic black shale, and the inverse distribution

patterns of Proterozoic lipids and kerogen are found worldwide. The former emphasized the characteristics of the marine sapropelic substance, which was also found in the Triassic and Tertiary strata (Wang 2000), and the latter emphasized the main contribution of primary prokaryote biomass.

It is worth investigating whether the ^{13}C enriched oils of TD2C and TZ62S in Tarim Basin are comparable with the inverse carbon isotope distribution patterns between Neoproterozoic lipids and kerogen. Their possible relevance with the reversed carbon isotope distribution patterns of lower palaeozoic black shale in South China should be probed as well.

3 Research viewpoints on the ^{13}C -enriched oils of TD2C and TZ62S

3.1 Thermo-genetic explanation

Compared with the study on the unusual organic carbon isotope distributions and their geochemistry significances in other geologic times and regions, there was not yet a general consensus about ^{13}C -enriched oils of TD2C and TZ62S. Zhang et al. (2004) considered that thermal cracking was the dominant diagnostic factor for the ^{13}C -enriched TD2C, as this oil had experienced high temperature thermal alteration of more than 200 °C. Nevertheless,

thermal alteration cannot explain the development of ^{13}C -enriched Oil TZ62S or gas condensate YN2, as the former is the normal black oil, which has good mobility (Xiao et al. 2005), and the latter, is crude oil whose light hydrocarbon component should be ^{12}C -enriched under thermal alteration. In fact the bulk stable carbon isotope ratio of condensate oil YN2 is $-27.7 \sim -27.1 \text{ ‰}$, much heavier than expected.

3.2 Protogenous mechanisms

With respect to the inverse carbon isotope distribution pattern of lipids and kerogen in Proterozoic sedimentary organics (Close et al. 2011), possible protogenous diagnostic mechanisms of ^{13}C -enriched Oils in Tarim Basin should be probed. Blue-green algae, diatom and sulfur green algae as the primary biomass that could make crude oils enrich in ^{13}C to various degrees (Stanley 2010; Close et al. 2011). Associated with the diverse tectonic geologic background in Tarim Basin, the thriving of some of these kinds of primary biomass would result in the ^{13}C -enriched crude oils occurring in some specific environment and preserved over the geologic evolution.

3.3 Constrained by multiple factors

Tarim Basin is a large-scale overlapped and composite basin. Oil and gas reservoir formation and distribution are closely related with multi-period tectonic movements of the basin, which is associated with the multi-period generation, migration, gathering, destroying and adjusting of hydrocarbons (Wang and Xiao 2004). These background processes complicated the related work and brought a lot of difficulties, especially to the work of oil-source correlation.

A single diagnostic factor cannot reasonably explain the ^{13}C -enriched oils such as TD2C and TZ62S in the Tarim Basin. Although the protogenous mechanisms are more effective in explaining the high degree inverse carbon isotope pattern between the bulk crude oils and kerogens, there still needs to be more study on the geochemical behavior of some special primary biomass first. Considering the complicated and long geologic evolution history, comprehensive factors are probably more effective and reasonable.

As it is widely known, CO_2 in the atmosphere and marine pools both finally entered into sediment through inorganic and organic circulation paths, with the isotope fractionation affected by the physical, chemical and biological environments. As shown in Fig. 1, the isotope ratio of dissolved inorganic carbon (DIC) in the ocean is sensitive to the temperature and partial pressure of carbon dioxide in the atmosphere (Pco_2). So the paleo-atmospheric temperature and Pco_2 have a crucial influence on the

isotopic equilibrium between DIC and carbonate. At the same time the sedimentary facies controlling the petrologic and biologic character can be used to assess the differences of physical, chemical and biotic environments along with sedimentation occurrence. In fact, research has shown that during the late Ediacaran and early Cambrian in Yangtze platform, $\delta^{13}\text{C}_{\text{Org}}$ values from shale- and chert-dominated basinal sections are 8–10 ‰ lower than those from the carbonate-dominated shelf sections (Jiang et al. 2011). Palaeo-latitude, palaeo- Pco_2 and sedimentary facies affect the carbon isotope fractionation of organics and inorganics to a great extent. Therefore, besides the post-depositional alteration occurring in the oil reservoirs or source rocks, the thriving of some special primary biomass restricted to the shelf in the Tarim Basin may be an important reason for the ^{13}C -enrichment of crude oils such as TD2C and TZ62S.

4 Enlightenment from the recent work

4.1 Heterogeneity of source rocks

As stated in Sect. 2.1, the unusual isotopic distribution can be usually attributed to the coevolution of local or global organic and inorganic carbon. Thus the isotopic stratum curves were studied from the Ya'erdang Mountain in northeastern Tarim Basin, NW China. As shown in Fig. 3, Cambrian source rocks are heterogeneous in relation to geological time. The stable carbon isotope compositions of kerogens ($\delta^{13}\text{C}_{\text{Ker}}$) and carbonate ($\delta^{13}\text{C}_{\text{Carb}}$) are generally consistent and follow a trend of ^{13}C enrichment with decreasing age nearly over the entire Cambrian. In addition, some different correlation relationships among $\delta^{13}\text{C}_{\text{Ker}}$, $\delta^{13}\text{C}_{\text{Carb}}$ and $\delta^{18}\text{O}_{\text{Carb}}$ were shown in different stages. It can be interpreted as a different primary factor leading to diversity of source rocks. In addition, the $\delta^{13}\text{C}_{\text{Ker}}$ of Aletonggou and Tuershakequn formations are ^{13}C -enriched along the profile, with a $\delta^{13}\text{C}_{\text{Ker}}$ ratio around -29.05 , -29.52 ‰ respectively (Fig. 3). This suggests that the contribution from this older source rocks is also worth concerning.

Furthermore, concerning the development of the exploration work on lower Palaeozoic source rocks in Tarim Basin, more and more researchers are tending to suggest that the lower Palaeozoic source rocks in Tarim Basin deposit in multiple environments. Consequently, the spatio-temporal heterogeneity of the lower Palaeozoic source rocks is probably another crucial factor for this issue. Combined with the heterogeneity of source rocks, the comprehensive diagnostic analyses including protogenous mechanisms and the thermal evolution explanation will finally resolve this issue.

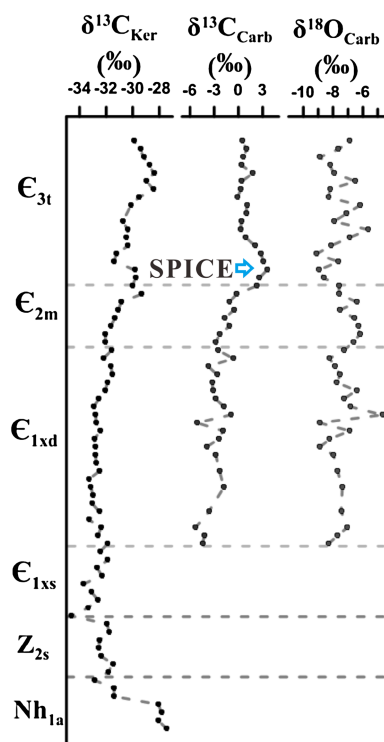


Fig. 3 Isotope stratum curves from the Ya'erdang Mountain in northeastern Tarim Basin, NW China. Nh_{1a}—Aletonggou formation, Z_{2s}—Shuiquan formation, E_{1xs}—Xishanbulake formation, E_{1xd}—Xidashan formation, E_{2m}—Moheershan formation, E_{3t}—Tuer-shankekun formation

4.2 Results from the stable hydrogen isotope analysis

Jia et al. (2013) found that crude oils of TD2C and TZ62S had relatively light δD values of both bulk oils and their *n*-alkanes, ranging from -145 to -142 ‰. These were distinctly different from the other crude oils from Tarim Basin. As the stable hydrogen isotope results can be used to elucidate the depositional environment of source rocks (Schimmelmann et al. 2004; Dawson et al. 2005; Li et al. 2011), a relatively light hydrogen isotope ratio may imply that the hydrogen isotopic fractionation level of palaeo-gulf waters corresponds with the contemporaneous cold event. It seems that the distinctly unusual inverse fractionation of stable carbon and hydrogen isotope of oils TD2C and TZ62S may suggest their protogenous nature of isotope composition signature.

4.3 Correlation between palaeoenvironment and the stable carbon/oxygen isotope ratios

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 eastern Manjiaer depression in the Tarim Basin had experienced a sea level change cycle from the Early Cambrian sea level rising to the Middle-Late Cambrian falling (Zhang et al. 2006a, b). The cycle corresponds to the varying in the inorganic and organic carbons isotope stratum curves of the outcrop at the Ya'erdang Mountain in Tarim Basin (Fig. 3). When the sea level rises in the early Cambrian, the ^{13}C -poor dissolved carbon dioxide from the deep Ocean was taken to the platform area. As a result, the depositional inorganic and organic carbons on the platform were ^{12}C -rich. When the sea level fell in the mid-late Cambrian, the depositional inorganic and organic carbons on the platform began to enrich ^{13}C . Besides, as Fig. 3 shows, the curve of $\delta^{13}C_{Carb}$ also records a Redlichiiid-Olenellid extinction carbon isotope excursion ("SPICE") at the beginning of late Cambrian (Zhu et al. 2004, 2006; Shabanov et al. 2008; Guo et al. 2010; Glumac and Walker 1998; Saltzman et al. 2000; Fan et al. 2011). The "SPICE", which is comparable worldwide, also corresponds to the "geological clod event" (Stanley 2010). As stated in 4.2, the crude oils represented by TD2C and TZ62S had relatively light δD values of both bulk oils and their *n*-alkanes, which inferred that they were connected with the contemporaneous cold event. Therefore, some special primary biomass controlled by regression, the cold event and the restricted platform are probably the fundamental reason why the crude oil represented by TD2C and TZ62S is ^{13}C -riched.

5 Summary and future work

Tarim Basin has a very complex depositional and tectonic geological background, which brings many difficulties when discussing the diagnostic factors of ^{13}C -enriched oils TD2C and TZ62S. Based on previous research work, this paper discusses multiple factors of stable carbon isotopic fractionation and coevolution among crude oil, kerogen and carbonate in the petroleum system. Palaeoclimate and palaeoenvironment lead to heterogenous distribution of Cambrian source rocks and the thriving of some special primary biomass in the Eastern Tarim Basin, NW China. Some of the upper Cambrian source rocks are more likely the source of ^{13}C -enriched oils such as TD2C and TZ62S. However, more studies are needed from the fields of petrology, organic geochemistry and biogeochemistry. Also some summary and further work concerning the following are needed:

- (1) Single genetic factor cannot explain well the diagnosis of ^{13}C -enriched oils such as TD2C and TZ62S. Multiple factors including protogenous mechanisms and thermal evolution would be effective.

- (2) Thriving of some special primary biomass restricted to the shelf in Tarim gulf is probably an important factor for the development of TD2C and TZ62S ¹³C-enriched crude oils.
- (3) The spatio-temporal heterogeneity of the lower Palaeozoic source rocks in Tarim Basin is probably another crucial factor for this issue, and the contribution from older source rocks needs concerning, too.
- (4) Relatively light hydrogen isotope ratios of the crude oils may imply their origin of palaeo-gulf water with the contemporaneous cold event. Thus, palaeoclimate research work will be helpful in understanding the unusual carbon and hydrogen isotope fractionation of oils TD2C and TZ62S.

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