# STUDY OF CRUDE OIL-SOURCE ROCKS CORRELATION IN THE PALEO-OIL RESERVOIRS OF THE SOUTHERN QIANGTANG DEPRESSION, CHINA

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Abstract. The Oinghai-Tibet Plateau is located in the east section of the Tethys region, which produces great amounts of oil and gas whose reserves are one of the most abundant in the world. The plateau includes two exploration areas: a Mesozoic marine basin and a Cenozoic continental facies. At present, the Qiangtang Basin in the Qinghai-Tibet Plateau is believed to be the marine basin with the most extensive oil and gas exploration prospects in the world; however, the thrust-nappe structure movement in the Cretaceous-Cenozoic Period caused massive surface exposure which damaged the oil reservoirs. In this paper, the paleo-oil reservoir exposure area was used as a window to reveal the massively exposed underground distribution of oil and gas reservoirs in the Southern Qiangtang Depression, and the reservoirs were analyzed in detail. The composition of biomarkers and the characteristics of carbon isotopes of crude oil in the reservoirs were identified via comparison of oil source rocks. Our findings indicated that the oil shale series source rock from the Biluocuo region of the Qiangtang Basin represents the main source rocks of the paleo-oil reservoirs.

*Keywords:* crude oil-source rock correlation, paleo-oil reservoir, Qinghai-Tibet Plateau, Southern Qiangtang Depression.

# 1. Introduction

The Qinghai-Tibet Plateau is the final frontier for land petroleum exploration in China and is located in the middle of the Tethys region, which produces the greatest amount of oil and gas whose reserves are one of the most

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abundant in the world. To the west there are the Persian Gulf oil-producing areas in the Middle East, and to the east there are the petroliferous basins of Southeast Asia. The Qinghai-Tibet Plateau has an area of 2,400,000 km<sup>2</sup> and accounts for approximately a quarter of China's land area. Four residual marine basins are distributed in the area – the Qiangtang Basin, the Selincuo-Biru Basin, the Changdu Basin and the Dingri-Gangba Basin, and more than 20 continental basin groups are represented by the Lunpola Basin, which forms the Mesozoic marine and Cenozoic continental exploration fields. In the marine field, the area of the Qiangtang Basin is the largest, and at present, it is considered one of the most promising oil-gas exploration basins in China.

Currently, approximately 200 oil shows have been observed in the Qiangtang Basin, which are mainly distributed from the Permian Rejuechaka Formation to the Pliocene Suonahu Formation. Within this area, the oil shows are chiefly dry bitumen. The liquid oil shows are mostly distributed in the paleo-oil reservoirs of the Buqu Formation in the Southern Qiangtang Depression [1–4]. However, these oil reservoirs were exposed to the surface via a large-scale movement of the thrust-nappe structure during the Cretaceous-Cenozoic Period [5–11]. Therefore, many scholars believe that the exploration potential of the Qiangtang Basin is poor. Nevertheless, based on the exploration history of oil and gas fields in China and abroad, the exposed oil springs and sands are the most direct evidence for underground reservoirs [12], and China's petroleum exploration practices offer further proof. For example, the discovery of dry oil springs on the surface provided an important clue for the first oilfield, the Yumen Oilfield, and represented a breakthrough in exploration. A large number of oil seeps can be found on the surface near Heivoushan in the Junggar Basin, and the Karamay Oilfield was discovered via exploration. Additionally, the exploration mode for Youshashan in the Qaidam Basin provides a reference because it has oilbearing strata exposed on the surface, and the Gasikule Oilfield was discovered on the footwall of a fault [13, 14]. Hence, paleo-oil reservoir exposure areas should be regarded as important because they provide a window that reveals the distribution of underground oil and gas reservoirs.

These massively exposed paleo-oil reservoirs in the Southern Qiangtang Depression were used as key objects in this study, and a detailed analysis of the composition of biomarkers and characteristics of carbon isotopes of crude oil was performed. By establishing a correlation between crude oil and source rock, in this work, the position of effective source rocks in the basin, the hydrocarbon-generating depression, and the proposed favorable sourcereservoir-caprock assemblage was identified, as well as the distribution and preservation conditions of favorable reservoirs, and the predicted areas and directions for oil and gas exploration were determined.

# 2. Geological background

The Qiangtang Basin is the largest Mesozoic marine residual basin on the Qinghai-Tibet Plateau, it is located on the northern Tibetan Plateau between the Hoh Xil Basin-Bayan Har Plateau and the Himalayas-Gandese Plate and covers approximately  $18 \times 10^4$  km<sup>2</sup>. The Qiangtang Basin is also the largest marine basin of the Tibetan Plateau. It appears to have an east-west trend and a rhombic shape and is divided into the Southern and the Northern Qiangtang Depression by a central uplift belt (Fig. 1). The crystalline basement in the Qiangtang Basin is a pre-Devonian system belonging to Qiangtang-Changdu strata. The Mesozoic strata exposed on the surface are mainly from the Triassic Tumengela Formation, and the Jurassic Quse, Sewa, Shaqiaomu, Buqu, Xiali and Suowa formations, however, a small amount comes from the Cretaceous Abushan Formation.

The Qiangtang Basin features good geological conditions for petroleum. The spatial-temporal relationship between the source rock, reservoir and caprock configuration indicates that the basin can be classified into four sets of Class I assemblages. For the Paleozoic assemblage, the few surface exposures and outcrops at the basin edges show that the assemblage source rocks are of high maturity and from the pyrolysis period. For the Cenozoic assemblage, the source rocks of the Niubao and Dingqinghu formations mainly developed continental basins on the Qinghai-Tibet Plateau. The Qiangtang Basin mostly contains red beds from that period, and the source rocks are undeveloped. Therefore, the Mesozoic Triassic and Jurassic assemblages have the greatest prospects for oil and gas exploration.



Fig. 1. Tectonic division of the Qiangtang Basin and geographical location of the research area.

## 3. Methods

33 samples were collected from the Tumengela, Quse, Sewa, Shaqiaomu, Buqu, Xiali and Suowa formations and analyzed to determine their organic geochemistry. Additionally, 30 oil samples were collected from the paleo-oil reservoir of the Buqu Formation and were subjected to organic geochemistry analysis (Table). All samples were first washed thoroughly with dichloromethane (DCM) to remove possible surface contamination and then ground and passed through a <200 mesh. All experiments were conducted at the Research Institute of Exploration and Development, PetroChina Huabei Oil Field, China.

Each sample was reacted with DCM for 72 h using a Soxhlet apparatus. The solvent was removed using rotary evaporation, and the residue was dissolved in cyclohexane. The extracts were weighed and fractionated. Asphaltenes were removed by centrifugal precipitation with chilled n-hexane (at least  $40 \times v/v$ ), and the remainder (maltenes) was separated into three

Lithology	Sample	Section	Series	Sample	Section	Series	Sample	Section	Series
	RP0-13S1	DBL	$J_{3}s$	MP-05S1	DBL	$J_2b$	BL-1	BLC	$J_1q$
	RP0-5S1	DBL	$J_3s$	GP055S3	GBXM	$J_2s$	BL-2	BLC	$J_1q$
Source rock	RP-4S1	DBL	$J_{3}s$	GP50S1	GBXM	$J_2s$	BL-3	BLC	$J_1q$
	RP0-1S1	DBL	$J_3s$	GP32S1	GBXM	$J_2s$	BL-4	BLC	$J_1q$
	DP20S1	DBL	$J_2 x$	GP28S1	GBXM	$J_2 sq$	BL-5	BLC	$J_1q$
	DP14S2	DBL	$J_2 x$	GP20S1	GBXM	$J_2 sq$	BL-6	BLC	$J_1q$
	DP012S1	DBL	$J_2 x$	GP08S1	GBXM	$J_2 sq$	BL-7	BLC	$J_1q$
	DP10S1	DBL	$J_2 x$	GBP05S1	GBXM	$J_1q$	ZLP9-S1	DPNL	$T_1t$
	DP003S1	DBL	$J_2 x$	GBP09S2	GBXM	$J_1q$	ZLP14-S1	DPNL	$T_1t$
	MP-20S1	DBL	$J_2b$	GBP10S2	GBXM	$J_1q$	ZLP7-S1	DPNL	$T_1t$
	MP-16S1	DBL	$J_2b$	GBP11S2	GBXM	$J_1q$	ZLP23-S2	DPNL	$T_1t$
	LP03-14S1	LP03	J <sub>2</sub> b	LP06-12S2	LP06	$J_2b$	ZP02-09S	ZP02	$J_2b$
	LP03-10S1	LP03	$J_2b$	ZP03-01S1	ZP03	$J_2b$	ZP02-08S	ZP02	$J_2b$
Crude oil	LP03-04S1	LP03	$J_2b$	ZP03-05S1	ZP03	$J_2b$	RP-08S1	RP	$J_2b$
	LP03-02S1	LP03	$J_2b$	AP01-12S1	AP01	$J_2b$	RP-15S2	RP	$J_2b$
	LP02-14S1	LP02	$J_2b$	AP01-12S2	AP01	$J_2b$	NP-08S1	NP	$J_2b$
	LP02-10S1	LP02	$J_2b$	AP02-04S1	AP02	$J_2b$	NP-18S1	NP	$J_2b$
	LP02-09S1	LP02	$J_2b$	AP02-08S1	AP02	$J_2b$	SP01-04S	SP01	$J_2b$
	KRP-01S1	LP02	$J_2b$	AP02-09S1	AP02	$J_2b$	SP01-02S1	SP01	$J_2b$
	KRP-07S1	KRP	$J_2b$	ZP01-14S1	ZP01	$J_2b$	SP02-05S1	SP02	$J_2b$
	LP06-03S1	LP06	$J_2b$	ZP01-12S1	ZP01	$J_2b$	SP02-07S	SP02	$J_2b$

Table. Crude oil and source rock samples for the biomarker and isotopic study

fractions using column chromatography over a silica gel and aluminum oxide. The saturated hydrocarbon fraction was eluted with n-hexane, the aromatic hydrocarbons were eluted with a mixture of hexane and DCM (4:1, v:v), and the NSO fractions (polar fractions containing nitrogen, sulfur and oxygen compounds) were eluted with a mixture of DCM and methanol (1:2, v:v). Activated Cu was added in normal amounts during the Soxhlet extraction to remove elemental sulfur. The fraction was further separated into straight-chain and branched/cyclic hydrocarbons using a 5 Å molecular sieve (Merck, Germany).

Gas chromatography-mass spectrometry (GC-MS) was performed using an Agilent 5975c mass spectrometer equipped with an Agilent 7890 gas chromatograph in full scan mode. For the saturated hydrocarbons, a 30 m HP-5MS fused silica column (0.25 mm i.d., 0.25 µm film thickness) with He as the carrier gas was used. The oven temperature was increased from 50 °C (1 min) to 310 °C (held for 30 min) at 3 °C/min. The transfer line temperature was 250 °C and the ion source temperature was 200 °C. The ion source was operated in the electron impact mode at 70 eV. Quantification of the compounds for calculating molecular parameters was performed by integrating the peak areas in the appropriate GC-MS chromatogram except for those used for  $\sum C_{21}$ -/ $\sum C_{21}$ +,  $C_{(21+22)}/C_{(28+29)}$ , Pr/Ph, Pr/nC<sub>17</sub>, Ph/nC<sub>18</sub>, CPI and OEP, which were integrated from the total ion chromatograms (TICs).

#### 4. Source rocks

The source rocks in the Southern Qiangtang Depression consist mainly of Triassic and Jurassic black rock series, such as shale, mudstone, limestone, etc. (Fig. 2). Triassic source rocks are predominantly coal-bearing strata of the Tumengela Formation deposited in delta plain-littoral swamps. Jurassic source rocks are mostly strata from the Lower Jurassic Sewa Formation, Middle Jurassic Quse, Shaqiaomu, Buqu and Xiali formations and the Upper Jurassic Suowa Formation. The Middle and Lower Jurassic rock series consist mainly of argillaceous source rocks deposited along the shelf basin, and the Middle and Upper Jurassic series are composed mostly of calcareous source rocks deposited in carbonate platforms or argillaceous source rocks deposited in tidal flats.

The Tumengela Formation development presents marine-terrigenous facies, which is mainly characterized by marlstone, mudstone and sandstone, with a coal-bearing series developed locally. The organic carbon content ranges from 0.63 to 1.50%, with an average of 1.02%; the organic matter type is mainly II<sub>2</sub>; the R<sub>o</sub> value varies between 1.46 and 1.78%, with an average of 1.62%; the rock pyrolysis peak temperature ( $T_{max}$ ) ranges from 419 to 552 °C at which it reaches the high-maturity evolution period.

Period	Formation	Thickness, m	Lithologic section	Source- reservoir- caprock assemblages	Source rock	Reservoir	Hydrocarbons	Samples Source rock
	Suonahu Fr. (N₂s)	212						
	Kangtuo Fr (N <sub>1</sub> k)	689		· · · · ·				
Tertiary	Dingqinghu Fr. (E <sub>3</sub> k)	560-1166						
	Niubao Fr. (E <sub>1-2</sub> k)	500-2885						
Cretaceous	Abushan Fr. (K <sub>1</sub> a)	982–1202		· · · · · · · · · · · · · · · · · · ·				
	Xueshan Fr. (J <sub>3</sub> x)	146–1604		••••				
Jurassic	Suowa Fr. (J <sub>3</sub> s)	1825			•			$\checkmark$
	Xiali Fr. (J <sub>2</sub> x)	789–1307						$\checkmark$
	Buqu Fr. (J <sub>2</sub> b)	104-860			•	R	* 🔺	$\checkmark$
	Shaqiaomu Fr. (J <sub>2</sub> sq)	1111			•			$\checkmark$
	Sewa Fr. (J <sub>2</sub> s)	1040						$\checkmark$
	Quse Fr. (J <sub>1</sub> q)	700			•			$\checkmark$
	Nadigangri Fr. (T <sub>3</sub> n)	527						
	Xiaochaka Fr. (T₃x)	451-690						
Triassic	Kangnan Fr. (T₂k)	101-640						
	Tumengela Fr. (T <sub>1</sub> k)	300-1300 >384	••••	•••				
	Rejuechaka Fr. (P <sub>2</sub> r)							
Permian	Longge Fr. (P <sub>1</sub> l) 3411				limestone	••• ••• ••• sand	  mudstone	dolostone
	Tunlonggongba Fr. (P <sub>1</sub> t)	860						
Carboniferous	Riwanchaka Fr. (C <sub>1</sub> t)	>458			marlstone	conglome	rate coal	basalt
Devonian	Chasangzu Fr. (D₃c)	> 312			+ + + + + granite	* oil	bitumen	bioclastic limestone

Fig. 2. Stratigraphic column of the Qiangtang Basin, source-reservoir-caprock assemblage and distribution of samples. (The abbreviation used: Fr. - Formation.)

The Quse Formation is a set of continental shelf deposits, and the source rocks are mainly distributed in the central region of the Southern Qiangtang Depression and consist primarily of dark mudstone and limestone. The mudstone has an organic carbon content of 0.40-0.51%, and that of limestone source rocks is 0.1-0.35%. The organic matter types are chiefly II<sub>1</sub> and II<sub>2</sub>, and the thermal evolution degree is mature.

The Sewa Formation is a set of continental shelf basin deposits, and the source rocks are mainly distributed in the central region of the Southern Qiangtang Depression. This formation consists predominantly of dark mudstone and shale that have a low organic carbon content, 0.40-0.83%. The organic matter types are mostly II<sub>1</sub> and II<sub>2</sub>, and the thermal evolution degree is mature.

The Shaqiaomu Formation is a set of continental shelf deposits, and the source rocks are mainly distributed in the central region of the Southern Qiangtang Depression. This formation consists predominantly of dark mudstone and sandy mudstone with a low organic carbon content, 0.4-0.67%. The organic matter types are chiefly II<sub>1</sub> and II<sub>2</sub>, and the thermal evolution degree is mature.

The Buqu Formation is a set of shallow continental shelf limestones deposited in the Jurassic transgression zenith, which is widely distributed throughout the Qiangtang Basin. The limestone source rocks are distributed throughout the basin, and the organic carbon content is 0.26–0.87%. The kerogen type is mainly I. The average Ro of limestone source rocks is 1.44%, and the  $T_{max}$  mainly ranges between 444 and 462 °C.

The Xiali Formation is composed mostly of tidal flat sand and mudstone, and has a maximum thickness above 1,000 m. The depocenter of mudstone source rocks is in the central section of the Southern Qiangtang Depression. The organic carbon content of the rocks is 0.31-3.01%, the kerogen type is predominantly II<sub>2</sub>, the average Ro is 1.41% and the average  $T_{max}$  441.5 °C.

The Suowa Formation is composed of deposits that formed during regression. The carbonate content of the rocks increases from east to west, and mudstone is mainly found in the east section of the basin. The formation is 300 m thick on average, and a maximum thickness of 600 m is observed in 114 locations. The organic carbon content of the source rocks is 0.11-1.11%, the kerogen type is mainly II<sub>1</sub> followed by I, and the thermal evolution degree is mature.

The oil shale series source rocks from the Biluocuo region developed in the midwest section of the Southern Qiangtang Depression belong to the Quse Formation. But the distribution of oil shale is very limited, and the evaluation of hydrocarbon source rocks is often neglected. The thickness of the oil shale series source rocks is up to 171.9 m, the organic carbon content is high and mainly varies between 1.87 and 26.12%. The kerogen types are predominantly II<sub>1</sub> and II<sub>2</sub>, and the thermal evolution degree is mature.

#### 5. Crude oil

The Qiangtang Basin was affected by the subduction of the Neo-Tethys Paleo-Ocean plate and the northward subduction and collision orogenesis of the Indian continent; thus, it was subject to large-scale thrust-nappe structure tectonic activities. As a result, the petroleum system of the basin was damaged, and numerous oil shows were exposed on the surface; however, they are mostly dry bitumen. Thus far, massive saccharoidal dolostone impregnated with liquid crude oil has only been found in the Longeni-Angdaercuo paleo-oil reservoir, which is characterized by long wet oil seepages and the strong smell of light crude oil [15-20]. The oil reservoir is 150 km long from east to west and 20 km wide from south to north (Fig. 3). Many oil shows have been found in the paleo-dolostone reservoir, which exhibited vellow, blue and white fluorescence during fluorescence titration. The organic carbon content is 0.03-0.09%, and the average content of chloroform bitumen "A" is 0.0027%. The chloroform bitumen "A" family consists of 5.60-50.58% saturated hydrocarbons, 21.10-34.10% aromatic hydrocarbons, 20.17–60.64% nonhydrocarbons and 2.69–33.77% asphaltenes. Although the massive exposure of the paleo-oil reservoirs reflects the damage inflicted on the original petroleum system of the basin, it also indicates that the Qiangtang Basin experienced a period where massive amounts of oil and gas were generated, migrated and accumulated. Therefore, oil and gas were formed in huge amounts. Although no liquid crude oil has been obtained from the Qiangtang Basin, a lot of oil-bearing dolostone samples were collected from the paleo-oil reservoirs exposed on the surface.



Fig. 3. Sampling profiles of source rock and oil-bearing dolostone samples in the Southern Qiangtang Depression.

Because of the poor geological drilling conditions, the paleo-oil reservoir exposure area offers an opportunity to investigate the distribution of underground oil and gas reservoirs and comprehensively analyze the accumulation conditions of paleo-oil reservoirs, which should enable a more precise prediction of the distribution of oil and gas resources in the basin.

# 6. Crude oil-source rocks correlation

To determine whether crude oil and source rocks in the same oil- and gasbearing basin are genetically related, various methods and biomarker parameters are used. Such methods have been employed for a systematic analysis of stable isotope data, and the results are correlated to identify the source of oil, and determine effective hydrocarbon generation layers, as well as the location of the hydrocarbon generation depression [21–27].

#### 6.1. Correlation between biomarkers in crude oil and source rocks

#### 6.1.1. Biomarkers

Comparative analysis of biomarkers is an intuitive and relatively effective method to elucidate whether there is a genetic relationship between crude oil and source rock. In this study, a quality spectrum that represents the relative strength of biomarkers in crude oil and source rocks (e.g., sterane m/z 217, m/z 372, m/z 386; hopane m/z 191, m/z 370, m/z 398; n-alkane m/z 85) was identified. First, terpanes and regular steranes in the samples were summarized and categorized. Next, the oil-bearing and source rock samples were compared based on compound characteristics. Comparison of the typical chromatograms of hopanes (m/z 191) and steranes (m/z 217) present in source rock and crude oil samples from the Southern Oiangtang Basin showed that crude oil had a similar distribution of terpanes fingerprints as the oil shale series source rock from the Biluocuo region. Regular tricyclic terpanes were rarely detected in either crude oil or oil shale series source rock extracts, which is a diagnostic characteristic to differentiate the oil shale series source rock from the other Jurassic source rocks. The C<sub>29</sub> steranes were found in abundance in the Triassic coal-bearing formation samples of the Southern Qiangtang Basin, which distinguishes the source rocks or crude oil from other source rocks or crude oils. This may be attributed to the elevated input of higher plants. Crude oil was characterized by low amounts of pregnane and relatively abundant steranes, which is also typical of Biluocuo oil shale. By comparison, the source rock samples from the Suowa, Xiali, Bugu, Shagiaomu, Sewa and Quse formations were rich in pregnane. Steranes and hopanes were rarely detected in the carbonate source rocks from the Suowa and Bugu formations but were found to be abundant in muddy source rocks (Fig. 4).



Fig. 4. Comparison of the chromatograms of biomarkers m/z 191 and m/z 217 of crude oil and source rock samples, Southern Qiangtang Depression.

## 6.1.2. Relative abundance of biomarkers

According to the theory of oil generation via kerogen pyrolysis, oil is derived from bitumen formed via kerogen pyrolysis of the source rock. Therefore, crude oil is certainly derived from more mature source rocks. The source rocks in the Southern Qiangtang Depression have broken through the oil generation threshold and entered a mature stage. Some kinds of sedimentary organic matter experienced thermal evolution and even reached the peak oil generation stage. All source rocks on the distribution map of  $C_{31}22S/(22R+22S)$  hopane and  $C_{30}\alpha\beta/(\beta\alpha+\alpha\beta)$  hopane have entered the oil generation threshold (Fig. 5a). The distribution of the  $C_{31}22S/(22R+22S)$ hopane ratio is relatively concentrated and has reached a balance. For the reason that the  $C_{30}\alpha\beta/(\beta\alpha+\alpha\beta)$  hopane ratio will continue to change regularly as the organic matter continues to further undergo thermal evolution, it can be used to determine the oil generation threshold, the oil generation peak and even the upper limit of the moisture zone. The diagram shows that the distribution of  $C_{30}\alpha\beta/(\beta\alpha+\alpha\beta)$  is quite decentralized, and, based on the kerogen oil generation theory, the source rocks in the coal-bearing rock of the Tumengela Formation and the mudstone type source rocks in the Xiali Formation are not active source rocks of the paleo-oil reservoir in the Bugu Formation because the maturity of the two sets of strata source rocks is lower than that of crude oil.

Traces of pristane and phytane were identified in all samples. Pristane and phytane are derived to a great extent from phytol side chains of chlorophyll-a or photosynthetic algae, although other sources are also known. Based on the n-alkane and isoprenoid contents, the primary organic matter sources are dominated by phytoplankton, especially algae, except for the source rock samples of the Tumengela Formation. Although Pr/Ph, Pr/nC<sub>17</sub> and Ph/nC<sub>18</sub> ratios have been used to infer depositional redox conditions, they are influenced by several other factors, such as thermal maturity, sources of biomolecules, lithology and diagenetic effects. Therefore, the low Pr/Ph ratios from 0.06 to 0.80 in most samples indicate anoxic to dysoxic depositional conditions except for the source rock samples of the Tumengela Formation. Furthermore, the generative kerogen and sedimentary environment of the oilbearing samples of the paleo-oil reservoir are similar to those of the source rocks in the oil shale series source rock from the Biluocuo region, and the Xiali, Quse and Sewa formations, but are obviously different from those of the source rocks in the Buqu, Suowa and Tumengela formations (Fig. 5b).

Tricyclic terpane is an important component of terpenoids in crude oil. The carbon number of tricyclic terpanes ranges from  $C_{19}$  to  $C_{54}$ , while the compound is highly resistant to biodegradation. Previously it has been shown that the source and preservation conditions of organic matter as well as carbon isotopes composition could be used as indicators of the composition characteristics of tricyclic terpanes as well as the origin of organic matter in source rocks, and for comparative studies of crude oil-source rocks correlation [28–30]. All source rocks in the Quse, Sewa and Shaqiaomu formations as well as the majority of the source rocks in the Xiali and Buqu formations contain high amounts of tricyclic terpanes, especially  $C_{19}$ – $C_{22}$  and  $C_{23}$ – $C_{26}$  homologues, while  $C_{28}$ – $C_{29}$  tricyclic terpanes are present in low abundance. However, the abundance of tricyclic terpanes in the oil-bearing samples of the paleo-oil reservoir is low, while  $C_{19}$ – $C_{22}$  and  $C_{28}$ – $C_{29}$  tricyclic

terpanes are present in similar, yet slightly lower amounts than  $C_{23}$ – $C_{26}$  homologues. The  $C_{23}$ – $C_{26}$  to  $C_{28}$ – $C_{29}$  tricyclic terpanes and terpane to sterane ratios in the oil shale samples from the Biluocuo region and the oil-bearing samples from the paleo-oil reservoir are low (Fig. 5c, 5d). Both ratios are indicative of that the source material of organic matter was primarily low-grade biological algae, with a minor contribution of terrestrial organic material. The above similar ratios suggest that the oil-bearing samples from the paleo-oil reservoir are genetically related to the oil shale samples from the Biluocuo region, whereas the genetic relationship with the other sets of source rocks is either poor or absent.

The presence of a homologous series of C<sub>27</sub>-C<sub>29</sub> regular steranes was observed in crude oil and source rock samples. In general, C<sub>29</sub> sterane is a major sterane derived from higher plants, although it can also represent significant inputs from marine algal sources. C<sub>27</sub> sterane is commonly associated with zooplankton and C28 sterane with chlorophyll-c-bearing dinosteranes are primarily while phytoplankton, derived from dinoflagellates. Nevertheless, Peters et al. [24] and Volkman [30] suggested to interpret sterane contents with caution because many algae are capable of synthesizing C<sub>29</sub> sterane, whereas C<sub>27</sub> and C<sub>28</sub> steranes may be derived from various sources. In this study, the source rocks and oil-bearing samples from the paleo-oil reservoir were divided into three types based on the relative abundance of  $C_{27}$ - $C_{28}$ - $C_{29}$  regular steranes. The  $C_{27}/C_{29}\alpha\alpha\alpha 20R$  ratio of the coal-bearing source rocks from the Tumengela Formation was extremely low, which indicates the suitability of using  $C_{29}\alpha\alpha\alpha 20R$  to differentiate coalbearing source rocks from the other types of source rocks. The distribution of C<sub>27</sub>-C<sub>29</sub> steranes in the other samples was relatively concentrated, while no significant difference in their content between the samples was observed. Most oil-bearing, oil shale and coal-bearing source rock samples had a low ratio of pregnane to aaaC<sub>29</sub> regular sterane (Fig. 5e, 5f). The contents of pregnane and homopregnane in the source rocks and crude oil of several salt lake facies basins of China are extremely high. In addition, the relative abundance of compounds of the same series was affected by maturity. Compared with other sets of source rocks, oil shale series source rocks exhibited a slightly higher degree of maturity which in turn differed from that of crude oil. At the same time, the overall maturity of crude oil was somewhat lower than that of source rocks. However, the abundance of pregnane in oil-bearing and oil shale series source rock samples was similar, being at the same time lower than that in the samples of several source rock sets. Moreover, pregnane was more abundant in mudstone source rocks than in limestone source rocks. Additionally, the distribution of homopregnane was similar to that of pregnane. Therefore, the relative abundance of pregnanes and homopregnanes in the source rocks and crude oil in the Southern Qiangtang Depression was controlled not by maturity but by the type of generative kerogen, which also better reflects the genetic relationship between crude oil and oil shale series source rocks.



Fig. 5. Relative content of saturated hydrocarbon biomarkers in the source rocks and crude oil of the Southern Qiangtang Depression: (a) relationship diagram of the relative ratio of hopane isomers in crude oil and source rocks; (b) distribution diagram of the relative abundance of isoprenoids in crude oil and source rocks; (c), (d) distribution diagrams of the relative abundance of tricyclic terpanes in crude oil and source rocks; (e) relationship diagram of the relative abundance of pregnanes and regular sterane in crude oil and source rocks; (f) relationship diagram of the relative abundance of pregnanes and tricyclic terpanes in crude oil and source rocks.

#### 6.2. Crude oil-source rocks correlation based on carbon isotopes

The carbon isotopes of crude oil and gas have a strong kerogen inheritance, which means that their compositions in homologous crude oil and gas samples are closely related. The organic matter of crude oil can be distinguished from that of oil source rocks by comparing their carbon isotope composition. This is because organic matter has a unique carbon isotope composition and the fractionation of crude oil isotopes caused by thermodynamic activity features kerogen inheritance. Therefore, carbon isotope composition can be used to establish a relationship between crude oil and source rocks.

In the Xiali Formation of the Qiangtang Basin, the chloroform bitumen "A" isotope content in Jurassic source rocks varies greatly, from -27.92 to -34.98%, being in the other strata relatively stable, between -27 and -29.63‰. The variation ranges and average values of chloroform bitumen "A" isotopes in the reservoir stratum oil seepage of dolostone and oil shale series source rocks from the Biluocuo region are quite similar, reflecting their genetic relationship (Fig. 6). Additionally, except for the source rocks in the Xiali Formation, both the chloroform bitumen and component isotopes of the source rocks and crude oil present a normal isotope distribution mode, namely,  $\delta^{13}C_{\text{saturated hydrocarbon}} < \delta^{13}C_{\text{chloroform bitumen}} < \delta^{13}C_{\text{aromatic hydrocarbon}}$ . The distribution diagrams of  $\delta^{13}C$  saturated hydrocarbon and  $\delta^{13}C$  aromatic hydrocarbon show that the Middle-Upper Jurassic source rocks are quite different from the Middle-Lower Jurassic ones. The isotopes content of saturated and aromatic hydrocarbons in the source rocks of the Suowa, Xiali and Bugu formations is relatively high while that in the source rocks of the Shaqiaomu, Sewa and Quse formations is low. Furthermore, the isotopes of the oil shale series source rocks from the Biluocuo region and crude oil show a relatively concentrated distribution in the Middle-Upper Jurassic source rocks as shown in the distribution map (Fig. 7). The line charts of the



Fig. 6. Distribution of chloroform bitumen "A" carbon isotopes in the oil seepage of the source rocks and oil-bearing dolostone of the Southern Qiangtang Depression. (The abbreviaton used: Fr. – Formation.)



Fig. 7. Comparison of the carbon isotopes distribution in the oil seepage of the source rocks and oil-bearing dolostone of the Southern Qiangtang Depression: (a) distribution line chart of average carbon isotopes in the group component; (b) distribution of isotopes of saturated and aromatic hydrocarbons.

distribution of average isotopes of saturated and aromatic hydrocarbons and chloroform bitumen "A" of the oil shale series source rock from the Biluocuo region and crude oil are nearly consistent, which further confirms the good genetic relationship between crude oil and source rocks.

# 6.3. Crude oil-source rocks correlation based on monomer hydrocarbon isotopes

Although the isotopes of chloroform bitumen "A" and group components of crude oil can well reflect the characteristics of kerogen, the mass number of isotopes may vary during the generation of hydrocarbons from source rocks and transportation of crude oil. Moreover, the same monomer hydrocarbon components may lead to the production of different isotopes, which will be reflected in the composition of monomer hydrocarbon isotopes. Therefore, the monomer hydrocarbon isotopes composition can also be used to establish a correlation between crude oil and source rocks.

Even though the carbon isotopes composition of chloroform bitumen "A" and isotope group components of Jurassic Buqu Formation source rocks are rather similar to those of crude oil, the curves of their monomer hydrocarbon isotopes considerably differ. The monomer hydrocarbon isotope values of the Buqu Formation source rocks are relatively high and vary from -25 to -28%. The monomer hydrocarbon isotope curve gradually declines before C<sub>24</sub>, while after C<sub>24</sub> it gradually ascends, but overall exhibits great variations. The monomer hydrocarbon isotope value of crude oil is relatively low, varying slightly around -30%; however, it may also considerably fluctuate, e.g., in case of C<sub>18</sub> and C<sub>29</sub> hydrocarbon monomers. Furthermore, the monomer hydrocarbon isotope components of the oil shale series source

rocks from the Biluocuo region and crude oil are similar, which indicates a good genetic relationship between crude oil and source rocks (Fig. 8). The monomer hydrocarbon isotope value of muddy source rocks in the Xiali Formation is quite different from that of the oil shale series source rocks from the Biluocuo region, and decreases continuously to a right-leaning oblique line.



Fig. 8. Distribution of monomer hydrocarbon isotopes in the source rocks and oilbearing dolostone of the Southern Qiangtang Depression. (The abbreviation used: Fr. – Formation.)

# 7. Conclusions

- 1. The distribution of terpanes and steranes in crude oil is similar to that in the oil shale series source rocks from the Biluocuo region, while regular tricyclic terpanes are rarely detected in either crude oil or oil shale series source rock extracts. In crude oil and oil shale series source rocks from the Biluocuo region, the  $C_{29}$ - $C_{26}/C_{28}$ - $C_{29}$  tricyclic terpane ratio is very low, the abundance of pregnane is low and that of steranes relatively high. Also, the  $C_{21}$  pregnane/ $C_{29}\alpha\alpha\alpha$  regular sterane ratio is low. All the above indices of Biluocuo source rocks differ from those of source rocks from other formations, in terms of biomarker fingerprints, and suggest a genetic relationship between crude oil and oil shale series source rocks from the Biluocuo region.
- 2. The  $\delta^{13}$ C values of either chloroform bitumen "A" or different bitumen fractions of crude oil are similar to those of oil shale series source rocks from the Biluocuo region, and the n-alkane carbon isotopic composition profiles of crude oil and the source rocks are similar as well. This means that the  $\delta^{13}$ C value of both is about -30‰, and these profiles follow a horizontal straight line, with a positive slope at C<sub>18</sub>. By contrast, the  $\delta^{13}$ C

profiles of other source rocks incline from upper right to lower left, with more negative  $\delta^{13}$ C values on average, and with no positive slope at C<sub>18</sub>. All of these carbon isotopic compositional characteristics suggest that crude oil is genetically related to the oil shale series source rocks from the Biluocuo region.

3. Considering the relative abundance of biomarkers, as well as carbon and monomer hydrocarbon isotope compositions of crude oil and source rocks, the oil shale series source rocks from the Biluocuo region represent the most likely source rocks of the paleo-oil reservoir in the Southern Qiangtang Basin. The distribution area of the oil shale series source rocks from the Biluocuo region coincides with that of the hydrocarbon generation depression of the Qiangtang Basin.

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