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Geochemical characteristics and genetic types of crude oils from Qinjiatun and Qikeshu oilfields in the Lishu Fault Depression, Songliao Basin, northeastern China

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Abstract The Qinjiatun and Qikeshu oilfields are new Mesozoic petroleum exploration targets in Lishu Fault Depression of Songliao Basin, northeastern China. Currently, researches on geochemistry of crude oils from Qinjiatun and Qikeshu oilfields have not been performed and the genesis of oils is still uncertain. Based on bulk analyses, the crude oils in the Qinjiatun and Qikeshu oilfields of Lishu Fault Depression from the Lower Cretaceous can be classified as three types. Type Ⅰ oils, from Quantou and Denglouku formations of Qikeshu oilfield, are characterized by high C_{24}tetracyclic terpane/C_{26}tricyclic terpanes ratios, low gammacerance/C_{30}hopane ratios, tricyclic terpanes/hopanes ratios, C_{29}Ts/C_{29}norhopane ratios and 17α(H)-diahopane/17α(H)-hopane ratios, indicating a brackish lacustrine facies. Type Ⅱ oils, from Shahezi Formation of Qikeshu oilfield show low C_{24}tetracyclic terpane/C_{26}tricyclic terpanes, high gammacerance/C_{30}hopane ratios, tricyclic terpanes/hopanes ratios, C_{29}Ts/C_{29} norhopane and C_{30}diahopane/C_{30}hopane ratios, thus suggesting that they originated from source rocks deposited in a weak reducing brackish lacustrine environment, or clay-rich sediments. Type Ⅲ oils, from some wells of Qikeshu oilfield have geochemical characteristics intermediate between those two types and may be mixture of type Ⅰ and Ⅱ oils.

Key words Songliao Basin; Lishu Fault Depression; crude oil; geochemistry; genesis

1 Introduction

The biological marker compounds of crude oils preserve molecular structure of various compounds that constitute the organisms. Biomarkers are widely used in the petroleum industry for studying oil/oil and oil/source rock correlations (Zhang Min et al., 2006; Li Hongbo et al., 2008; Mohammed et al., 2011) and source rock attributes, including depositional environment, kerogen type and maturity for migrated oil of uncertain origin (Moldowan et al., 1985; Peters and Moldowan, 1993; Peters et al., 2005).

Lishu Fault Depression is located in the southeast uplift of Songliao basin, northeastern China, covering total area of about 3100 km^2. Through 20 years of petroleum exploration, nine oilfields and a number of oil bearing structures have been found in this region with the proven reserves of about 4435×10^4 t. Despite the relatively long history of oil exploration, several important issues regarding aspects of Lishu Fault Depression petroleum system still remain uncertain. For example, no oil/oil and oil/source rock correlations have been reported and geochemical studies on Lishu Fault Depression are relatively few. Recently, reports on source rock evaluation (Zhang Jun, 2010), hydrocarbon accumulation timing (Zhou Zuoming et al., 2012) and dynamic evaluation of oil-gas resources (Zhang Xi et al., 2013) appear, but very few evaluate the biomarker distributions of crude oils (Chen Xiaohui, 2012). So, this study is focused on the crude oils from the Qinjiatun and Qikeshu oilfields of Lishu Fault Depression, and systematically analyzes the geochemical characteristics of the crude oils. Types of crude oils are also distinguished.

2 Geological setting

Songliao Basin is a large-scale Mesozoic-Cenozoic continental petroliferous basin in northeastern China. The Lishu Fault Depression is located in southern part of the southeast uplift of the Songliao Basin, which developed from Late Jurassic to Early Cretaceous, belonging to a separate hydrocarbon depression. Qinjiatun oilfields are situated in eastern slopes of Lishu Fault Depression within Qinjiatun north-east trending large nose structure, where Quantou and Denglouku formations are the main oil-producting intervals. The Qikeshu oilfield is seated in the central uplift belt edge of Lishu Fault Depression, east of Shiwu oilfield and west of Qinjiatun oilfield.

3 Sampling and experimental method

A set of twenty-seven oil samples were collected from Qinjiatun and Qikeshu oilfields. Twenty-two samples were collected from Qinjiatun oilfield, including SN106, SN107, SN121, SN122, SN142, SN143 and SN106 blocks, which are concentrated in Quantou Formation and Denglouku Formation. Other 5 samples were collected from Shahezi Formation of Qikeshu oilfield, including SW8, SW9, and SW11 wells oils.

Samples were analyzed by gas chromatography-mass spectrometry (GC-MS) to detect the distribution of saturated hydrocarbons. A HP 5973 mass spectrometer coupled with a HP 6890 GC and equipped with a 30 m (0.25 mm i.d., 025 μm in film thicknesses) HP-5MS fused silica capillary column was used for GC-MS analysis. The GC for the analysis was temperature-programmed as being heated for 1 minute at 50℃, then increasing from 50 to 100℃ at a rate of 20℃/min, and from 100 to 315℃ at a rate of 3℃/min and at 315℃ for 18 minutes. Helium was used as the carrier gas with a rate of 1.0 mL/min and the ionization source was operated at 70 eV.

4 Bulk properties of crude oils

The vast majority of crude oil samples are black; a few of them are yellow and mainly from Quantou Formation of Qinjiatun Oilfield and Shahezi formation of Qikeshu oilfield. Those oil samples show wide variations in the wax content, ranging from 1.91% to 39.56%, with the density of 0.83–0.90 g/cm^3 at 20℃. The viscosity of crude oils is concentrated in the 6.26–24.77 mPa.s (50℃), and the freezing points are all between 11 and 24℃. And those samples are low in sulfur (<0.1%), reflecting that those oils probably derived from lacustrine depositional environment, or clay-rich sediments (Tissot and Welte, 1984).

5 Results and discussion

5.1 *n*-Alkanes and isoprenoid alkanes

As seen in Fig. 1, *n*-alkanes of the crude oils from Qinjiatun and Qikeshu oilfields mostly present a unimodal to slightly bimodal distribution pattern with a predominance of medium molecular weight compounds (nC_{19}, nC_{21} or nC_{23}). Typically, the crude oil from SW11 well displays a unimodal hydrocarbon pattern with the short-chain C_{10} *n*-alkane dominating, which shows the typical condensate oil characteristics. Accordingly, the investigated samples are characterized by OEP and CPI values of 1.0–1.2, nC_{21}^-/nC_{22}^+ ratios generally higher than 1.0 and Pr/Ph ratios between 0.86 and 1.76, indicating that those oils, as a whole, show a relatively higher maturity and corresponding source deposited under weak-reducing to reducing sedimentary conditions (Didyk et al., 1978).

5.2 Sesquiterpenes

The drimane series are often ascribed to input from prokaryotes bacteria, and the relative abundance of rearranged drimanes is closely related to the oxidation-reduction conditions of the depositional environment (Peters and Moldowan, 1993). As shown in Fig.1, most crude oil samples present a low content of diadrimanes and a strong dominance of C_{15} drimanes, whose diaC_{15}/C_{15} ratios are between 0.41 and 0.69. In particular, the oil sample from SW11 well displays a strong predominance of rearranged drimanes, and the corresponding ratio is 3.35. The special drimanes distribution characteristics of SW11 wells crude oil probably imply a relatively weak reducing depositional setting.

5.3 Tricyclic terpanes and hopanes

The terpane biomarkers of the oil samples have the feature of abundant pentacyclic terpanes, and relatively low concentrations of tricyclic terpanes. The carbon number distribution is characterized by the highest peak at C_{23}, suggesting a terrigenous environment (Aquino et al., 1983). Tricyclic terpanes are generally believed to be a contribution of organic matter from algae or microorganisms, while the significant content of a C_{24} tetracyclic terpane is correlatable with low saline, non-marine conditions and higher plant material (Aquino et al., 1983). The ratios of C_{24}tetracylic/C_{26}tricyclic terpane and tricyclic terpanes/hopanes of Qinjiatun oilfield are in the ranges of 0.4–0.5 and 0.1–0.15, respectively. By contrast, oils from the Qikeshu oilfield exhibit lower C_{24} tetracylic/C_{26}tricyclic terpane ratios, mostly in the range of 0.14–0.25, whilst the tricyclic terpanes/hopanes ratios of these oils are relatively higher, ranging from 0.47 to 4.17. It was suggested that crude oils from Qinjiatun oilfield have a larger contribution of terrigenous organic material.

The most important feature of terpane distributions for those oils is the significant variation in the abundance of 17α(H)-diahopanes relative to other terpenoid components. There are two extremes of terpane distributions between Qinjiatun and Qikeshu oilfields as shown in the m/z 191 mass chromatograms (Fig. 4). As seen from Table 1 and Fig. 3, crude oils from Qinjiatun oilfield are dominated by 17α(H)-hopanes, whose gammacerane/C_{30}-hopane ratios vary from 0.29 to 0.44, with low abundance of C_{29}Ts and C_{30} diahopane. Nevertheless, the hopanes distribution of the crude oils from Qikeshu oilfield shows a dominance of 17α(H)-diahopanes and 18α(H)-norneohopanes, with high contents of C_{29}Ts and C_{30} diahopane. Their gammacerane/C_{30}-hopane ratios range from 0.46 to 0.50. It is generally believed that high content of diahopanes indicates catalytic effect of clay minerals under oxic-suboxic conditions (Sinninghe et al., 1995; Zhu Yangming et al., 2007). Therefore, the relative high abundance of 17α(H)-diahopanes of Qikeshu oilfield oils illustrates that the oils deposited under a more oxydic conditions than the oils from Qinjiatun oilfield .

Most oils are mature oils as indicated by C_{31}22S/(22S+22R) (0.58–0.6) and Ts/(Ts+Tm) (0.47–0.77) homohopane isomerized parameters.

5.4 Steranes

The distribution of steranes is best studied with GC/MS by monitoring ion m/z 217 (Fig. 4). Huang Wenyen and Meinschein (1979) proposed that a predominance of C_{29}sterols (steranes) indicate a strong contribution from organic matter of higher plants, whereas a dominance of C_{27} steranes suggests a dominance of plankton. All those crude oil samples present a general higher abundance of C_{29} regular steranes, as seen in Fig. 4, illustrating a strong contribution of organic matter from terrigenous higher plants.

The sterane maturity parameters of ααα C_{29}20S/(20S+20R) and C_{29}ββ/(ββ+αα) for all these oil samples (Table 1) individually range from 0.48 to 0.60 and 0.35–0.54, respectively. Both of them are close or reach their equilibrium values (Mackenzie et al., 1984), indicating the attributes of mature oils.

Diasterane/sterane ratios are commonly used to distinguish carbonate-from clay-rich source rocks. Oils from Qinjiatun and Qikeshu oilfields exhibit a medium to high abundance of diasteranes (Table 1), which suggests clastic source rocks. On the other hand, the relatively high abundance of diasteranes in those samples from Qinjiatun oilfield implies generation of those oils from source rocks lean in clay minerals. It should be noted, however, that high diasterane/sterane ratios could be brought from high thermal maturity (Seifert and Moldowan, 1978). The maturity parameters based on αααC_{29}20S/(20S+20R) and C_{29}ββ/(ββ+αα) for the oil samples (Table 1) indicate that the high diasterane ratios of oils from Qinjiatun oilfield are not simply due to thermal maturity effects.

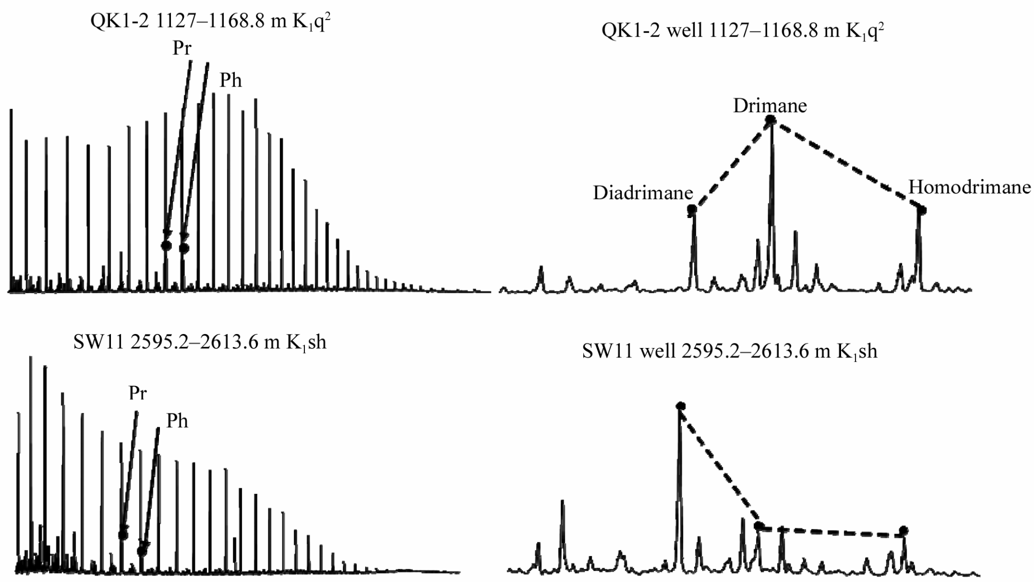

Fig.1. Distribution of *n*-alkanes and drimane series in representative oils from Qijiatun and Qikeshu oilfields (m/z 85 and m/z 191).

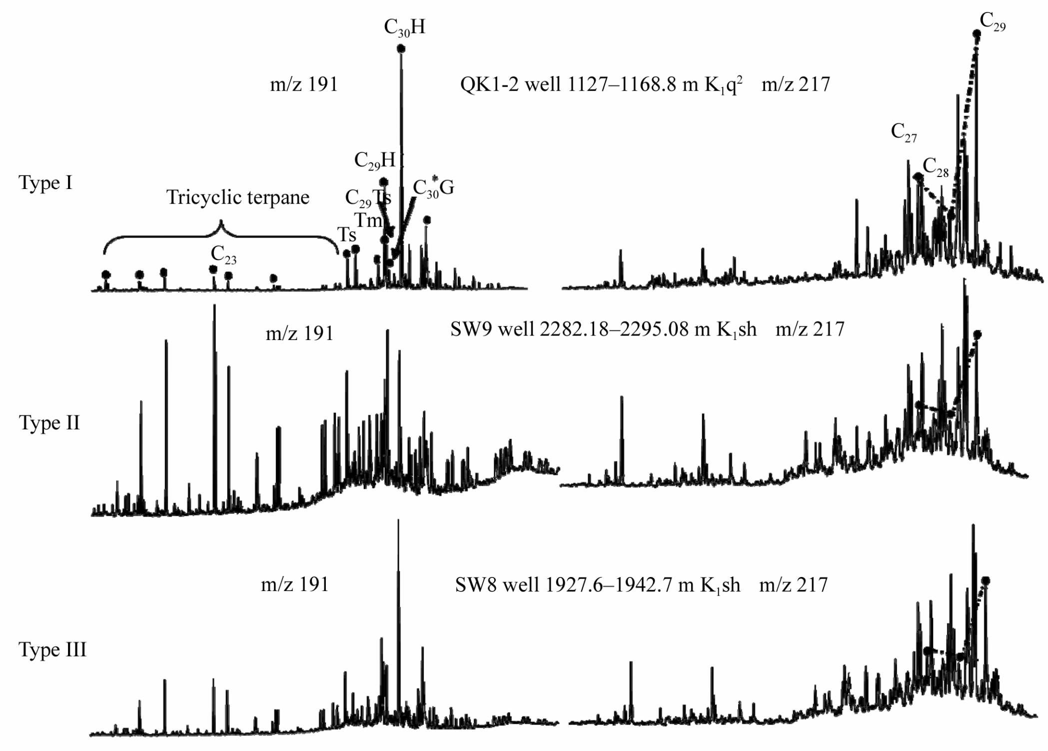

Fig. 2. Distribution of steranes and terpanes in representative oils from Qijiatun and Qikeshu oilfields.

Table 1 Geochemical parameters in typical crude oils samples from Qinjiatun and Qikeshu oilfields

Area	Well No.	Depth (m)	Formation	1	2	3	4	5	6	7	8	9
	SW8	1927.56–1942.7	K_1sh	0.25	0.47	0.46	1.62	0.34	0.78	0.30	0.41	0.59
	SW8–1	1865.6–1867	K_1sh	0.25	0.47	0.46	1.64	0.30	0.79	0.32	0.34	0.59
Qikeshu	SW8–2	1962.8–2000.3	K_1sh	0.25	0.47	0.50	1.62	0.32	0.74	0.31	0.38	0.59
	SW9	2282.18–2295.08	K_1sh	0.14	1.40	0.69	3.33	1.22	1.56	0.31	0.41	0.60
	SW11	2595.2–2613.6	K_1sh		4.17		4.69	10.6	1.62	0.81		0.56
	QK1–2	1127–1168.8	K_1q^2	0.50	0.11	0.28	0.89	0.10	0.46	0.42	0.34	0.52
	QK121–1	1001.3–1060.4	K_1d	0.41	0.12	0.25	1.44	0.14	0.59	0.45	0.39	0.53
	QK121–4	1024–1109.5	K_1d	0.40	0.12	0.25	1.45	0.14	0.56	0.45	0.37	0.52
	QK121–6	1069–1088	K_1d	0.40	0.13	0.26	1.46	0.14	0.52	0.51	0.31	0.50
	QK121–7	1009.4–1235	K_1q^2	0.41	0.13	0.23	1.50	0.14	0.56	0.50	0.34	0.50
	QK122–12	1008.8–1026.5	K_1q^2	0.41	0.10	0.35	1.42	0.12	0.51	0.46	0.32	0.49
	QK122–14	1013.8–1026	K_1q^2	0.42	0.11	0.25	1.43	0.12	0.50	0.45	0.37	0.52
	QK122–16	1003.6–1013	K_1q^2	0.41	0.11	0.24	1.42	0.12	0.50	0.44	0.32	0.48
	QK122–20	1011–1023	K_1q^2	0.42	0.11	0.23	1.38	0.12	0.52	0.40	0.31	0.49
	QK122–21	1062.9–1067.9	K_1q^2	0.41	0.11	0.22	1.37	0.12	0.50	0.39	0.30	0.48
	QK122–8	1015–1023.5	K_1q^2	0.42	0.11	0.21	1.42	0.11	0.52	0.45	0.31	0.49
Qinjiatun	QK129–1	945.6–1013.4	K_1q^2	0.43	0.12	0.24	1.47	0.13	0.52	0.38	0.37	0.53
	QK142–20	1016.2–1033	K_1q^2	0.41	0.12	0.27	1.43	0.14	0.52	0.41	0.35	0.53
	QK142–3	1130–1144	K_1q^2	0.40	0.11	0.25	1.52	0.12	0.49	0.43	0.32	0.49
	QK142–4	1053.4–1055.2	K_1q^2	0.39	0.11	0.27	1.43	0.14	0.55	0.42	0.34	0.49
	QK142–7	1132–1065	K_1q^2	0.42	0.12	0.21	1.46	0.13	0.49	0.44	0.34	0.49
	QK142–8	953–995.7	K_1q^2	0.39	0.11	0.31	1.34	0.15	0.64	0.43	0.36	0.54
	QK5–13	1439–1447	K_1q^2	0.43	0.11	0.27	0.90	0.08	0.44	0.42	0.32	0.54
	QK1–3	1448.6–1467.6	K_1q^2	0.53	0.10	0.39	0.91	0.10	0.49	0.43	0.31	0.48
	QK3–1	1441.2–1446.6	K_1d	0.52	0.15	0.22	1.83	0.19	0.65	0.46	0.34	0.49
	SN107A	1331.8–1338.2	K_1sh	0.34	0.12	0.26	1.31	0.15	0.61	0.45	0.38	0.55
	QK143–1	1410–1433.4	K_1q^2	0.44	0.12	0.27	1.31	0.16	0.60	0.45	0.35	0.53
	QK21	1906.6–1926.6	K_1q^2	0.45	0.47	0.46	1.62	0.34	0.78	0.46	0.35	0.50

Note: 1. C_{24} tetracylic/C_{26} tricyclic terpane; 2. tricyclic terpanes/hopanes; 3. gammacerane/C_{30}hopane; 4. Ts/(Ts+Tm); 5. C_{30}diahopane/C_{30}hopane; 6. C_{29}Ts/C_{29}hopane; 7. C_{27}diasterane/C_{27}sterane; 8. C_{27}/C_{29} regular steranes; 9. C_{29} sterane20S/(20S+20R).

6 Classification of crude oils

Among the biomarker compounds, C_{29}Ts/C_{29} hopane and C_{30} diahopane/C_{30} hopane are good indicators of sedimentary environment, as well as effective parameters of oil-to-oil or oil-to-source rock correlations (Wang Chunjiang et al., 2000). It can be seen from Fig. 3 that crude oils in the Qinjiatun and Qikeshu region can precisely be divided into three types. Type Ⅰ oils are mainly distributed in Quantou and Denglouku formations of Qinjiatun oilfield which are represented by crude oil collected from QK1-2 well (Fig. 2). Type Ⅱ oils are located in Shahezi Formation of Qikeshu oilfield, including SW9 and SW11 oils; Type Ⅲ oils are also from Qikeshu oilfield, including SW8-1, SW8-2, SW8 oils, and mixed oils.

Distinguishing biomarker ratios and indices showing differences in the oil types are listed in Table 2. Different types of crude oils represent different genesis. Type Ⅰ oils are characterized by high C_{24}tetracyclic terpane/C_{26}tricyclic terpanes ratios, low gammacerance/C_{30}hopane ratios, tricyclic terpanes/hopanes ratios, C_{29}Ts/C_{29}norhopane ratios, and 17α(H)-diahopane/17α(H)-hopane ratios, indicating brackish lacustrine facies. Type Ⅱ oils show low C_{24}tetracyclic terpane/C_{26}tricyclic terpanes, high gammacerance/C_{30}-hopane ratios, tricyclic terpanes/hopanes ratios, C_{29}Ts/C_{29}norhopane and C_{30}diahopane/C_{30}hopane ratios, thus suggesting that they originated from source rocks deposited in a reducing-slight brackish lacustrine environment, or clay-rich sediments. Type Ⅲ oils have geochemical characteristics intermediate between those two types and may be the mixture of type Ⅰ and Ⅱ oils.

7 Conclusions

Based on the detailed analysis and description of the geochemical characteristics of crude oils, it can be found that, as a whole, oils from Qinjiatun-Qikeshu region are originated from source rocks deposited lacustrine environment, in the maturity stage, and have a large contribution of higher plants.

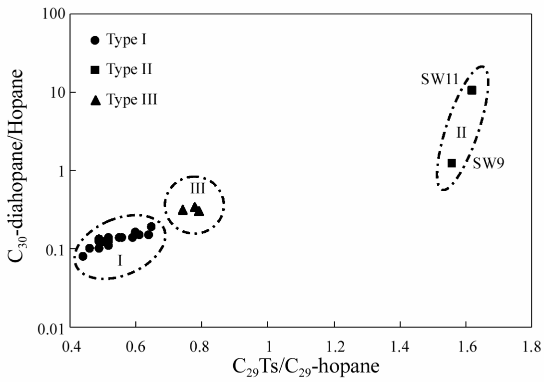

Fig. 3. Cross plot about parameters for dividing the oil types of crude oils. C_{29}Ts. 18α(H)-30-norneohopane; C_{29}Hopane. 17α(H)-30-norhopane; C_{30}-diahppane. C_{30}-17α(H)-diahopane; hopane. C_{30}-17α(H)-hopane.

Table 2 Distinguishing biomarker parameters for different type of crude oils from Qinjiatun-Qikeshu oilfields

Classification of oils	Parameter and compound	Type I oil	Type II oils	Type III oils (mixed oils)
Physical property	Color	Black	Yellow-tawny	Black
Classification parameter	Ts/Tm	<1.83	>3.33	Intermediate between I and II
	C_{29}Ts/C_{29}norhopane	<0.65	>1.56	Intermediate between I and II
	C_{30}diahopane/C_{30}hopane	<0.19	>1.22	Intermediate between I and II
	Gammacerane/C_{30}-hopane	<0.39	>0.69	Intermediate between I and II
	Tricyclic terpanes/hopane	<0.15	>1.40	Intermediate between I and II
Distribution pattern	Sesquiterpene distribution	C_{15}diadrime<drimane	C_{15}diadrimane >drimane	Intermediate between I and II
	Hopane distribution	Ts>Tm	Ts>>Tm	Ts>Tm
		C_{29}norhopane >C_{29}Ts >C_{30} diahopane	C_{29} norhopane <C_{29}Ts <C_{30} diahopane	C_{29}norhopane >C_{29}Ts ≤C_{30}diahopane
	Sterane distribution	C_{27}R>C_{28}R <C_{29}Rsterane	C_{27}R>C_{28}R <<C_{29}Rsterane	C_{27}R >C_{28}R <C_{29}Rsterane
Distribution area	Distribution region	Qinjiatun oilfields	SW9 and SW11wells of Qikeshu oilfield	SW8, SW8-1and SW8-2 wells of Qikeshu oilfield

According to the assemblage characteristics of biomarkers in crude oils and their geochemical implications, crude oils from Qinjiatun and Qikeshu oilfields of Lishu Fault Depression can be roughly classified as three types. The three types of crude oils are different in regional distribution. Laterally, Type Ⅰ crude oils occurred in Qinjiatun oilfield, Type Ⅱ and mixed crude oils distributed in Qikeshu oilfield. Vertically, Type Ⅰ oils were located in Quantou and Denglouku formations; Type Ⅱ oils appear in the lower layer of Shahezi Formation; and Type-Ⅲ (the mixed crude oils) are in the upper layer of Shahezi Formation.

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