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ORIGINAL ARTICLE

Oil geochemistry derived from the Qinjiatun–Qikeshu oilfields: insight from light hydrocarbons

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Abstract A suite of 27 oils from the Qinjiatun–Qikeshu oilfields in the Lishu Fault Depression of the Songliao Basin was analyzed using whole oil gas chromatography. In combination with the relative distribution of C_{27}, C_{28}, and C_{29} regular steranes, detailed geochemical analyses of light hydrocarbons in oil samples revealed crude oils characterized by the dual input of lower aquatic organisms and higher terrestrial plants. Several light hydrocarbon indicators suggest that the liquid hydrocarbons have maturities equivalent to vitrinite reflectances of around 0.78%–0.93%. This is consistent with the maturity determination of steranes C_{29} 20S/(20S + 20R) and C_{29} $\alpha\alpha\beta/(\alpha\alpha\alpha + \alpha\beta\beta)$. Crude oils derived from the two distinct oilfields likely both have source rocks deposited in a lacustrine environment based on light hydrocarbon parameters and on higher molecular weight hydrocarbon parameters. The results show that light hydrocarbon data in crude oils can provide important information for understanding the geochemical characteristics of the Qinjiatun–Qikeshu oils during geologic evolution.

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Keywords Light hydrocarbons · Crude oil · Lishu Fault Depression · Geochemistry characteristic

1 Introduction

Light hydrocarbon, a significant part of petroleum and natural gas, contains considerable geological-geochemical information, and thus plays an important role in the study of hydrocarbon accumulation. Schaefer and Leythaeuser (1980) used capillary columns gas chromatography to analyze hydrocarbons (C_2–C_8). Light hydrocarbons have received increasing attention as biomarkers for geochemical application. Light hydrocarbon parameters, such as heptane value, paraffin index, and methylcyclohexane ($MCyC_6$) index, may indicate original source, thermal maturity, and various secondary processes (Thompson 1979; Snowdon and Powell 1982; Mango 1987; George et al. 2002). Due to conventional chromatographic analysis of saturated hydrocarbons (starting with C_{13} hydrocarbons), light hydrocarbons are generally defined as between C_4 and C_{12}. However, scholars have used various definitions. Cheng et al. (1987) mainly analyzed composition of C_1–C_8 compounds, proposing an exponential relationship between paraffin index and heptane value. Dai (1993) suggested that light hydrocarbon refers to C_5–C_{10}, with boiling points below 200 °C. Thompson (1987) investigated the compositional regularities among isomeric groups of light hydrocarbons (C_2–C_8 compounds). A light hydrocarbon definition of C_1–C_9 was used to explore their origin by Mango (1997); Odden (1999) extended the range to C_{13} to study the natural and artificial generation of light hydrocarbons.

Some variations notwithstanding, the study of light hydrocarbon has focused on C_1–C_8 compounds. Light hydrocarbon analysis provides a host of invaluable information including maturity effect, biodegradation, and evaporation fractionation. Thompson (1983, 1987) and Mango (1990a, b, 1994) have reported on possible mechanisms controlling the composition of light hydrocarbons in petroleum; studies of molecular composition and distribution characteristics of light hydrocarbon have been successfully applied to genetic types and oil maturity evaluation.

Despite a relatively long history of oil exploration, the Lishu Fault Depression petroleum system remains comparatively under studied. Reports on the identification of source rocks (Zhang 2010), the dynamic evaluation of oil–gas resources (Zhang et al. 2013b), and the distribution of saturated hydrocarbon (Chen 2012; Zeng et al. 2014) have been published, but few papers evaluate the light hydrocarbons of crude oil within the system. Therefore, this study is focused on the distribution of light hydrocarbons, exploring the geochemical characteristics of organic matter input and thermal maturity of crude oil in the study area by C_6–C_8 compound composition.

2 Geological background

The Songliao Basin in northeastern China is one of the largest continental petroliferous basins, with a total area of about 260,000 km^2. Four major tectonic episodes in the Mesozoic and Cenozoic controlled the tectonic evolution and sedimentary filling of the basin (Li et al. 2013; Zhang et al. 2013a), which is divided into four structural belts including the central structural belt, northern slope belt, southeastern slope belt, and Sangshutai sag belt. The Lishu Fault Depression is a faulted basin with abundant oil and gas resources in the southeastern uplift of the Songliao Basin, regarded as an oil-bearing system. The Qinjiatun Oilfield is located in the Qinjiatun structure, situated in the eastern slope of the southeast uplift of the Songliao Basin (Fig. 1), and its main source strata are the Quantou and Denglouku Formations. The most advantageous source rock is the dark mudstone in the Shahezi and Yingcheng Formations. Based on oil-source rock correlation, Chen (2012) reported that the biomarker fingerprints of the Qinjiatun–Qikeshu oils resemble source rocks from the Shahezi and Yingcheng Formations with vitrinite reflectances (Ro) ranging from 0.7% to 2.0%. The Shahezi oil–gas reservoir in the Qikeshu oilfield is located at the edge of the central uplift belt in the southern part of the Songliao Basin, adjacent to the western Shiwu oilfield and eastern Qinjiatun oilfield. The main reservoirs in the Qikeshu oilfield are the Shahezi and Yingcheng Formations. The Qikeshu oilfield mainly developed in fan deltaic facies in the upper Shahezi Formation, with a lithology of fine sandstone and siltstone. The reservoir of the Yingcheng Formation is mainly located in the first and third sections, and the sand body was deposited in a braided river system.

3 Experimental methods

3.1 Samples

A suite of 27 crude oils was collected from the Qinjiatun–Qikeshu oilfields in the Lishu Fault Depression, of which 22 were derived from the Qinjiatun oilfield, and five from the Qikeshu. The Qinjiatun oils were selected from the Quantou, Denglouku, and Shahezi Formations, while the Qikeshu oils were exclusively from the Shahezi. Gas chromatography and gas chromatography–mass spectrometry (GC and GC–MS, respectively) were used for the analysis of oil samples in this study. A summary of well distribution in the study area is presented in Fig. 2.

3.2 Gas chromatography and gas chromatography–mass spectrometry

GC was performed with a 6890 N equipped with a HP-PONA fused silica capillary column (50 m × 0.20 mm i.d., film thicknesses 0.3 μm). GC was programmed to start at 300 °C, with the program initially holding 10 min at 35 °C, then increasing from 35 to 300 °C at a rate of 4 °C/min, and with a final hold of 50 min. Helium was used as the carrier gas at a rate of 1.0 mL/min.

GC–MS was carried out with a HP 5973 mass spectrometer, coupled to a HP 6890 GC equipped with a HP-5MS fused silica capillary column (30 m × 0.25 mm i.d., film thicknesses 0.25 μm). GC was programmed to start at 50 °C for 1 min, increase to 100 °C at a rate of 20 °C/min, and from 100 to 310 °C at a rate of 3 °C/min with a final hold of 18 min. Helium was used as the carrier gas with a rate of 1.0 mL/min and the ionization source operated at 70 eV. Saturated hydrocarbons were analyzed using GC–MS in full scan mode.

4 Results and discussion

4.1 Physical properties

The oil samples in the study area were mainly black, though some were yellow. The crude oils had density ranging from 0.83 to 0.90 g/cm^3 and wax contents between 19.1% and 39.56%, classifying them as high-wax crude oils. Sample viscosities were concentrated in the range of 6.26–24.77 mPa s (50 °C), with freezing points between

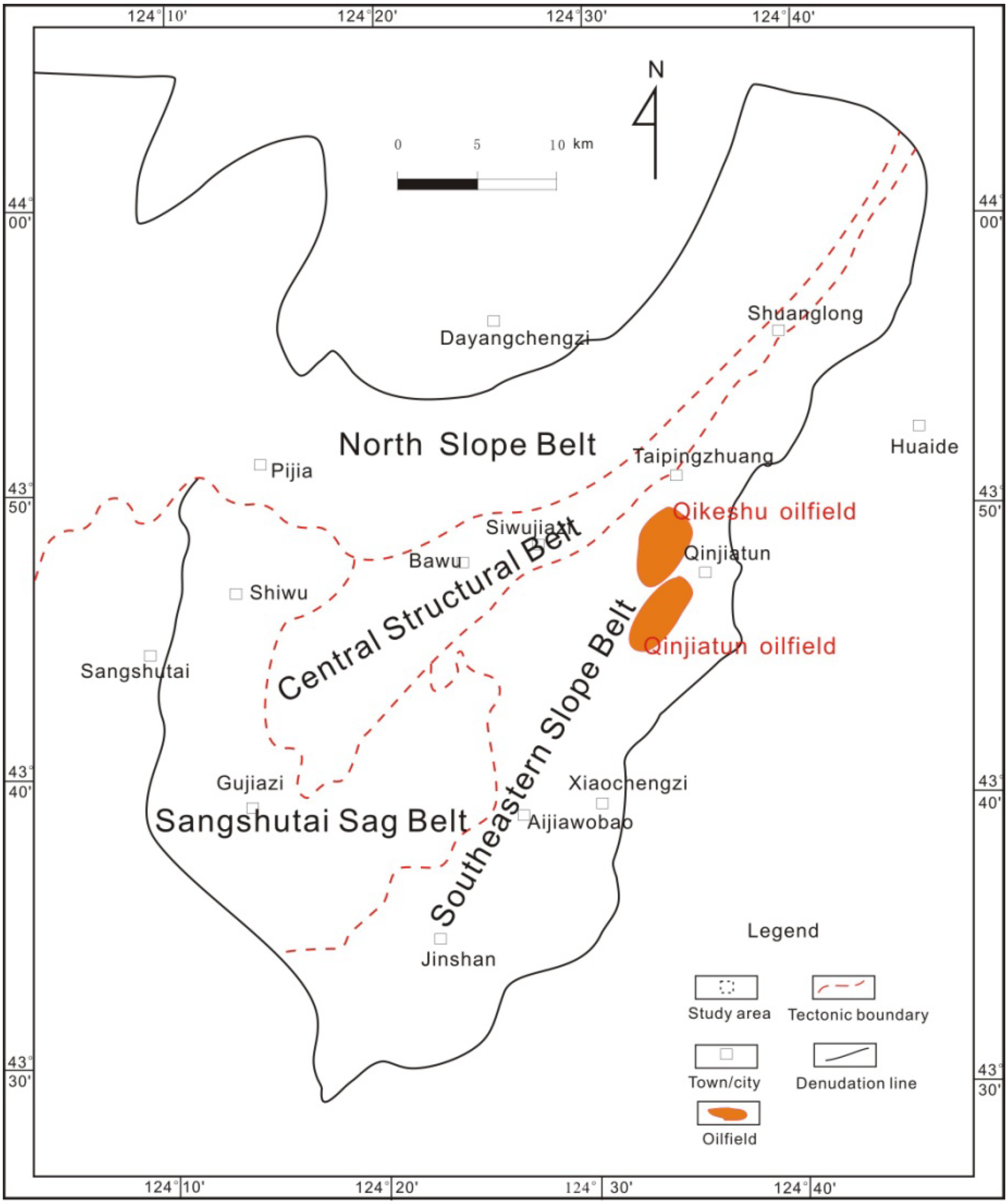

Fig. 1 Location of the Qinjiatun–Qikeshu oilfields in the Lishu Fault Depression of the Songliao Basin

11 and 24 °C. The sulfur contents were mostly below 0.1%, classifying the samples as low sulfur crude oils.

Light hydrocarbons, referring to the easily volatile fractions, include C_1–C_8 hydrocarbons in this study. Owing to the volatility of C_1–C_5 hydrocarbons at normal ground temperatures, C_6–C_8 compounds were analyzed to study the geochemical characteristics of crude oils from the Qinjiatun–Qikeshu oilfields in combination with higher molecular weight hydrocarbon based parameters.

4.2 Source of the light hydrocarbon fraction

Owing to different thermal stability and organic matter input, crude oils from different sedimentary sets contain various light hydrocarbon composition and relative contents. Light hydrocarbon composition can be used to determine the type of crude oils. Isoalkanes and cycloalkanes of C_6–C_8 hydrocarbons are delineated in Fig. 3. Some Qinjiatun and Qikeshu samples coincide,

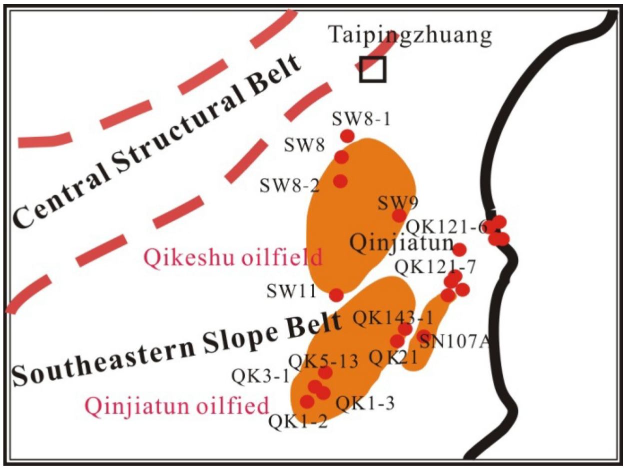

Fig. 2 Distribution of oil samples from the Qinjiatun–Qikeshu oilfields in the Lishu Fault Depression

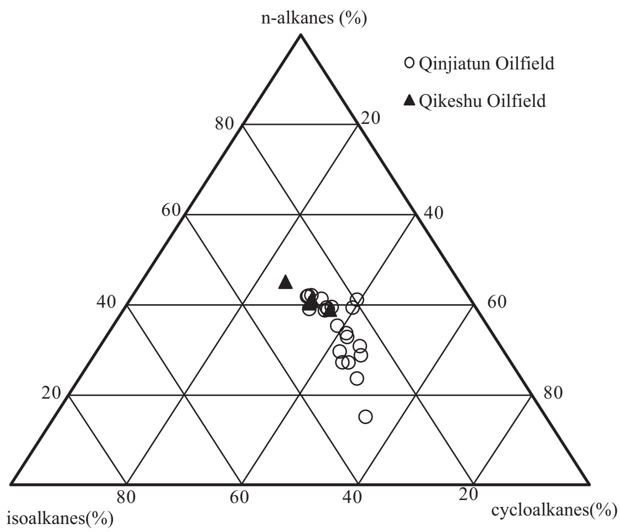

Fig. 3 Triangular diagram of C_6–C_8 *n*-alkanes, isoalkanes, and cycloalkanes in crude oils from the Qinjiatun–Qikeshu oilfields

indicating that some crude oils from the two oilfields contain similar geochemical characteristics. However, samples from the Qinjiatun oilfields were characterized by abundant cycloalkanes in the range of 30.09%–53.57%, with *n*-alkanes ranging from 15.24% to 42.26%, whereas Qikeshu oilfield samples had higher abundances of *n*-alkanes (ranging from 39.04% to 45.26%). Leythaeuser et al. (1983) reported that light hydrocarbons derived from sapropelic kerogen are rich in *n*-alkanes, while light hydrocarbon fractions derived from humic kerogen are rich in isoalkanes and aromatic hydrocarbons. In addition, terrestrial source matter is also characterized by abundant cycloalkanes. This suggests that the Qinjiatun–Qikeshu oilfields may contain terrestrial organic matter.

In addition, light hydrocarbon based parameters (Table 1) can be used to determine the source of crude oils in the study area.

In this study, C_7 light hydrocarbons include *n*-heptane (nC_7), $MCyC_6$, and dimethylcyclopentane ($DMCyC_5$). Different compositions of C_7 compounds often indicate different original source input as reported by Peters et al. (2005). In general, nC_7 is mainly derived from algae and bacterial lipids, is very sensitive to maturation, and could act as a good maturity index. $MCyC_6$ mainly comes from the lignin, cellulose, and carbohydrate of higher plants; contains relatively stable thermodynamic properties; and could act as a good indicator of original source input. In addition, its prominent existence is one of the characteristics of light hydrocarbon in coal-type gas. $DMCyC_5$ is mainly derived from the lipid compounds of aquatic organisms and is affected by maturation. Moreover, its existence is one of the characteristics of light hydrocarbon in oil-type gas. The relative compositions of nC_7, $MCyC_6$, and $DMCyC_5$ can be used to differentiate crude oils from various sources.

As shown in Fig. 4, C_7 compositions in crude oils from the Qinjiatun–Qikeshu oilfields are generally similar, and the contents of $DMCyC_5$ are relatively low. The contents of nC_7 and $MCyC_6$ in oils from the Qinjiatun oilfield are less than 40% and more than 50%, respectively. On the whole, the light hydrocarbon compositions indicate that the oils from the Qinjiatun–Qikeshu oilfields may have the characteristics of a dual input from aquatic organisms and terrigenous plants. Moreover, the Qinjiatun oils contain higher terrestrial organic matter input, consistent with the analysis of C_6–C_8 *n*-alkanes, isoalkanes, and cycloalkanes.

Hu et al. (1990) presented a formula to differentiate depositional environment and original source of hydrocarbon generation. The formula for calculating a $MCyC_6$ index of studied samples is $MCH = (MCyC_6 \times 100\%)/(MCyC_6 + nC_7 + DMCyC_5)$. It is proposed that MCH less than 35% suggests a semi-deep to deep lake facies, MCH in the range of 35%–50% indicates a shallow to semi-deep lake facies, MCH from 50% to 65% suggests shore to shallow lake facies, and MCH more than 65% indicates a variety of swamp facies. MCH was also used to determine the different types of crude oils and successfully establish three categories: marine, lacustrine, and coal-type oils (Zhang et al. 1999). Based on the standard by Hu et al. (1990), the source rocks of crude oils of the Qinjiatun and Qikeshu oilfields were deposited in shallow to semi-deep lake and shore to shallow lake facies, respectively.

On the classification and genesis of light hydrocarbons in crude oil, the Thompson and Mango theories are the most cited (Thompson 1979, 1983, 1987; Mango 1987, 1990a, b, 1994, 1997). Mango studied 2258 crude oil samples from all over the world (mainly North America)

Table 1 Hydrocarbon parameters of crude oils from the Qinjiatun–Qikeshu oilfields

Oilfield	Well	Depth (m)	Formation	Heptane (%)	Isoheptane (%)	C_7 composition (%)		
						nC_7 (%)	$MCyC_6$ (%)	$DMCyC_5$ (%)
Qikeshu	SW8	1927.56–1942.7	Shahezi	25.76	1.48	12.44	44.43	43.13
	SW8-1	1865.6–1867	Shahezi	28.31	1.41	12.77	44.58	42.65
	SW8-2	1962.8–2000.3	Shahezi	28.80	1.40	12.98	43.46	43.56
	SW9	2282.18–2295.08	Shahezi	0.00	0.00	4.29	51.20	44.50
	SW11	2595.2–2613.6	Shahezi	31.93	2.82	8.53	43.41	48.06
Qinjiatun	QK1-2	1127–1168.8	Quantou	31.67	2.70	11.11	43.65	45.23
	QK121-1	1001.3–1060.4	Quantou	17.32	1.73	12.94	63.08	23.98
	QK121-4	1024–1109.5	Quantou	22.05	1.35	11.84	60.25	27.91
	QK121-6	1069–1088	Quantou	11.42	1.93	14.69	68.42	16.90
	QK121-7	1009.4–1235	Quantou	14.95	1.88	12.79	66.39	20.82
	QK122-12	1008.8–1026.5	Quantou	36.52	2.71	10.43	39.45	50.12
	QK122-14	1013.8–1026	Quantou	34.65	2.46	10.29	42.67	47.04
	QK122-16	1003.6–1013	Quantou	36.20	2.79	10.49	39.58	49.93
	QK122-20	1011–1023	Quantou	36.04	2.71	10.57	49.65	39.78
	QK122-21	1062.9–1067.9	Quantou	16.39	1.62	16.42	60.47	23.12
	QK122-8	1015–1023.5	Quantou	14.43	1.73	15.91	63.74	20.35
	QK142-20	1016.2–1033	Quantou	31.90	2.33	10.77	44.95	44.29
	QK142-3	1130–1144	Quantou	31.67	2.36	10.83	44.11	45.06
	QK142-4	1053.4–1055.2	Quantou	31.60	2.34	11.14	43.89	44.98
	QK142-7	1132–1065	Quantou	5.65	1.57	17.21	74.61	8.19
	QK142-8	953–995.7	Quantou	31.96	2.32	10.49	44.87	44.65
	QK5-13	1439–1447	Quantou	26.88	1.75	13.51	50.56	35.93
	QK1-3	1448.6–1467.6	Denglouku	21.38	1.92	14.91	54.72	30.37
	QK3-1	1441.2–1446.6	Denglouku	22.57	1.80	14.04	54.87	31.09
	SN107A	1331.8–1338.2	Denglouku	28.74	2.20	8.84	52.90	38.26
	QK143-1	1410–1433.4	Denglouku	32.09	2.58	10.21	43.90	45.89
	QK21	1906.6–1926.6	Shahezi	21.03	2.14	11.63	58.88	29.49

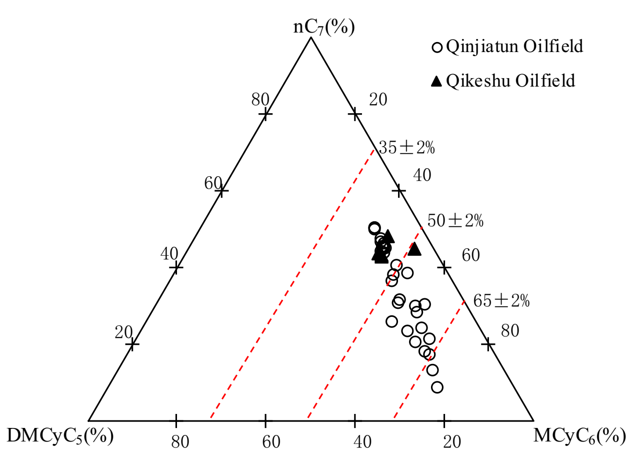

Fig. 4 Triangular diagram of C_7 light hydrocarbons in crude oils from the Qinjiatun–Qikeshu oilfields

and found that four different heptane compounds contained an interesting variation. Mango (1987) first reported the relative ratio of $K_1 = (2\text{-MH} + 2,3\text{-DMP})/(3\text{-MH} + 2,4\text{-DMP})$ remains invariant. It was suggested that K_1 could be used to determine whether crude oils and source rocks have a common origin. Figure 5 shows that K_1 values of Qinjiatun–Qikeshu crude oils oilfields are in the range of 0.72–1.40, but the majority are around 1.0, indicating similar organic matter input and uniform depositional conditions.

Together, the distribution of $MCyC_6$ index and K_1 of crude oils in the study area indicates that the source rocks of crude oils from the Qinjiatun–Qikeshu oilfields were mainly deposited in a shallow lake.

Selected geochemical results of the 26 oils are depicted Fig. 6. The relative distribution of C_{27}, C_{28}, and C_{29} regular steranes is represented in the form of a ternary diagram to show the original source of oils from the Qinjiatun–Qikeshu oilfields. Source input of organic matter undoubtedly has a strong influence on the sterane distribution of rock extracts and oils although there has been considerable debate as to the environmental significance of the

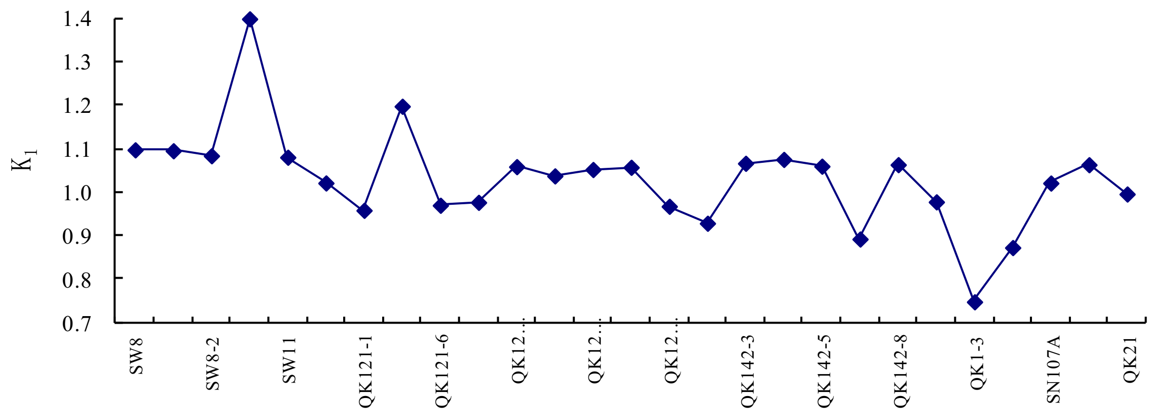

Fig. 5 K_1 values in crude oils from the Qinjiatun–Qikeshu oilfields

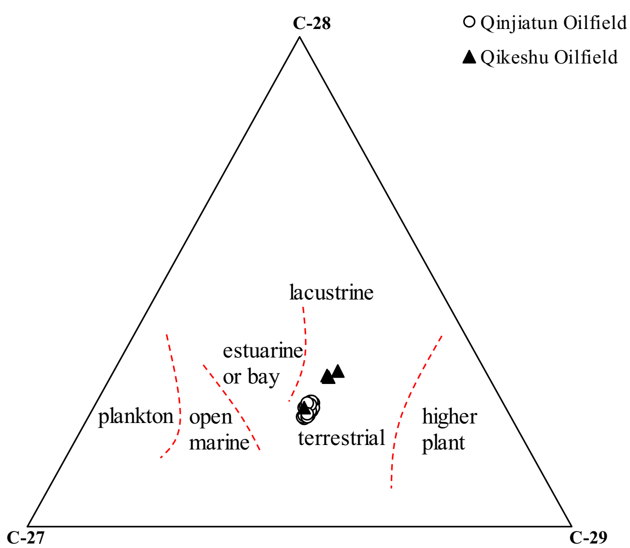

Fig. 6 Relationship between sterane compositions, source input, and depositional environment for crude oils from the Qinjiatun–Qikeshu oilfields (from Huang and Meinschein 1979)

distribution. The original classification of Huang and Meinschein (1979) related C_{27} steranes to strong algal influence and C_{29} to strong higher plant influence. Figure 6 suggests a close genetic relationship between the Qinjiatun and Qikeshu oils and agrees with the geochemical characteristics determined by other biomarkers (Zeng et al. 2014).

4.3 Maturity of crude oils based on light hydrocarbons

Light hydrocarbon based parameters such as heptane value and isoheptane value were applied to determine the maturity of crude oils from the Qinjiatun–Qikeshu oilfields. Thompson (1983) used plots of heptane versus isoheptane ratios as indicators of source, thermal maturity, and biodegradation. According to the different datasets in the plots, oils with heptane ratios from 18 to 22, 22 to 30, and > 30 were called normal, mature, and supermature, respectively. Philippi (1975) found that the degree of alkylation of light hydrocarbon increased with increasing maturity when studying the California basin. Thompson (1987, 1988) described evaporation fraction to account for certain-hydrocarbon distributions.

Generally speaking, the values of heptane and isoheptane are not only controlled by maturity, but also influenced by the original material type. Under the same maturity condition, heptane values of the oils generated from the sapropelic type kerogen are higher than those from the humic type kerogen. In addition, secondary alteration such as biodegradation and water washing could also change the heptane value. Heptane and isoheptane ratios could be used to define broad oil classifications. Because the parameters have such a strong source influence, their use in defining thermal maturity is limited to homogeneous suites of oils that did not experience significant reservoir fractionation or alteration.

The heptane and isoheptane contents of crude oils from the Qinjiatun oilfield were mainly distributed between 11.42% and 36.52%, and 1.35% and 2.79%, respectively. The sample from well QK142-7 had a lower value of heptanes with 5.75%. The heptane and isoheptane contents of crude oils from the Qikeshu oilfield were mostly less than 30% and 1.5%, respectively. The crude oil from well SW11 contained the highest heptane and isoheptane, which indicates that this crude oil was generated at a relatively higher maturity stage.

Based on Mango's light hydrocarbon parameters (1994), Bement et al. (1995) proposed that the ratio of 2,4-dimethylpentane to 2,3-dimethylpentane (2,4-DMP/2,3-DMP) can effectively reflect the oil generation temperature. Furthermore, the formula calculating the maximum temperature is $Tmax = 140 + 15 \times \ln(2,4\text{-DMP}/2,3\text{-DMP})$, and the maximum temperature can be converted into the corresponding vitrinite reflectance,

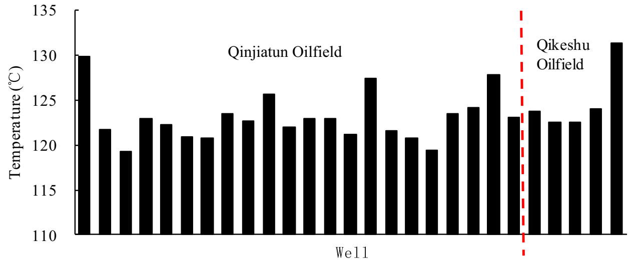

Fig. 7 Generated temperature of crude oils from the Qinjiatun–Qikeshu oilfields

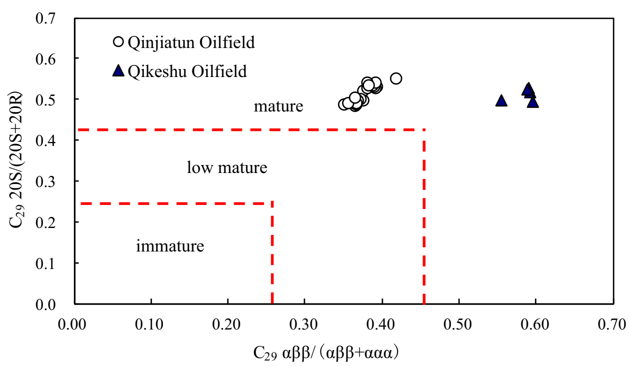

Fig. 8 Cross-plot of C_{29} 20S/(20S + 20R) and C_{29} αββ/(ααα + αββ) in crude oils from the Qinjiatun–Qikeshu oilfields

Rc = 0.0123 × Tmax − 0.6764. This calculation method of thermal maturity is almost unaffected by secondary alteration such as evaporation fractionation and biodegradation, resulting in a good correspondence with increasing maturity.

The calculated temperature of oil generation from the Qinjiatun oilfield was in the range of 118.1 to 130 °C, with an average of 122.9 °C, and corresponding Rc ranging from 0.78% to 0.92%, with an average of 0.83%. Similarly, the calculated temperature from the Qikeshu oilfield was 122.5 to 131.3 °C, with an average of 125.14 °C, and corresponding Rc of 0.83% to 0.93%, with an average of 0.86% (Fig. 7). It can be inferred that the studied crude oils are at mature stage, with Qikeshu oils having a similar maturity to the Qinjiatun oils.

Plots of 20S/(20S + 20R) versus αββ/(ααα + αββ) for the C_{29} steranes are particularly effective in describing the thermal maturity of crude oils. Figure 8 shows the correlation of thermal maturity parameters based on apparent isomerization of asymmetric centers in the C_{29} steranes for the oils generated from the Qinjiatun–Qikeshu oilfields. 20S/(20S + 20R) of the crude oils was between 0.48 and 0.60, with an average value of 0.52. αββ/(ααα + αββ) was in the range of 0.35–0.54, with an average value of 0.4. Figure 8 suggests Qinjiatun–Qikeshu oils are mature-stage, supporting the previous discussion based on light hydrocarbon parameters.

5 Conclusions

Geochemical characteristics of light hydrocarbon in crude oils from the Qinjiatun–Qikeshu oilfields were studied in combination with high molecular weight hydrocarbons. The results indicate that the crude oils from the Qinjiatun–Qikeshu oilfields contain higher contents of cycloalkanes and *n*-alkanes than isoalkanes of C_6–C_8 light hydrocarbon compounds. In addition, nC_7 contents in the crude oils were mostly less than 40%, and $MCyC_6$ contents more than 50%, indicating the dual input of lower aquatic organisms and higher terrestrial plants. Moreover, K_1 values near 1.0 serve as a good indicator of uniform sedimentary environment, which is consistent with the distribution of C_{27}, C_{28}, and C_{29} regular steranes. In combination with heptane and isoheptane values, maturity parameters of light hydrocarbons indicated Rc values in the range of 0.78%–0.92%, with an average value of 0.83%, indicating mature stage oils. This is supported by thermal maturity parameters based on C_{29} sterane isomerization. On the whole, the crude oils from the Qinjiatun–Qikeshu oilfields originated from lacustrine source rocks, at the mature stage, and had a dual organic matter input.

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