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Characteristics and Evaluation of the High-Quality Source Rocks of Cretaceous Continental Shale Oil in Tonghua Basin, China

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Abstract: The presence of shale oil in the Cretaceous Hengtongshan Formation in the Tonghua Basin, drilled by the well TD-01, has been discussed in this geological investigation for the first time. To evaluate the high-quality source rocks of Cretaceous continental shale oil, the distribution characteristics and the evolution of the ancient environment, samples of shale were systematically analyzed in terms of sedimentary facies, organic geochemistry, and organic carbon isotopic composition. The results demonstrate that a TOC value of 1.5% represents the lower-limit TOC value of the high-quality source rocks. Source rocks have an aggregate thickness of 211 m and contain abundant organic matter, with TOC values of 2.69% on average and a maximum value over 5.44%. The original hydrocarbon-generative potential value (S_1+S_2) is between 0.18 mg/g and 6.13 mg/g, and the R_o is between 0.97% and 1.40%. The thermal maturation of the source rocks is relatively mature to highly mature. The $\delta^{13}C$ value range is between −34.75‰ and −26.53‰. The ratio of saturated hydrocarbons to aromatic hydrocarbons is 1.55 to 5.24, with an average of 2.85, which is greater than 1.6. The organic types are mainly type II_1, followed by type I. The organic carbon source was C_3 plants and hydrophytes. The paleoclimate of the Hengtongshan Formation can be characterized as hot and dry to humid, and these conditions were conducive to the development of high-quality source rocks. A favorable paleoenvironment and abundant organic carbon sources provide a solid hydrocarbon generation base for the formation and accumulation of oil and gas in the shale of the Tonghua Basin.

Key words: evaluation of high-quality source rocks, Cretaceous continental shale oil, formational environment, Tonghua Basin

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1 Introduction

Shales bear abundant mineral resources and unconventional petroleum resources, often containing substantial organic matter required for a potential source rock. However, due to intense tectonic deformation, shale can also act as a fractured reservoir. The oil found at high concentrations in shale is called “shale oil.” There is not yet a widely accepted definition of “shale oil.” Some scholars think that shale oil can be considered mature oil specifically in the shale, and others consider shale oil to be equivalent to tight oil on a broader scale. Shale oil is an important component of the unconventional petroleum resource of shale (Regtop et al., 1982; Rovere et al., 1983; Guo and Zhu, 1995; Aamir et al., 2017; Blaizot, 2017; Cui Jingwei et al., 2017; Liu et al., 2017). With continued experience in petroleum exploration, petroleum workers have gradually realized that the study of source rocks is an important aspect of petroleum systems research on shale oil, (Peters, 1986; Farouk et al., 2016; Arora et al., 2017; Silva et al., 2017; Synnott et al., 2017). The scale and distribution range of highly abundant, high-quality source rocks play direct roles in controlling the size and distribution of shale oil. Therefore, the evaluation of high-quality source rocks is an important research direction.

The Tonghua Basin is a typical continental lacustrine basin, and the following issueshave become important research goals: whether the Tonghua Basin has a good material base and stable lacustrine sedimentary environment and whether the Tonghua Basin experienced conditions favorable for the formation of shale oil with a high exploration value. Although basic geological studies, such as detailed gravitational and magnetic exploration, have been conducted in some areas of the Tonghua Basin recently (Wang Yubo et al., 2011), these studies suffered from limited exploration, weak research, unclear resource potential, unclear exploration direction and uncertain petroleum resource prospects. As the first geological survey well in the Tonghua Basin, the TD-01 well represents an important breakthrough in research on the Tonghua Basin. Evidence of petroleum was detected for the first time in the drilled strata of the Hengtongshan Formation, and oily cores were successfully recovered. These findings revealed for the first time that shale is present in the strata of the Hengtongshan Formation. The shale contains high organic matter with characteristic of low porosity and permeability (Fig. 1; Table 1). The shale provides a high-quality source rock and contains tight reservoirs of shale oil. The above findings provided the

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Fig. 1. Photographs of shale cores in the Hengtongshan Formation, Tonghua Basin.

Table 1 Porosity and permeability of shale in the Hengtongshan Formation of the Tonghua Basin

Sample No.	Lithology	Depth (m)	Porosity (%)	Permeability (mD)
TD1-83	shale	417.70	2.2	0.0501
TD1-192	shale	248.50	2.4	0.0113
TD1-227	shale	193.10	2.0	0.0114
TD1-231	shale	188.35	6.8	0.0195
TD1-235	shale	161.15	2.7	0.0256

Note: Test method is mercury intrusion porosimetry (MIP).

basis for studying the characteristics and reconstructing the formational environment of the high-quality source rocks in the Hengtongshan Formation and provided more precise information for use in researching the shale oil in this formation. Therefore, organic geochemical tests were conducted on the newly discovered source rocks from the Hengtongshan Formation, and the high-quality source rocks and their formational environment were studied in combination with the geological drilling data (Wang Dandan et al., 2017). The aim was to promote a breakthrough in petroleum exploration in the Tonghua Basin as well as in other Mesozoic residual basin groups in eastern Jilin Province.

2 Geological Settings

The Tonghua Basin is located in southeastern Jinlin Province and stretches from Tonghua City in the east to Wangqingmen Town of Liaoning Province in the west and from Sanyuanpu Town of Liuhe County in the north to Tonghua County in the south. The basin formed in the middle Jurassic to late Cretaceous and experienced the Mesozoic Indosinian movement and Yanshan movement. Because of the effects of subduction of the Pacific Plate beneath the Euro-Asian continent, the development of the Tonghua Basin was controlled by a northeast-trending fault zone from Sanyuanpu to Xiangzi, which formed an asymmetric synclinal basin with extension to the northeast (Xu Min et al., 2013). The basin can be divided into two secondary tectonic units, i.e., the Sankeyushu-Sanyuanpu depression and the Yingebu uplift (Han Xinpeng et al., 2013). The well TD-01 is located in the southeastern Sankeyushu depression (Fig. 2). The lithology of the basin basement includes various Archaean and Proterozoic metamorphic rocks. For example, the Proterozoic Anshan group is a metamorphic series composed mainly of migmatite, granulite, and other similar lithologies (Jiang Weiwei et al., 2006). The sedimentary cover layer is composed of pre-Mesozoic marine sedimentary rocks and Mesozoic continental classic sedimentary rocks. The Cretaceous Yingzuilazi Formation, Xiahuapidianzi Formation and Hengtongshan Formation are composed of

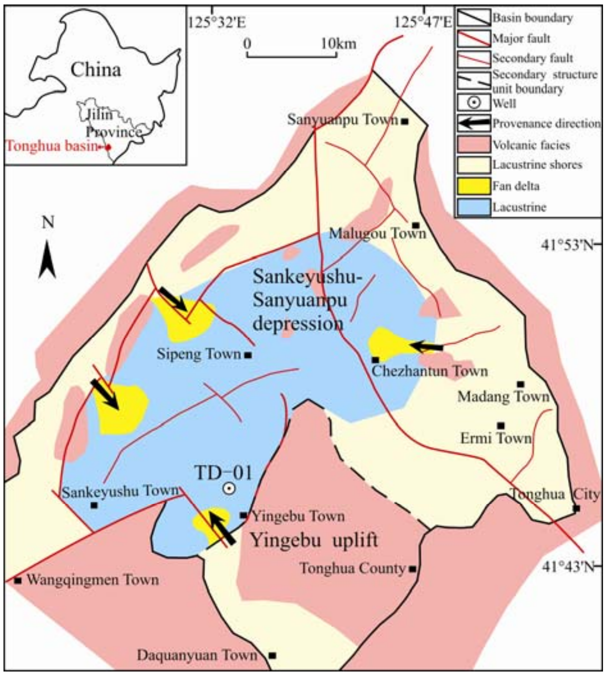

Fig. 2. Distribution of tectonic features and sedimentary facies of the Hengtongshan Formation and the studied well of the Tonghua Basin (modified from Han Xinpeng et al., 2013).

mudstones, shales and sandstones (Li Dongjin et al., 1988).

3 Samples and Methods

The cores of the whole Well TD-01 were observed, classified, described and recorded on site. Ninety-seven black shale samples in the Hengtongshan Formation of the Tonghua Basin were collected at an average interval of 5 meters from depth 59 m to 599 m. Organic geochemical tests, i.e., rock pyrolysis, TOC value analysis, organic carbon isotopic tests and so on, were performed on 97 samples to further analyze the characteristics and distribution of high-quality source rocks in the Hengtongshan Formation (da S. Ramos et al., 2015; Cao et al., 2017; Liang et al., 2017; Baiyegunhi Christopher et al., 2018). Sixty-six samples were undertaken for organic carbon isotope and TOC value by using the Vcrio PYRO Cube elemental analyzer and the Isoprime 100 isotope mass spectrometer with the Chinese petroleum industry standard SY/T 5238-2008 at the National-Local Joint Engineering Laboratory of In-situ Conversion, Drilling and Exploitation Technology for Oil Shale. According to Chinese national standard GB/T 18602-2012 and petroleum industry standards SY/T 5124-2012, SY/T 5118 -2005, SY/T 5119-2008 to complete the rock pyrolysis analysis of 44 samples, R_o value analysis of 31 samples, bitumen “A” analysis of 24 samples and group component analysis of 10 samples respectively at the Geochemistry Laboratory of Yangtze University.

4 Results

4.1 Classification standards for high-quality source rocks

High-quality source rocks usually feature high contents of certain organic matter types and have medium or high maturities. Different scholars follow different classification standards for high-quality source rocks (Table 2) (Peters, 1986; Jarvie, 1991; Zheng Hongju et al., 2007; Pang Xiongqi et al., 2009; Chen Jianping et al., 2012). The lower limit of the total organic carbon (TOC) values of the high-quality source rocks was determined using the method developed by Wang Zhensheng (2014), which improved upon the plot method of Lu Shuangfang (2012). The lower-limit TOC value was assigned to the inflection point in the curve between the chloroform bitumen “A” content and the TOC value at which further increases in the TOC value do not produce significant increases in the chloroform bitumen “A” content.

Table 2 The evaluation criterion of source rocks in the Hengtongshan Formation, Tonghua Basin

Name	TOC (%)				
Peters	Poor 0–0.5	Fair 0.5–1.0	Good 1.0–2.0	Very good >2.0	
Chen Jianping	Poor 0–0.5	Fair 0.5–1.0	Good 1.0–2.0	Very good 2.0–5.0	Excellent >5.0
Zheng Hongju			Effective 1.0–2.0	High quality 2.0–5.0	
Pang Xiongqi	Poor 0–0.5	Fair 0.5–1.0	High quality II 1.0–2.0	High quality I >2.0	
Tonghua Basin		General quality 0.5–1.0	Medium quality 1.0–1.5	High quality ≥1.5	

As the chloroform bitumen “A” content ceases to increase significantly and the curve flattens when the TOC value of the samples of Hengtongshan Formation reaches 1.5% (Fig. 3), the lower-limit TOC value of the high-quality source rocks is set to 1.5%. Meanwhile, the R_o values of the source rock samples are between 0.97% and 1.40%, with an average of 1.10% (Fig. 4), indicating a mature stage for the source rocks (Wang Libo et al., 2006; Ding Wenlong et al., 2011; Jing et al., 2014). Therefore, it is scientifically reasonable to set the lower-limit TOC value of the high-quality source rocks to 1.5%.

4.2 Distribution of high-quality source rocks

There are five layers of high-quality source rocks in the form of black shales in the semi-deep to deep lacustrine facies in the middle and upper portions of the formation and shales in the fan delta facies of the lower portion of the formation. The aggregate thickness of the high-quality source rocks is 211 m, representing approximately 73.3% of the total thickness of the source rocks. The middle layer has the largest single-layer thickness of 100 m, and the thickness of the other layers is approximately 30 m (Fig. 5).

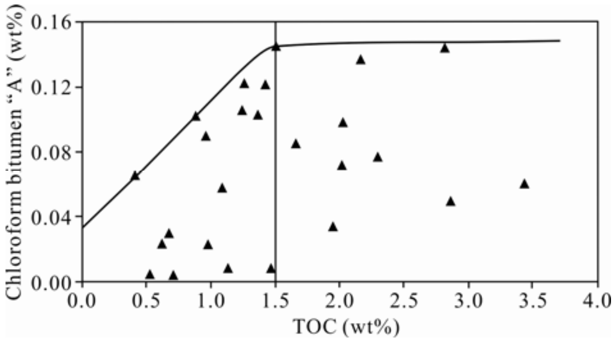

Fig. 3. Relationship between chloroform bitumen “A” and TOC in the source rocks of the Hengtongshan Formation, Tonghua Basin.

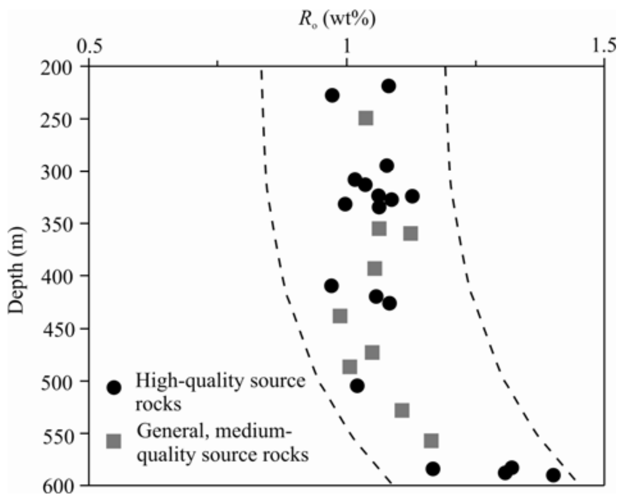

Fig. 4. Relationship between R_o and depth of source rocks in the Hengtongshan Formation, Tonghua Basin.

4.3 Geochemical characteristics of high-quality source rocks

The test results showed that the TOC values of the high-quality source rocks were between 1.51% and 5.44%, and the average was 2.69%. The original hydrocarbon-generative potential values (S_1+S_2) were 0.18–6.13 mg/g, with an average of 3.23 mg/g. The R_o value was 0.97%–1.40%, with an average of 1.10%, representing source

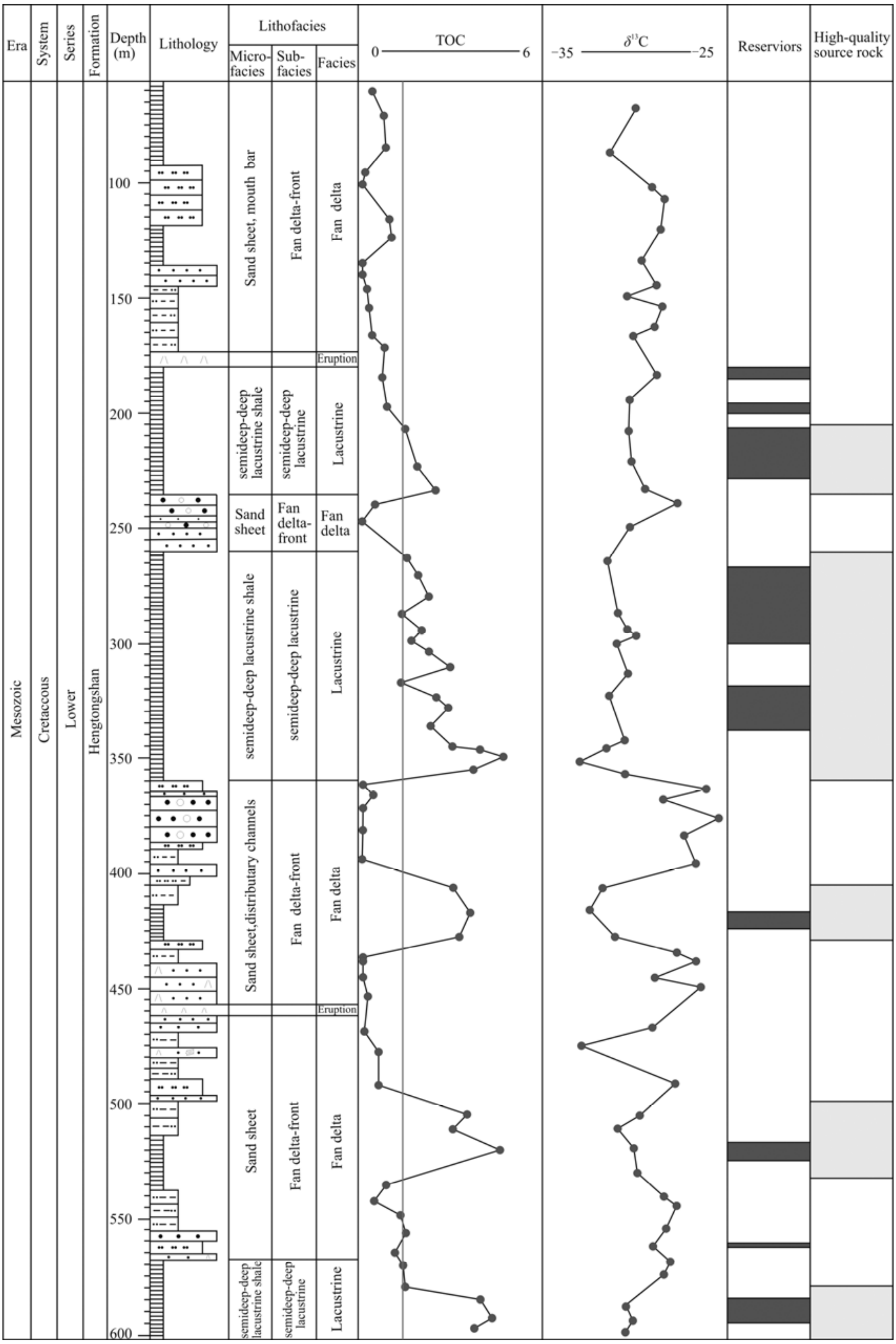

Fig. 5. Comprehensive stratigraphic column of the Hengtongshan Formation, Tonghua Basin.

rocks in the mature or highly mature stage (Gao et al., 2018). Compared with similar shale oil in other basins, the parameters of the high-quality source rocks in the Hengtongshan Formation are approximate (Table 3) (Jing et al., 2014; Liu Bo et al., 2014; Gao et al., 2017; Li Shizhen et al., 2017). The plot of pyrolysis T_{max} vs. production index (PI) shows source rocks in the main stage of hydrocarbon generation (Liu et al., 2018) (Fig. 6). The relative content of saturated hydrocarbon fractions is dominant in the high-quality source rocks. The ratio of saturated hydrocarbons to aromatic hydrocarbons is 1.55–5.24, with an average of 2.85, which is greater than 1.6. The organic matter type is mainly composed of humic-sapropelic kerogen (II_1), followed by sapropelic kerogen (I) (Table 4) (Liu Zhaojun et al., 2009). The carbon isotopic values ($\delta^{13}C$) of the kerogen in the high-quality source rocks are −26.53‰–−34.75‰, with an average of −29.94‰. The organic matter types are mainly kerogen type II1 followed by kerogen type I based on the kerogen $\delta^{13}C$ classification standard of organic matter types (I: −35‰–−30‰, II_1: −30‰–−27.5‰, II_2: −27.5‰–−25‰, III: >−25‰) given by Huang Jizhong et al. (1988) and Chen Mengjin et al. (2007). These results are consistent with the conclusions of the rock pyrolysis analysis (Fig.7). (Liu Chunlian et al., 2010; Sun et al., 2014; Gajica et al., 2017; Abarghani et al., 2018).

Based on the organic geochemical test results and stratigraphic sequence, the high-quality source rocks is divided into two portions by depth 360 m: The high-quality source rocks in the bottom and top of the Hengtongshan Formation. The thick layer of high-quality source rocks present in the top of the Hengtongshan Formation in the Tonghua Basin is composed of black shale. The high-quality source rocks are characterized by large thicknesses, good continuity and high organic matter contents (with TOC values greater than 2.42%). The kerogen types are mainly type II_1, followed by type I (Fig. 7). The average value of R_o is 1.05%, indicating that the source rocks are in the mature stage and that the high-quality source rocks have excellent hydrocarbon generation potential.

Table 3 Comparison of parameters of the shale oil in different basins

Basin/areas	Strata	TOC (%)	R_o (%)	S_1+S_2 (mg/g)	T_{max} (°C)
Liaohe western depression	Shahejie Formation	2.55	0.60		440
Sognliao Basin	Qingshankou Formation	2.13	1.00	13.61	448
Junggar Basin	Baijiantan Formation	1.53	0.93	2.35	444
Tonghua Basin	Hengtongshan Formation	2.69	1.10	3.23	459

The high-quality source rocks at the bottom of the Hengtongshan Formation are mainly distributed in units of interbedded sandstones and shale in fan delta facies, and a few of them are associated with semi-deep to deep lacustrine facies. Their lithologies are mainly black shale

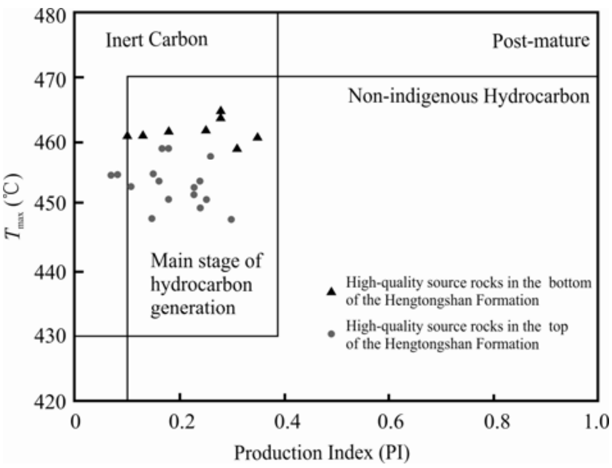

Fig. 6. Plot of pyrolysis T_{max} vs. production index (PI) of the high-quality source rocks in the Hengtongshan Formation, Tonghua Basin.

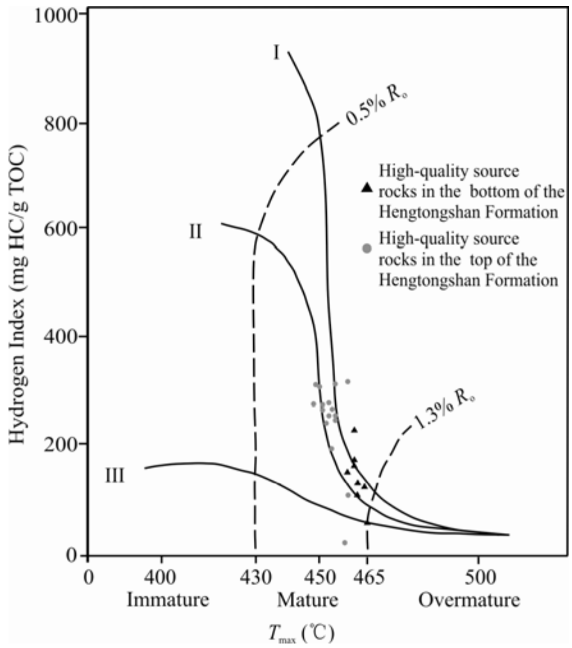

Fig. 7. Van Krevelen diagram for the high-quality source rocks in the Hengtongshan Formation, Tonghua Basin.

Table 4 Group compositions of the high-quality source rocks in the Hengtongshan Formation, Tonghua Basin

Portion	Sample No.	Saturated hydrocarbon (%)	Aromatic hydrocarbon (%)	Nonhydrocarbon (%)	Asphaltine (%)
Bottom	TD1-75	43.03	26.63	11.46	9.29
	TD1-77	58.08	19.62	8.46	3.46
	TD1-82	58.82	17.65	11.76	17.65
	TD1-89	64.54	17.17	7.48	5.82
Top	TD1-116	54.65	18.82	9.52	3.85
	TD1-120	64.61	12.33	7.36	1.79
	TD1-124	44.63	28.78	12.20	10.24
	TD1-128	53.96	25.22	10.26	4.99
	TD1-137	56.65	20.23	9.83	2.02
	TD1-156	49.24	22.66	10.27	3.63

and a few gray-black silty mudstone, and they are relatively thin. However, the organic matter contents are high (with TOC values greater than 3.25%), and the organic matter types are mainly type I and II_1 (Fig. 7). The average value of R_o is 1.11%, indicating a high maturity. Consequently, the quality of these rocks is better than the top rocks, but the thickness is thinner.

5 Discussions

5.1 Sedimentary environment

The quality of source rocks was controlled by the characteristics of the sedimentary environment (Pederson and Calvert, 1990; Lin Junfeng et al., 2015; Cheng Chen and Gao Liang, 2017). According to the observations of the TD-01 drill cores, the lithologies of the source rocks in the Hengtongshan Formation are mainly gray-brown shale and black shale. Among these source rocks, the high-quality source rocks are generally black shale and silty mudstone (Fig. 8a).

Analysis of the original sedimentary structure preservation revealed continuous horizontal bedding of shale in the high-quality source rocks in the top of the Hengtongshan Formation (Fig. 8b). The sedimentary environment featured calm and oxygen-poor conditions (Tao et al., 2013; Gao et al., 2017; Li et al., 2017). Well-preserved animal and plant fossils have been found in outcrops of the high-quality source rocks section and in the drill cores (Fig. 8c), indicating that the sedimentary environment was a typical stable semi-deep to deep lacustrine environment (Cheng Lixue et al., 2013).

The stratigraphic continuity of the high-quality source rocks is poor in the bottom of the Hengtongshan Formation, as deposition of shale rocks were disrupted or destroyed by sandstone (Fig. 8d), indicating that the sedimentary environment became less stable. The study of the tectonic evolution of the basin suggests that large-scale wet alluvial fan facies formed along a fault zone via flooding and perennial running water at that time. In conjunction with fault activity, the alluvial fan facies entered the lake basin, forming an alluvial fan-fan delta facies (Fig. 1). Considerable terrigenous organic matter

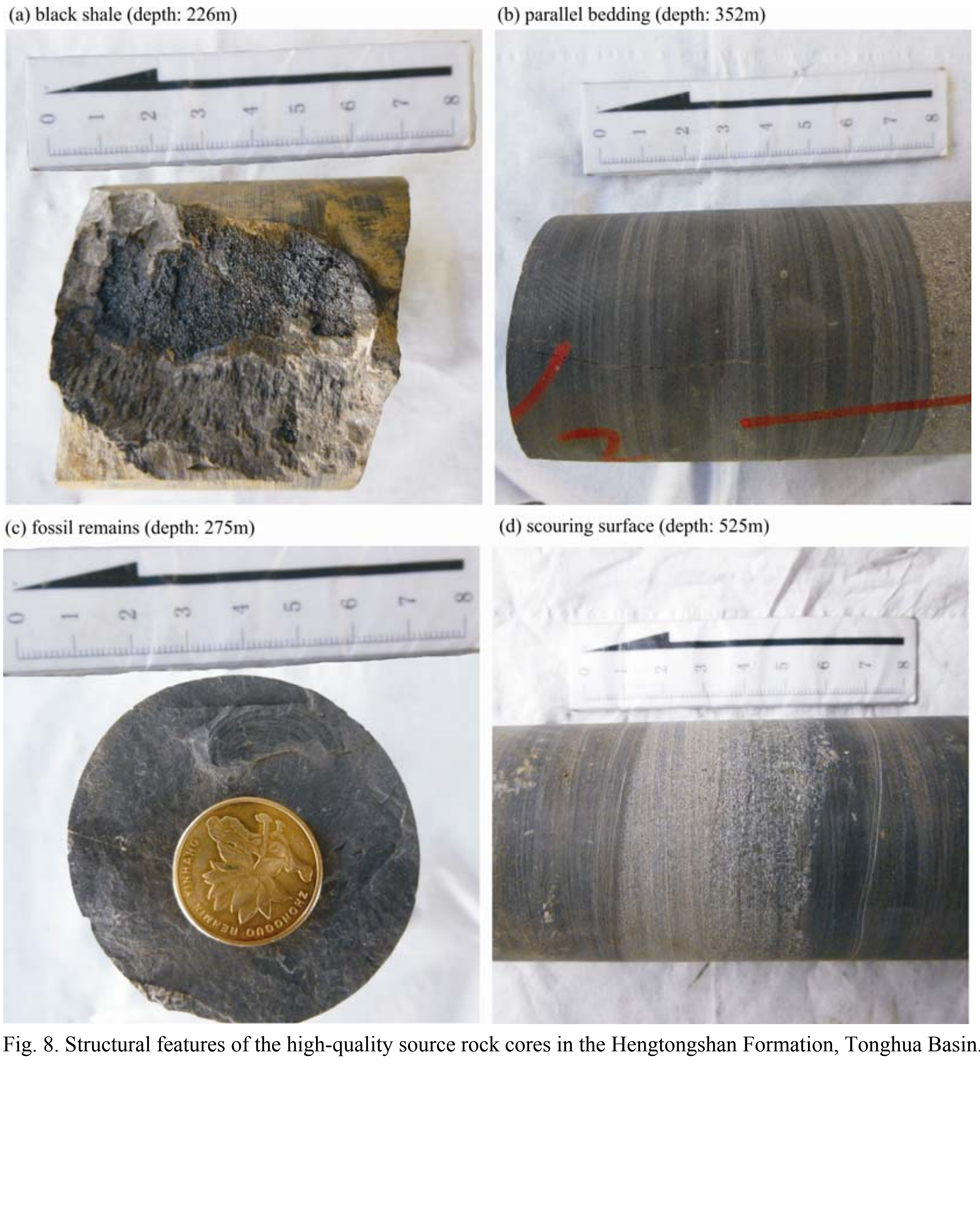

Fig. 8. Structural features of the high-quality source rock cores in the Hengtongshan Formation, Tonghua Basin.

was injected into the lake basin, resulting in unusually high organic matter contents (Song et al., 2017). However, the geochemical data from the collected samples indicate that the TOC contents of the samples are greater than 3.25% on average; hence, the organic matter was not completely destroyed by later tectonic activity or alteration. The combination of the semi-deep to deep lacustrine facies and the fan delta facies in the Hengtongshan Formation in the study area created a sedimentary environment conducive to forming high-quality source rocks.

5.2 Paleoclimatic environment

The $\delta^{13}C$ values of organic carbon have been widely used as a proxywhen reconstructing paleoenvironmental evolution (Chen Lan et al., 2017; Wei et al., 2018; Xu et al., 2018). The ranges of $\delta^{13}C$ values differ for different organic carbon sources. Terrestrial vegetation can be classified into three types—C_3, C_4 and CAM—according to the photosynthetic pathways of different plants (Attendorn and Bown, 1988; Liu Zhaojun et al., 2009), and the corresponding $\delta^{13}C$ ranges are −34‰–−23‰, −22‰–−6‰ and −20‰–−10‰, respectively (Luo Chao et al., 2008). Aquatic plants are more complex, and they can be divided into submerged plants and floating plants (including phytoplankton, floating-leaved plants and emergent aquatic plants) with different $\delta^{13}C$ values (Pearson and Coplen, 1978). Submerged plants commonly adopt HCO_3^- as a carbon source, and the $\delta^{13}C$ values range from −12‰–−20‰. Floating plants use CO_2 from the air as the carbon source, and their $\delta^{13}C$ values are close to those of C_3 plants and are thus more negative than those of submerged plants (Aravena et al., 1992; Gao et al., 2016). The $\delta^{13}C$ values decrease when the water temperature decreases and when the climate is cool and humid. The $\delta^{13}C$ values increase when the water temperature increases and when the climate is warm and dry (Zhao Xinwen et al., 2014).

According to the analysis of the Hengtongshan Formation samples obtained from well TD-01, the $\delta^{13}C$ values range from −34.86‰–−26.01‰. Therefore, the organic carbon sources were mainly terrigenous C_3 plants and aquatic floating plants. Furthermore, based on the distribution of the $\delta^{13}C$ values, the climate in the Tonghua Basin during the deposition of the Hengtongshan Formation changed from hot and dry to warm and humid and then back to hot and dry.

The lower portion of the Hengtongshan Formation features semi-deep to deep lacustrine facies deposits, relatively high TOC contents in the shales, and minor fluctuations in the $\delta^{13}C$ curve. Therefore, the syndepositional paleoclimatic conditions were cool and humid and the water temperature was low and stable. In contrast, the fluctuation range of the $\delta^{13}C$ values in the fan delta sedimentary environment is large and the $\delta^{13}C$ values in the non-high-quality and high-quality source rocks sections are relatively high and low, respectively. The abrupt negative trend indicates that the water effect weakened and that the sedimentation effect strengthened under a warm and humid climate during this period. In the middle Hengtongshan Formation, the $\delta^{13}C$ values of the thick layer of high-quality source rocks in the semi-deep to deep lacustrine sedimentary system exhibit an obvious negative trend and then stabilize. The stable $\delta^{13}C$ values indicate that the Tonghua Basin changed from fan delta to lacustrine facies sedimentation quickly because of tectonic activity. The sedimentary environment was very stable, and the climate type was warm and humid. The negative trend in the $\delta^{13}C$ values indicates that the CO_2 content in the air increased, which was beneficial to the growth of aquatic organisms. Saprophytic algae account for the majority of the maceral composition, indicating that the organic carbon sources varied primarily from terrigenous fragments to aquatic floating plants. The variation in the $\delta^{13}C$ values decreases in the upper Hengtongshan Formation, indicating that the organic carbon sources changed from aquatic plants to terrigenous fragments and that the climate was warm and humid.

Overall, the climate of the Tonghua Basin during the deposition of the Hengtongshan Formation changed from hot and dry to warm and humid. The development of high-quality source rocks was controlled by this change in the ancient environment. During the formation of the high-quality source rocks, the climate was humid and the temperature was mild. Hence, aquatic plants flourished in the lake, while higher plants flourished in the surrounding continental areas.

6 Conclusions

(1) The thick, high-quality source rocks of the Hengtongshan Formation in Tonghua Basin were detected for the first time in well TD-01. According to the organic geochemical test results, the lower-limit TOC value of the high-quality source rocks was determined as 1.5% according to the plot method.

(2) The high-quality source rocks of the Hengtongshan Formation in the Tonghua Basin are mainly thick black shales associated with semi-deep to deep lacustrine facies in the upper portions and shales associated with fan delta facies in the lower portion. The aggregate thickness of the high-quality source rocks is 211 m. These rocks have TOC values between 1.51% and 5.44%, with an average value of 2.69%. The original hydrocarbon-generative potential value (S_1+S_2) is 0.18–6.13 mg/g, with an average of 3.23 mg/g. The R_o value is 0.97%–1.40%, with an average of 1.10%, indicating a mature to highly mature stage. The $\delta^{13}C$ values of the kerogen in the high-quality source rocks range from −26.53‰–−34.75‰, with an average of −29.94‰. The organic matter types are mainly kerogen type II_1, followed by kerogen type I.

(3) The sedimentary environment of the high-quality source rocks was relatively stable. The climate was humid, and the temperature was mild. These conditions were favorable for the generation of high-quality hydrocarbon source rocks.

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