Geochemical characteristics and hydrocarbon expulsion of source rocks in the first member of the Qingshankou Formation in the Qijia-Gulong Sag, Songliao Basin, Northeast China: Evaluation of shale oil resource potential

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Abstract
In recent years, there has been significant progress in shale oil exploration in the first member of the Qingshankou Formation (K2qn1) in the Qijia-Gulong Sag, Songliao Basin, Northeast China: It shows good prospects for shale oil. However, the recognized lack of the geochemical and hydrocarbon generation and expulsion characteristics of K2qn1 source rocks limits an accurate evaluation of shale oil resource. This study systematically investigated the geological and geochemical characteristics, hydrocarbon generation and expulsion, and shale oil potential of the K2qn1 source rocks. The results show that the K2qn1 mudstones were mainly deposited in the semideep and deep lacustrine facies under reducing and weak reducing conditions. Compared with the southern Gulong Sag, the northern Qijia Sag has a higher salinity, more abundant prosperous aquatic organisms, and a greater paleoproductivity. The K2qn1 source rocks are pervasive and continuous in the entire sag, with maximum thicknesses greater than 110 m. They have a higher organic matter (OM) abundance (2.40% of the average TOC), are dominated by type I and II1 kerogen, and are mature (0.8%-0.1.3% VR), which indicate that they are good to excellent source rocks and have significant hydrocarbon generation potential. The threshold and peak hydrocarbon expulsion values for marlstone source rocks are 0.85% VR and 0.95% VR, respectively. The volumes of hydrocarbons generated and expelled from the K2qn1 source rocks are 121.8 × 108 t and 46.9 × 108 t, respectively, with a retention efficiency of 61.5%. The in-place and recoverable resources of shale oil are 74.9 × 108 t and (12.0-13.5) × 108 t, respectively, indicating that the entire sag has a significant shale oil potential, especially the Qijia Sag.

KEYWORDS
first member of the Qingshankou Formation, geochemical characteristics, hydrocarbon expulsion, Qijia-Gulong Sag, shale oil potential, source rocks
Shale oil refers to petroleum retained (unexpelled) in micropores within organic-rich shales.\textsuperscript{1,2} Shale oil has been successfully massively developed in North America and has been becoming a new hotspot after shale gas in global unconventional hydrocarbon exploration and development.\textsuperscript{2,3} Unlike shale oil in North America which is found in marine deposits, shale oil in China is mainly distributed in continental Mesozoic and Cenozoic shale strata.\textsuperscript{4-6} Out of 41 countries in the world, China has the third largest recoverable shale oil resources, at 32 billion barrels.\textsuperscript{7} In recent years, Chinese petroleum geologists have shown great interest and have heavily invested in the exploration and development of shale oil in the Songliao Basin, the Bohai Bay Basin, the Ordos Basin, and the Nanxiang Basin.\textsuperscript{5,8-12}

The Songliao Basin is the largest of the petroliferous basins in China.\textsuperscript{13} As conventional oil yields gradually decrease, replacement resources are need urgently. Shale oil is important for increasing oil reserves and production and for the sustainable development of the Daqing and Jilin Oilfield.\textsuperscript{11,14} In fact, the shales in the first member of the Qingshankou Formation (K\textsubscript{2}qn\textsuperscript{1}) were previously sampled for test production in 1990 and 1992 and yielded average single-well daily shale oil productions of 5.8 and 4.8 bbl, respectively.\textsuperscript{11} Since 2017, as knowledge of shale (oil) geology has gradually increased and development technologies have improved increasingly, shale oil has attracted more attention and enthusiasm for prospective exploration in the Songliao Basin. Three exploratory wells (SYY1, SYY2, and JYY1HF) were drilled successively (another well, SYY3, will be drilled soon); of these, SYY1 is located in the Qijia-Gulong Sag and has achieved commercial oil flows. Studies and exploration have revealed that the lacustrine shale oil offers a good prospect for exploration; therefore, it has become an important target for future exploration in the Songliao Basin, especially in the Qijia-Gulong Sag with thick and organic-rich shales.\textsuperscript{1,6,8,11,15,16}

Source rock is a key factor that controls the formation and enrichment of shale oil resources.\textsuperscript{1,17} In the Qijia-Gulong Sag, the K\textsubscript{2}qn\textsuperscript{1} mudstones are the source rocks for shale oil. The sedimentary environment, thickness and
| Stratigraphic unit                          | Series       | Form.      | Stages  | Member | Thickness (m) | Geologic time (Ma) | Lithology profile        | Sedimentary facies                  | Oil layer | Source | Reservoir | Caprock |
|-------------------------------------------|--------------|------------|---------|--------|---------------|-------------------|------------------------|-------------------------------------|-----------|--------|-----------|---------|
| Qingshankou Formation (K. qn)             | Upper Cretaceous (K.) | Santonian | K.2y<sup>†</sup> | 0       | 150           | 85                | Retrogradational Delta    | Retrogradational Delta Progradational Delta | Sartu (S) |        |           |         |
|                                          |              | Coniacian  | K.2y<sup>†</sup> | 0       | 60            | 87                | Initial lake flooding surface Lowstand delta | Putaohua (P) |        |           |         |
|                                          |              | Turonian-Cenomanian | K.4n<sup>†</sup> | 0       | 290           |                    | Deltaic plain distributary channel | Progradational Delta | Gaotaizi (G) |        |           |         |
|                                          |              | Qingshankou Formation (K. qn) | K.4n<sup>†</sup> | 0       | 260           |                    | Progradational Delta | Delta front mouth bar |                     |        |        |         |
|                                          |              |            |         |        |               |                    | Retrogradational Delta | Delta front mouth bar | Deep lake, semi-deep lake |        |        |         |
|                                          |              |            |         |        |               |                    | The maximum flooding surface | Shallow-water Delta Fluvial swamp facies Fluvial facies | Fuyu (F) |        |           |         |

**FIGURE 2** Source-reservoir-caprock assemblages of the Qingshankou Formation in the Qijia-Gulong Sag, Songliao Basin
distribution, and geochemical features have been studied for many years. However, these investigations for the K2qn1 source rocks were still not sufficient for the following reasons: (a) Geochemical characteristics were conducted for only a single index: Only the total organic carbon (TOC) content was used to evaluate organic matter (OM) abundance and only the vitrinite reflectance (VR) was used to evaluate OM maturity; an evaluation using multiple indicators will be more reliable; (b) the Qijia Sag and the Gulong Sag were usually regarded as a whole to study the characteristics of their source rocks, but there are differences between these two sags; and (c) the hydrocarbon generation and expulsion characteristics—including expulsion intensity, efficiency and expelled hydrocarbon quantities—were evaluated systematically based on source rock characteristics and a modified conceptual model for generation and expulsion. These results were then used to estimate the shale oil resource potential in the Qijia-Gulong Sag.

2 | GEOLOGICAL SETTING

The Songliao Basin, located in Northeast China, is a large Mesozoic-Cenozoic superimposed continental sedimentary basin that trends NE (Figure 1A). It contains six first-order structural units: the Central Depression, the Northern Plunge, the Northeastern Uplift, the Southeastern Uplift, the Southwestern Uplift, and the Western Slope (Figure 1B), which can be further divided into 32 second-order structural units. The Qijia-Gulong Sag, which contains the northern Qijia Sag and the southern Gulong Sag, is situated on the western part of the Central Depression, is bordered by the Daqing Placanticline to the east and the Longhupao terrace to...
the west (Figure 1C), and encompasses an area of more than 5000 km². Its deepest position is in the southern part of the Gulong Sag.

In the Qijia-Gulong Sag, there are three types of exploration wells: Jin, Gu, and Ying. These wells were drilled in the continental Mesozoic-Cenozoic strata with a maximum thickness of 7000 m, which unconformably overlies a Paleozoic basement, and consist of Jurassic, Cretaceous, Paleogene, Neogene, and Quaternary rocks. The Cretaceous strata have been further subdivided into the Lower Cretaceous (the Huoshiling, Shihezi, Yingcheng, Denglouku, Quantou formations) and the Upper Cretaceous (the Qingshankou, Yaojia, Nenjiang, Sifangtai, and Mingshui formations) in ascending order. The Qingshankou Formation (K2qn) can be further subdivided into first (K2qn1), second (K2qn2), and the third member (K2qn3), from top to bottom, according to their lithology. K2qn1 contains dark-gray mudstones with a few thin interlayers of oil shale and muddy siltstone. These dark mudstones are rich in organic matter and are the best source rocks in the Songliao Basin, particularly in the Qijia-Gulong Sag. For conventional and tight oil/gas, the Qingshankou Formation contains multiple source-reservoir-caprock assemblages (Figure 2). However, K2qn1 is the most petroliferous layer of shale oil in the Qijia-Gulong Sag; therefore, it is the focus of this study.

### RESULTS AND DISCUSSION

Three types of exploration wells, Jin, Gu, and Ying, are denoted J, G, and Y, respectively, for example, J87, G40, and Y38; of these, all Jin wells and several Gu wells (G7 and G700 series, G2 and G200 series) are located in the northern Qijia Sag, and Ying wells and the majority of the Gu are situated in the southern Gulong Sag. The geological, geochemical, and hydrocarbon generation and expulsion characteristics of the source rocks in the Qijia Sag and Gulong Gag were interpreted and discussed, respectively, and their differences were compared.

#### 4.1 Geological characteristics of the K2qn1 source rocks

#### 4.1.1 Sedimentary environment

The Songliao Basin, including the Qijia-Gulong Sag, experienced four main stages of tectonic evolution pre-rift with thermal uplift, syn-rift (fault subsidence), depression, and structural inversion. The Q2qn was deposited in a depression with the lacustrine facies in the Central Depression, especially in the Qijia-Gulong Sag. A rapid and large-scale lake transgression formed the K2qn1 strata, which developed thick, dark-gray, organic-rich mudstone, and oil shale of semideep and deep lake facies. The sag experienced lake regression and a decrease in the lake area during the deposition of the K2qn2 and K2qn3 strata.

#### DATA AND METHODS

Many exploration wells have been drilled in the Songliao Basin, including in the Qijia-Gulong Sag, and relative data of mudstone source rocks are abundant. Therefore, the majority of the data used in this work were obtained from the Research Institute of Exploration and Development, PetroChina Daqing Oilfield Company, including TOC content (265 data points), rock-eval pyrolysis values (265 data points), chloroform bitumen “A” values (71 data points), VR values (122 data points), organic elements (63 data points), and gas chromatography of saturated hydrocarbon (204 data points). Some gas chromatography-mass spectrometry (GC-MS) data were obtained from public references. The thickness and maturity distribution figures for the K2qn1 were modified from Feng.

| Well | Sag | Depth/m | Layer | Tm/ppm | T1/ppm | T1/(T1 + Tm) | C30 Hopane/ppm | Gammacerane/ppm | Pr/Ph | Pr/nC17 | Ph/nC18 | 4-Methyisterane/ppm | Dinosterane/ppm | Aryl isoprenoids/ppm |
|------|-----|---------|-------|--------|--------|-------------|----------------|----------------|-------|--------|--------|-------------------|-----------------|---------------------|
| J57  | Qijia | 1848.24 | K2qn1 | 29.41  | 47.13  | 0.62        | 293.71         | 175.6          |       |        |        |                   |                  |                     |
| G204 |       | 2391.25 | K2qn1 | 55.16  | 10.57  | 0.16        | 47.73          | 15.8           |       |        |        |                   |                  |                     |
| G18  | Gulong| 2338.06 | K2qn1 | 4.88   | 0.83   | 0.16        | 2.94           | 0.7            |       |        |        |                   |                  |                     |
| G138 |       | 1944.10 | K2qn1 | 2.55   | 0.59   | 0.19        | 5.81           | 1.4            |       |        |        |                   |                  |                     |
| Y51  |       | 2261.56 | K2qn1 | 6.58   | 1.40   | 0.18        | 7.26           | 2.2            |       |        |        |                   |                  |                     |
| Y78  |       | 2319.51 | K2qn1 | 0.89   | 0.69   | 0.44        | 3.50           | 0.8            |       |        |        |                   |                  |                     |
(0.06-0.25) and the Ph/nC18 ratios (0.09-0.28) in the Gulong Sag are obviously lower than the 0.15-1.12 and 0.08-1.01 in the Qijia Sag (Figure 3B, Table 1). These results indicate mainly reducing-weak reducing environments; however, the reducing conditions in the southern Gulong Sag are stronger than those in the Qijia Sag (Figure 3). In addition, the carbon number distribution and the n-alkane composition are key indicators for sources of OM.28,33 For continental lacustrine sediments, low carbon numbers (nC20- or nC21-), middle carbon numbers (nC21-nC25), and high carbon numbers (nC26+) reveal that bacteria and algae, aquatic plants, and terrestrial plants, respectively, were the sources.28,34,35 In the Qijia Sag, the terrigenous/aquatic ratios (TAR) ((nC27 + nC29 + nC31)/(nC15 + nC17 + nC19)) and the ΣC23+/ΣC21- values are 0.13-11.11 and 0.07-5.95, and average 1.17 and 1.90, respectively. The main peak carbon is nC21, nC23. Specifically, the maximum values (large average values) of the TAR, ΣC22+/ΣC21-, and the main peak carbon (11.11, 5.95, nC29) all occur in the north part of the Qijia Sag (Jin8, 9, 86, 87, 95 wells); these values are larger than those in the north part of Qijia Sag (Table 1). These results reveal that the OMs in the Qijia Sag are mainly derived from aquatic plants, with some terrestrial plants in the north part of Qijia Sag. In the southern Gulong Sag, the TAR and the ΣC22+/ΣC21- values range from 0.02 to 0.36 and from 0.02 to 0.81,—with average values of 0.16 and 0.26, respectively—which are much lower than those in the Qijia Sag; the main peak carbons are nC13, nC15 (Table 1), which show that bacteria-algae and aquatic plants sources are dominant. Referring to the sterane data of Feng et al27 (Table 2), the proportion (not absolute values) of C27 steranes in the Gulong Sag (G18, G138, Y51, and Y78) is greater than the proportion of C29 steranes in the Qijia Sag (J57 and G204), which also suggests that the provenances of these compounds are mainly lower aquatic organisms, followed by several higher plants in the Qijia Sag. Using comprehensive analyses of sedimentary conditions and sources of OM, the provenance of the rocks is the north of the Qijia-Gulong Sag, which is consistent with previous results studied based on heavy minerals, sandstone percentages, and biomarkers.19,36 In addition, a higher content of terpane (such as Tm, Te, and C30 Hopane) and sterane (eg, regular sterane and 4-methylsterane) in the Qijia Sag reveals more prosperous aquatic organisms and higher paleoproduc-tivity than in the Gulong Sag.27

Gammacerane, dinosterane, and aryl isoprenoids are important markers for salinity and water column stratification during source rock deposition.28,30-32 The gammacerane, dinosterane, and aryl isoprenoids values are 15.8-175.6 ppm, 2.1-12.3 ppm, and 0.3-3.28 ppm, respectively, in the Qijia Sag, much greater than the corresponding values (0.7-2.2 ppm, 0.46-0.58 ppm, and 0-1.61 ppm, respectively) in the
Gulong Sag (Table 2), revealing a higher bottom water salinity of K2qn1 sedimentary environment in the Qijia Sag and a lower salinity in the Gulong Sag.

4.1.2 Distribution and thickness

The K2qn1 is the development strata of the best source rocks in the Songliao Basin. The distribution and thickness of the K2qn1 source rocks were studied based on a comprehensive analysis of well and sedimentary facies data and published references. The Qijia-Gulong Sag is one of the most favorable sedimentary sags for source rocks in the Songliao Basin, and the K2qn1 mudstones/shales are pervasive and continuous in this sag. Influenced by the sedimentary center, the K2qn1 organic-rich source rocks are thickest in the northern and southern areas and thinner in the middle part of the Qijia-Gulong Sag, but very thin in the northern and southwestern margins. The thickness of the source rocks is generally 20-80 m. In the northern Qijia Sag, the maximum thickness, up to more than 110 m, is located in the area of wells G202-G2-J3. There are

**FIGURE 5** Organic matter (OM) abundance of the K2qn1 source rocks in the Qijia-Gulong Sag, Songliao Basin

**TABLE 3** Evaluation standard for organic matter abundance of source rocks

| Type of source rocks | TOC/% | $S_1 + S_f$ (mg HC/g rock) | Chloroform bitumen “A”/ppm |
|----------------------|-------|---------------------------|-----------------------------|
| Excellent            | >2.0  | >20                       | >2000                       |
| Good                 | >1.0  | >6.0                      | >1000                       |
| Fair                 | 1.0-0.6 | 2.0-6.0                  | 500-1000                    |
| Poor                 | 0.6-0.4 | 0.5-2                   | 100-500                     |
| Non                  | <0.4  | <0.5                     | <100                        |

**TABLE 4** Other geochemical data from the K2qn1 source rocks in the Qijia-Gulong Sag

| Sag               | TOC/%       | $S_1$/ (mg HC/g rock) | $S_f$/ (mg HC/g rock) | $S_1 + S_f$/ (mg HC/g rock) | $CO_2$/ (mg HC/g rock) | COI/(mg HC/g rock) | HI/(mg HC/g rock) |
|-------------------|-------------|----------------------|----------------------|------------------------------|------------------------|-------------------|-------------------|
| Qijia-Gulong      | 0.48-3.2/1.40a | 0.01-10.81/1.80 | 0.15-37.56/10.66 | 0.10-9.05/0.68 | 0.02-41.58/12.30 | 27.38-1140.47/454.48 |
| Qijia             | 0.55-3.2/1.26a | 0.02-10.81/1.48 | 0.28-37.56/13.50 | 0.01-9.05/0.74 | 0.28-41.58/14.74 | 27.38-1140.47/549.00 |
| Gulong            | 0.48-5.9/2.09a | 0.01-5.65/2.15 | 0.15-19.22/7.00 | 0.14-8.49/0.62 | 0.33-23.38/9.15 | 28.74-567.47/333.06 |

*aMaximum value-minimum value/average value.*
three thickness centers in the southern Gulong Sag, which are distributed in the areas of well G50-G822-G842, Y8-Y942, and A151-GS1; the maximum thicknesses are 80 m, 90 m, and 110 m, respectively (Figure 4).

4.2 | Geochemical characteristics of K2qn1 source rocks

4.2.1 | Organic matter (OM) abundance

The OM abundance of the K2qn1 source rocks was analyzed using TOC, the hydrocarbon generation potential ($S_1$ + $S_2$), the pyrolysis yields ($S_2$), and the chloroform bitumen “A” content$^{28,37,38}$ (Table 1). In fact, the light hydrocarbon (C5-C14, liquid hydrocarbon) in pyrolysis yields ($S_1$) and “A” are easy to evaporate to loss during coring to surface and during sample storage and handling before testing. The evaluation of OM abundance and shale oil resources using the present $S_1$ or “A” values will underestimate the OM abundance and the resource potential. Therefore, it is necessary to restore the light hydrocarbon loss to obtain original oil content before OM abundance and resource evaluation.$^{2,39-42}$ Xue et al$^{40}$ used the northern Songliao Basin as an example and conducted evaporative loss calibrations using rock-eval results and kerogen kinetics. The loss recovery coefficients of the $S_1$ and “A” values were 4.2 and 1.2, respectively; the authors of this study thought that the value of 1.2 for “A” was reasonable and the value of 4.2 for $S_1$ was larger than the actual value. Therefore, we used a value of 1.2 for the loss recovery coefficient to obtain the original “A” in the Qijia-Gulong Sag. For the $S_1$ restoration, we adopted the method proposed by Pang et al.$^{39}$

The TOC of the K2qn1 source rocks spans from 0.48% to 8.31% and averages 2.40%, with 97.7% of 265 samples exceeding 1.0% (lower limit of good source rocks) and 58.5% exceeding 2.0% (lower limit of excellent source rocks) in the Qijia-Gulong Sag (Figure 5A,B). The $S_1$ + $S_2$ values of the K2qn1 source rocks are between 0.28 mg HC/g rock and 41.58 mg HC/g rock, with an average value of 12.30 mg HC/g rock. Although the $S_2$ evaluation standard of source rocks used by Peters and Cassa$^{44}$ is different from SY/T 5735-1995,$^{43}$ the $S_2$ values suggest that most of the K2qn1 mudstones have generative hydrocarbon potential of good and excellent source rocks, in accordance with the TOC content (Figure 5D). Almost all

| OI (mg HC/g rock) | Chloroform bitumen “A”/ppm | $T_{max}/^\circ C$ | VR/% | H/C | O/C |
|-------------------|-----------------------------|-----------------|------|-----|-----|
| 0.39-505.26/35.09 | 1108-13162/4824             | 403-455/444     | 0.82-1.61/0.94 | 0.62-1.66/1.23 | 0.03-0.25/0.07 |
| 0.39-505.26/36.44 | 1366-13162/5295             | 423-455/445     | 0.82-1.14/0.90 | 1.07-1.66/1.34 | 0.03-0.25/0.07 |
| 5.10-292.20/33.43 | 1108-7265/3203               | 403-454/443     | 1.06-1.61/1.15 | 0.62-1.04/0.77 | 0.06-0.12/0.08 |
TOC data with >4.0%, S1 + S2 > 20 mg HC/g rock, and S2 > 20 mg HC/g rock come from the Qijia Sag, which indicates that the OM abundance of source rocks in the Qijia Sag is greater than those in the Gulong Sag (Table 4). This is probably because the abundance of aquatic organisms and a greater salinity value in the Qijia Sag result in higher paleoproduction and better preservation of OM.27,45-47

Based on the test TOC values of many wells and considering the sedimentary facies, the TOC distribution of the K2qn1 source rocks was evaluated (Figure 6). The TOC of more than 70% of the area of the Qijia-Gulong Sag is greater than 2.0%, developing excellent source rocks. In the Qijia Sag, the high-value zones are distributed in the areas of well J75-G705 (TOC > 3.0%), J27-J47-J3 (TOC > 3.5%, maximum value >5.0%), and G202 (TOC > 3.5%). In the Gulong Sag, the areas of well G50-G822 and A151-GS1 have higher TOC values, >3.0% and >2.5%, respectively. These results also indicate that the TOC in the Qijia Sag is larger than that in the Gulong Sag. The TOC distribution is closely related to the distribution of source rock thicknesses (Figure 4) and sedimentary facies.

4.2.2 | Types of OM

OM (kerogen) is generally classified into four types: type I, type II1, type II2, and type III. OM type can be determined using organic elemental analyses and rock-eval pyrolysis data.37,48,49

OM types were confirmed based on elemental C, H, and O and their ratio.37,50,51 The H/C and O/C ratios were 0.62-1.66 and 0.03-0.25, respectively (Figure 7A). The kerogens are dominated by type I and type II1, with a few type II2 (Figure 7A).

For the rock-eval pyrolysis method, the cross-plots of the hydrogen index (HI) vs oxygen index (OI), HI vs Tmax, and S2 vs TOC could be used to classify the OM types.37,48,52,53 The HI values range from 27.38 mg HC/g TOC to 1140.47 mg HC/g TOC and are mostly more than 300 mg HC/g TOC. The OI values are mainly 0.39-505.26 mg CO2/g TOC, but the majority are less than 100 mg CO2/g TOC (Figure 7B). The Tmax values are from 403°C to 455°C (Figure 7C). The S2 values oscillate from 0.15 mg HC/g TOC to 37.56 mg HC/g TOC, and the TOC values are 0.48%-8.31% (Figure 7D). These results show that the samples can be categorized mainly as type I and type II1 kerogens, with some type II2 and III kerogens, which agrees with

**FIGURE 7** OM types of the K2qn1 source rocks in the Qijia-Gulong Sag, Songliao Basin
the results obtained from the organic element and other scholars.5,11,15,16,18,20 Because of their higher salinities, the source rocks have greater HI and H/C values in the Qijia Sag (Table 4) and contain mainly type I kerogens, which are better than the type II1 kerogen in the Gulong Sag. In short, the K2qn1 source rocks are oil-prone and beneficial to the formation of shale oil.

4.2.3 | Thermal maturity of OM

In this work, the VR, pyrolysis $T_{\text{max}}$, odd/even predominance ratio (OEP), and carbon preference index (CPI) were used to determine the organic matter maturity of source rocks.28,37,38,54,55

The critical values of VR for immature, lowly (or early) mature, mature, highly mature, and over mature stages are <0.5%, 0.5%-0.7%, 0.7%-1.3%, 1.3%-2.0%, and >2.0%, respectively.37 For $T_{\text{max}}$, the range of maturity (oil window) and the high maturity (gas/condensate) are at 430°C-455°C and 455°C-470°C, respectively, for type I and II1 kerogen.38 The VR values are 0.82%-1.29%, except for one sample, which had a value of 1.61% (Figure 8A). The VR values gradually decrease from the present center of the Qijia Sag and the Gulong Sag to their margins. The most mature area is located in the center (well area of G50-G822-G48) of the southern Gulong Sag and is more than 1.6% (at the highly mature stage); however, the high-maturity area (that generates condensate gas) is very small (Figure 9, Table 4). The maturity in the center of the Qijia Sag is less than 1.3% and still in the mature stage. The $T_{\text{max}}$ values span from 403°C to 455°C (Figure 8B). The CPI values of these samples range from 0.50 to 1.50, and the OEP values are 0.79-1.47 (Figure 8C, Table 1); this also reveals that the K2qn1 source rocks are mainly at the mature stage within the oil window. Thus, the K2qn1 source rocks are favorable to shale oil enrichment.

4.3 | Hydrocarbon generation and expulsion characteristics of the K2qn1 source rocks

4.3.1 | Hydrocarbon generation and expulsion conceptual model

A conceptual model was proposed by Pang et al.23 to estimate the quantities of hydrocarbon generated and expelled. We
used and improved this model by restoring the original $S_1$ and hydrocarbon generation potential according to the mass balance (Figure 10).

The $[S_1 + S_2]/$TOC $\times$ 100 ratio, called the hydrocarbon generation potential index ($GPI$), represents the total potential of generated hydrocarbons. With increasing burial and thermal evolution of the source rocks, the generated hydrocarbons first experience retention (including water solution and adsorption) and subsequently discharge in quantity (mostly in the free phase) from the source rocks if the amount of generated hydrocarbon outnumbers the maximum retention of the source rocks. The $VR$ or depth at which hydrocarbons begin to be expelled is the hydrocarbon expulsion threshold. The $GPI$ reaches its maximum value ($GPI^0$) at the hydrocarbon expulsion threshold. Before hydrocarbon expulsion, the $GPI$ is defined as the original hydrocarbon generation potential index ($GPI_o$). The $GPI_o$ reveals both the hydrocarbon generation and the retention potential of source rocks. The remaining hydrocarbon generation potential index ($GPI_i$) indicates the decreased hydrocarbon generation potential after hydrocarbon expulsion. This $GPI_i$ is present after expulsion, while the original is not. Therefore, it is necessary to restore the $GPI_o$.

The $GPI_o$ was restored based on the mass balance. The organic carbon (or TOC) in a source rock includes ineffective carbon and effective carbon. The part of the organic carbon or TOC that cannot generate hydrocarbons is the ineffective carbon, in which its absolute mass remains unchanged both before and after hydrocarbon expulsion. Therefore, $GPI_o$ can be restored using Equations (1) and (2), as following:

$$k = \frac{1 - 0.83GPI_r/1000}{1 - 0.83GPI^0/1000}$$

$$GPI_o = k \cdot GPI^0$$

where $k$ is a restored coefficient; 0.83 is the average carbon content in hydrocarbons; $GPI_r$ is the residual hydrocarbon generation potential index, mg HC/g TOC; $GPI_o$ is the original hydrocarbon generation potential index, mg HC/g TOC; and $GPI^0$ is the hydrocarbon generation potential index at hydrocarbon expulsion threshold, mg HC/g TOC.

The difference between the $GPI_o$ and the $GPI_r$ is the hydrocarbon expulsion ratio ($q_e$), which represents the hydrocarbon expulsion quantity per unit of organic carbon (Figure 10). $q_e$ can be calculated using Equation (3). The hydrocarbon expulsion efficiency ($P_{ef}$) is the ratio of the hydrocarbon expulsion to its generation, as shown in Equation (4). The hydrocarbon expulsion velocity ($V_e$) refers to the variations in the hydrocarbon expulsion ratios with a $VR$ increase of 0.1%, and can be obtained from Equation (5), as follows:

$$q_e(VR) = GPI_o(VR) - GPI_r(VR)$$

$$P_{ef} = \frac{q_e(VR)}{GPI_o(VR)} \times 100$$

$$V_e = \frac{\Delta q_e(VR)}{\Delta VR}$$

where $q_e$ is the hydrocarbon expulsion ratio, mg HC/g TOC; $P_{ef}$ is the hydrocarbon expulsion efficiency, %; and $V_e$ is the hydrocarbon expulsion velocity, (mg HC/g TOC)/(0.1% $VR$).

In the original of hydrocarbon generation and expulsion model, after hydrocarbon expulsion, the $GPI_o$ remains constant (at $GPI^0$, the hydrocarbon generation potential index at the hydrocarbon expulsion threshold) while the $VR$ increases. However, in this improved model, the $GPI_o$ gradually increases as the $VR$ increases, which agrees with the actual model of hydrocarbon generation proposed by Tissot and Welte. Therefore, this improved model can more accurately reflect the characteristics of hydrocarbon generation and expulsion in source rocks.

The hydrocarbon generation and expulsion intensity ($I_g$ and $I_e$) are defined to be the amounts of hydrocarbon generation and expulsion per area in the same layer of source rock. Once we established an envelope curve of data points...
of \((S_1 + S_2)/\text{TOC}\) \times 100 in Figure 10, the hydrocarbon generation and expulsion ratios \((q_g \text{ and } q_e)\), intensities \((I_g \text{ and } I_e)\) and amounts \((Q_g \text{ and } Q_e)\) were calculated using Equations (3) and (6)-(9), respectively.

\[
I_g = \int_{V_R_{1}}^{V_R} 10q_g(VR) \cdot H \cdot \rho \cdot \text{TOC} \cdot d(VR) \tag{6}
\]

\[
I_e = \int_{V_R_{2}}^{V_R} 10q_e(VR) \cdot H \cdot \rho \cdot \text{TOC} \cdot d(VR) \tag{7}
\]

\[
Q_g = \int_{V_R_{1}}^{V_R} 10^7 q_g(VR) \cdot H \cdot A \cdot \rho \cdot \text{TOC} \cdot d(VR) \tag{8}
\]

\[
Q_e = \int_{V_R_{2}}^{V_R} 10^7 q_e(VR) \cdot H \cdot A \cdot \rho \cdot \text{TOC} \cdot d(VR) \tag{9}
\]

where \(I_g\) is the hydrocarbon generation intensity, \(t/km^2\); \(I_e\) is the hydrocarbon expulsion intensity, \(t/km^2\); \(q_g\) is the hydrocarbon generation ratio, \(mg \text{ HC/g TOC}\); \(Q_g\) is the hydrocarbon generation quantity, \(t\); \(VR\) is the vitrinite reflectance, \%; \(V_R_{1}\) is the hydrocarbon generation threshold, \%; \(V_R_{2}\) is the hydrocarbon expulsion threshold, \%; \(H\) is the thickness of source rocks, \(m\); \(\text{TOC}\) is the total organic carbon contents, \%; \(\rho\) is the bulk density of source rocks, \(g/cm^3\); and \(A\) is the area of source rocks, \(m^2\).

### 4.3.2 Hydrocarbon generation and expulsion model of the K2qn1 source rocks

A hydrocarbon generation and expulsion model for the K2qn1 source rocks was established based on the TOC values, rock pyrolysis parameters, burial depths, and VR values (Figure 11). In this study, the actual measured VR values were used for the 122 samples in Figure 8A, and the VR...
values were calculated using the fitting equations for the VR and the depth from those samples without actual measured values.

The massive hydrocarbon generation threshold of the K2qn1 source rocks in the Qijia-Gulong Sag was 0.5% VR (Figure 11A), which is consistent with the results of previous studies. The threshold of hydrocarbon expulsion was 0.85% VR (Figure 11B). The hydrocarbon expulsion velocity was the largest at the hydrocarbon expulsion peak, corresponding to 0.95% VR (Figure 11C). When the hydrocarbon generation and expulsion thresholds were reached, the hydrocarbon generation and expulsion ratios increased rapidly but the degree of increase gradually reduced, in particular, during the late stage (Figure 11C). The maximum velocity of expulsion can reach 190.2 mg HC/g TOC/0.1% VR at 0.95% VR. With increasing thermal evolution (from 0.5% to 1.6% VR), the expulsion efficiency increased from 0% to 69.1%; in contrast, the hydrocarbon retention efficiency in the source rocks decreased gradually to 30.9% (Figure 11D).

4.3.3 Hydrocarbon generation and expulsion intensity, and amount of the K2qn1 source rocks

The differences in the intensities of hydrocarbon generated and expelled are controlled by comprehensive variations in the thickness, thermal maturation, and OM abundance of the K2qn1 source rocks in the Qijia-Gulong Sag, as shown in Figures 4-9. For the better OM type, a larger hydrocarbon generation potential, and a higher thermal evolution of source rocks, their hydrocarbon expulsion extensity and efficiency are higher. The intensities of hydrocarbon generation and expulsion were calculated using Equations (6) and (7) (Figures 12 and 13).

With a better OM type and a higher OM abundance, the highest hydrocarbon generation intensities for the K2qn1 source rocks in the area of well J27-J47-J3-G202 in the Qijia Sag are up to 700 × 10^4 t/km², greater than (400-500) × 10^4 t/km² in the areas of well G50-G822 and A151-GS1 in the Gulong Sag (Figure 12). All the K2qn1...
source rocks have reached the expulsion threshold. The hydrocarbon expulsion intensity decreases rapidly around from high-value areas (roughly corresponding to the thickness and TOC center), which is consistent with the variation in the hydrocarbon generation intensity. The maximum intensities of hydrocarbon expulsion in the areas of well J27-J47-J3 (in the Qijia Sag) and G50-G822 (in the Gulong Sag) are both greater than $300 \times 10^4 \, \text{t/km}^2$ (Figure 13), because there are more mature hydrocarbons ($VR > 1.4\%$) in wells G50-G822.

The cumulative amounts of generated and expelled hydrocarbon were determined multiplying the hydrocarbon generation and expulsion intensity by the area of the source rocks using Equations (8) and (9). Currently, the total quantities of generated and expelled hydrocarbon from the K$_2$qn$^1$ source rocks are $121.8 \times 10^8 \, \text{t}$ and $46.9 \times 10^8 \, \text{t}$, respectively, which indicate the hydrocarbon expulsion efficiency is 38.5%. Therefore, 61.5\% of generated hydrocarbons remain in the K$_2$qn$^1$ source rocks, indicating an enormous retained hydrocarbon potential.

### 4.4 | Shale oil evaluation

#### 4.4.1 | Shale oil potential

The K$_2$qn$^1$ dark mudstones are widely distributed and thick, abundant in organic matter (generally 1\%-5\% of TOC), oil-prone type I and II, and are at a stage mature (0.8\%-1.3\% VR, oil window) in the Qijia-Gulong Sag. Moreover, these mudstones are composed of clay minerals (6.1\%-39.1\%, average 23.5\%), quartz (10.9\%-57.2\%, average 44.7\%), plagioclase (5.1\%-33.2\%, average 18.2\%), followed by carbonate (1.1\%-50.5\%, average 7.2\%), and pyrite (1.0\%-12.8\%, average 5.9\%) and thus have a stronger brittleness. The reservoir spaces, mainly including intergranular pores, intracrystalline pores, dissolved pores (owing to higher calcite and dolomite contents), organic pores, and microfractures, are abundant. These conditions reveal that a huge shale oil potential occurs in the Qijia-Gulong Sag.

Shale oil is defined as the oil that self-generated and was retained in the shale in a self-reservoir. Therefore,
Remarkably, the hydrocarbon generation and expulsion control the accumulation, enrichment, and distribution of shale oil.\textsuperscript{11,59} The hydrocarbon retention intensity is the difference between the hydrocarbon generation and the expulsion intensities. The areas of wells J75-G705, J27-J47-J3-G202, and A151-GS1 still have high hydrocarbon retention intensities, greater than \(400 \times 10^8 \text{t/km}^2\) (Figure 14), which are basically consistent with the distribution of the hydrocarbon generation intensities (Figure 12). These areas are sweet spots for shale oil. However, the area of well G50-G822 has a lower intensity of hydrocarbon retention because of its higher maturity and greater expulsion intensity. The difference between the amounts of hydrocarbon generation and expulsion is the maximum quantity of residual oil. As shown by the analyses above, the quantities of hydrocarbons generated and expelled from the K\(_2\)qn\(^1\) source rocks in the Qijia-Gulong Sag are \(121.8 \times 10^8 \text{t}\) and \(46.9 \times 10^8 \text{t}\), respectively. The shale oil resource potential is \(74.9 \times 10^8 \text{t}\).

### 4.4.2 Discussion of shale oil potential

In this study, an improved model of hydrocarbon generation and expulsion was established based on material balance. The K\(_2\)qn\(^1\) source rocks data used are derived from core samples that were sufficient and widely and evenly distributed across the whole Qijia-Gulong Sag. In addition, the concept (hydrocarbon generation potential method) is clear and simple and reduces human biases. Therefore, this model is more reasonable and appropriate and can objectively reflect the overall hydrocarbon generation and expulsion characteristics of source rocks during resource evaluation.

The volumetric method is effective to evaluate in-place resources of shale oil in the early stages of geological prospecting. When assessing the in-place resources in tight marlstone oil using the volumetric method, the thickness, area, oil content, and maturation are key parameters, as shown in Equation (10):

\[
Q = S h p q
\]

(10)

where \(Q\) is the amount of geological resources in place, kg; \(S\) is the distribution area of the source rocks, \(m^2\); \(h\) is the oil shale thickness, m; \(p\) is the density of the mudstone, \(2.6 \text{t/m}^3\); and \(q\) is the original residual hydrocarbon content (oil content) \(\text{mg HC/g rock}\).

In this method, it is most critical to accurately determine the oil content.\textsuperscript{1,4,40} As analyze above, the original chloroform bitumen “A” content was used to represent oil content and was obtained using the 1.2 of the loss recovery coefficient.\textsuperscript{40} The “A” distribution was evaluated (Figure 15), which enabled a better corresponding relationship with the residual hydrocarbon intensity (Figure 14). Based on the “A” distribution, the source rock thickness, and distribution, the shale oil resources in the Qijia-Gulong Sag were evaluated using Equation (10). The in-place resource of shale oil is approximately \(31.9 \times 10^8 \text{t}\), which is less than \(74.9 \times 10^8 \text{t}\) determined by the improved hydrocarbon generation model. One important reason for the differences in these two methods is that the residual shale oil resources (\(74.9 \times 10^8 \text{t}\)) include kerogen-cracking hydrocarbons (\(S_2\)). With the pyrolysis data, the average value of \(S_2\) is 10.66 mg HC/g rock, which is much greater than the 1.64 mg HC/g rock value for \(S_1\). Therefore, the quantities of residual oil in source rocks are regarded as the maximum potential of shale oil. In situ conversion processing of shale oil using heating technology is currently being researched and field tested by many large oil companies.\textsuperscript{64,65} This technology can create kerogens and residual oils from medium-low mature shale by underground cracks or hydrogenation reactions, and can generate light crude oil, which will greatly increase shale oil reserves and enhance recovery. Therefore, the maximum amount of shale oil calculated using the improved hydrocarbon generation model is possibly more reasonable and can be exploited in the future. At present, the recovery coefficient of shale oil in the Chinese continental basin is only 16%-18%,\textsuperscript{5} and the corresponding recoverable resource is \((12.0-13.5) \times 10^8 \text{t}\).

### 5 CONCLUSIONS

1. The K\(_2\)qn\(^1\) organic-rich mudstones/shales mainly developed in semideep and deep lake facies with reducing and weak reducing sedimentary conditions. Compared with the southern Gulong Sag, the northern Qijia Sag has higher salinity, more abundant aquatic organisms, and a larger paleoproductivity. The K\(_2\)qn\(^1\) source rocks are pervasive and continuous in the entire sag. Their thickness is generally 20-80 m, and they are thicker in northern and southern areas and thinner in the middle area. The maximum thicknesses are both up to 110 m in the Qijia Sag and the Gulong Sag.

2. The K\(_2\)qn\(^1\) source rocks have higher OM abundance (the TOC ranges from 0.48% to 8.31%, with an average value of 2.40%), are dominated by type I and type II kerogen, and are at the maturity stage, with 0.8%-1.3% VR. The source rocks in the Qijia Sag have greater OM abundance, a better OM type, and a lower OM maturity than those in the Gulong Sag. The overall evaluation showed that the dark mudstones in the Qijia-Gulong Sag are good to excellent source rocks and have enormous hydrocarbon generation potential.

3. The threshold and peak values of hydrocarbon expulsion from the K\(_2\)qn\(^1\) source rocks are at 0.85% VR and 95% VR.
respectively. The largest intensity values of hydrocarbon generation in the area of well J27-J47-J3-G202 in the Qijia Sag are up to $900 \times 10^4$ t/km²; these are greater than the $(400-500) \times 10^4$ t/km² in the areas of well G50-G822 and A151-GS1 in the Gulong Sag. The maximum intensities of hydrocarbon expulsion in the areas of well J27-J47-J3 and G50-G822 are greater than $300 \times 10^4$ t/km².

4. The amounts of hydrocarbon generation and expulsion from the K2qn1 source rocks are $121.8 \times 10^8$ t and $46.9 \times 10^8$ t, respectively. The hydrocarbon retention efficiency is 51.3%. The maximum amount of retained oil (shale oil) in-place resource is $74.9 \times 10^8$ t, and the maximum amount of recoverable resource is $(12.0-13.5) \times 10^8$ t, which indicates a significant shale oil potential in the Gulong Sag, especially in the Qijia Sag, Songliao Basin, Eastern China.

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CONFLICT OF INTEREST
The authors declare no competing financial interest.

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