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Organic Geochemical Evidence for the Formation of Condensate from Coaly Source Rocks in the Wumaying Buried Hill of the Huanghua Depression, Bohai Bay Basin

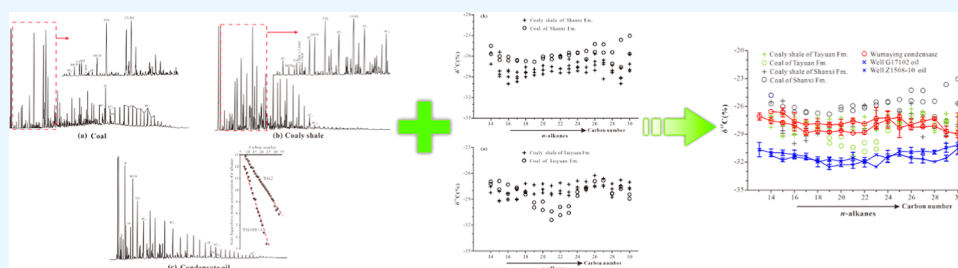Da Lou,* LE Wang, Rong Luo, Jinjun Xu, Yongge Sun, Feipeng Zhang, Yonggang Zhao, Hui Wang, and Jiacheng Ge

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ABSTRACT: Although oil and gas from coaly source rocks have been widely discovered worldwide, the role of oil generated from coal measures in marine-continental coaly deposits during the Carboniferous-Permian period in the Bohai Bay Basin has long been a subject of debate. The recent discovery of a condensate reservoir in the Wumaying buried hill within the Huanghua Depression of the Bohai Bay Basin offers new potential insights into this issue. In this study, we employed organic geochemical methods to explore the possibility of the Carboniferous-Permian coal deposit being a primary source of the condensate. The distribution of light hydrocarbons and the biomarker assemblage indicate that the condensate did not undergo significant secondary alterations such as thermal cracking, gas invasion fractionation, or biodegradation. The hydrocarbon generation potential of the Carboniferous-Permian coaly source rocks suggests that they could be an important contributor to the formation of condensate. High pristine/phytane ratios (1.0−7.5), an abundant presence of benzene series, and the dominance of C_{29} steranes (>50%) within the condensate could be indicative of coaly organic matter. These features are comparable to those found in coaly source rocks. Moreover, the stable carbon isotopic compositions of n-alkanes in the condensate, ranging from −26.0 to −30.0‰, correlate well with those from coaly mudstone (−25.4 to −30.0‰). This suggests that the condensate of the Wumaying buried hill may predominantly originate from the Carboniferous-Permian coaly mudstone. When integrated with the geological background, the results distinctly demonstrate that the Carboniferous-Permian coaly source rocks have significantly contributed to the formation of the condensate reservoir in the Wumaying buried hill. This provides an essential reference for future exploration of oil and gas resources derived from the carboniferous-Permian coaly source rocks in the Bohai Bay Basin.

1. INTRODUCTION

Coal, as a distinctive sedimentary organic rock, encapsulates abundant information pertinent to the evolution of Earth's ecosystem and its deep-climatic environments.[1,2] Simultaneously, coal serves as a crucial fossil fuel, supplying substantial energy for both human existence and industrial advancement. The significance of additional resources, particularly hydrocarbons derived from coal, within coal measures has garnered increasing attention, leading to a transformative perception and role for coal.[3] Notably, the Carboniferous-Permian coal measures, widely deposited in numerous basins globally, possess a significant potential for hydrocarbon generation.[4] The cumulative natural gas expulsion of Carboniferous-Permian coaly source rocks in the Ordos basin was 237.9 ×

10^{12} m^3, and the oil and gas in the Donetts basin were mainly derived from Carboniferous-Permian coaly source rocks (oil equivalent was slightly higher than 1.57 × 10^8 t).[5−7]
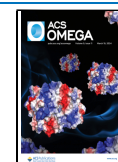
In the Bohai Bay Basin, Carboniferous-Permian coaly source rocks exhibit immense petroleum potential. These rocks have yielded an impressive natural gas resource estimated at 2.84 × 10^{11} m^3 in the Huanghua Depression.[8] Furthermore, the
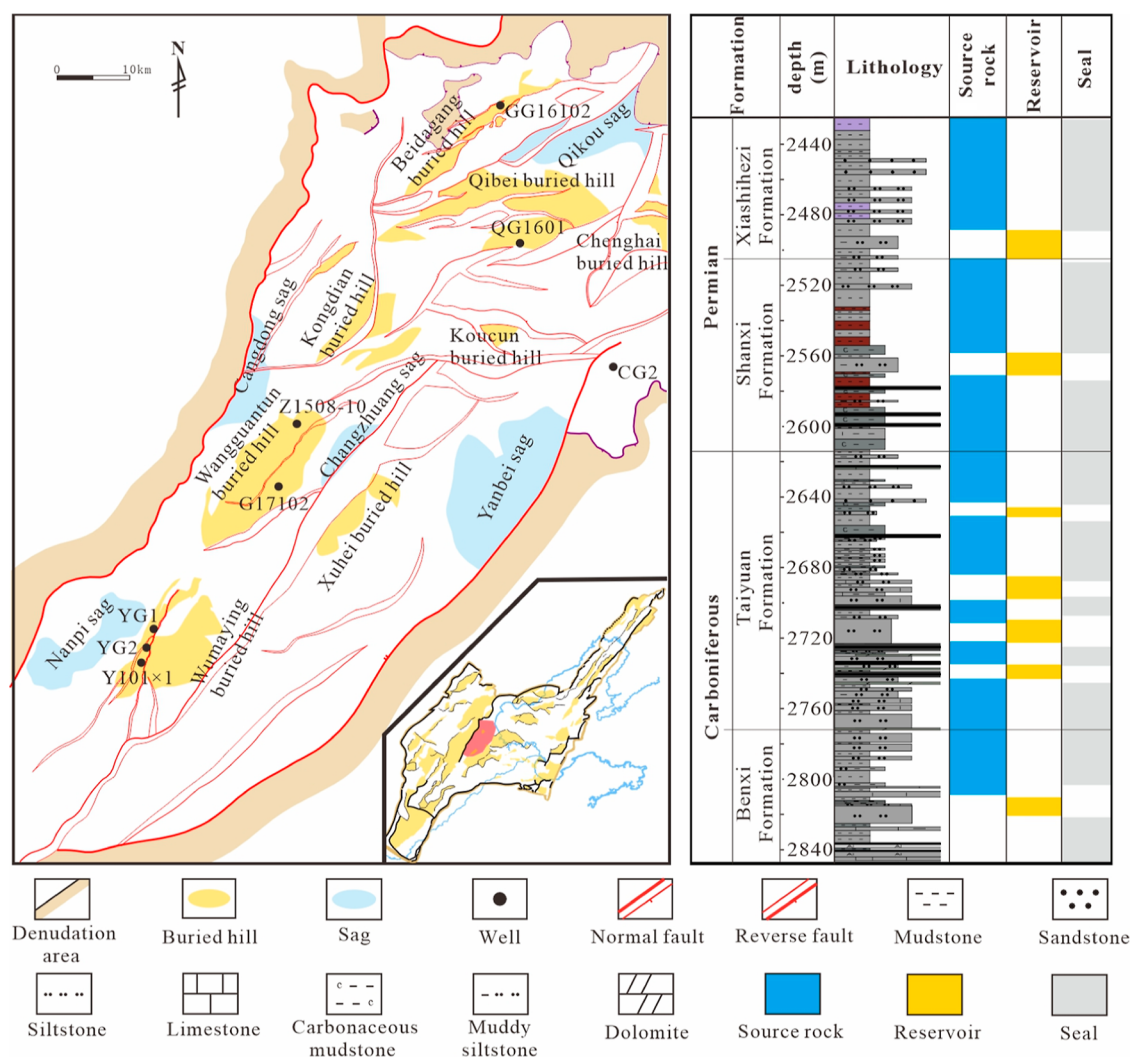
Figure 1. Structural map and stratigraphic profile of Wumaying buried hill.

discovery of additional oil and gas reservoirs in other depressions, originating from Carboniferous-Permian coaly source rocks, underscores their efficacy as a prolific source of hydrocarbon generation.[9−11] A previous study showed that the Carboniferous-Permian coaly source rocks (Taiyuan Formation [Fm.] to Shanxi Fm.) in the Bohai Bay Basin are deposited in the transitional environment from marine to continent, including coal, coaly shale, and carbonaceous mudstone.[6,12,13] The coal-bearing strata of the Taiyuan Fm. were formed in the peat flat under the epicontinental barrier island sedimentary system, and under the influence of frequent transgression and retrogression, abundant hydrocarbon-generating materials were accumulated and preserved under anoxic conditions.[14,15] The coal-bearing strata of the Shanxi Fm. were characterized by a peat swamp coal-accumulating environment in a shallow water delta plain. Coaly shale accumulation occurred in delta plain sediments. Both of them evolved regularly in time and space. Controlled by the sedimentary environment and organic matter supply, the macerals of coaly source rocks were mainly composed of vitrinite, including collodetrinite, collotelinite, and corpogelinite, followed by the lowest inertinite and a few liptinite contents.[16−19] The vitrinite content of coaly source rocks in the Qikou Sag can reach 80%, and the inertinite content of coaly source rocks in Beidagang

buried hill was generally 20%,[18] indicating that coaly source rocks had a rich hydrocarbon-generating material foundation.

In recent years, the Dagang Oilfield had achieved continuous breakthroughs in oil and gas exploration from Carboniferous-Permian coaly source rocks in the Huanghua Depression, with the Wumaying buried hill serving as a notable example. In the early stages, high-yield industrial gas flow was obtained in well WS1 in the southern part of the thrust belt on the western margin of Wumaying buried hill, but the exploration was stopped because of the rich hydrogen sulfide. Since 2018, the Dagang oilfield has been focusing on the exploration of Carboniferous-Permian clastic buried hill reservoirs. Significant breakthroughs were achieved in wells YG1 and YG2 in the western thrust belt as well as in well WT1 in the eastern anticlinal belt. High oil and gas flow was obtained, and no hydrogen sulfide was detected. The Wumaying buried hill was located in the depositional center of the Carboniferous-Permian coaly source rocks, and the genesis of its oil and gas reservoir was related to these coaly source rocks. It was considered another significant discovery in the Bohai Bay Basin following the Suqiao coal-formed oil and gas field. This underlined the promising exploration prospects of the petroleum system in the Bohai Bay Basin, with the Carbon-

iferous-Permian coaly source rocks serving as the primary source rocks.

Large-scale marine-continental coaly deposits were developed during the Carboniferous-Permian in the North China platform, and the main source of natural gas in the Ordos Basin was considered the Carboniferous-Permian coaly source rocks. However, the oil and gas reservoir of the Suqiao buried hill, discovered in the early 1980s and primarily sourced from typical Carboniferous-Permian coaly source rocks, remains a subject of controversy regarding its petroleum origins. This debate has been particularly energized by the recent breakthroughs in deep exploration at wells XL1 and ND1. It was clear that the fourth member of Shahejie Fm. and the first member of Koingdian Fm. were the main source rocks in the regional deep layer.

Therefore, in view of petroleum geology, there was the possibility of deep hydrocarbon supply to form the Suqiao buried hill condensate reservoir, which increased the uncertainty about the understanding of the Wumaying buried hill reservoir as the source of coaly source rocks. Considering that, the study dissected the molecular isotope geochemistry and biomarker characteristics of crude oil in detail and made a good comparison with the Carboniferous-Permian coaly source rocks. The paper attempts to falsify the genetic types of condensate oil in the Wumaying buried hill reservoir from a geochemical point of view, so as to provide an important reference for oil and gas exploration in other areas of the Bohai Bay Basin.

2. GEOLOGICAL SETTING

Huanghua Depression was an important oil and gas-bearing area in the central Bohai Bay Basin, which was an asymmetrical long and narrow faulted basin with SW converging and NE spreading, with a total area of about 1.7×10^4 km^2 (Figure 1).[20,21] The Carboniferous-Permian strata were mainly composed of Benxi Fm., Taiyuan Fm., Shanxi Fm., and Shihezi Fm. (Figure 1b),[22] in which coaly source rocks were deposited in the Taiyuan Fm. and Shanxi Fm. (Figure 1b). Taiyuan Fm. mainly consisted of peat flat and lagoon sediments, containing thin layers of sandstone, limestone, and coaly source rock, and a relatively thick layer of black shale, which belonged to marine-continental deposits. The Shanxi Fm. was characterized by delta front and coastal swamp sediments, including carbonaceous shale, sandstone of intermediate layer, thin layer coal, and dark coaly shale of delta facies.[18,23,24] Most measured results of previous drilling samples showed that the total organic carbon (TOC) content of coal was 20.5−75.0 wt %, with an average of 45.6 wt % (S_1+S_2 varying from 60.00−300.00 mg/g). The TOC content of coaly shale changed between 0.25 and 14.83 wt % (average value: 4.32 wt %). The overall quality of coaly shale and coal ranged from poor to good.[25,26]

3. SAMPLES AND METHODS

The Carboniferous-Permian coaly source rock samples were mainly collected from well GG16102L, well CG2, and well QG1601, including 19 coal, 11 carbonaceous mudstone, and 66 coaly shale. The crude oil samples consisted of wells YG1, YG 2, Y101 × 1, G17102, and well Z1508-10.

3.1. Determination of Total Hydrocarbon Components in Crude Oil. An Agilent 5977A gas chromatograph with a DB-1MS (60 m × 0.32 mm × 0.25 μm) elastic quartz capillary chromatographic column was used. The heating program was set at an initial temperature of 30 °C with a constant temperature for 15 min and then heated at a rate of 3 °C/min and up to 310 °C with a constant temperature for 30 min. Nitrogen was used as a carrier gas with a flow rate of 1.0 mL/min. The inlet temperature was 290 °C. The split ratio was 10:1, and the detector temperature was 300 °C. The identification of light hydrocarbon compounds was based on the retention time and determined by a comparison of the literature.

3.2. Extraction and Separation of Extractable Organic Matter from Coaly Source Rocks. Using the mixed solution of dichloromethane and methanol as solvent, the extractable organic matter (chloroform asphalt A) in the coaly source rock was extracted by the Soxhlet extraction method, and the element sulfur was removed by the activated copper sheet. The composition of chloroform asphalts "A" or crude oil was separated by column chromatography of silica gel and alumina, and the asphaltene was precipitated and filtered by petroleum ether (30−60 °C). The asphaltene was removed through silica gel and alumina columns (3:1, V/V) and eluted with petroleum ether (30−60 °C), benzene, and methanol to obtain the saturated hydrocarbon component, aromatic hydrocarbon component, and polar component, respectively. The saturated hydrocarbons were further separated into n-alkanes, isomeric alkanes, and cycloalkanes by urea complexation.

3.3. Analysis of Biomarkers in Source Rock and Crude Oil. An agilent-5977A gas chromatograph with a hydrogen flame ionization detector (FID) was used to analyze saturated hydrocarbons and aromatic hydrocarbons directly and calculate the content and ratio of compounds. The DB-1MS (60 m × 0.32 mm × 0.25 μm) column and the DB-5MS (60 m × 0.32 mm × 0.25 μm) column were used, respectively. The carrier gas was high-purity nitrogen, and the flow rate was 1 mL/min. The temperatures of the injection port and FID are 290 and 300 °C, respectively. The temperature increase procedure for saturated hydrocarbon detection was that the initial temperature was 60 °C, lasting for 2 min. Then, the temperature was raised to 220 °C at a heating rate of 3 °C/min and increased to 295 °C at a heating rate of 2 °C/min with maintaining for 30 min. The heating program for aromatic hydrocarbon detection was as follows: the initial temperature was 60 °C for 2 min. Afterward, the temperature was heated to 295 °C at a heating rate of 3 °C/min and maintained for 30 min.

An agilent 7890B-5977A chromatographic mass spectrometer was used to analyze isomeric/cycloalkanes and aromatics with a DB-1MS (60 m × 0.32 mm × 0.25 μm) column and a DB-5MS (60 m × 0.32 mm × 0.25 μm) column, respectively. The temperature of the injection port at the chromatography was 290 °C, and the temperature of the auxiliary heater was 280 °C. The ion source of the mass spectrometry was an electron bombardment source (EI, 70 eV) with a temperature of 230 °C. The sample was injected in non-shunt mode, the carrier gas was high purity helium, and the flow rate was 1 mL/min. Isomeric/naphthenic hydrocarbon component chromatographic heating program: the initial temperature was 60 °C with a 2 min duration and increased to 220 and 295 °C at a heating rate of 3 and 2 °C/min separately, and the final temperature was maintained at 30 min. The ion source adopted full scan plus selective ion scanning mode, and the selected ions were m/z = 123, 183, 191, 205, 217, and 218.
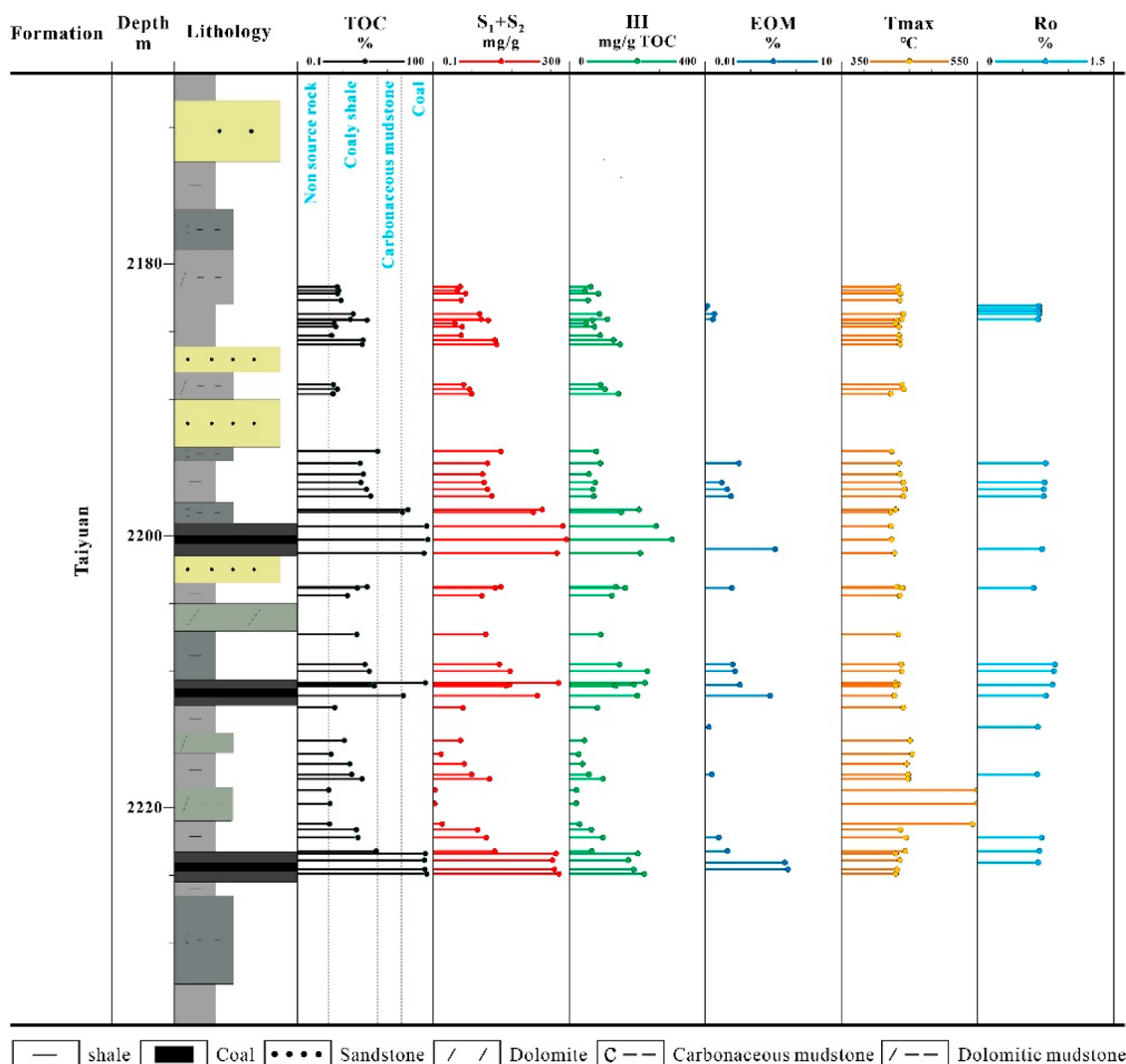
Figure 2. Comprehensive profile of organic geochemical characteristics of Coaly source rock in well GG16102 of Taiyuan Fm. Notes: TOC means total organic carbon; $S_1 + S_2$ means hydrocarbon generation potential; HI means hydrogen index; EOM means extractable organic matter; T_{max} means the maximum temperature of kerogen degradation during pyrolysis; and R_o means vitrinite reflectance.

Aromatics components chromatographic heating program: the initial temperature was 60 °C for 2 min, heated up to 300 °C at a heating rate of 3 °C/min, and the constant temperature was kept for 30 min. The ion source was selected as a full scan mode.

3.4. Stable Carbon Isotope Analysis of *n*-Alkanes in Coaly Source Rocks and Crude Oils. The monomeric carbon isotope analysis of *n*-alkanes was carried out by Seam Field Trace Ultra gas chromatography-isotope ratio mass spectrometry (MAT-253). The gas chromatograph was equipped with a DB-1MS capillary column (60 m × 0.32 mm × 0.25 μ m). The initial temperature was 50 °C holding for 1 min, raised to 120 °C at a rate of 1.5 °C/min, and then increased up to 300 °C at a rate of 5 °C/min, maintaining 25 min. The non-shunt injection mode was adopted, the carrier gas was high-purity helium (flow rate 1.2 mL/min), and the inlet temperature was 290 °C. All samples were tested in parallel to ensure the repeatability of the data. External standard samples were tested six times to control the status of the instrument. The standard sample was a group of *n*-C$_{16}$ to *n*-C$_{30}$ standards (Indiana University, Bloomington, USA). The

test accuracy was less than ±0.5‰ and followed the VPDB standard.

4. RESULTS AND DISCUSSION

4.1. Organic Geochemical Characteristics of Carboniferous-Permian Coaly Source Rocks. *4.1.1. Bulk Characters.* The coaly source rocks of Taiyuan Fm. were mainly developed in the tidal flat environment under the background of regression, which was more stable than that of the delta plain (Shanxi Fm. coaly source rocks).[27] Therefore, the development of the coal seam was relatively stable laterally, and 3−4 layers of main productive coal seams could be seen on the well profile (Figure 2), which was considered to be the main source rock for oil and gas generation. The results of TOC and Rock-eval analysis from coaly source rocks of Shanxi Fm. and Taiyuan Fm. showed that the highest hydrogen index (HI) of the coaly organic matter can reach 336.00 mg/g TOC, containing mainly type II$_2$ organic matter with TOC distribution in the range of 22.3 to 76.4 wt % and high oil generation potential ($S_1 + S_2$ between 67.68 and 254.97 mg/ g). All parameters indicated a robust foundation for the hydrocarbon supply from coaly source rock (Figure 2). The
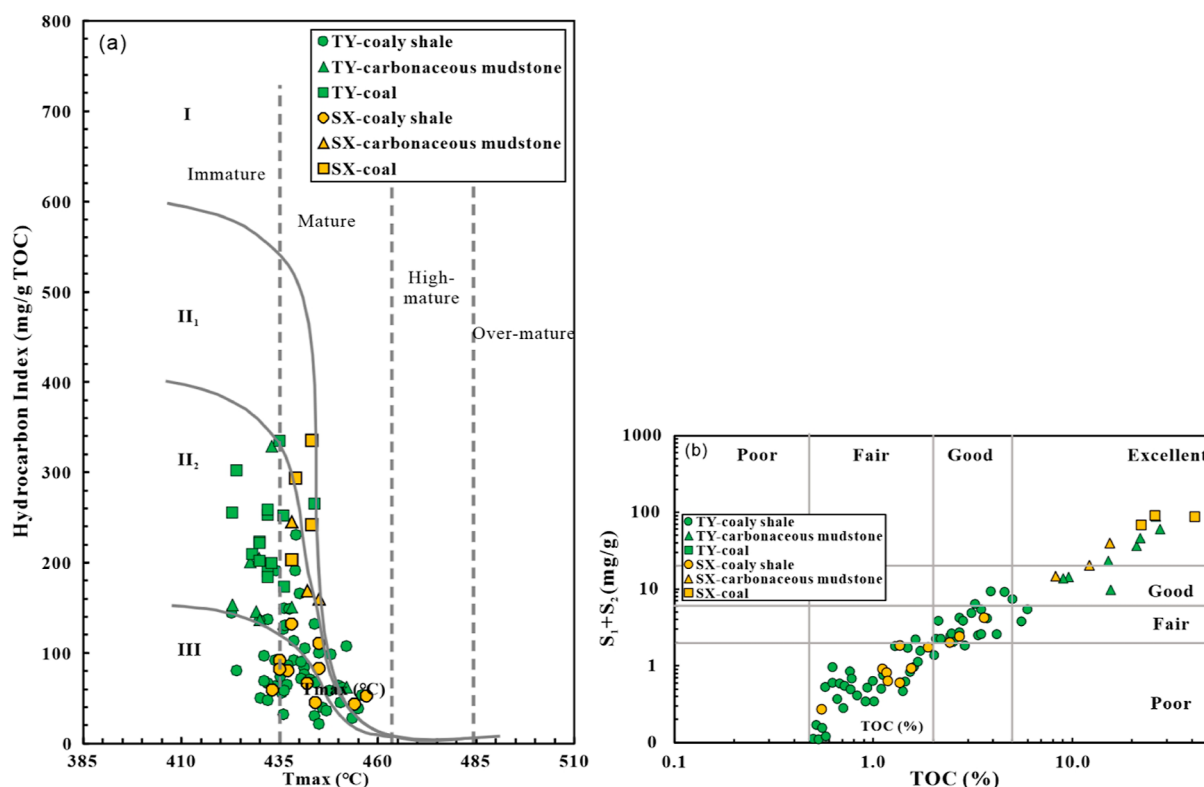
Figure 3. Organic matter types and hydrocarbon potential of Carboniferous-Permian coaly source rocks.

coaly shale was characterized by low TOC content (less than 5.97 wt %, average value of 1.93 wt %) and corresponded to the low HI characteristic (HI:20.32−230.69 mg/g TOC), which was mainly composed of type III kerogen (Figure 3a). The hydrocarbon generation potential of coaly shale was not higher than 9.30 mg/g (Figure 3b). The kerogen type and hydrocarbon generation potential of carbonaceous mudstone were better than those of coaly shale but worse than coal, indicating better hydrocarbon supply capacity (Figures 2 and 3).

4.1.2. Biomarker Distribution. Variations were observed in the distribution of biomarkers among the three types of extractable organic matter in rocks. Ranging from dark coaly shale and carbonaceous mudstone to coal deposits, the main peak carbon number of n-alkanes in extractable organic matter exhibited a trend of backward shift. Concurrently, there was an increasing trend in the relative content of medium- to long-chain n-alkanes and in the Pr/Ph ratios. The changing trend typically reflected the increase in the contribution of terrestrial higher plants to sedimentary organic matter and the enhancement of oxidation in the sedimentary environment during the development from dark coaly shale to coal (Figure 4). The distribution of steranes in extractable organic matter showed that the dominance of C_{29} regular steranes was more prominent, and the relative contents of C_{27} regular steranes, rearranged steranes, pregnane, and homopregnane decreased, reflecting the decrease of input from lower aquatic organisms such as algae and the weakening of catalytic effects from clay minerals. The relative content of tricyclic terpanes decreased progressively from dark coaly shale to carbonaceous mudstone and then to coal deposits. Additionally, trace gammacerane compounds were significantly detected in the extractable organic matter of coal shale (Figure 4).

The composition of saturated hydrocarbon biomarkers presented different characteristics within coaly source rocks. Trace gammacerane and 18α (H)-22, 29, 30-trinorhopane (Ts) series compounds were detected in coal, carbonaceous mudstone, and coaly shale. C_{29} and C_{30} hopanes were distributed with similar abundance, accompanied by a high content of C_{29} steranes and abundant terpenoids (Table 1). Coal exhibited a higher Pr/Ph ratio and a lower Pr/nC_{17}, Ph/nC_{18} ratio compared to carbonaceous mudstone and coaly shale (Figure 5). Coaly shale demonstrated a noticeable predominance of low-carbon n-alkanes and contained more rearranged steranes, pregnane, tricyclic terpanes, and methyl-hopane (Figures 4 and 5). The compositional characteristics of saturated hydrocarbon biomarkers from carbonaceous mudstone showed a transitional nature from coal to coaly shale. These characteristics were more inclined toward coaly shale in the Taiyuan Fm. and more similar to coal in the Shanxi Fm. (Figure 4, Table 1).

4.1.3. Light Hydrocarbons in Coaly Rocks. The results showed that the light hydrocarbon composition of coaly source rock in the Taiyuan Fm. was characterized by a high content of benzene series, including benzene, toluene, xylene, C_3-benzene, C_4-benzene, etc. The relative contents of cyclohexane and methylcyclohexane were significantly lower than those of the benzene series. The heavy hydrocarbons displayed comparability with the saturated hydrocarbon fractions obtained by Soxhlet extraction, characterized by a high Pr/Ph ratio and high-carbon-number n-alkanes with a slight odd−even predominance (Figure 6). The distribution characteristics of light hydrocarbons in the extractable organic matter of Taiyuan Fm. coal were consistent with the results from pyrolysis gas chromatography of most coal samples, often marked by high concentrations of light aromatic hydrocarbons and phenolic compounds (Figure 6a).
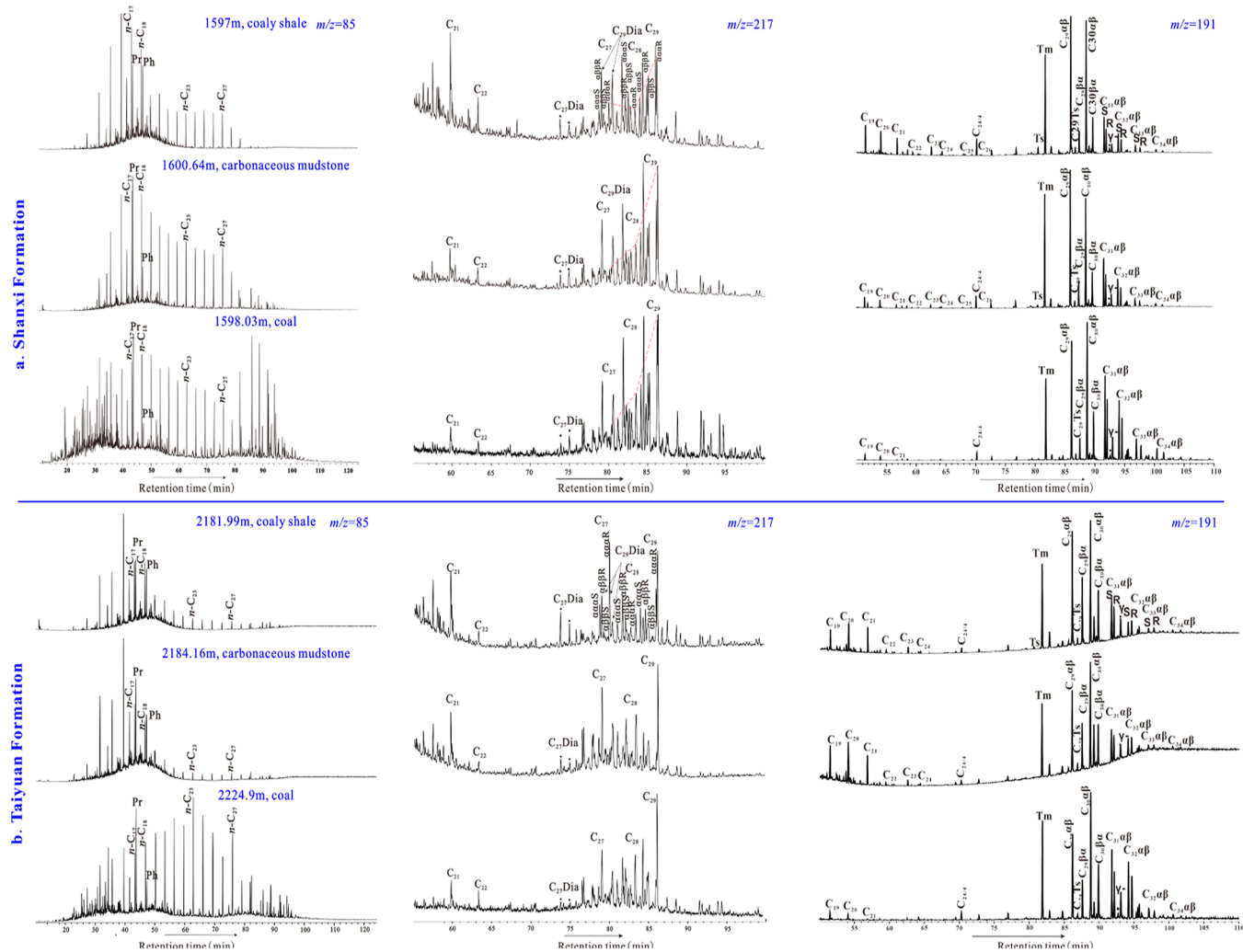
Figure 4. Distribution characteristics of biomarkers in extractable organic matter of coaly source rocks in the Taiyuan Fm., well GG 16102, and well CG2.

Table 1. Differences of Saturated Hydrocarbon Composition in the Coaly Source Rocks of Taiyuan-Shanxi Fm

lithology	common features	inherent features
coal	(1) $C_{29}ab$-norhopane and $C_{30}ab$−hopane equilibrium distribution	high Pr/Ph ratio
	(2) trace gammacerane	enrichment of medium chain length of high-carbon-number n-alkanes
	(3) trace Ts series compounds	C_{29} steranes > C_{27} steranes
	(4) C_{29} sterane dominance	detection of trace tricyclic terpanes
	(5) abundance of terrestrial diterpanes	abnormal enrichment of methyl hopane
carbonaceous mudstone		more inclined to coaly shale in the Taiyuan Fm. and more similar to coal in the Shanxi Fm. during the transitional stage
coaly shale		medium Pr/Ph ratio
		advantage of n-alkanes with low carbon numbers
		C_{29} steranes > C_{27} steranes, C_{27} regular steranes were significantly detected
		high relative content of rearranged sterane and pregnane
		tricyclic terpanes were significantly detected
		significantly detection of ethyl hopane

Compared with the extractable organic matter of coal, light hydrocarbons in the coaly shale of Taiyuan Fm. not only had the inherent characteristics of a high benzene series and a high Pr/Ph ratio but also contained significantly higher relative contents of cyclohexane and methyl cyclohexane. In the total hydrocarbon chromatogram, low-carbon-number hydrocarbons were dominant, and the relative content of high-

carbon-number n-alkanes decreased in the coaly shale significantly (Figure 6b). The increase in the relative content of cyclohexane and methyl cyclohexane in light hydrocarbon molecules of extractable organic matter in coaly shale indicated an increase in the input contribution of sapropelic organic matter, which accorded with the actual sedimentary background. The biggest difference between the coaly shale and
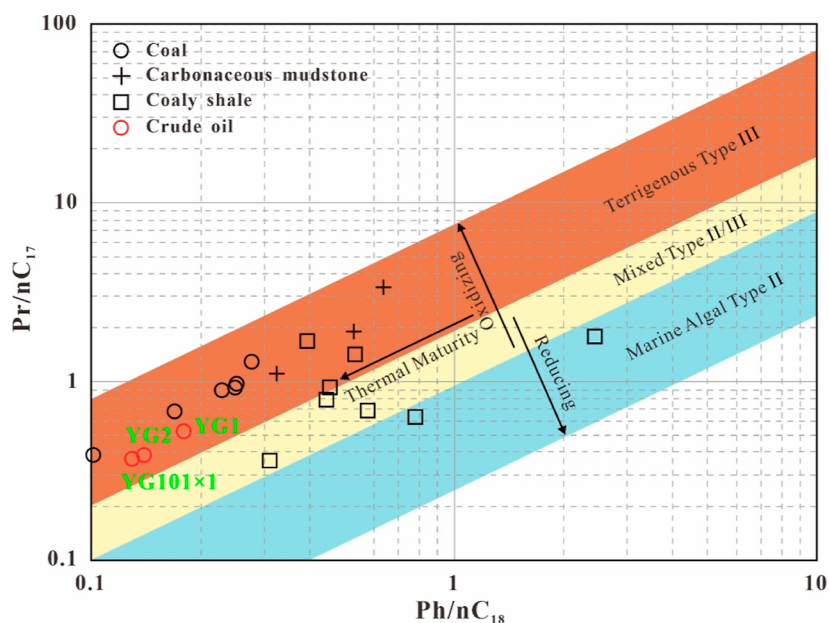
Figure 5. Characteristics of the Pr/Ph ratio in the sedimentary organic matter of coaly source rock in the Shanxi Fm.
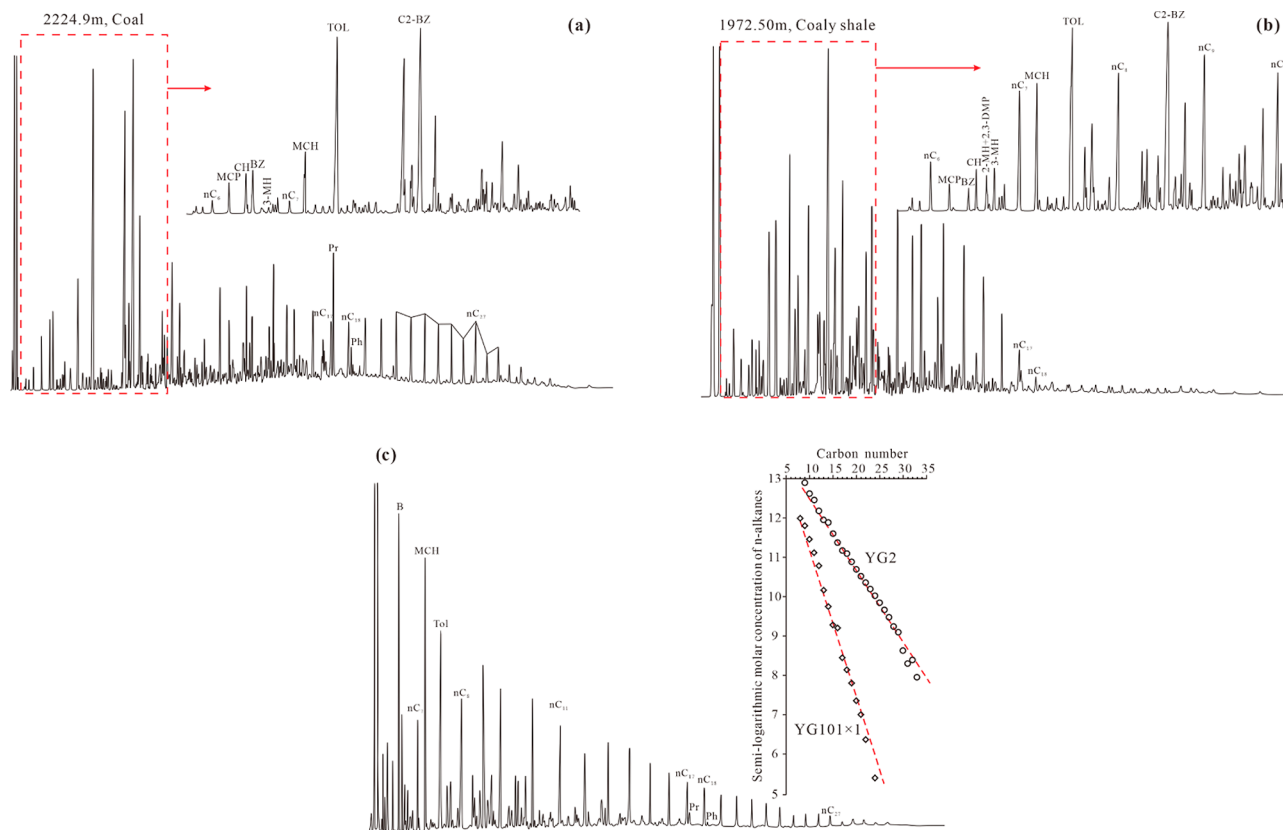

Figure 6. Characteristics of total hydrocarbon chromatography and semilogarithmic distribution of *n*-alkane mass concentration in Carboniferous-Permian coaly source rock and condensate of wells YG2 and YG1001 × 1 (a,b) are light hydrocarbons in the extractable organic matter of Taiyuan Fm. coal and coaly shale, separately; (c) light hydrocarbons in condensate of wells YG2 and YG1001 × 1.

coal sedimentary environments was that the former water body had a certain depth and was easy to develop with a certain algae productivity. Moreover, the relative content of C_{27}–C_{28}–C_{29} regular sterane in the extractable organic matter of coal was significantly higher than that in coaly shale (Figure 6).

4.1.4. Stable Carbon Stable Isotopes of n-Alkanes. Some differences in the distribution patterns of *n*-alkane molecular

carbon isotopes existed in the extractable organic matter of Taiyuan Fm. coal and coaly shale. The carbon isotope composition of *n*-alkanes in extractable organic matter of coaly shale basically spread horizontally with the increase of carbon chain (Figure 7), which was significantly different from the carbon isotope distribution pattern of *n*-alkanes in sedimentary organic matter of freshwater lacustrine.[28,29] With
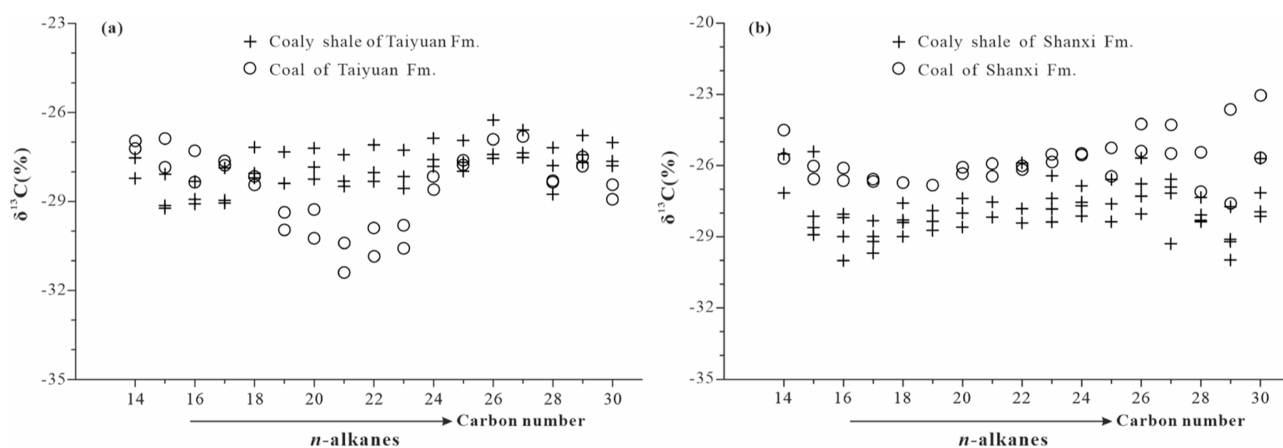
Figure 7. Monomeric carbon isotopic distribution of *n*-alkanes in the extractable organic matter of coaly source rocks of Taiyuan (a) and Shanxi Fm. (b).

Table 2. Physical Properties of Condensate-Light Oil

well	density of 20 °Cg/cm³	kinematic viscosity of 50 °C mPa·s	freezing point °C	wax content %	sulfur content %	crude oil
YG1	0.8091	1.38	≤30			condensate-light oil
YG2	0.7739	0.67		3.96	0.03	
YG101 × 1	0.7810	0.92	−20			

the increase of the *n*-alkane carbon chain, the carbon isotope became lighter and formed an oblique line. However, the carbon isotope distribution pattern of *n*-alkanes in coaly shale was similar to that of saline facies and marine sedimentary organic matter,[30,31] which was also consistent with the previous studies that the coaly deposits of Taiyuan Fm. in North China were developed under the background of sea regression. The carbon isotopes of *n*-alkanes in the extractable organic matter of Taiyuan Fm. showed a ship-shaped distribution with the increase of carbon chain, and the carbon isotopes were significantly rich in $\delta^{12}C$ within the carbon number range of medium chain length ($nC_{19}-nC_{23}$) (Figure 7a). The medium-length chain of long carbon-number *n*-alkanes in the extractable organic matter of coal mainly came from benthic algae with a relatively low niche. The significant negative deviation of carbon isotope may be related to the carbon microcirculation of the swamp environment; that is, the algae with a lower niche used CO_2 produced by organic matter degradation as the carbon source, whose carbon isotope was significantly lighter than atmospheric CO_2.[32,33] The production of these CO_2 may be mainly related to chemical autotrophs, and the detection of high abundance hoppers in coal confirmed the activity of chemical autotrophs.[34]

The molecular carbon isotopes of *n*-alkanes in the extractable organic matter of Shanxi Fm. coal and coaly shale were similar in distribution pattern, showing flat and gentle characteristics, which were consistent with the characteristics of marine-continental sedimentary organic matter (Figure 7b). The molecular carbon isotope of *n*-alkanes in the coal extractable organic matter of Shanxi Fm. coal was about 1‰ heavier than that of coaly shale, which was a common phenomenon observed in all samples. The most significant difference from Taiyuan Fm. coal was that the carbon isotope distribution pattern of *n*-alkanes in the extractable organic matter of Shanxi Fm. coal did not exhibit a ship shape. Furthermore, the extractable organic matter from Shanxi Fm. coal was also rich in 2α- and 3β-methyl hopane series. This variance could be attributed to the distinct sedimentary

environment of the Shanxi Fm. coal. The coal of Shanxi Fm. was formed in the swamp facies of the delta plain under the background of sea retrogression during Taiyuan Fm. deposition, which belonged to a high swamp. The environment was completely different from the tidal flat sedimentary environment of Taiyuan Fm., which was dominated by low swamp, the water body was relatively deep, and it was easy to develop low niche algae such as benthic algae, which provided a user for the degradation of organic matter to produce CO_2. Therefore, although abundant 2α- and 3β-methyl hopane series were detected in the extractable organic matter of Shanxi Fm. and Taiyuan Fm. coal, the supply of sedimentary organic matter, mainly in Shanxi Fm. coal, depended on terrestrial plant input due to the lack of conditions conducive to algae breeding. This was reflected in the molecular carbon isotope composition of *n*-alkanes, which was richer in $\delta^{13}C$ than that of the extractable organic matter of coaly shale (Figure 4). The coal of the Shanxi Fm. was obviously richer in heavy carbon isotopes than coaly shale (including carbonaceous mudstone, Figure 7), which could have been caused by the relatively deeper sedimentary environment of the coaly shale, which is more suitable for the reproduction of lower aquatic organisms such as algae. Compared with coal and carbonaceous mudstone, the input of algae in the coaly shale of the Shanxi Fm. was significantly increased. The relatively high concentration of atmospheric carbon dioxide that dissolved in deeper water bodies during the deposition of coaly shale eventually led to lighter carbon isotopes of organic matter derived from phytoplankton than those originating from terrestrial higher plants.

4.2. Biomarkers in Condensate from Wumaying Buried Hill. *4.2.1. Condensate Properties.* Industrial oil and gas flow in Xiashihe Fm. was discovered in wells YG 1, YG 2, and YG 101 × 1 in the Wumaying buried hill. The density and kinematic viscosity of condensate were 0.7739−0.8091 g/cm³ under 20 °C and 0.67−1.38 mPa·s under 50 °C, with less than 5% of wax content and less than 0.1% of sulfur content (Table 2). The properties revealed low wax and low sulfur condensate.
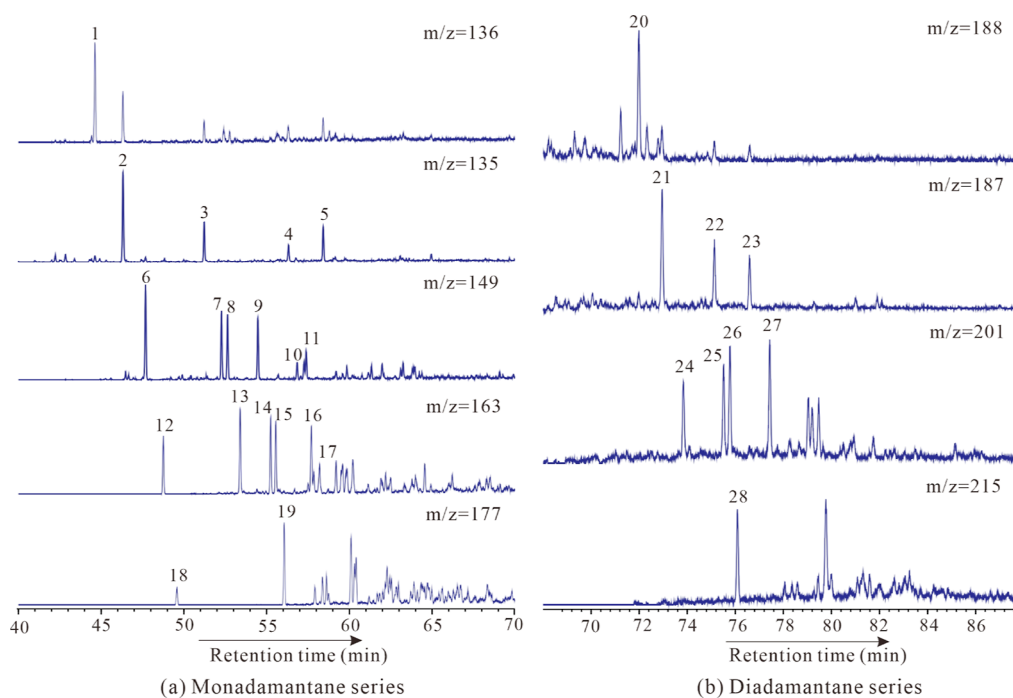
Figure 8. Mass chromatogram of monoadamantane and diadamantane series compounds in the condensate oil of Wumaying Buried hill (1) monoadamantane; (2)1-methyl monoadamantane; (3) 2-methyl monoadamantane; (4) 1-ethylmonoadamantane; (5) 2-ethylmonoadamantane; (6) 1,3-dimethylmonoadamantane; (7) 1,4-dimethylmonoadamantane (cis isomer); (8) 1,4-dimethylmonoadamantane (trans isomer); (9) 1,2-dimethylmonoadamantane; (10) 2,6-dimethyladamantane + 2,4-dimethyladamantane; (11) 3-methyl-1-ethyl monoadamantane; (12) 1,3,5-trimethyl monoadamantane; (13) 1,3,6-trimethyl monoadamantane; (14) 1,3,4-trimethyl monoadamantane (cis isomer); (15) 1,3,4-trimethyl monoadamantane (trans isomer); (16) 1,2,3-trimethyl monoadamantane; (17) 3,5-dimethyl-1-ethylmonoadamantane; (18) 1,3,5,7-tetramethylmonoadamantane; (19) 1,25,7-tetramethylmonoadamantane; (20) diamantane; (21) 4-methyl diamondane; (22) 1 methyl diamondane; (23) 3-methyl diamondane; (24) 4,9-dimethyldiamondane; (25) 1,4-dimethyldiamondane + 2,4-dimethyldiamondane; (26) 4,8-dimethyldiamondane; (27) 3,4-dimethyldiamondane; and (28) 1,4,9-trimethyldiamondane.

The total hydrocarbon chromatography of condensate showed that medium- and low-carbon-number alkanes were dominant, with the highest carbon number being nC_{33}, and the content of alkanes above the nC_{25} chain was relatively low. The high content of the benzene series and methylcyclohexane in light hydrocarbon components was one of its main characteristics (Figure 6c).

4.2.2. Characteristics of Saturated Hydrocarbon Composition in the Condensate Oil. According to its physical properties, the crude oil of the Wumaying buried hill reservoir was a condensate-light oil. The whole hydrocarbon chromatography showed that medium- and low-carbon-number alkanes were dominant, with few of the above-mentioned nC_{25} chain alkanes and the highest carbon number reaching nC_{33}. Light hydrocarbon components were characterized by the high content of the benzene series and methylcyclohexane (Figure 6c). Two possibilities could cause the high content of benzene-series compounds in crude oil. First, coaly organic matter made it easy to produce crude oil of high benzene series content.[35,36] Second, the crude oil of the paleo-reservoir was cracked to form a light oil reservoir of high aromatic hydrocarbon component content by cracking, cyclization, and aromatization, including benzene series.[37,38] The high benzene series content of Wumaying buried hill crude oil should be mainly related to the parent material of hydrocarbon generation. Compared to normal alkanes, the relative contents of isoprenoid alkanes, as indicated by lower Pr and Ph content and a higher Pr/Ph ratio of about 3.2, were observed in wells

YG1, YG2, and Y101 × 1. The corresponding values of Pr/nC_{17} and Ph/nC_{18} were also relatively low in these wells.

The semilogarithmic distribution of the molar mass of *n*-alkane content in crude oil can usually reflect whether the crude oil has experienced secondary processes such as mixing, evaporative fractionation, and biodegradation.[39] For crude oil from a single source and not affected by secondary alteration, The semilogarithmic distribution of molar mass of *n*-alkane content in crude oil showed a linear relationship with the distribution of carbon number, namely, it was approximately a straight line on the plane diagram. Based on the chromatographic analysis of crude oil, it was found that the semilogarithmic distribution of *n*-alkane molar mass in the Wumaying buried hill condensate-light oil trended linearly (Figure 6c). This indicated a single source and suggested that the oil did not undergo significant secondary alteration.

4.2.3. Adamantane Index of Condensate and Its Maturity Indication. The concentration of benzene and toluene in condensate oil was controlled by source rock and thermal maturity. Additionally, evaporation fractionation led to the enrichment of the monoaromatic hydrocarbons. Furthermore, the stripping of Carboniferous-Permian coaly source rocks around the reservoir was also a possible factor in the increased concentrations of benzene and toluene in the condensate oil. The maturity of condensate needed to be confirmed to analyze the reason for the high concentration of monoaromatic hydrocarbons effectively.

Normally, with an increase of the thermal evolution degree, kerogen would degrade and release a large amount of *n*-

Table 3. Molecular Parameters of Light Hydrocarbons in Deep Condensate Oil of Wumaying Buried Hill[a]

well name	C_6-C_9 n-alkanes/ C6−C9 (%)	C_6-C_9 cycloalkane/ C6−C9 (%)	C_6-C_9 aromatic/ C_6-C_9 (%)	methyl cyclohexane index (%)	cyclohexane index (%)	heptane (%)	isoheptane (%)
YG1	47.26	29.09	23.65	59.63	38.36	43.67	1.41
YG2	35.52	28.05	36.44	62.42	44.89	43.57	1.54
Y101 × 1	53.27	28.49	18.24	60.36	24.57	48.04	2.19
average value	45.35	28.54	26.11	60.80	35.94	45.09	1.71

[a]Note: methylcyclohexane index (%) = MCH × 100%/(MCH + T-1, 3-DMCP + C-1, 3-DMCP + T-1, 2-DMCP + nC7).
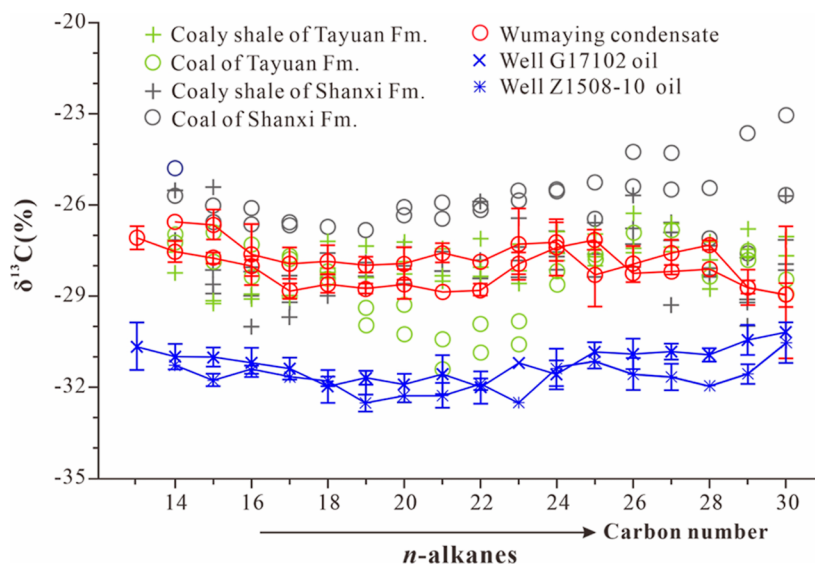

Figure 9. Monomeric carbon isotope comparison curve of n-alkanes in Wumaying buried hill crude oil, well G1702, well Z1508-10, and coaly source rocks.

alkanes, resulting in a gradual decrease in the values of Pr/n-C_{17} and Ph/n-C_{18}. Therefore, the closer to the lower left corner of the Pr/n-C17 vs Ph/n-C_{18} graph, the higher the maturity of the sample. The condensate samples from the Wumaying buried hill were distributed very closely and located in the lower left corner of the graph, indicating the same biogenic characteristics and a higher degree of thermal evolution (Figure 5).

Nineteen monoadamantanes and nine diadamantane series compounds were detected in the Wumaying buried hill condensate (Figure 8). According to the methyl-monoada-mantane index (MAI) and methyl-diadamantane index (MDI) formulas, the MAI ratios of YG1, YG2, and Y101 × 1 well condensates were calculated and distributed between 62 and 69%, with an average of 66%. The MDI ratios ranged from 46 to 51%, with an average of 49%. According to the corresponding relationship between adamantane index and vitrinite reflectance,[40] the equivalent vitrinite reflectance (Rc) of Wumaying buried hill condensate was about 1.2−1.3% R_o, indicating a high thermal maturity. This result was consistent with the maturity of the Wumaying buried hill condensate obtained by using isoheptane−heptane values and Pr/nC_{17}−Ph/nC_{18} values.

4.3. Geochemical Evidence for the Coal-Derived Genesis of Condensate in the Wumaying Buried Hill.
4.3.1. Direct Evidence of High Iso-alkanes and Benzene Series. One of the main characteristics of crude oil in the Wumaying buried hill condensate reservoir was the high content of gaseous hydrocarbons, the high benzene series and cycloalkane series, and the high Pr/Ph ratio in the medium-

length chain of hydrocarbons (Figure 6c). The high Pr/Ph ratio and benzene series content may indicate that the condensate came from a set of source rocks developed in an oxidizing environment, while low Pr/nC_{17} and Ph/nC_{18} values indicated that the source material of crude oil was mainly terrigenous organic matter (Figure 5). According to the regional petroleum geological background, the sedimentation of Carboniferous-Permian coaly source rocks in the area was accorded with the characteristic, indicating the possibility of hydrocarbon supply from Carboniferous-Permian coaly source rocks in Wumaying buried hill condensate reservoir (Figure 6b). However, the crude oil derived from coaly source rocks should be characterized by a high Pr/Ph ratio, trace gammacerane, high C_{27}Tm compounds, a dominant distribution of C_{27} steranes in regular steranes, and terrestrial diterpanes. Trace steroidal terpane biomarkers in the condensate oil were detected but presented uncertainty, making it difficult to effectively clarify oil-source correlations. Furthermore, although terrestrial diterpanes were commonly detected in the extractable organic matter of Carboniferous-Permian coaly source rocks, most of them had undergone cracking due to the high thermal maturity of the condensate (equivalent vitrinite reflectance 1.1−1.2% R_o) and could not be used for oil-source correlation analysis.

4.3.2. Cyclohexane Evidence for Condensate Originating from Coaly Source Rocks. The methyl-cyclohexane index of the YG1 and YG2 wells' condensate in Wumaying buried hill was 59.63 and 62.42%, respectively. The index for the Y101 × 1 well was 60.36%, positioned in the middle with an average of 60.81%. Using 50 ± 2% as a threshold, the methyl-cyclohexane

index of Wumaying buried hill condensate displayed a clear source of humic organic matter. The cyclohexane index (CH × 100%/(CH + MCP + nC_6)) could also reflect the parent material type of the source rocks. Normally, the cyclohexane index of sapropelic (type I, type II) source rocks was less than $27 \pm 2\%$, and that of humic (type III) source rocks was more than $27 \pm 2\%$. The cyclohexane index of condensate in the YG1 and YG2 wells of Wumaying buried hill was 38.36 and 44.89%, respectively. The cyclohexane index of condensate in well Y101 × 1 was 24.57%, with an average of 35.94% (Table 3). All of the indexes from the three wells indicated a hydrocarbon contribution from humic organic matter.

4.3.3. Correlation of Stable Carbon Isotopes of n-Alkanes. As a new technology developed in the 1990s, molecular carbon isotope has been proven to be an effective method of oil–oil and oil-source correlation analysis.[41,42] The comparison indicated that the distribution of molecular carbon isotopes of *n*-alkanes in extractable organic matter was similar between the condensate, coaly shale, and carbonaceous mudstone of Taiyuan Fm. (Figure 9). A large number of studies have revealed that within the range of the main oil generation window, the carbon isotope fractionation effect of *n*-alkane molecular carbon isotopes due to different thermal maturity was less than 2‰, which means that *n*-alkane molecular carbon isotopes can be used to study oil-source correlation from the immature to highly mature condensate stage (Figure 9). For condensate and light oil, *n*-alkanes were the main components, and their identification was very important for the search for source rocks. The results of the molecular carbon isotope distribution of *n*-alkanes in Paleogene source rocks such as well G17102 and well Z1508-10 were added to realize a more accurate oil-source correlation.

The results showed that the molecular carbon isotope composition of *n*-alkanes in the Wumaying buried hill condensate was approximately horizontal (Figure 9). Combined with the distribution of high-content benzene series in crude oil, the near-horizontal model of carbon isotope distribution of *n*-alkanes in crude oil is likely to reflect a coastal swamp environment. It was difficult to develop this distribution pattern of source rocks deposited in marine, saline lakes, deep freshwater lakes, and shallow river-lake swamp environments. The molecular carbon isotope distribution of *n*-alkanes in Wumaying buried hill condensate was very consistent with that of Carboniferous-Permian coaly shale rather than the extractable organic matter of coal. The molecular carbon isotope of *n*-alkane in tertiary crude oil was significantly enriched in the $\delta^{12}C$ isotope compared with Wumaying buried hill condensate. Therefore, the molecular carbon isotope distribution of *n*-alkanes in crude oil was a good indicator to clarify the oil-source correlation between Carboniferous-Permian coaly shale and Wumaying buried hill condensate. The investigation provided new insight into the source discrimination in coaly related oil.

5. CONCLUSIONS

The coaly source rocks exhibited significant hydrocarbon generation potential, with the Taiyuan Fm. slightly surpassing the Shanxi Fm. It was observed that coal tended to generate more hydrocarbons than carbonaceous mudstone and dark coaly shale.

Variations were noted in the composition and concentration of biomarkers in the extractable organic matter from different lithotypes within the Carboniferous-Permian coaly source

rocks. Common characteristics included a high content of C_{29} $\alpha\beta$-hopane, trace amounts of gammacerane and the Ts series, a dominance of C_{29} steranes, and an abundance of terrestrial diterpanes. Compared to coaly shale, the extractable organic matter from coal exhibited a higher Pr/Ph ratio, a relatively higher concentration of medium-chain length *n*-alkanes, and more C_{29} steranes than C_{27} steranes. In contrast, coaly shale was characterized by a moderate Pr/Ph ratio, a predominance of low-carbon-number *n*-alkanes, C_{29} steranes exceeding C_{27} steranes (with significant detection of C_{27} steranes), and a relatively high abundance of rearranged steranes and pregnanes.

The condensate of the Wumaying buried hill was predominantly derived from coaly organic matter, which contributed to hydrocarbon generation. This was substantiated by the results of molecular carbon isotope comparisons of *n*-alkanes in crude oil. The isotope distribution of saturated hydrocarbon monomers proved to be an effective tool for discriminating the genesis of condensate-light oil.

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