Research Article

Climate-Provenance Effect on the Organic Matter Enrichment of the Chang 9 Source Rocks in the Central Ordos Basin, China

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Received 19 May 2021; Accepted 5 July 2021; Published 2 August 2021

Academic Editor: Bo Liu

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The Upper Triassic Chang 9 organic-rich sediments have been considered as effective hydrocarbon source rocks for the Mesozoic petroleum system in the Ordos Basin. Previous studies on the Chang 9 member mostly focused on the influence of their paleoproductivity and paleoredox conditions on the organic matter (OM) enrichment, whereas there are few studies on the influence of the paleoclimate condition and sediment provenance on the OM enrichment. In this study, a series of geochemical analyses was performed on the Chang 9 core samples, and their hydrocarbon generation potential, paleoclimate condition, and sediment provenance were assessed to analyze the effect of paleoclimate-provenance on OM enrichment. The Chang 9 source rocks are characterized by high OM abundance, type I−II OM type, and suitable thermal maturity, implying good hydrocarbon generation potential. Based on the C-values and Sr/Cu ratios, the paleoclimate condition of the Chang 9 member was mainly semihumid. In addition, the Th/Co vs. La/Sc diagram and negative δEuN indicate that the Chang 9 sediments were mainly derived from felsic source rocks. Meanwhile, the paleoweathering intensity of the Chang 9 member is moderate based on moderate values of CIA, PIA, and CIW, which corresponds to the semihumid paleoclimate. The relatively humid paleoclimate not only enhances photosynthesis of the primary producer, but also promotes chemical weathering intensity, leading to suitable terrestrial clastic influx to the lacustrine basin, which is beneficial for OM enrichment.

1. Introduction

The Ordos Basin, as the main continental petroliferous basin in central China, shows tremendous potential for oil and gas resources in the Mesozoic petroleum system [1, 2]. The hydrocarbon resource in the Ordos Basin is mainly derived from the Yanchang Formation, which is considered as the most important hydrocarbon source rock for the Mesozoic petroleum system, especially for the Chang 7 member [3–5]. In the past, the dark mudstone and shale in the Chang 8 to Chang 4+5 members have been recognized as the main source rocks [1, 3]. However, in recent years, an increasing number of studies suggest that the Chang 9 organic-rich sediments have a great contribution to the oil and gas resource enrichment [6–8]. Previous studies have suggested that the Chang 9 sediments contain high OM abundance and mixed biological sources with a thermal maturity of 0.66–1.05%, which exhibits excellent hydrocarbon generation potential [6, 8, 9]. The Chang 9 sandstones were good petroleum reservoirs with abundant intergranular and dissolution pores [6, 9]. The porosity and permeability range from 3.20% to 14.72% and from $0.09 \times 10^{-3}$ to $3.50 \times 10^{-3} \mu m^2$, respectively [10, 11]. In addition, numerous studies on oil-source correlation have indicated that oils in the Chang 8–Chang10 oil-bearing intervals have the contribution from the Chang 9 source rocks [10, 11]. In consideration of the important role of the Chang 9 organic-rich source rocks in hydrocarbon accumulation, the formation mechanism of the Chang 9 organic-rich source rock has been studied [6, 9]. Previous studies on the Chang 9 OM enrichment mechanism were mainly based on the paleoproductivity and paleoredox conditions, and the results have shown that the Chang 9 organic-rich sediments were deposited in a suboxic semideep lacustrine environment of fresh-brackish water mass with...
occasional volcanic activity [6, 12]. The paleoproductivity is regarded as the major factor controlling the formation of Chang 9\textsuperscript{th} black shales [6, 12]. However, the work on paleoclimate-provenance and its effect on OM enrichment of the Chang 9 member is still lacking, which restricts the establishment of an OM enrichment model.

In order to illustrate the relationship between OM abundance and paleoclimate-provenance of the Chang 9 member, a series of geochemical analyses including total organic content (TOC), rock pyrosis, X-ray fluorescence (XRF), and inductively coupled plasma mass spectrometry (ICP-MS) analyses was performed on core samples to analyze the hydrocarbon generation potential, paleoclimate condition, and sediment provenance of the Chang 9 sediments. This study not only fills the gap of the study on OM enrichment of the Chang 9 source rock, but also directs the future exploration of the Ordos Basin.

2. Geologic Setting

The Ordos Basin, located in central China, is the second largest petroliferous basin crossing five provinces with an area of 370,000 km\textsuperscript{2} [2, 13] (Figure 1). Based on the tectonic framework, the Ordos Basin can be divided into six first-order structural units: the Yimeng uplift, the Weibei uplift, the West Jinshinian fold belt, the Yishan slope, the Tianhuan depression, and the western margin thrust belt [14] (Figure 1). The Ordos Basin is a multicycle superimposed basin with sedimentary stability, depression-migration, and obvious torsion [14]. As a part of the North China Craton Basin, the Ordos Basin was a marginal ocean basin before the Late Triassic [13]. However, the Ordos Basin gradually transformed from the North China Craton Basin to an inland basin at the Late Triassic due to the influence of the Indosinian movement [1, 15]. Since then, the sedimentary facies changed from marine facies to continental facies, and the continental clastic deposits replaced marine deposits gradually [3, 16–18]. Especially at the Late Triassic period, the collision of the North China Block and the Yangtze Block led to the suture of two blocks and the uplift of the Qingling Mountains [14]. The intense tectonic process led to a sufficient depositional accommodation for a set of fluvial-lacustrine sediments with a thickness of approximately 4,000 m [18, 19]. The Yanchang Formation recorded the occurrence, development, and extinction process of the lake [16]. Based on the cycled lithologic alternations of sandstone and mudstone, the Yanchang Formation can be subdivided into 10 members, named Chang 1 to Chang 10 [20, 21] (Figure 1). The ninth member, the Chang 9 member, represents the expansion period of the lacustrine member, which results in
subaqueous fan deposits and fluvial delta sandstone and mudstone deposits from shallow to semideep water mass in the Longdong area [19, 22]. In particular, a set of thick-bedded black shale was deposited at the top of the Chang 9 member, named “Lijiapan” shale, which is characterized by high OM abundance, type I OM, and appropriate maturity [3, 6, 9].

In this study, the D81 well penetrates the whole Chang 9 member, which provides a good chance to reconstruct the paleoclimate and analyze sediment provenance. Based on the rhythmicity of lithology, the Chang 9 member can be divided into three subunits, named Unit I, Unit II, and Unit III (Figure 2). Sandstone, mudstone, and shale with occasionally lamellated tuff on the top are the main lithologies of the Chang 9 member in the D81 well (Figure 2). The shales are scattered and distributed into three subunits, which are characterized by terrestrial plant fossils (Figure 2). In addition, lenticular-shaped siltstone was distributed in shale layers, indicating that gravity flow deposit was developed in the Chang 9 member.

### 3. Sampling and Methods

In order to reconstruct the paleoclimate condition and analyze sediment provenance of the Chang 9 member, a total of seventeen core samples (including nine shales and mudstones) from the D81 well were selected to systematically investigate the hydrocarbon generation potential, reconstruct paleoclimate condition, and analyze sediment provenance during the deposition of the Chang 9 source rock through TOC, rock pyrolysis, XRF, and ICP-MS analyses. The location of the selected samples from the D81 well is shown in Figure 1. The studied samples were divided into three units from the bottom to the top of well D81 (Figure 2).

All selected core samples were crushed to powder under 200 meshes with agate mortar in order to avoid contamination. Then, powder samples were blended with hydrogen chloride (10%) for two hours to eliminate inorganic carbon completely. After that, deionized water was used to clear hydrogen chloride. After drying the residues, the Leco CS230 analyzer was used to analyze the TOC and total sulfur content ($S_T$). Rock pyrolysis analysis was performed on the powdered samples by an OGE-VI oil evaluation station.

Trace elements were determined by a Jena PlasmaQuant MS inductively coupled plasma mass spectrometer (ICP-MS) at the Institute of Geochemistry, Chinese Academy of Sciences [19, 22]. About 0.0500 grams of the powdered sample were placed in a PTFE bomb, and 0.6 ml of HF and 3 ml of HNO$_3$ were added. The sealed bombs were then placed in

![Figure 2: Geochemical profiles of the Chang 9 samples from the D81 well: (a) TOC, %; (b) C-value; (c) Sr/Cu; (d) Rb/Sr; (e) CIA; (f) CIW; (g) PIA; (h) La/Sc; (i) Th/Co; (j) δEu$_N$; (k) Ti/Al.](image-url)
an electric oven and heated to 185°C for about 24 h. After cooling, the bombs were heated on a hot plate to evaporate to dryness. 200 ng of Rh was added as an internal standard, and then 2 ml of HNO3 and 4 ml of water were added. The bomb was again sealed and placed in an electric oven at 135°C for about 5 hours to dissolve the residue. After cooling, the final dilute factor is about 3000 for ICP-MS measurement. The accuracies of the ICP-MS analyses are estimated to be better than ±5-10% (relative) for most elements. The sample preparation procedure for major elements (except SiO2) is the same as the trace elements and measured by ICP-OES (Agilent 720) with a dilute factor of 1000. The technique of alkali fusion was used for sample digestion for SiO2 measurement. 0.05 g of sample was dissolved with 0.25 g NaOH at 700°C in a Ag crucible for about 30 min. After cooling, the content was dissolved with hot water and acidified with 5 ml HCl; finally, make up to 250 ml for ICP-OES measurement. About 1 g of sample was used for LOI. The sample was weighed in a porcelain crucible and heated at 900°C in a muffle furnace for about 1 h; the weighted difference before and after ignition is the LOI. The analytical accuracies are estimated to be ±2% (relative) for major oxides present in concentrations greater than 0.5% and ±5% (relative) for minor oxides present in concentrations between 0.1 wt.% and 0.5%.

4. Results and Discussions

4.1. Organic Geochemical Characteristics. The TOC, S1, and S2, and data from rock pyrolysis analyses are listed in Table 1. The black shales in three units are characterized by high TOC varying between 3.79% and 28.96% (Unit I black shales averaging 6.8%, Unit II shales averaging 14.4%, and Unit III averaging 4.31%), whereas the gray mudstones have low TOC ranging from 0.37% to 2.04% (averaging 0.96%). A majority of samples in three units show low S2 values below 1% except Unit I black shales (1.06%–1.46%, averaging 1.27%) (Table 1).

The $T_{\text{max}}$ value is commonly used as a maturity proxy. Samples with $T_{\text{max}} < 435°C$, 435 to 445°C, 445 to 450°C, and 450 to 470°C are considered to be immature, early oil window, peak oil window, and late oil window, respectively [23–25]. Noteworthily, an $S_2$ value below 0.5 mg/g is unreliable and can result in erroneous $T_{\text{max}}$ values; hence, samples with $S_2 < 0.5$ mg/g were removed while discussing the OM type and maturity. The $T_{\text{max}}$ values of all samples range from 444°C to 458°C (averaging 453°C) in this study, revealing a peak oil window stage. Also, according to the formula proposed in Ref. [23] to convert $T_{\text{max}}$ values to EqVRo (EqVRo = 0.018 × $T_{\text{max}}$ – 7.16), the EqVRo values of the Chang 9 samples in well D81 fall between 0.83% and 1.08% (averaging 0.99%) indicating that they are in the peak oil window.

The HI values of black shales (328 mg/g–774 mg/g, averaging 550 mg/g) are much higher than those in gray mudstones (120 mg/g–230 mg/g, averaging 157 mg/g). The HI vs. $T_{\text{max}}$ diagram exhibits that the OM of the black shales and the gray mudstones belongs to type I and type II, respectively (Figure 3(a)). All black shales are very good to excellent hydrocarbon source rocks in the $S_2$ versus TOC plot, whereas the majority of the gray mudstones fall into fair to good quality fields (Figure 3(b)).

4.2. Paleoclimate Condition. Climate can affect the geochemical properties of lacustrine deposition via controlling epi-gene actions and terrigenous material influxes [26, 27]. The climatic conditions can be effectively separated into five types by the C-values (Figure 2). The calculation formula is $C =$
value = (Fe + Mn + Cr + Ni + V + Co)/(Ca + Mg + Sr + Ba + K + Na) (elements in ppm). A higher C-value represents a more humid climate. A C-value below 0.2, 0.2–0.4, 0.4–0.6, and 0.6–0.8 and above 0.8 imply arid, semiarid, semiarid to semihumid, semihumid, and humid climate, respectively [28]. The Chang 9 samples with C-values of 0.38–1.37 (averaging 0.72) were substantially deposited under semihumid conditions (Figures 2 and 4 and Table 2). However, the paleoclimates of three units have different variation scopes. The Unit III samples have the lowest and widely fluctuating C-values from 0.38 to 0.85 (averaging 0.65), implying semiarid-semihumid to semihumid environments (Figure 4). The C-values in the Unit II samples are highest and widely fluctuating between 0.73 and 1.37 (averaging 0.97), implying a transition from the semiarid-semihumid to semihumid paleoclimate of the Unit III interval to the humid paleoclimate of the Unit II interval (Figure 4). The Unit I samples, with relatively stable C-values of 0.59 to 0.80 (averaging 0.70), were deposited under a semihumid environment (Figure 4).

Contrary to C-value, Sr/Cu and Rb/Sr ratios as effective climate indicators decrease with more humid conditions. A humid climate has the Sr/Cu and Rb/Sr values of 1–10 and <0.5, while the Sr/Cu and Rb/Sr values > 10 and >0.5 mean a hot and arid climate [29, 30]. Nearly all studied samples have the Sr/Cu values below 10 except for two samples from the Unit III interval, implying that the Chang 9 period had a semihumid climate and the paleoclimate of the Unit III interval had an occasional fluctuation (Figure 4). The Rb/Sr ratios of the Chang 9 member vary from 0.33 to 0.63 with an average of 0.45, which indicates a semihumid condition (Figure 2).

4.3. Paleoweathering Intensity. The weathering intensity of the source region can influence the mineral components of detrital rocks. This is because immobile elements like Ti and Al can be retained in clastic rocks during weathering, whereas unstable elements are prone to be lost. The CIA value, defined by \( \frac{\text{Al}_2\text{O}_3/(\text{Al}_2\text{O}_3 + \text{CaO}^* + \text{Na}_2\text{O} + K_2\text{O}_\text{corr})}{100} \), is a widely utilized proxy for estimating chemical weathering conditions [31]. High CIA values of 80–100 reflect an intense weathering of parent rocks, whereas low CIA values of 50–60 imply a weak weathering condition [34]. The CIA values of Chang 9 sediments vary between 64.11 and 78.31 (Table 2; averaging 70.44), showing a moderate degree of chemical weathering in the source region. Moreover, the plagioclase index of alternation (PIA) and the chemical index of weathering (CIW) also are effective parameters to determine paleoclimate conditions, which can be calculated via the following formulas, respectively:

\[
\text{PIA} = \frac{\text{Al}_2\text{O}_3 - K_2\text{O}}{\text{Al}_2\text{O}_3 + \text{CaO}^* + \text{Na}_2\text{O}} \times 100
\]

and

\[
\text{CIW} = \frac{\text{Al}_2\text{O}_3 - K_2\text{O}}{\text{Al}_2\text{O}_3 - K_2\text{O} + \text{CaO}^* + \text{Na}_2\text{O}} \times 100
\]

The PIA and CIW values of the Chang 9 sediments are in the range of 70.4–86.9 and 74.7–88.7, respectively, which also indicate a moderate degree of chemical weathering during the Chang 9 stage (Table 2; Figure 2).

The paleoweathering degree of parent rocks can be intensified by both humid paleoclimate and tectonic uplift activities [26, 37]. Intense weathering conditions are commonly formed under warm and humid climates with strong rainfall.
and continental runoff [38]. Combining the CIA and PIA with CIW values, the paleoweathering intensity during the Chang 9 stage was moderate, which resulted in appropriate terrestrial detrital input for OM accumulation (Figures 2 and 4).

4.4. Sediment Provenance Analysis. Because of low solubility and short retention time in water, some elements like REE, Th, Co, Sc, Zr, and Ti are more stable and easily retained during weathering, deposition, and diagenesis [39, 40]. These stable elements in sedimentary rocks are mostly derived from terrestrial detrital sources and hence can be credibly applied to infer mineral compositions of source rocks [41].

The igneous rocks can be classified into silicic and basic rocks according to the silica content. Silicic rocks are characterized by high La/Sc and Th/Co ratios relative to basic rocks [41, 42]. The La/Sc and Th/Co ratios of Chang 9 samples vary from 1.84 to 3.90 (averaging 2.80) and from 0.23 to 4.43 (averaging 0.83), respectively, indicating that the sedimentary rocks of Chang 9 are mainly derived from silicic rocks.
Moreover, Eu anomalies in sedimentary rocks are commonly considered to be inherited from the provenance and commonly used for determining source rocks \[43–45\]. Eu anomalies can be calculated by the following formula: $\delta_{Eu}^N = \frac{Eu}{Sm \times Gd}^{1/2}$ (where subscript $N$ represents the chondrite normalized value). Generally, evident negative $\delta_{Eu}^N$ ($<0.95$) can occur in felsic rocks, significant positive $\delta_{Eu}^N$ (>1) can appear in hydrothermal fluids, whereas almost no $\delta_{Eu}^N$ exists in mafic source rocks \[27\]. The significant negative $\delta_{Eu}^N$ (0.53–0.99, averaging 0.70) demonstrates that the studied samples are mostly from felsic rocks and not affected by hydrothermal input. This conclusion is consistent with the TiO$_2$ vs. Zr bivariate diagram (Figure 5(b)). The other study also confirmed that there is no hydrothermal activity during the Chang 9 period \[46\].

4.5. Clastic Influx. Clastic influx can affect sedimentation rate (SR) and OM enrichment in sediments. Generally, increasing detrital influx can accelerate SR and decrease OM biodegradation efficiency. However, excess detrital influx will dilute OM concentration in the sediments \[47\]. As observed in both marine and lacustrine sediments, the preservation effect results in the increase of TOC with the SR at below the threshold of 5 cm/ka, after which the dilution effect leads to the decrease of TOC \[48–50\]. Elements like Ti and Al are considered to mainly originate from clastic rocks. Thus, the Ti/Al ratio is selected to serve as an effective proxy of detrital influx. High Ti/Al usually means more detrital input and high SR \[51–53\]. The Chang 9 samples show no evident correlation between the Ti/Al ratio and TOC content. However, the TOC values initially rise as the Ti/Al ratios increase, and then decline after the Ti/Al values reach about 0.045 (Figure 6), implying that only moderate clastic input is the most helpful for OM enrichment.

The Ti/Al values of Unit I black shales (0.036–0.043, averaging 0.04) in the studied area are below and closest to the optimum value of 0.045 (Table 2 and Figure 6), implying the most beneficial clastic input for OM preservation during the Chang 9 period. The Unit II black shales generally show high Ti/Al ratios (0.036–0.051, averaging 0.045), mostly above 0.045, indicating a high detrital influx and dilution effect. The Unit III black shales generally have fluctuant Ti/Al ranges (0.035–0.049, averaging 0.041), mostly lower than that of Unit II black shales, representing a low detrital influx and preservation effect. The gray mudstones, except for two samples with the lowest Ti/Al values, exhibit high Ti/Al

![Figure 5: The crossplots of La/Sc vs. Th/Co and TiO$_2$ vs. Zr of the Chang 9 samples.](image-url)

![Figure 6: Crossplot of Ti/Al vs. TOC of the Chang 9 samples.](image-url)
values (0.044–0.053, averaging 0.049), indicating the highest clastic input and dilution effect.

4.6. The Effect of Paleoclimate-Provenance on OM Enrichment. The Indosinian movement caused the uplift of the Qinling Orogenic belt and subsidence of the Ordos basin during the Late Triassic. The Yin Mountain in the north and the Qinling Orogenic belt in the south were considered as primary source regions for the Upper Triassic fluvial-lacustrine-deltaic sediments in the Ordos basin [1]. Previous researches have manifested that there was an accelerated tectonic subsidence during the Chang 9 period, leading to an increase of lake depth, which is consistent with the maximum lake depth in the late Chang 9 period. The increase of lake area and depth caused a decrease of detrital rock influx and the suitable SR for OM preservation in a deep lake. Additionally, the basin periphery frequently underwent volcanic activities during the Late Triassic [46]. Volcanic tuffs from the basin margin have been discovered in the Unit I interval (Figure 2), which can provide large quantities of nutrient elements (like P, Fe, Mo, and Cu) for primary producer booming during its deposition [46].

Apart from tectonic and volcanic activities, paleoclimate condition and sediment provenance also affect the OM enrichment of the Chang 9 sediments. The paleoclimate condition of the Chang 9 member was mostly semihumid, which resulted in moderate chemical weathering and moderate to high terrestrial clastic influx to the lacustrine basin. In addition, the sediment provenance analysis indicates that the provenance of the Chang 9 member is mainly derived from felsic rocks. The felsic rocks can provide less essential nutrimental elements for algae growth than the mafic rocks. But high clastic input is beneficial for higher plants input and moderate clastic influx enhance OM preservation. In general, intense tectonic activity, volcanism, felsic source rocks, semihumid paleoclimate, and moderate to high terrestrial clastic influx jointly control the OM accumulation for the Chang 9 sediments.

5. Conclusion

In this study, a series of geochemical analyses was carried out on the Chang 9 organic-rich sediments and utilized to evaluate the hydrocarbon generation potential, reconstruct paleoclimate condition, analyze the sediment provenance, and illustrate their relationship with OM accumulation. The black shales in the Chang 9 member are characterized by high OM abundance, type I–II kerogen, and appropriate maturity, indicating good hydrocarbon generation potential. Three intervals of the Chang 9 member exhibit obvious OM heterogeneity, and the Unit II interval contains the highest OM abundance. The C-value and Sr/Cu and Rb/Sr ratios suggest that the Chang 9 member was mostly deposited under a semihumid condition. The chemical weathering was moderate based on CIA, CIW, and PIA values. Additionally, the sediment provenance analysis suggests that the provenance of the Chang 9 member mainly is felsic rock, which can provide less essential elements for algae growth. The semihumid climate not only promotes the boom of the primary producer but also leads to moderate to high terrestrial detrital clastic input, accelerating terrigenous OM accumulation.

Data Availability

The (data type) data used to support the findings of this study are included within the article.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

This work was supported by the project from PetroChina Changqing Oilfield Company (No. ZLZX2020-02-01-01).

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