

SEDIMENTARY AND GEOCHEMICAL CHARACTERISTICS OF OIL SHALE IN THE PERMIAN LUCAOGOU FORMATION IN THE SOUTHEASTERN JUNGGAR BASIN, NORTHWEST CHINA: IMPLICATIONS FOR SEDIMENTARY ENVIRONMENTS

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Abstract. *In this study, the sedimentary and geochemical characteristics of oil shale in the Lower and the Upper Member of the Permian Lucaogou Formation in the southeastern Junggar Basin, Northwest China, were investigated. Mineral and bulk chemical compositions, sedimentary facies and distribution of oil shale show that in the Lower Member it is rich in organic matter, but in the Upper Member, organic-poor. The distribution area of deep lake subfacies and oil shale in the Lower Member is larger than that in the Upper Member. The sedimentary modes of these two members were estimated using geochemical parameters, such as Sr/Cu, the chemical index of alteration (CIA), Ni/Co, V/Sc and $(C_{27} + C_{28})/C_{29}$ regular steranes. The organic-rich oil shale in the Lower Member of the Lucaogou Formation formed in the more humid climate and under oxidative conditions, its organic matter derived mainly from algae. On the other hand, the organic-poor oil shale in the formation's Upper Member formed in the humid climate and under more oxidative conditions, while the input of land plants to its organic matter was high and that of algae low.*

Keywords: *sedimentary characteristics, geochemical characteristics, Permian oil shale, Lucaogou Formation, Junggar Basin.*

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1. Introduction

Oil shale of the Middle Permian Lucaogou Formation is found in the south-eastern part of the Junggar Basin, Northwest China [1, 2]. Earlier studies suggest that lake facies are the main sedimentary facies of the Lucaogou Formation. Previously the formation model of the Lucaogou Formation oil shale has been reconstructed [3–5]. The formation's oil shale measures can be divided into two: the Lower Member and the Upper Member [5]. However, the sedimentary environments of these two members have not been systematically described yet. This study aims to analyze the sedimentary (mineral and bulk chemical compositions, sedimentary facies) and geochemical characteristics of oil shale, revealing the paleoclimate, redox conditions and organic matter input to the oil shales of the Lower and the Upper Member of the Lucaogou Formation. This study will provide a basis for the further exploration and exploitation of oil shale resources in the southeast of the Junggar Basin.

2. Geological setting

The Junggar Basin is one of the largest sedimentary basins in Northwest China with an area of about 137,000 km². Two other basins, Santanghu and Turpan-Hami, are situated next to the Junggar Basin at its eastern and southern margins, respectively. The study area is located in the area between the Tianshan Mountains and the Junggar Basin (Fig. 1).

Because of the tectonic activity during the Mesozoic and the Cenozoic, a series of multiple anticlines and multistage thrust faults were developed under compressive stresses. The basement of the Junggar Basin was formed due to accretion of oceanic rocks and island arcs [1, 6]. Carboniferous, Permian, Triassic, Jurassic, Neogene and Quaternary strata are widely exposed on the surface. The Permian sequence is mainly exposed in the western area, but some outcrops can be found in the eastern area too (Fig. 1).

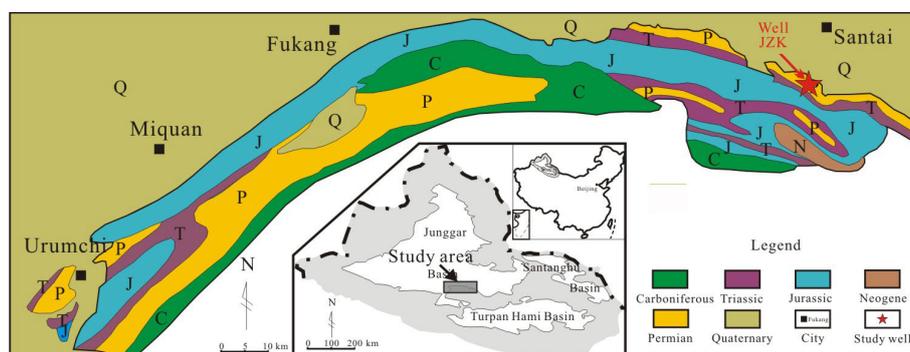


Fig. 1. Stratum distribution map of the southeast of the Junggar Basin (modified from [7]).

The southeastern part of the Junggar Basin was a big sedimentary depression during the time when the sediments of the Lucaogou Formation were deposited [6]. The reported outcrops revealing the Lucaogou Formation include the Yaomoshan, Tianchi, Sangonghe, Dahuangshan and Shichanggou outcrops [1, 5, 6]. The Lucaogou Formation is represented by oil shale, mudstone, calcareous mudstone, dolostone, calcareous siltstone, siltstone, fine-grained sandstone and medium-grained sandstone. Based on the continental sequence stratigraphy division principles, the Lucaogou Formation is divided into two complete third-order cycles corresponding to the Lower Member and the Upper Member, respectively [5–8] (Fig. 2a).

3. Material and tests

Previous studies often used oil shale samples from outcrops to study the sedimentary environments [3–6, 8]. However, the samples from outcrops are severely weathered, making the analytical results unreliable. Supported by the Oil and Gas Survey Center, China Geological Survey, twenty oil shale samples with no signs of weathering damages were taken from the oil shale exploration well JZK drilled in the eastern part of the study area (Fig. 1). The collected samples were mostly gray or black oil shale.

All the samples were studied by total organic carbon content (TOC, wt%) analysis, solvent extraction, fractionation, and gas chromatography-mass spectrometry (GC-MS). TOC analysis was conducted using the LECO CS-400 instrument [9]. The thin rock sections were examined under a polarizing microscope. All the samples were crushed, solvent extracted, fractionated and analyzed by GC-MS at the Organic Geochemistry Laboratory of the Department of Earth and Planetary Sciences, Macquarie University, Australia. In addition, the mineral and chemical compositions of representative samples were analyzed. X-ray diffractometry analysis (XRD) was performed using an XRDA Philips PW1830 diffractometer system with $\text{Cu-K}\alpha$ radiation on pulverized samples (≤ 200 mesh). Elemental composition was measured at the Beijing Research Institute of Uranium Geology, China. Trace element contents (Sr, Cu, V, Sc) were determined by a high resolution inductively coupled plasma mass spectrometer (HR-ICP-MS). Major element concentrations (Al_2O_3 , Na_2O , K_2O , CaO) were determined using a Thermo Scientific X-series inductively coupled plasma mass spectrometer (ICP-MS), following the Chinese National Standard method DZ/T 0223-2001 [10].

4. Sedimentary characteristics

4.1. Mineral and bulk chemical composition characteristics

The bulk chemical compositions of oil shales in the Lower and the Upper Member of the Lucaogou Formation differ. The oil shale in the Lower

Member is rich in organic matter (TOC averages 12.4%). The amount of terrigenous clastics is relatively low (Fig. 3l). In contrast, the oil shale in the Upper Member is poor in organic matter (TOC averages 3.2%) (Fig. 3n). Microscopically, terrigenous clastics are abundant in it. As shown by the plot of HI vs T_{max} (Fig. 2d), most data points from the Lower Member oil shale fall within the Type I kerogen area, suggesting that the organic matter of the Lower Member oil shale derived mainly from aquatic organisms. Some oil shale samples from the Upper Member fall within the Type II kerogen area, and others, within the Type I kerogen area. This suggests that the organic matter of the Upper Member oil shale derived mostly from land plants.

The mineral compositions of oil shales of the Lower and the Upper Member of the Lucaogou Formation also differ. The mineral composition of the Lower Member oil shale is represented by quartz (26.7–36.3%, avg 31.78%), feldspars (38.8–58.2%, avg 46.53%) and dolostone (8.4–34.5%, avg 21.73%) (Fig. 2b). Clay minerals, calcite and pyrite were not detected in this oil shale. On the other hand, the mineral composition of the Upper Member oil shale is represented by quartz (26.9–38.5%, avg 34.13%), feldspars (6.5–27.5%, avg 15.93%), clay minerals (15.3–47.2%, avg 32.48%), calcite (6.8–19.7%, avg 11.53%), dolostone (0–19.5%, avg 4.88%) and pyrite (0–4.3%, avg 1.08%) (Fig. 2c). The differences in mineral

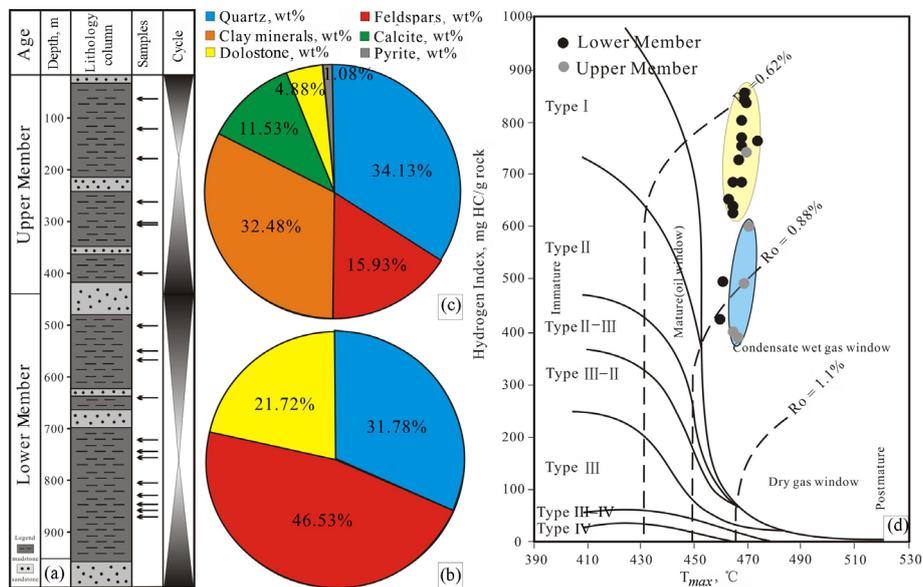


Fig. 2. Lithology column, sampling locations, mineral composition and kerogen types characteristics of oil shale from well JZK: (a) lithology column and sampling locations of well JZK; (b) mineral composition of the Lower Member oil shale; (c) mineral composition of the Upper Member organic-poor oil shale; (d) plot of HI vs T_{max} outlining kerogen types of oil shale of the Lucaogou Formation (HI and T_{max} data in (d) come from [11]).

composition between the oil shales of the Lower and the Upper Member may result from different sedimentary environments.

4.2. Sedimentary facies characteristics

Sedimentary facies of the study area were analyzed based on data obtained through drilling dozens of wells. The main sedimentary facies of the Lucaogou Formation in the study area are delta facies, shallow lake subfacies and deep lake subfacies. The delta facies include underwater distributary channel (UDC) and underwater distributary interchannel (UDIC) micro-facies (Fig. 3a). The typical sedimentary structures of UDC are scour surface (Figs. 3c–3d), liquefaction sandstone vein, massive bedding and teared mudstone (Fig. 3b). The main rock types in UDC are medium-grained sandstone and fine-grained sandstone (Fig. 3a). In UDIC, the predominant rock types are siltstone and mudstone (Fig. 3a). Horizontal bedding is a common sedimentary structure in UDIC (Fig. 3e). Shallow lake subfacies include muddy lake and sandy lake microfacies (Fig. 3f). The principal rock types of muddy lake microfacies are mudstone and silty mudstone (Fig. 3g). Horizontal bedding is often observed in this kind of microfacies (Fig. 3g). The prevailing rock types in sandy lake microfacies are medium-grained sandstone, fine-grained sandstone and siltstone (Fig. 3f). The common sedimentary structures of sandy lake microfacies are lenticular bedding, parallel bedding (Figs. 3h–3i), liquefaction sandstone vein and massive bedding (Fig. 3j). Deep lake subfacies are the chief sedimentary subfacies in the study area. The main rock types are black mudstone, oil shale and silty mudstone. Horizontal bedding (Fig. 3o) and massive bedding (Fig. 3m) are the common sedimentary structures of this kind of microfacies.

Sedimentary facies distribution characteristics of the Lower Member are shown in Figure 4a. Delta plain subfacies were not found in the study area and the exact distribution of delta plain subfacies remains yet unclear. However, the distribution of the other sedimentary subfacies is well defined. Delta front and delta plain subfacies were distributed in the southwest area, while the area of distribution of delta front subfacies in the eastern Jimusaer region was smaller. Shallow lake subfacies were distributed as a strip in the study area. Deep lake subfacies were distributed along the NW-SE direction and cover the largest area in the studied basin. However, the distribution of the sedimentary facies of the Upper Member is a little different from that of the Lower Member (Fig. 4b). In the Upper Member, delta front and delta plain subfacies were distributed in the southwest area. Compared with the Lower Member, the distribution area of delta front and shallow lake subfacies next to the Jimusaer region in the Upper Member is relatively large, while that of deep lake subfacies in the study area is relatively small. Generally, oil shale was mainly formed in deep lake subfacies [3, 4]. The distribution area of the sedimentary facies fit for the formation of oil shale in the Lower Member is larger than that in the Upper Member.

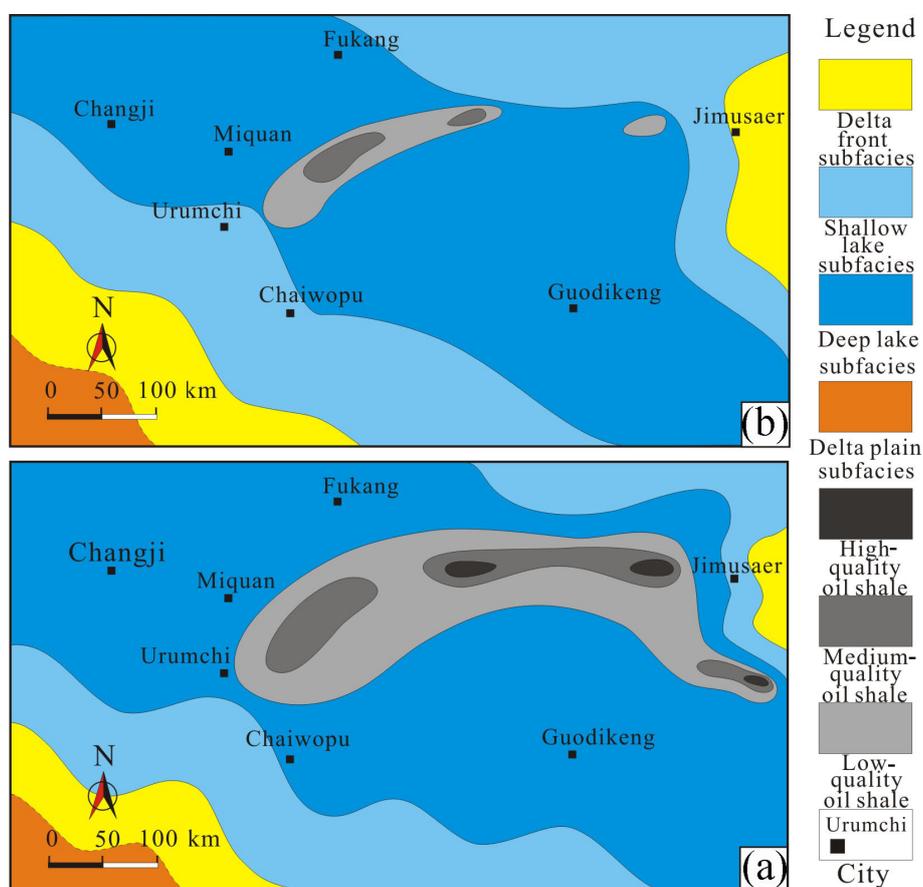


Fig. 4. Distribution features of sedimentary facies and oil shale of the Lucaogou Formation in the southeast of the Junggar Basin: (a) distribution features of sedimentary facies and oil shale of the Lower Member in the Lucaogou Formation; (b) distribution features of sedimentary facies and oil shale of the Upper Member in the Lucaogou Formation.

4.3. Oil shale distribution characteristics

Because of the tectonic uplift, the oil shale sequence in the southeastern Junggar Basin was severely eroded, and therefore oil yield data about oil shale in Chaiwopu and Guodikeng areas could not be obtained for investigation. However, the existing oil yield data can still reveal the distribution features of oil shale in the southeast part of the Junggar Basin. The formation of oil shale of medium quality ($5\% < \text{oil yield} < 10\%$) and poor quality (oil yield $< 5\%$) took place mostly in the Lower Member, while high-quality oil shale (oil yield $> 10\%$) was developed at the time of deposition of the Lower Member too (Fig. 4a). However, the distribution area of oil shale in the Upper Member obviously decreased (Fig. 4b). Medium- and poor-quality oil shale was mainly distributed in the western areas of the basin, while some

amount of poor-quality oil shale was formed in its eastern areas as well. No formation of high-quality oil shale occurred during that period. The above suggests that the distribution area of oil shale in the Lower Member is larger than that in the Upper Member.

5. Geochemical characteristics

Sedimentary environmental parameters, such as paleoclimate, redox conditions and organic matter input, can control the formation of oil shale [12, 13]. Geochemical indices are used to characterize ancient sedimentary environments [3, 4, 14]. Different geochemical indicators were used in this study to reconstruct the sedimentary environments of the Lower and the Upper Member of the Lucaogou Formation in the study area.

5.1. Paleoclimate

Paleoclimate is considered an important controlling factor for oil shale deposition [15, 16]. Sr/Cu ratio is a significant indicator of paleoclimate. Generally, a low Sr/Cu ratio (1.3–5.0) reflects a warm humid climate, a high Sr/Cu ratio (> 20.0) suggests a hot arid climate [17, 18]. The Sr/Cu ratio in the Lower Member oil shale (1.35–6.00, avg 3.86) is lower than that in the Upper Member oil shale (4.30–12.86, avg 9.02), indicating that the climate during the deposition of the Lower Member oil shale was relatively humid, while during the deposition of the Upper Member oil shale it became somewhat drier, yet not completely dry. The chemical index of alteration (CIA) = $100 \times \text{Al}_2\text{O}_3 / (\text{Al}_2\text{O}_3 + \text{Na}_2\text{O} + \text{K}_2\text{O} + \text{CaO}^*)$ is regarded as an important indicator of paleoclimate too. Generally, a high CIA value reflects a relatively humid climate, while a low CIA value is indicative of a relatively dry climate [19–21]. Some studies have even suggested that the CIA value ranging from 50 to 65 indicates a dry climate during low chemical weathering, the CIA value varying from 65 to 85 implies a warm and humid climate during moderate chemical weathering, while the CIA value between 85 and 100 mirrors a hot and humid climate during strong chemical weathering [19–22]. The CIA in the Lower Member is in the range of 79–86 (avg 82), while in the Upper Member it varies between 64 and 78 (avg 70) (Fig. 5a). The above CIA values give evidence of that the oil shale of both the members was deposited in a warm and humid climate, however, the climate during the deposition of the Lower Member was more humid.

5.2. Redox conditions

Redox conditions are an important influencing factor for the preservation and accumulation of organic matter and may control its degree of oxidation [23, 24]. On the other hand, pristane/phytane (Pr/Ph) ratio has been shown to be a significant indicator of redox conditions [25, 26]. However, this

parameter can be used only for the samples of rock that has not undergone biodegradation. According to Li [6], the Lucaogou Formation oil shale has suffered from biodegradation, therefore in the current work, the Pr/Ph ratio was neglected. Nickel (Ni) and cobalt (Co) are important indicators of redox conditions as well. Generally, Ni is more abundant in anoxic conditions, while Co is usually present in abundance in oxidative conditions. The Ni/Co values < 5.0 in sediments indicate oxic conditions, whereas Ni/Co > 7.0 reflects deposition in an anoxic environment. The Ni/Co values between 5.0 and 7.0 are indicative of deposition in the dysoxic environment [22, 27–29]. The low Ni/Co ratio (3.43–5.18, avg 4.12) argues for oxidative conditions in the Lower Member. The lower ratio of Ni/Co (1.87–3.26, avg 2.46) in the Upper Member oil shale also reflects the oxidative conditions (Fig. 5b), suggesting that its depositional environment was more oxidative than that in the Lower Member. In addition, vanadium (V) and scandium (Sc) are important indicators of redox conditions too. Generally, V is more abundant in anoxic conditions, while Sc is usually abundant in oxidative conditions. The V/Sc values < 9.1 correspond to normal environments (oxidative conditions), whereas V/Sc ≥ 9.1 corresponds to low-oxygen depositional environments (low-oxygen conditions) [29, 30]. The high V/Sc ratio (7.66–11.03, avg 8.97) argues for low-oxygen or oxidative environments during the deposition of the Lower Member, yet not becoming anoxic conditions. On the other hand, the low V/Sc ratio (6.13–7.01, avg 6.59) also reflects the oxidative conditions in the Upper Member. The above suggests that oil shales of the Lower and the Upper Member of the Lucaogou Formation were deposited in the oxidative conditions. However, the depositional conditions of the Upper Member oil shale were more oxidative than those of the Lower Member oil shale.

5.3. Organic matter input

The organic matter of oil shale derived mainly from terrigenous plants, lake algae, phytoplankton and zooplankton [2, 31]. Typically, regular steranes are derived from sterols, known as constituents of the cell membranes in all eukaryotes. Algae are believed to be the predominant primary producers of C₂₇ sterols, while C₂₉ sterols are more typically associated with land plants [32–35]. However, C₂₈ steranes are suggested to be derived from phytoplankton and zooplankton. The high proportion of C₂₇ and C₂₈ steranes indicates the contribution of lake organisms, while the abundance of C₂₉ steranes gives evidence of the contribution of land plants [36]. So, based on the amounts of C₂₇, C₂₈ and C₂₉ regular steranes, the input of organic matter to sediments can be determined. From the ternary diagram outlining the relative proportions of C₂₇, C₂₈ and C₂₉ steranes, the Lower Member oil shale derived predominantly from algae. The contribution of land plants to the Upper Member oil shale is relatively high (Fig. 5d). The total proportion of

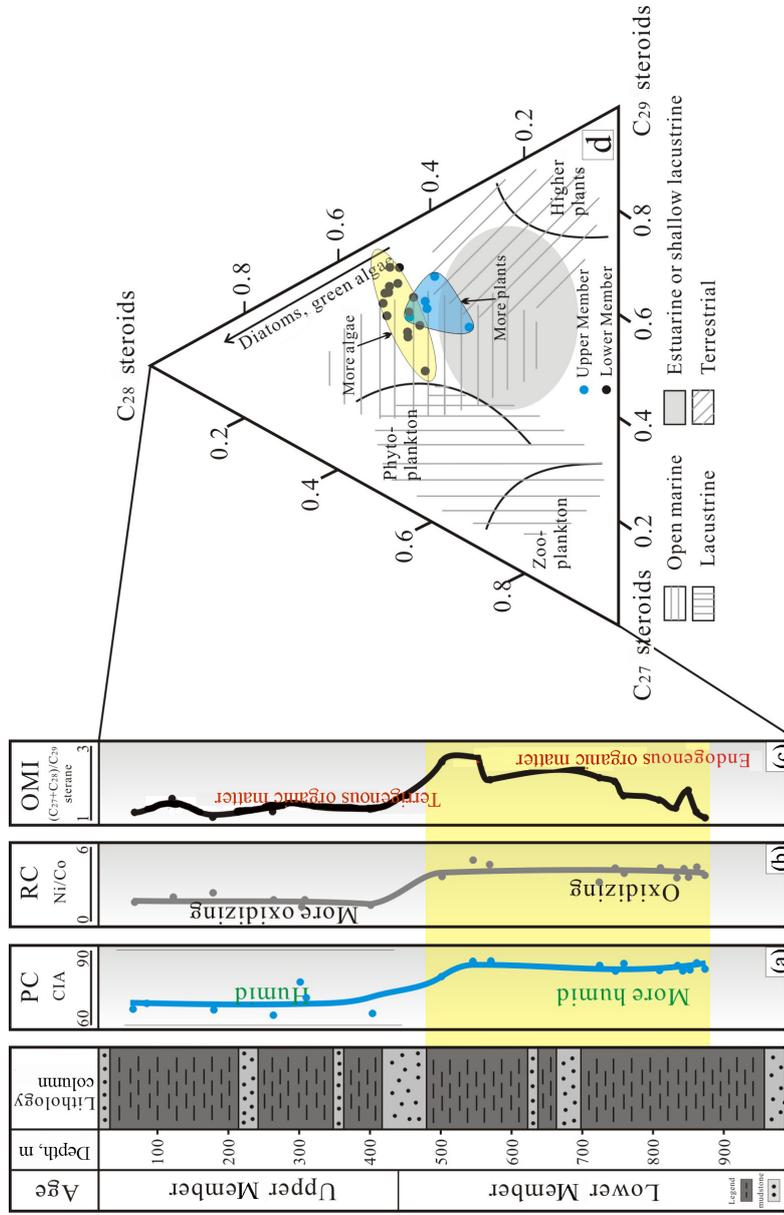


Fig. 5. (a)–(c) Vertical evolution features of paleoclimate, redox conditions and organic matter input of oil shale in well JZK; (d) ternary plot of relative proportions of C₂₇, C₂₈ and C₂₉ steranes for the Lucaogou Formation (paleoenvironmental and source interpretation according to [36]). (The abbreviations used: PC – paleoclimate, RC – redox conditions, OMI – organic matter input.)

C_{27} and C_{28} steranes in the Lower Member oil shale samples (0.54–0.72, avg 0.63) is higher than in the Upper Member ones (0.58–0.64, avg 0.47). At the same time, the relative proportion of C_{29} steranes in the Upper Member oil shale samples (0.36–0.46, avg 0.41) is lower than in those of the Lower Member (0.28–0.46, avg 0.36). The Lower Member oil shale samples are characterized by higher $(C_{27} + C_{28})/C_{29}$ steranes ratios (1.18–2.63, avg 1.82) as compared to the Upper Member oil shale samples (1.16–1.77, avg 1.42) (Fig. 5c). These results suggest that the organic matter of the Lower Member oil shale derived predominantly from algae. However, the proportion of land plants in the Upper Member oil shale samples is higher than in the Lower Member oil shale samples.

6. Sedimentary environment modes

Investigators have proposed different sedimentary environment modes of shale deposition [37–40]. Based on the analysis of sedimentary and geochemical characteristics, the sedimentary environment modes of oil shales of the Lower and the Upper Member of the Lucaogou Formation were found which can shed light on the differences between these two oil shales.

During the deposition of the Lower Member of the Lucaogou Formation, the distribution area of deep lake sufacies was large (Fig. 4a). The climate was more humid, while the lake water column represented an oxidative environment. In such an environment, the terrigenous organic matter input was low. In addition, in the lake there deposited organic matter of mostly algal origin, which led to the formation of organic-rich oil shale (Fig. 6a).

On the contrary, the distribution area of deep lake subfacies at the time of deposition of the Upper Member was smaller than that during the accumulation of the Lower Member (Fig. 4b). The climate was drier, but still humid. The lake water column was more oxidative. In such an environment, the terrigenous organic matter input was relatively high. Moreover, lesser algal organic matter was deposited in the oxidative water conditions, as a result, organic-poor oil shale was formed (Fig. 6b).

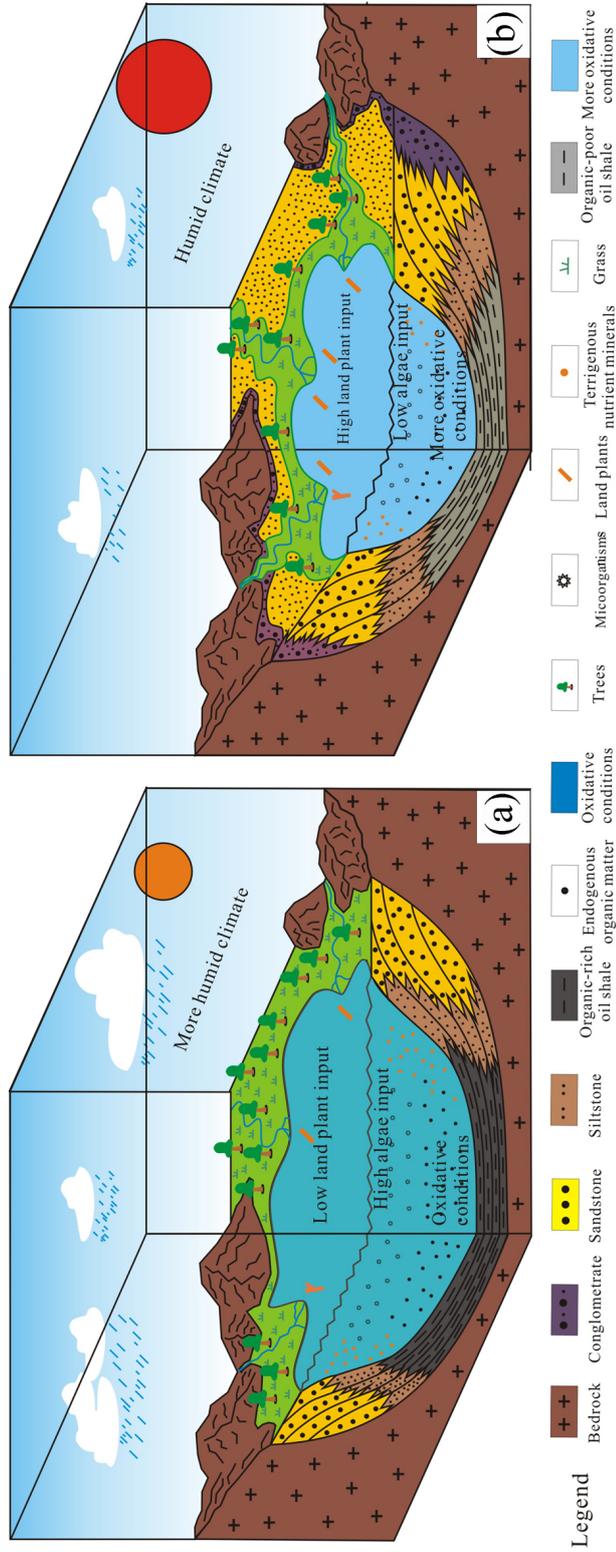


Fig. 6. (a) Ancient sedimentary environment mode of the organic-rich oil shale in the Lower Member of the Lucaogou Formation; (b) ancient sedimentary environment mode of the organic-poor oil shale in the Upper Member of the Lucaogou Formation.

7. Conclusions

1. The characteristics of oil shales of the Lower and the Upper Member of the Lucaogou Formation are different. Total organic carbon content shows that the Lower Member oil shale is richer in organic matter. Minerals of the Lower Member oil shale include quartz, feldspars and dolostone, while those of the Upper Member oil shale are represented by quartz, feldspars, clay minerals, calcite, dolostone and pyrite.
2. The main sedimentary facies of the Lucaogou Formation are delta facies and lake facies. The sedimentary facies distribution characteristics of the Lower and the Upper Member of the Lucaogou Formation are a little different, the distribution area of deep lake subfacies and oil shale of the Lower Member is larger than that of the Upper Member.
3. Geochemical indicators such as Sr/Cu, chemical index of alteration, Ni/Co and V/Sc show that the paleoclimate at the time of deposition of the Lower Member was more humid than that during the accumulation of the Upper Member. However, the sedimentary environment of the Upper Member was more oxidative than that of the Lower Member.
4. Based on $(C_{27} + C_{28})/C_{29}$ regular steranes values, the organic matter of the Lower Member oil shale is mainly of algal origin, while the organic matter of the Upper Member oil shale is derived mostly from land plants.
5. The constructed sedimentary environment models indicate that the organic-rich oil shale of the Lower Member was formed in the more humid climate and under oxidative conditions. In contrast, the organic-poor oil shale of the Upper Member was formed in the humid climate and under more oxidative conditions.

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