



Depositional Setting and Enrichment Mechanism of Organic Matter of Lower Cretaceous Shale in Ri-Qing-Wei Basin in the Central Sulu Orogenic Belt

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A suite of source rock consists of mudstone and shale, with great thickness and continuous deposition was found in the well LK-1 in Lingshan island in Ri-Qing-Wei basin. In order to evaluate the hydrocarbon generation prospects of these source rock and find the mechanism of organic matter enrichment, shale samples were selected from the core for TOC (total organic carbon) and element geochemistry analysis. The results show that organic matter abundance of the source rocks are generally high with average TOC content of 1.26 wt%, suggesting they are good source rocks. The geochemical features show that the sedimentary environment is mostly anoxic brackish water to salt water environment with arid to semiarid climate condition. The enrichment mechanism of organic matter varied with the evolution of the basin, which was divided into three stages according to the sedimentary characteristics. In the initial-middle period of rifting evolution (stage 1 and early stage 2), paleoproductivity is the major factor of OM-enrichment reflecting by high positive correlation between the TOC contents and paleoproductivity proxies. While with the evolution of the rift basin, redox condition and terrigenous clastic input became more and more important until they became the major factor of OM enrichment in the middle stage of rift evolution (stage 2). In the later stage of rift evolution (latest stage 2 and stage 3), besides terrigenous clastic input, the effect of paleoclimate on OM-enrichment increased gradually from a minor factor to a major factor.

Keywords: Lingshan Island, organic matter enrichment, shale, Cretaceous, depositional setting

INTRODUCTION

As the most important type of source rock, shale is widely distributed in almost every depositional setting in different tectonic settings (Verma and Armstrong-Altrin, 2013). Whatever depositional setting the shale belongs to, the Total Organic Carbon (TOC) content is the primary indicator to judge the oil and gas potential. Almost each big oil and gas field all over the world owes a suite of shale or mudstone as source rock, with huge thickness and high TOC content. Previous researches (Yan et al., 2018; Gallego-Torres et al., 2007; Mort et al., 2007) indicate that restricted basin sedimentary conditions are good for organic matter (OM) enrichment and there are two typical models for OM enrichment: the paleoproductivity model and the preservation model. The paleoproductivity model

emphasizes that large amount of OM enriched for the higher productivity in the sedimentary water. Large amount of OM consume oxygen in the water, leading to an anoxic environment which is good for preservation of OM (Sageman et al., 2003; 8; Tang et al., 2020). While the preservation model stresses the importance of the sea level fluctuation and the restricted effect of basin, proving an anoxic environment for OM accumulation (Arthur et al., 1998; Mort et al., 2007; Tang et al., 2020). In addition, the terrigenous clastic input is another important factor controlling the OM enrichment in recent study (Tang et al., 2020). Changes in sedimentary environment would affect the OM enrichment. The geochemistry of shale could reflect the sedimentary environment such as Ba, P, Cu for paleoproductivity (Paytan and Griffith, 2007; Algeo et al., 2011), Mo, U, V for redox condition and Al, Ti, Zr for terrigenous clastic input (Tribouillard et al., 2006).

There were a lot of Meso-Cenozoic basins developed in eastern China as the respond to the subduction of western Pacific plate. With the development of petroleum exploration, more and more petroliferous basins were found such as Bohai Bay basin, Songliao basin (Wang et al., 2020). However, the oil and gas potential of Jiaolai basin which is the best record with relatively complete Cretaceous strata, is not ideal. Recently, the Lingshan island scientific drilling (Well LK-1) uncovered there was a rifting basin in the central Sulu orogenic belt, beside the Jiaolai basin. Although a lot of researchers have studied Lingshan island in different aspects, the oil and gas potential is still unclear. This study focuses on the core sample of LK-1, using TOC content and element contents of shale samples to reveal the mechanism of organic matter enrichment.

GEOLOGICAL SETTING

Ri-Qing-Wei basin is located in the central Sulu orogenic belt, eastern coastal Shandong, China. It is a long and narrow late Mesozoic rift basin and is distributed along the NE-SW direction. The northwest boundary is Wulian-Qingdao faults and Jimo-Muping faults, while the southeast boundary is Qianliyan faults. Lingshan island is located in the south Ri-Qing-Wei basin (Figure 1). The strata in Lingshan island represents the stratigraphic sequence of Ri-Qing-Wei basin. The stratigraphic sequence is divided into two groups: 1) Laiyang group, the older one which is consisted by a series of gravity flows' deposition (Lü et al., 2011; Zhong et al., 2012; Yang and van Loon, 2016; Yang et al., 2017; Liang et al., 2018; Yang et al., 2018), mainly developed in the west island, including several outcrops (Figure 2A) such as Beilishi, Qiancengya, Chuanchang, Dengta et al., and 2) Qingshan group, the younger one which is mainly dominated by volcanic deposition and a suite of terrestrial clastic deposits (Wang et al., 2015; Zhou et al., 2017; Zhou et al., 2018).

In order to uncover the main layer sequence of the Laiyang group, we set up the Lingshan Island Scientific Drill (Well LK-1). The depth of LK-1 is 1,350 m, and the total thickness of layers of shale and mudstone is about 550 m. This discovery further confirms the existence of Ri-Qing-Wei basin. In the layers of Laiyang group which are presented by the core of LK-1

(Figure 2B), various kinds of gravity flows and large amounts of soft sediments deformation structures are found. Also, a lot of markers representing deep-water sedimentary environment are observed, such as iron nodule, vitrain and plant debris. According to the lithology and sedimentary characteristics of LK-1, Yuan et al. (2019) show that the sedimentary strata of Laiyang group developed fan delta, subaqueous fan, turbidite fan and delta from the bottom to the top. In addition, the evolution of Ri-Qing-Wei basin could be divided into four stages (Yuan et al., 2019): initial stage of the basin rifting, the stage of basin expansion and subsidence, the stable stage of deep subsidence and the stage of basin uplift.

MATERIALS AND METHODS

Eighty-two samples of shale of LK-1 were selected to do TOC (total organic carbon) analysis. Thirty samples of the batch above and other 22 samples were selected to do major and trace element analysis.

TOC (total organic carbon) content analyses were done in Northeast Petroleum University. The TOC contents were measured with acid-dissolution method. Diluted hydrochloric acid were used to removing the inorganic carbon compound of the samples' powder. After, samples were burned in the high temperature oxygen. Carbon content of CO₂ produced during the burning process are all from the organic carbon of samples. The contents of CO₂ were measured by thermal conductivity detector, to get the TOC contents. The detailed analytical method is described by Liu et al. (2019).

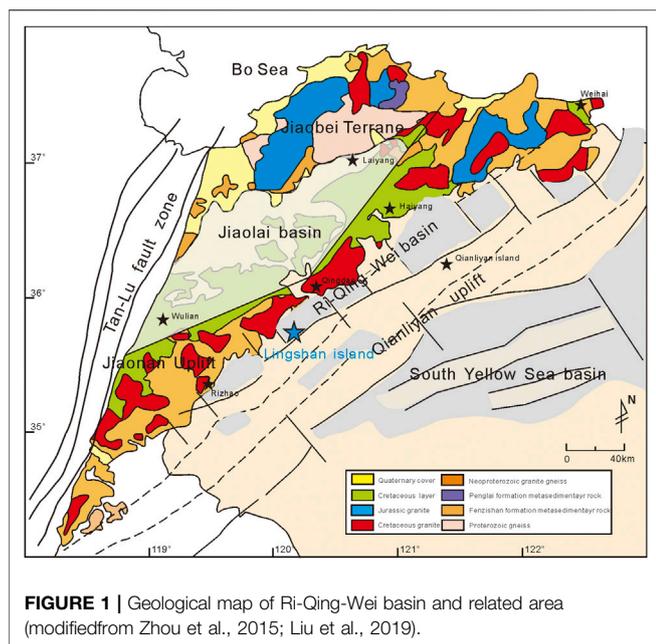
Chemical analyses were conducted in the laboratory of Eighth Institute of Geology and Mineral Exploration of Shandong Province, China. Fresh rock pieces without cracks were crushed to 200 meshes before experiment. Major oxides were analyzed by X-ray fluorescence spectrometry (XRF; equipment model is Axios) with the relative standard derivation (RSD) of major oxides less than 5%. Trace elements were analyzed by ICP-MS (equipment model is ICAP Qc), and detailed procedures for trace elements analysis are as described by Liang et al. (2000). The relative standard deviations (RSD) of trace elements are within 5%. The procedures for major and trace element analyses are described by Kimura (1998) and Liang et al. (2000), respectively.

RESULTS

The TOC, major element, trace element and rare element contents of LK-1 shales analyzed in this study are listed in the **Supplementary Table S1**.

TOC Contents

The TOC contents of collected samples of Laiyang group from LK-1 are generally high, with a mean TOC content at about 1.26 wt%. More than 70% of samples' TOC content are greater than 1 wt%. The TOC contents are different within different depth of LK-1. In the depth ranges below, TOC content of most samples are greater than 1 wt%: 145–246 m (average TOC,



1.39 wt%), 392–453 m (1.74 wt%), 589–665 m (1.35 wt%), 696–776 m (1.46 wt%), 833–919 m (1.85 wt%), 1,031–1,136 m (1.40 wt%), 1,175–1,301 m (1.45 wt%).

Major Elements

The element composition of the shales of Laiyang group are dominated by SiO_2 (51.90–75.12 wt%, average 61.09 wt%), Al_2O_3 (8.68–18.58 wt%, average 14.86 wt%) and CaO (1.21–17.56 wt%, average 7.12 wt%). Besides, these shales also contain relative low concentrations of Fe_2O_3 (average 5.22 wt%), K_2O (average 3.59 wt%), MgO (average 3.42 wt%), Na_2O (average 3.22 wt%). Other major element contents (MnO , P_2O_5 , TiO_2) are all lower than 1 wt%.

The contents of SiO_2 , Al_2O_3 , Fe_2O_3 , K_2O and MnO of our samples are close to PAAS (Taylor and Mclennan, 1985), while the contents of CaO and P_2O_5 show much higher than the corresponding contents in PASS (Figure 3). The higher concentration of CaO may be related to the calcite veins which widely spread in the core. The higher content of P_2O_5 may reflect the higher paleoproductivity in the sedimentary water in Laiyang period, because P is an important nutrient element to life.

A negative correlation can be seen between SiO_2 and Al_2O_3 contents, and between Al_2O_3 and CaO contents (Figure 4). No obvious correlation could be observed between Al_2O_3 and P_2O_5 . These indicate Si, Ca, P of samples are mainly effected by sedimentary environment, while the effect of terrigenous clastic input is relative slight. However, samples show a positive correlation between Al_2O_3 and TiO_2 contents. This indicates Ti in samples were mainly controlled by terrigenous clastic input.

Trace Elements

Different trace element owes different sensitivities to specific geological event, so sedimentary environment could be

reconstructed by suitable trace elements. Selecting authigenic element composition could help reflecting the sedimentary water conditions better and also its variation during the geological period.

Element enrichment factors were used to see the enrichment or depletion of an element through comparing with selected standard value. In this study, the trace elemental compositions of the samples were compared with the composition of PAAS (Taylor and Mclennan, 1985) through Al-normalization to identify significant deviations. Element enrichment factors of X (X_{EF}) were calculated by $X_{EF} = (X/Al)_{\text{Sample}} / (X/Al)_{\text{PAAS}}$. $X_{EF} > 1$ represents the element X was relative enriched in Sample compared to X in PAAS, while $X_{EF} < 1$ represents the situation of depletion. The X_{EF} of Co, Pb, Ba, Sr, Hf, U are obviously larger than 1 showing these trace elements are enriched in samples than PAAS. While the X_{EF} of V, Cr, Ni, Sc, Cu, Cs are much lower than 1, indicating these elements are depleted. As to the elements Li, Rb, Th, Nb and Zr, behaved similar to PAAS for their X_{EF} values are close to 1 (Figure 3).

Rare Earth Elements

Both sedimentary water condition and terrigenous clastic input affect the rare earth elements in shale. Because of the stability of rare earth elements after they were absorbed from the sedimentary water and preserved in sedimentary rock, they are often used to reflect the paleo-environment conditions (Sholkovitz et al., 1994; Xia et al., 2015). The $\sum\text{REE}$ of shales of Laiyang group from LK-1 range from 115.69 to 334.03×10^{-6} , with an average value of 225.19×10^{-6} , larger than $\sum\text{REE}$ contents in UCC (148.14×10^{-6}) and PAAS (184.77×10^{-6}). The ratios of $\sum\text{LREE} / \sum\text{HREE}$ of our samples are 3.48–6.25 with a mean value of 5.10. These show shales of Laiyang group from LK-1 are enriched in LREE, relative depleted in REE.

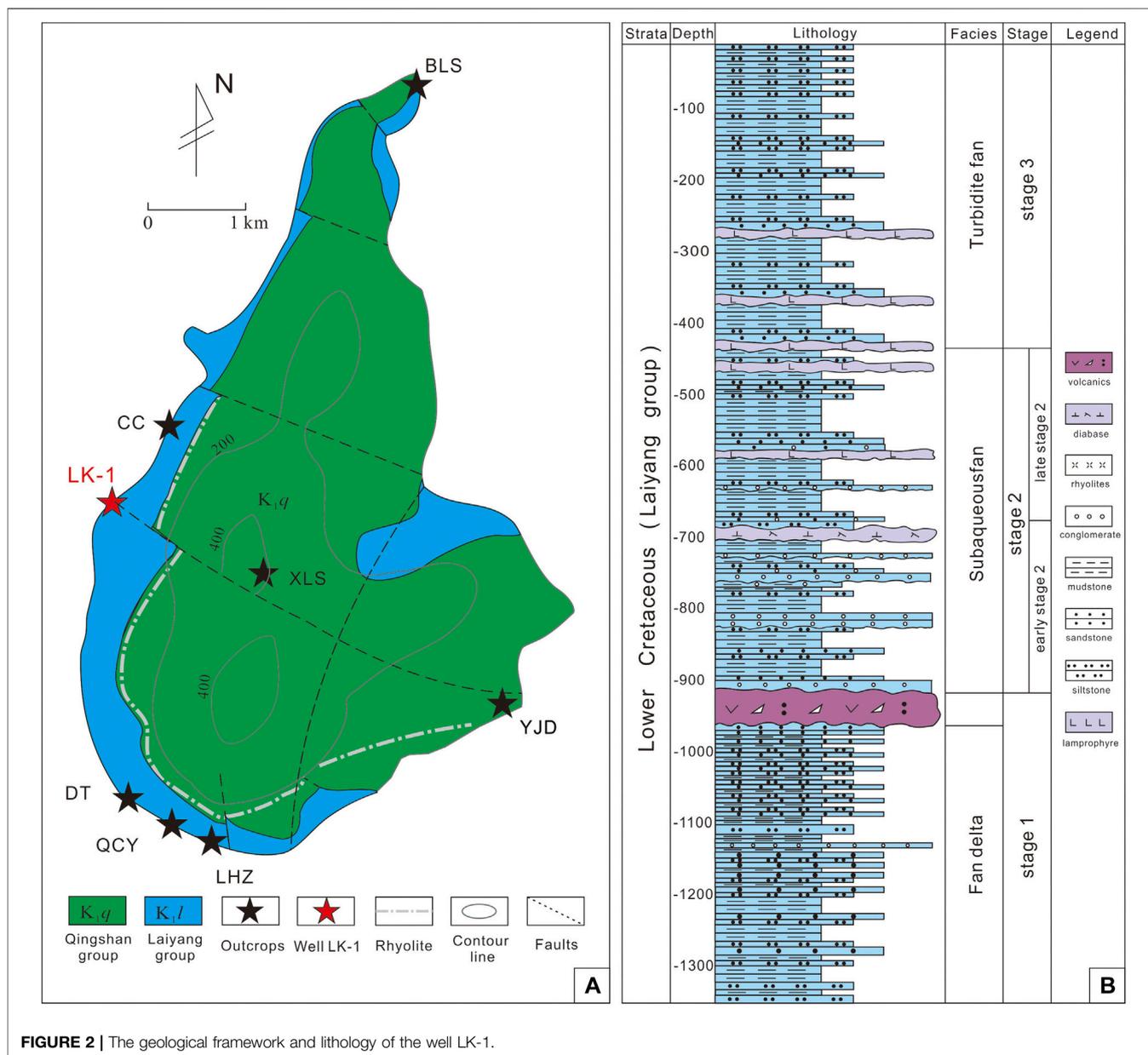
The trends of variation of LREE and HREE contents match each other in vertical direction of well LK-1 (Supplementary Table S1). At the beginning, from the depth 1,312–1,032 m, the ratio of $\sum\text{LREE} / \sum\text{HREE}$ varies from 4.72 to 5.69, does not change much. The ratio show much variation (3.48–6.25) from the depth 887–412 m. From the depth of 412–24 m, the ratio does not show much fluctuation, demonstrating the similar situation as the beginning. The different variation of $\sum\text{LREE} / \sum\text{HREE}$ may be related to the terrigenous clastic input and sedimentation rate.

DISCUSSION

The calculations of proxies are represented in Supplementary Table S1.

Paleoproductivity Proxies

The enrichment of organic matter depends on whether there were enough organic matter sources in the geological period. The sources of organic matter mainly come from two parts: besides the partly organic matter provided by terrigenous clastic input, the biological productivity in sedimentary water is another key factor for organic matter enrichment (Authur and Sageman, 1994). The higher biological productivity in sedimentary

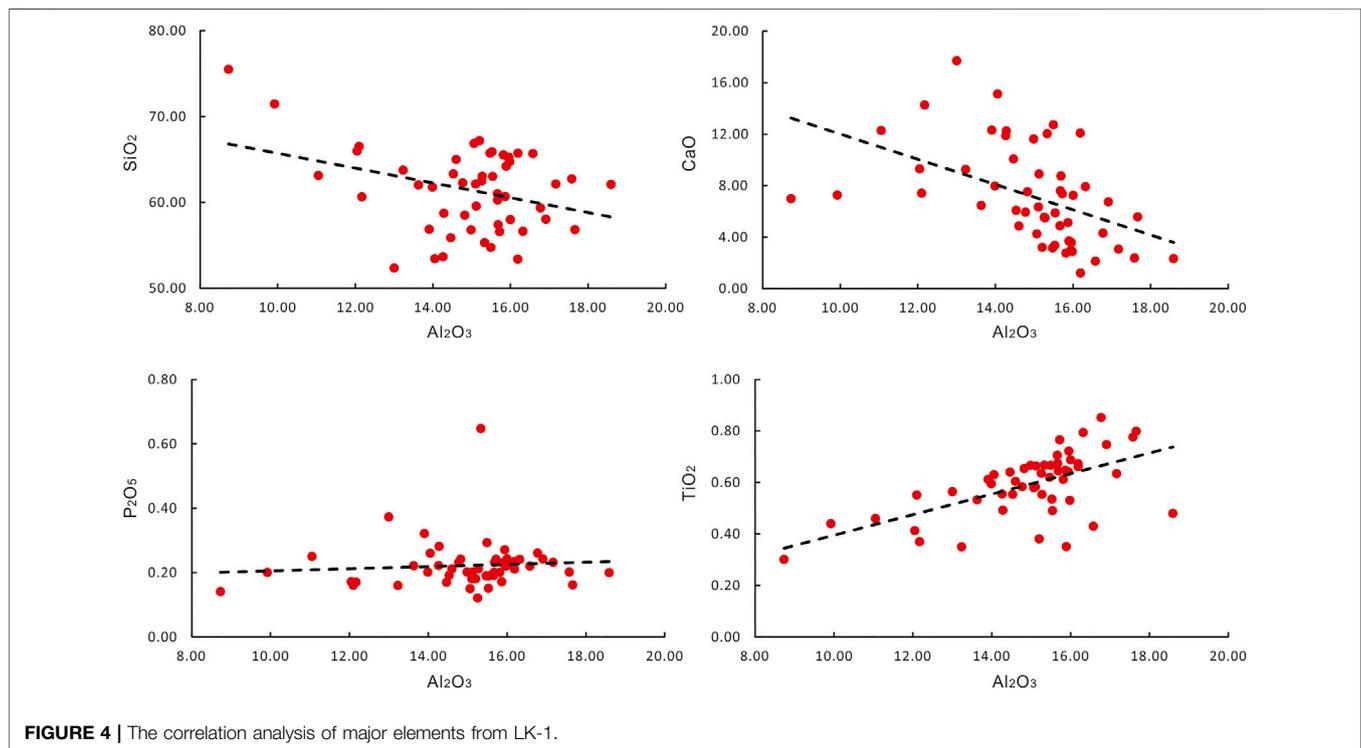
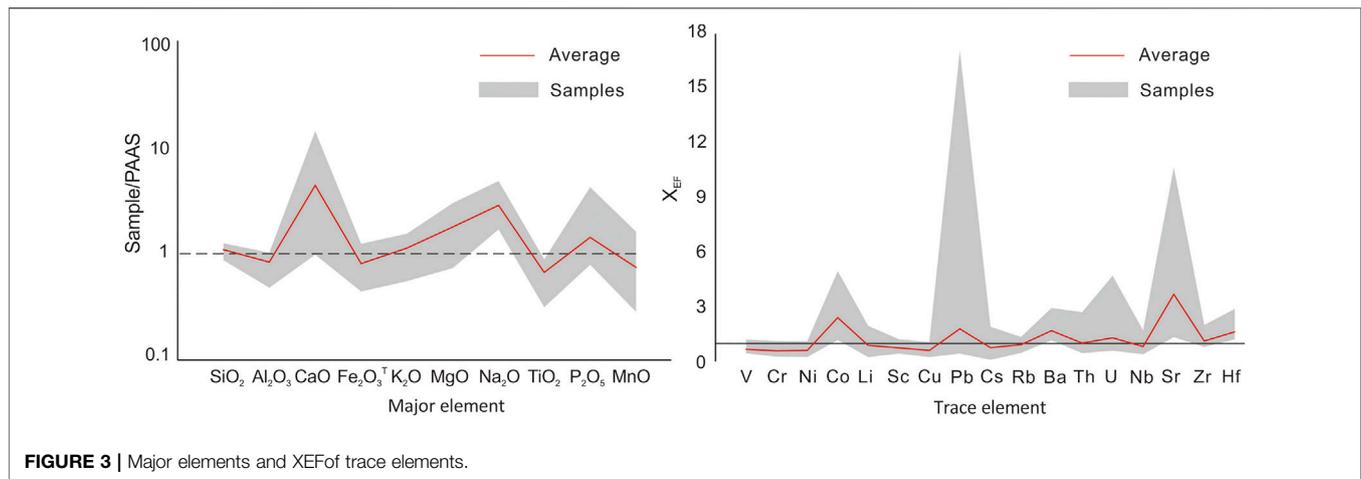


water, the better sedimentary condition for organic matter enrichment. Elements such as Ba, P, Al and Ni were widely used as paleoproductivity proxies. In this study, we select P and biogenic Ba (Ba_{bio}) content to discuss the paleoproductivity of Laiyang group.

Element P is necessary for organisms to survive and reproduce in water and the content of P element could reflect the paleoproductivity. In addition, the P of organisms will transfer into sediments after their death (Shen et al., 2015). In this study, P of samples are divided by Al and Ti to avoid the effect of terrigenous clastic input, so P/Al and P/Ti could reflect the paleoproductivity more precisely. The P/Ti and P/Al of shales of Laiyang group from LK-1 vary with the depth, behaving similar trends. The ratio of P/Ti ranges from 0.14 to 0.71 with a mean

value of 0.28 and the ratio of P/Al ranges from 0.007 to 0.035 with a mean value of 0.012. These two ratios are generally larger than the ones of PAAS (P/Ti = 0.12, P/Al = 0.007) according to **Figure 5**, indicating the high level of paleoproductivity in Laiyang period.

Biogenic Ba content (Ba_{bio}) has been widely used as an important paleoproductivity proxy. Also the Ba of samples were affected by terrigenous clastic as other elements, so avoiding the effects of terrigenous clastic is the first step to do. Murray and Leinen (1996) proposed the method to calculate the biogenic Ba content (Ba_{bio}): $Ba_{bio} = Ba_{Sample} - Al_{Sample} \times 0.0077$. The Ba_{bio} values of shales of Laiyang group from LK-1 range from 11.85 to 767.05 ppm with a mean value of 283.99 ppm. The trend of Ba_{bio} does not match the trends of P/Ti and P/Al perfectly

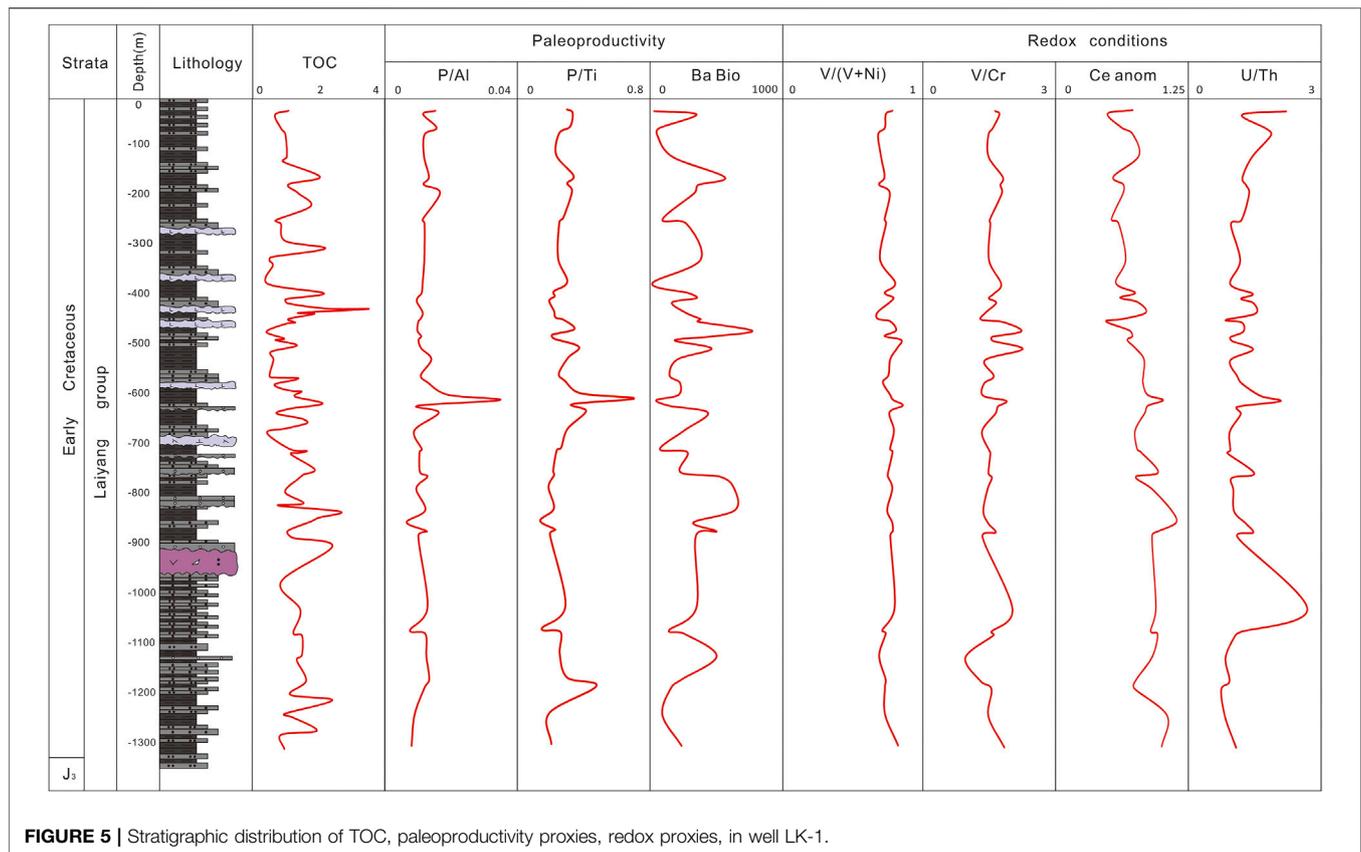


because the content of Ba may be affected by the redox condition and the hydrothermal activity.

Redox Proxies

Redox condition of sedimentary water is directly related to the preservation of organic matter, so the study of redox condition is very important. $V/(V + Ni)$ and Ce_{anom} are widely used to judge the redox condition (Abanda and Hannigan, 2006; Raiswell, 1988; Wang et al., 2014). $V/(V + Ni) < 0.46$ represents oxic environment, $0.46-0.57$ represents semi-anoxic environment, >0.57 represents anoxic environment. Moreover, when the value of $V/(V + Ni)$ is larger than 0.54 but less than 0.82,

anoxic and a less strongly stratified water column is indicated. If the value of $V/(V + Ni)$ is larger than 0.84, it indicates the presence of H_2S in a strongly stratified water column (Hatch and Leventhal, 1992). To avoid the potential analytical errors of single element concentration, other reliable indices such as V/Cr , Ce_{anom} and U/Th (German and Elderfield, 1990; Jones and Manning, 1994) are also used in this study. Values of V/Cr above two represent anoxic of depositional conditions, and the closer this value gets to 1, the more oxic the depositional condition it was. Ce anomaly was defined and applied by German and Elderfield (1990) as a paleoredox indicator: Ce anomaly $(Ce/Ce^*) = 3(Ce/Ce_{shale}) / [(2La/La_{shale}) + (Nd/Nd_{shale})]$,



the value of REE of the shale as normalized standard is from De Baar et al. (1985). As **Figure 5** shows, the values of $V/(V + Ni)$ of our samples do not show obvious fluctuation with the depth, ranging from 0.68 to 0.86 with a mean value of 0.75. This indicates the depositional condition is anoxic environment with a less strongly stratified water column. The values of Ce_{anom} of our samples range from 1.09 to 1.23 with a mean value of 1.16. According to the application of Ce_{anom} from German and Elderfield (1990), the depositional conditions in Laiyang period is an anoxic environment. The values of U/Th range from 0.70 to 2.69 with a mean value of 1.26, indicating the depositional conditions varied between anoxic and dysoxic environment (Jones and Manning, 1994). The values of V/Cr of shales from LK-1 range from 0.97 to 2.26 with a mean value of 1.58. Although the ratio of V/Cr might be affected by the carbonate content (Jones and Manning, 1994), its trend with the depth shows similarity as the trend of $V/(V + Ni)$. Combined those geochemical evidences above with widely distributed pyrite across the whole layers of Laiyang group, the redox condition of Laiyang period is mainly anoxic environment.

Paleosalinity and Paleoclimate Proxies

Paleosalinity is an important indicator to reflect the sedimentary water, and also could help to judge the marine and terrestrial environment. The ratio of Sr/Ba and the Value m ($=100 \times MgO/Al_2O_3$) were selected as the indicators to reflect the paleosalinity. The ratio of $Sr/Ba < 0.5$ represents freshwater environment, $0.5 <$

$Sr/Ba < 1$ represents brackish water and $Sr/Ba > 1$ represents salt water (He et al., 2019). Zhang et al. (1988) proposed the value m ($=100 \times MgO/Al_2O_3$) as an indicator to interpret the paleosalinity: $m < 1$ represents fresh water, $1 < m < 10$ represents transitional environment and $m > 10$ represents marine environment.

The ratios of Sr/Ba of shales of Laiyang group from LK-1 (**Figure 6**) range from 0.22 to 1.89 with a mean value of 0.68 and the value m range from 10.43 to 43.57 with a mean value of 23.27. These indicate the paleosalinity condition of depositional water of Laiyang period is almost brackish water to salt water in a marine environment.

CIA (Chemical Index of Alteration), C-value and Sr/Cu were selected to interpret the paleoclimate condition of Laiyang period. Because the weathering degree of rock is different under different climate condition, the CIA were successfully applied to reflect the paleoclimate condition (McLennan, 1993): 50 to 65 (arid), 65 to 85 (semiarid to semimoiest), 85 to 100 (moist). The element Sr, Cu, Mg, Ca are sensitive to the paleoclimate, so the ratios of Sr/Cu and Mg/Ca were used as indicators of paleoclimate in sedimentary period. The ratio of $Sr/Cu > 10$ was explained to stand for the arid and hot climate condition, while $1 < Sr/Cu < 10$ indicates warm and moist climate condition. The trace elements Fe, Mn, Cr, V, Ni and Co have been proved to be relatively enriched in sediments formed under moist conditions. In contrast, the elements of Ca, Mg, K, Na, Sr and Ba exhibit higher concentrations under arid conditions because the evaporation prompts saline minerals to

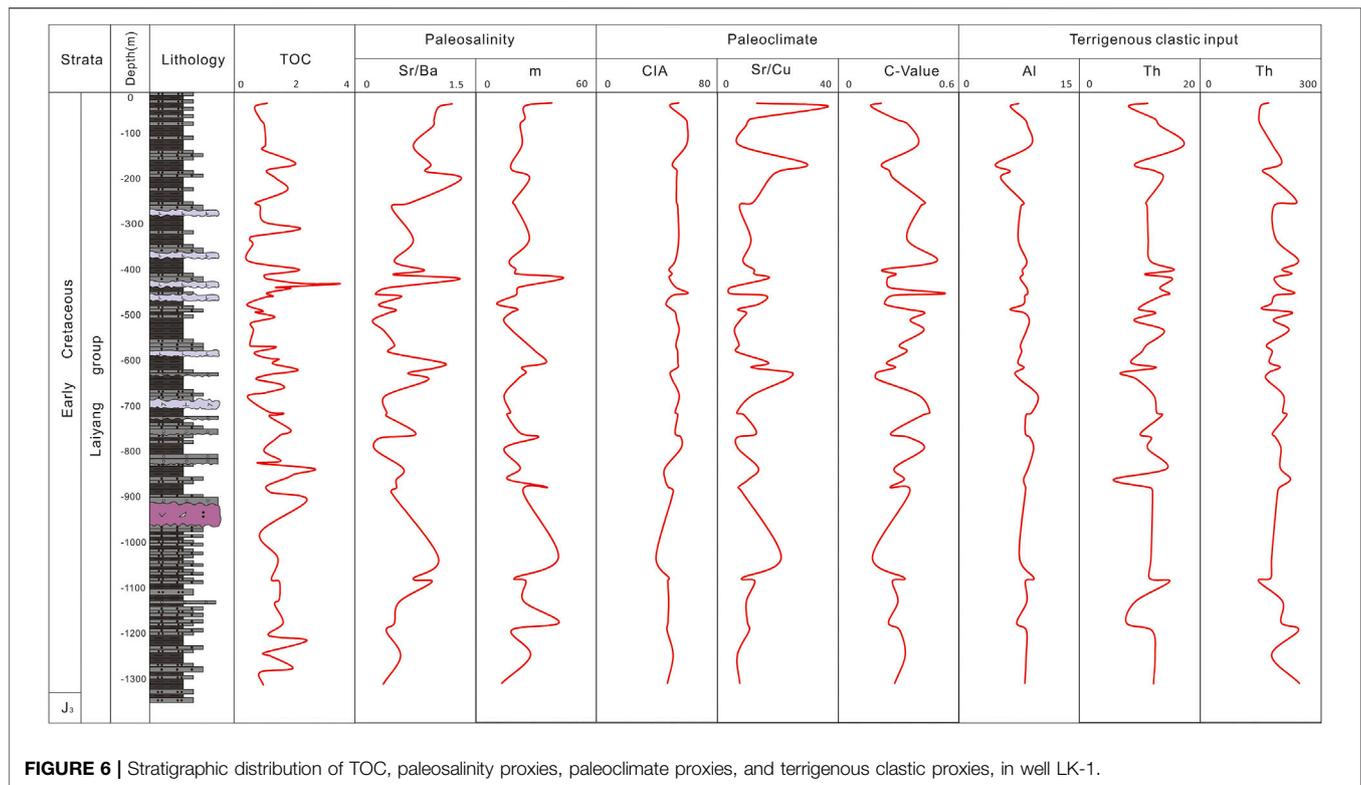


FIGURE 6 | Stratigraphic distribution of TOC, paleosalinity proxies, paleoclimate proxies, and terrigenous clastic proxies, in well LK-1.

precipitate (Cao et al., 2012). The C-value [$C\text{-value} = \frac{\Sigma(\text{Fe} + \text{Mn} + \text{Cr} + \text{Ni} + \text{V} + \text{Co})}{\Sigma(\text{Ca} + \text{Mg} + \text{Sr} + \text{Ba} + \text{K} + \text{Na})}$] has been applied successfully in evaluating the palaeoclimatic conditions (Cao et al., 2012; Fu et al., 2016; He et al., 2019).

In this study (Figure 6), the CIA of shales of Laiyang group range from 39.36 to 60.44 with an mean value 51.26, indicating an arid paleoclimate in Laiyang period. The C-values range from 0.16 to 0.53, reflecting dominant arid to semiarid climatic conditions at the time of deposition. The ratio of Sr/Cu range from 8.46 to 73.59 with a mean value of 25.12, showing an arid to semiarid paleoclimate condition.

Terrigenous Clastic Proxies

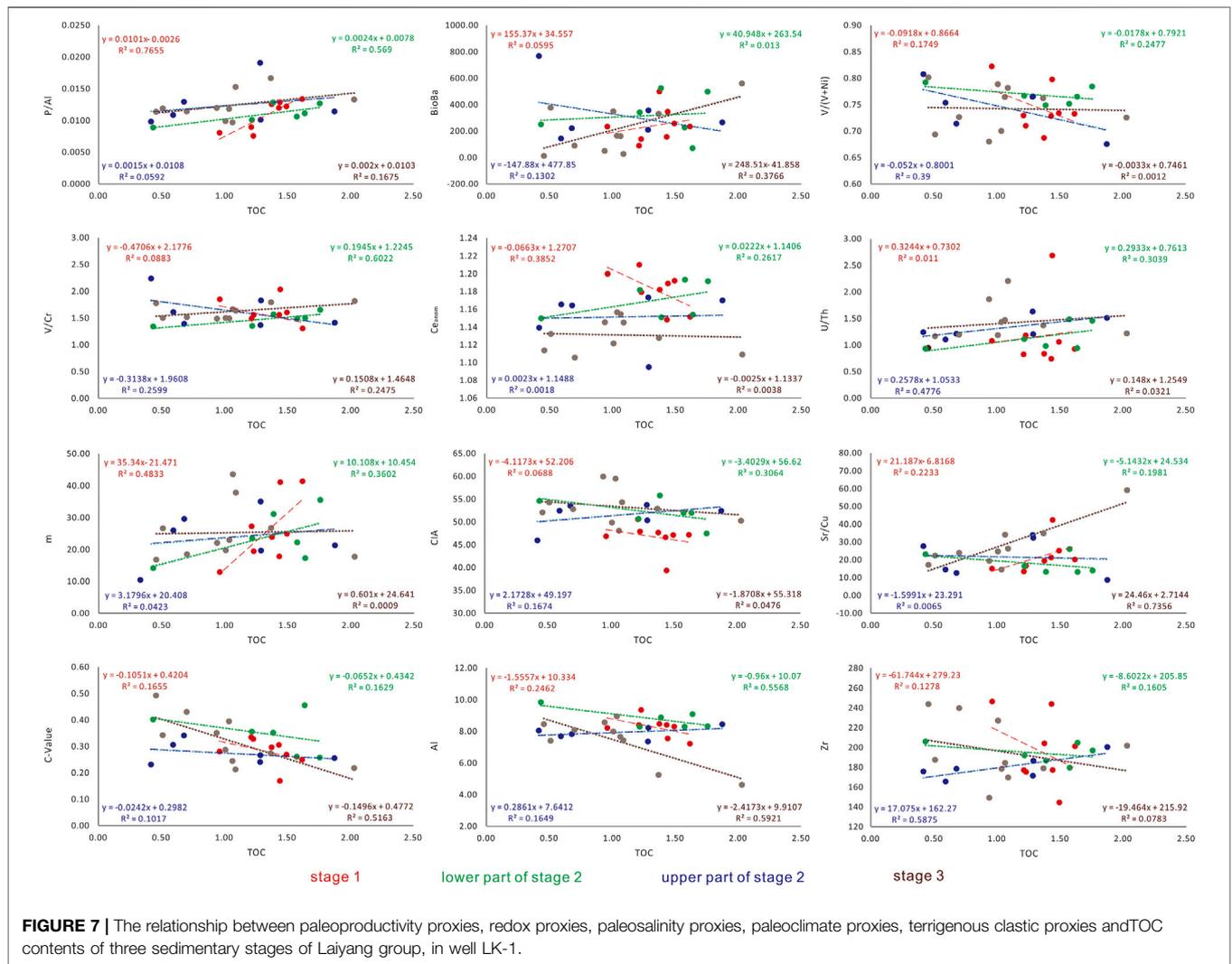
The concentration of Al, Ti, Zr, Th in sediments are mainly affected by terrigenous clastic. The latter diagenesis and weathering had little effect on their preservation. So, these elements were used to reflect the situation of terrigenous clastic input during the deposition period (Murphy et al., 2000; Tribouvillard et al., 2006). In sedimentary rock, element Ti, Zr and Th are persevered in the heavy minerals such as ilmenite, zircon or in the clay minerals, while Al are mainly persevered in the clay minerals (Young and Nesbitt, 1998). The contents of Al, Ti, Th and Zr vary in a wider range among the shales of Laiyang group: Al (4.62–9.84 wt% with a mean value of 7.97 wt%), Ti (0.18–0.51 wt% with a mean value of 0.36 wt%), Zr (144.51×10^{-6} to 246.31×10^{-6} with a mean value of 191.29×10^{-6}) and Th (5.64×10^{-6} to 28.30×10^{-6} with a mean value of 12.04×10^{-6}). From the trends of variation of Al, Zr, Th, the condition of terrigenous clastic

input varied during the Laiyang period. In the first sedimentary period, the change in terrigenous clastic input was quite small. The condition of terrigenous clastic input raised and then reduced for two times, showing an intermediate level. In the second sedimentary period, the change became much more fluctuant and reached the peak in the middle of this period. This may be related to the continuous input water as the rift developed. The level of terrigenous clastic input reduced at the beginning of the third sedimentary period, then raised gradually.

Controlling Factors of OM Enrichment

From the 15 different indicators above, the basic change in sedimentary condition of Laiyang period in Lingshan island could be concluded. The sedimentary environment is almost brackish water to salt water in a marine environment with redox condition of mainly anoxic. The paleoclimate condition is dominated by arid to semiarid climatic conditions. Paleoproductivity kept at a relatively high level in the whole Laiyang period, compared to PAAS. In order to find the relationship between TOC contents and those indicators of different factors and also find the key indicators in different stages, correlation analysis method were applied (Figure 7).

In the first stage, the index of P/Al is highly positive correlated to the TOC contents with $R^2 = 0.7655$. This suggests the paleoproductivity is the key controlling factors of OM enrichment in the initial stage. Although $C_{e\text{anom}}$ is slightly negative correlated with the TOC contents ($R^2 = 0.3852$), the correlation coefficients of $V/(V + \text{Ni})$ and V/Cr are all less than



0.2. Thus, the redox condition may be a minor factor of OM enrichment. The paleosalinity condition should be another minor factor of OM enrichment as well, reflecting by its medium positive correlation between *m* and TOC contents ($R^2 = 0.4833$). The correlation coefficients of all the other indexes versus TOC contents are less than 0.3, suggesting very little effect on OM enrichment. In the second stage, no obvious pattern were seen from the correlation analysis between the selected indexes with TOC contents if this period was regarded as a whole. This is because the variation of the factors of the upper part and the lower part of the second stage follow different trends, suggesting the controlling factors should be different between the upper and lower parts. In the lower part, the index of P/Al is medium positive correlated to the TOC contents ($R^2 = 0.569$), suggesting the paleoproductivity is one of the major controlling factor of OM enrichment. The indicators of redox condition show correlation with the TOC content at different level (V/Cr with $R^2 = 0.6022$, U/Th with $R^2 = 0.3039$, Ce_{anom} with $R^2 = 0.2617$), together suggesting the redox condition is another major controlling factor of OM enrichment

in this period. The effect of terrigenous clastic in the lower part is significant, because the Al content is medium negative correlated to the TOC content ($R^2 = 0.5568$). The indicators of paleosalinity and paleoclimate show slightly correlated to the TOC contents (*m* with $R^2 = 0.3602$ and CIA with $R^2 = 0.3064$), suggesting both the paleosalinity and paleoclimate condition are minor factors of OM enrichment. While in the upper part, only the index of Zr is medium positive correlated to the TOC contents with $R^2 = 0.5875$. But the indexes of Al and Th are almost not correlated to the TOC contents with $R^2 = 0.1649$, 0.201, respectively. The indexes of redox condition show medium to slight correlation with the TOC contents: U/Th with $R^2 = 0.4776$, V/(V + Ni) with $R^2 = 0.39$ and V/Cr with $R^2 = 0.2599$. These together indicate the terrigenous clastic input and redox condition are the major factors of OM enrichment of the upper part of the second stage of Laiyang period. In the third stage, the indexes of Sr/Cu, C-value and Al are highly to medium correlated to the TOC contents with $R^2 = 0.7356$, 0.5163 and 0.5921, respectively, suggesting paleoclimate condition is the major factor and the terrigenous clastic input is the minor factor of OM enrichment.

TABLE 1 | Factor of OM-enrichment in different stages.

Period	Factor of OM-enrichment	Grade	Represented proxies and correlation coefficient
Stage 1	Paleoproductivity	Major	P/Al with $R^2 = 0.7655$, positive
	Paleosalinity	Minor	m with $R^2 = 0.4833$, positive
	Redox condition	Minor	$C_{e_{anom}}$ with $R^2 = 0.3852$, negative
Lower part of Stage 2	Paleoproductivity	Major	P/Al with $R^2 = 0.569$, positive
	Redox condition	Major	V/Cr with $R^2 = 0.6022$, positive
	Terrigenous clastic input	Major	Al with $R^2 = 0.5568$, negative
	Paleosalinity	Minor	m with $R^2 = 0.3602$, positive
	Paleoclimate	Minor	CIA with $R^2 = 0.3064$, negative
Upper part of Stage 2	Terrigenous clastic input	Major	Zr with $R^2 = 0.5875$, positive
	redox condition	Major	U/Th with $R^2 = 0.4776$, positive V/(V + Ni) with $R^2 = 0.39$, negative; and V/Cr with $R^2 = 0.2599$, negative
Stage 3	Paleoclimate	Major	Sr/Cu with $R^2 = 0.7356$, positive C-value with $R^2 = 0.5163$, negative
	Terrigenous clastic input	Major	Al with $R^2 = 0.5921$, negative
	Paleoproductivity	Minor	BioBa with $R^2 = 0.3766$, positive

The paleoproductivity also show slightly to not positive correlated to the TOC contents, reflecting by the indexes of BioBa ($R^2 = 0.3766$), P/Ti ($R^2 = 0.1836$) and P/Al (0.1675). However, the other indicators are not obviously correlated to the TOC contents.

Above all, the controlling factors of OM enrichment varied during the three sedimentary periods (Table 1). This may be affected by the different sedimentary microfacies of the shale in different depth. Such as the geochemical features of shale from the “Te” part of turbidite may be closely related to the terrigenous clastic input, while the one from deep lake/marine should be much more affected by the paleoproductivity, redox condition and paleosalinity condition. In addition, the paleoclimate didn't change much during Laiyang period. Thus, the evolution of the rift basin which controlled the sedimentary facies should be the primitive factor controlling the OM-enrichment of shale.

CONCLUSION

The TOC contents of shale samples of Laiyang group from the well LK-1 are generally higher than 1 wt%, indicating that they are good source rocks. The factors controlling OM-enrichment varied in different sedimentary period of Laiyang period, recorded by the geochemical features of the shale samples. In the early period of rifting evolution (the stage 1 and the early stage 2), paleoproductivity is the major factor of OM-enrichment. The role of redox condition and terrigenous clastic input became more and more important. When the rift evolution entered the late period (the late stage 2 and the stage 3), terrigenous clastic input is the most important factor

controlling the OM-enrichment.(Mcmanus et al., 1998)., (Raiswell et al., 1988)

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/**Supplementary Material**, further inquiries can be directed to the corresponding authors.

AUTHOR CONTRIBUTIONS

All authors listed have made a substantial, direct, and intellectual contribution to the work and approved it for publication.

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SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: <https://www.frontiersin.org/articles/10.3389/feart.2021.808916/full#supplementary-material>

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Conflict of Interest: Author HZ is employed by the company Xianhe Oil Production Plant of Shengli Oilfield Company, SINOPEC; and Author ML is employed by the company Shengli Oil Production Plant of Shengli Oilfield Company, SINOPEC.

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