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Research article

Organic matter enrichment mechanism of Youganwo Formation oil shale in the Maoming Basin

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ABSTRACT

The Youganwo Formation oil shale located in the Maoming Basin represents a large commercially valuable lacustrine oil shale resource and a potential bio-shale gas reservoir in South China. With the aim of deepening the understanding of factors that influence organic matter enrichment, this research conducted a geochemical investigation to reconstruct the depositional paleoenvironment of bioproductivity, preservation and dilution. Youganwo Formation oil shale is mainly deposited in semi-deep to deep-lake environments with relatively warm and humid paleoclimate in the subtropical-temperate zone. The total organic carbon (TOC) content (1.46–11.85%), S_2 values (4.79–115.80 mg HC/mg rock) and HI (328–1040 mg HC/mg TOC) indicate that the oil shale has a good oil source rock potential. TOC content, ($S_1 + S_2$) values and vitrinite reflectance values show that its marginally mature organic matter (OM) belongs to kerogen type I-III with good oil-generating potential.

A 3rd order sequence was identified in the Yougnwo formation. Subsequently, the multiple factors including bioproductivity, preservation and dilution that control the OM enrichment of oil shale within system tracts were discussed. Moderate-quality oil shales ($O_{y\text{-}1}$) were developed in the transgressive systems tract (TST) in an oxidizing condition with abundant detrital input. High-quality oil shales ($O_{y\text{-}2}$) were deposited during the high-stand systems tract (HST) with increased accommodation space, improved preservation conditions, warm and humid climate, higher water bioproductivity and minimum detrital matter input. During the regressive systems tract (RST, $O_{y\text{-}3}$), higher detrital matter input and fresher water led to lower TOC values. Among these multiple factors, dilution condition was the major one that influences OM abundance and variation on the basis of sufficient organic matter input. Thus, OM enrichment models of $O_{y\text{-}1}$, $O_{y\text{-}2}$ and $O_{y\text{-}3}$ sub-members were established. The OM enrichment and quality in oil shale were controlled by the combined effect of bioproductivity, preservation, and dilution.

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1. Introduction

Oil shale is a potential energy resource alternative that can substitute crude oil [1,2]. It is deposited in more than 33 countries worldwide, and has a large *in situ* resource reserve that accounts for approximately 400 billion tons of oil [3]. As a special unconventional resource, oil shale is characterized by its high proportion of organic matter (OM) and minerals, low calorific value, high ash content, and geological concentration in a few of the major oil shale basins in Estonia, the United States and China [3,4,5].

China is now actively engaged in the geological research and exploration of oil shale [6]. A total of 47 basins in China host oil shale layers and the total number of identified reserves now rank fourth worldwide [3]. The Songliao Basin, Ordos Basin, Junggar Basin, Qaida Basin, Qiangtang Basin, Maoming Basin and Fushun Basin are the predominant basins that contain oil shale with commercial potential [7]. Among these, the Maoming Basin is located in economically developed areas of Southern China, where energy resources are relatively scarce.

The Paleogene Youganwo Formation and Neogene Shangcun Formation are two oil shale bearing formations in the Maoming Basin [8,9]. The former has been proven to be better in quality than the latter [9]. The biomarker characteristics [10,11], pyrolysis characteristics [12] and reservoir utilization approaches [8,13–15] of oil shale in the Maoming Basin have been studied and reported. However, geological investigations on the quality of oil shale and influencing factors of OM enrichment mechanism receive less attention.

The OM enrichment mechanism, which is influenced by a variety of factors during oil shale deposition, controls the quality of oil shale for utilization. Uncovering the OM enrichment mechanism of the Youganwo Formation is a key to investigating and assessing oil shale resource in the Maoming Basin. In addition to the commercial utilization of oil shale, the study of sedimentary geological characteristics can also provide valuable information on paleoenvironmental and global paleoclimate changes during the Cenozoic Paleogene period [14]. The aim of the present study was to investigate the geochemical and paleoenvironment characteristics of Youganwo Formation oil shale in the Maoming Basin. In addition, the present study also aimed to reveal the influencing factors of OM enrichment and establish a model for its mechanism. A drill core was conducted at the M − 1 well to prepare the samples for the experiments. In the present study, the reservoir characteristics and key influencing factors of OM enrichment in the Youganwo Formation were investigated, which is beneficial for deeply understanding high-quality oil shale formation mechanism, and providing a basis for effective evaluation and exploitation of oil shale resources as well as potential biogenic shale gas in the Maoming Basin.

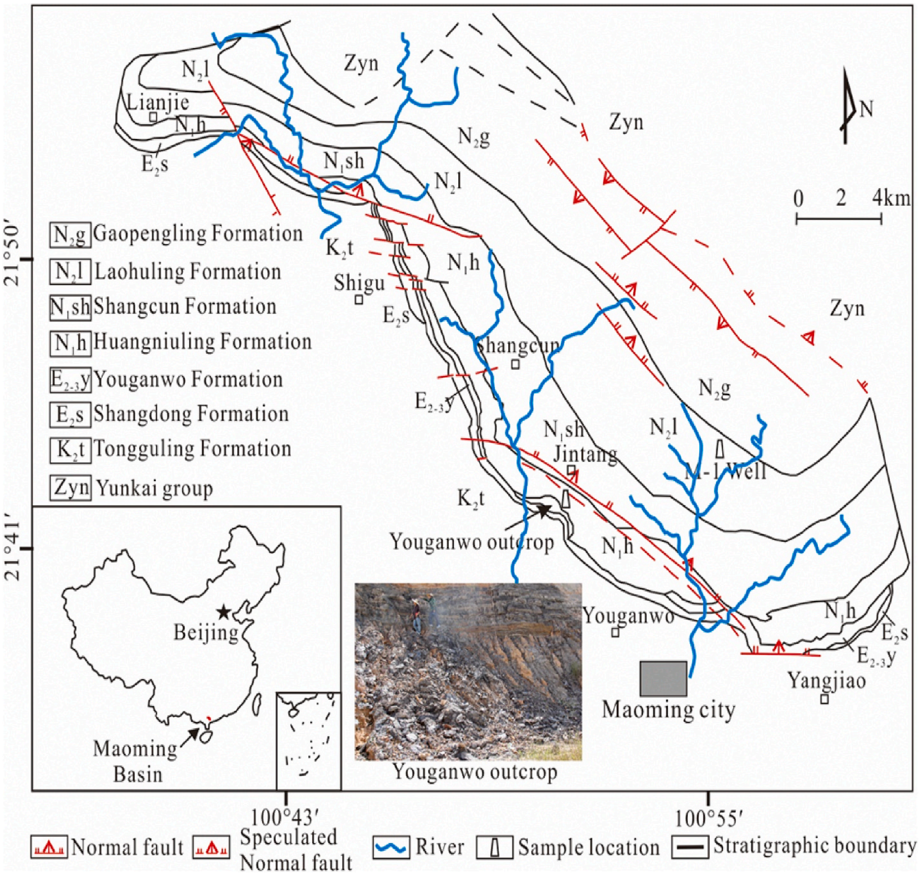

Fig. 1. Generalized map of the location of the research area and M − 1 well (Modified after [9]).

2. Geological setting and experimental methods

2.1. Geological setting

The Maoming Basin is a half-graben basin located at the southeastern margin of the Yunkai uplift (belonging to the West-Guangdong uplift) in the Southern China orogenic belt, which covers an area of approximately 400 km^2 (Fig. 1). Specifically, the Maoming basin is located between the Wuchuan-Sihui fault zone and Xinyi-Lianjiang fault zone, and is approximately 44 km in length and 4–14 km in width. This basin is a symmetric diagonal basin developed on the basement of the Mesozoic Nansheng Basin, with red terrigenous clastic rocks and igneous rocks from the Tongguling Formation of Early Cretaceous. Outcrops and oil shale mines are located at the south-western edge of the basin, with an exposed area of approximately 4 km^2. In the section, the basin has a half-graben shape with a monoclinic structure. On the south-eastern edge, the basin is fault-bounded, and controlled by the Wuchuan-Sihui fault and Gaopengling fault. As a result, the basin has a wider south wing, and is steeper and deeper on the north. The Maoming basin has experienced multi-phased tectonic evolutions, including the Caledonian movement, the Indosinian movement, the Yanshan movement and the Himalaya movement, and NE stratigraphic dips were thus formed with dip angles ranging within 4–10°.

As shown in the generalized stratigraphic column (Fig. 2), the Mesozoic strata, Triassic strata and Jurassic strata are not found in the Maoming basin. The Cretaceous strata of the Shigu group, Xitangling Formation, Tongguling formation, Paleogene strata of the Shangdong formation, Youganwo formation, Neogene strata of the Huangniuling formation, Shangcun formation, Laohudong formation, Gaopengling formation and Quaternary strata are deposited on the basement of the Sinian Yunkai group, respectively. Among these, the Youganwo formation mainly consists of oil shale and OM rich shale interbedded with sandstone and lignite. The exploitation of Youganwo formation oil shale in the Maoming Basin is mainly conducted by open pit mining and near-to-surface underground mining. The three major mining areas of Shigu, Jintang and Yangjiao are located in the west, middle and east of the basin, respectively. The thickness of the Youganwo formation ranges within 19.2–46.5 m in open pit mines. The majority of the Youganwo Formation oil

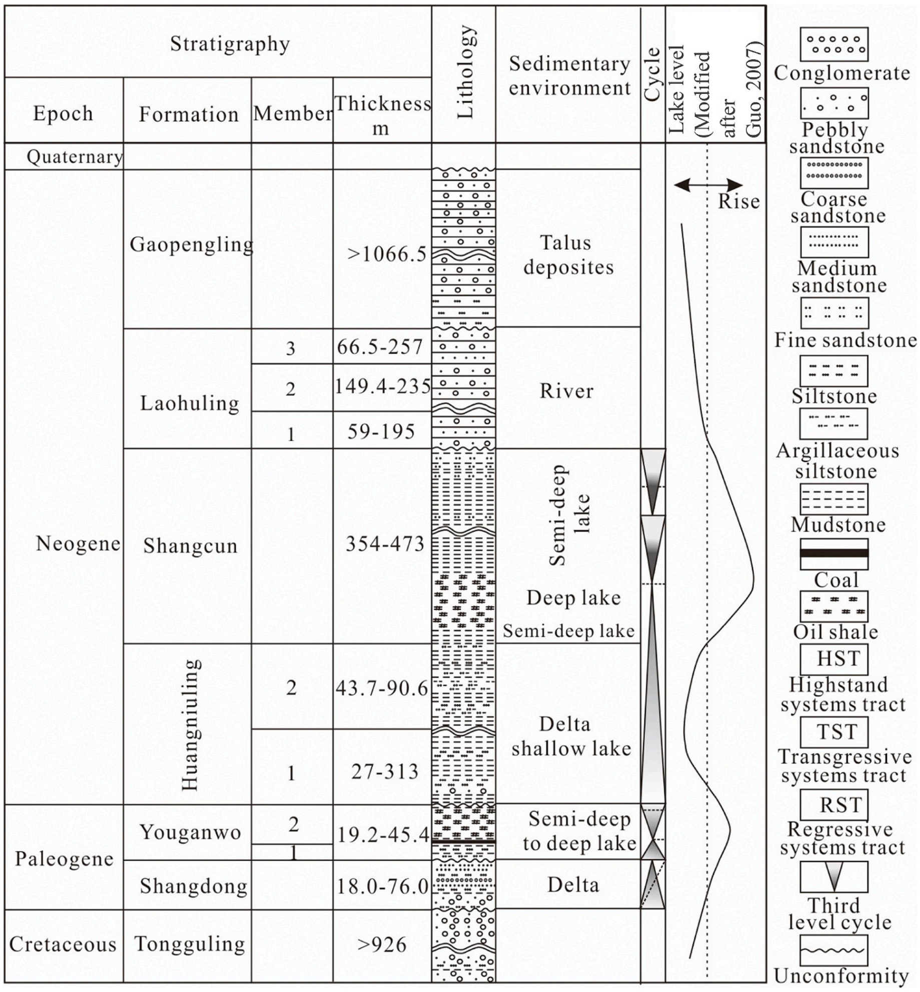

Fig. 2. Generalized stratigraphic column of the Maoming Basin.

shale is black or brown in color, and its oil content ranges from 5 to 9% [9,10]. Furthermore, 1–3 layers of lignite coal seams are developed in the Youganwo formation, while a layer of conglomerate with a thickness of approximately 0.5 m is developed at the bottom of the Youganwo formation.

2.2. Experimental methods

A total of 29 samples were collected from the M − 1 well (N21°44′20″, E110°57′29″) and Youganwo outcrop (Fig. 1). A geochemical study was conducted to determine the characteristics of the paleo-sedimentary environment. The paleoclimate, paleo-redox condition, paleosalinity, origin of the OM, bioproductivity and detrital matter input were studied based on the geochemical data and sporo-pollen test, in order to provide information for the further discussion on influencing factors of OM enrichment in Youganwo Formation oil shale. A total of 21 samples obtained from the M − 1 well were used in bulk for the inorganic geochemical analysis.

The sporo-pollen test was conducted in Nanjing Institute of Geology and Palaeonotology, Chinese Academy of Sciences (CAS). The total organic carbon (TOC) test and Rock-Eval pyrolysis test were conducted at the Lanzhou Center for oil and gas resources of the Chinese Academy of Sciences (CAS) to determine the OM characteristics. Samples for TOC test were decontaminated and grinded to 80 meshes, and divided into two groups. One group was decontaminated with 5% hydrochloric acid to remove carbonate minerals, and then subjected to organic carbon test with CS-344 carbon/sulfur analyzer. Rock-Eval 6 apparatus (Vinci Technologies) was used for

Fig. 3. Field sedimentary characteristics of the Youganwo Formation on the Youganwo Outcrop.

rock pyrolysis analysis. 20 mg of shale samples were weighed and put into a small porcelain crucible. Before the experiment, the organic matter in the crucible was removed in a muffle furnace with the temperature 850 °C according to the procedure as follows. The initial temperature of the oxidation furnace was set at 300 °C and kept constant for 3 min. And then, the temperature was operated to rise to 650 °C at the rate of 25 °C/min, and the temperature was kept constant for 3 min. The initial temperature of the cracking furnace was set at 300 °C, and kept constant for 1 min. Then, it was operated to increase to 850 °C at the rate of 20 °C/min, and kept constant for 5 min. Mean random reflectance (%R_o) of organic matter was measured using an Axioskop 40 photometer system following the experimental procedures of Taylor et al. (1998) [16].

The samples used for inorganic elemental analysis were crushed to particle sizes less than 100 μm. After being dried and cooled to room temperature, 0.500 g samples were weighed and put into a poly tetra fluoroethylene beaker, and then 1 ml deionized water was added for wetting. The samples were decomposed with HNO_3+HF mixed acid, and then their metal elements, including main and trace elements, were tested by VISTA MPX Inductively Coupled Plasma-Atomic Emission Spectroscopy (ICP-AES). In the rare earth element test, sodium peroxide melting method was used to decompose samples firstly, and then samples were dissolved and precipitated using HNO_3. X SERIES 2 Inductively Coupled Plasma Mass Spectrometer (ICP-MS) was used to determine the element at last. The relative error of element analysis was less than 5%. The inorganic elemental analysis was conducted in the Research Institute of Petroleum Exploration & Development (RFPED).

3. Results

3.1. Sedimentary environment and lithological characteristics

Sedimentary environment is one of the key influencing factors of the quality of hydrocarbon source and reservoir [17]. As revealed by outcrops and drill cores, the oil shale of the Youganwo Formation in the study area was deposited in a lacustrine environment, which can be divided into.

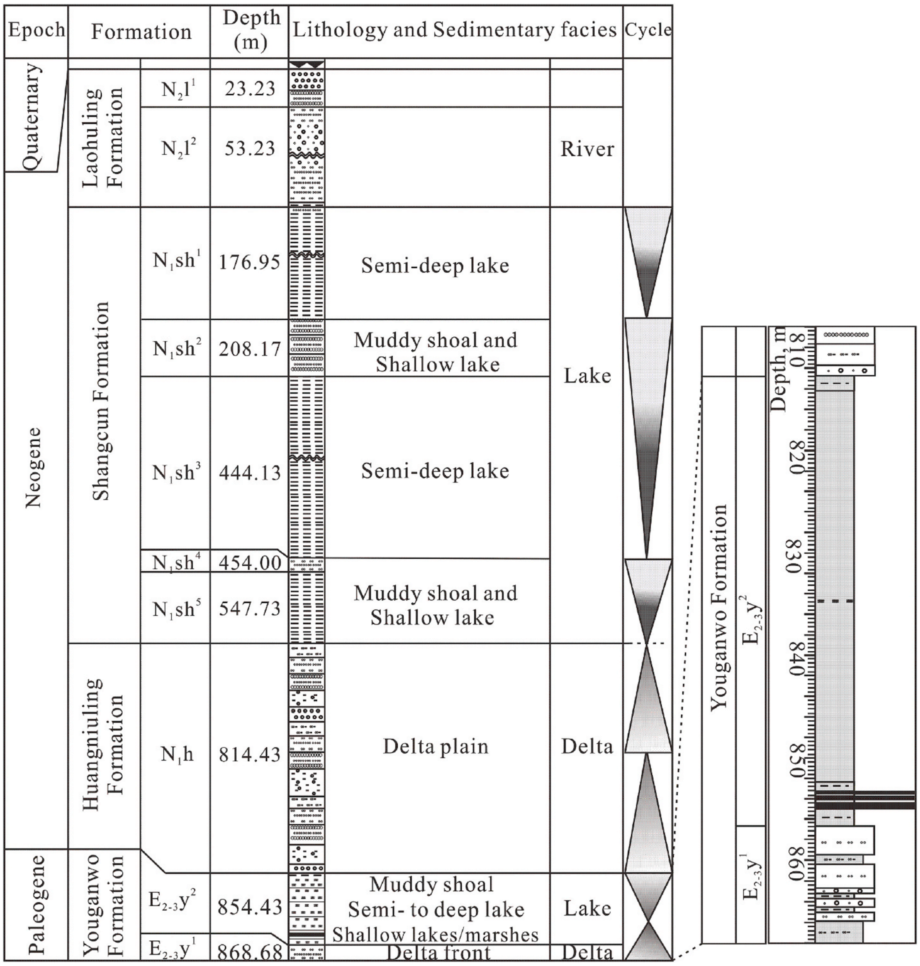

Fig. 4. Lithostratigraphic columnar sections and sedimentary environments revealed by M − 1 well.

Two major subfacies: swamp subfacies and lake subfacies. Lake subfacies can be further divided into three microfacies: semi-deep lake microfacies, deep-lake microfacies and shallow-lake microfacies (Figs. 3 and 4). Oil shale was mainly deposited in semi-deep to deep-lake microfacies (Fig. 3a–d). Organic-rich mudstone (with sandstone interlayers) of shallow-lake microfacies had lower oil content (Fig. 3d).

Delta subfacies were found on the edge of the basin, showing fluviation on the lacustrine depositions, such as the underwater distributary river microfacies shown in Fig. 3e and f. The vertical distribution of microfacies also illustrates that semi-deep to deep-lake microfacies was the predominant sedimentary environment for the formation of oil shale in the Youganwo Formation (Fig. 4).

The Youganwo Formation can be divided into two members of $E_{2\text{-}3}y^1$ and $E_{2\text{-}3}y^2$ (Fig. 4). In $E_{2\text{-}3}y^1$, the delta subfacies were deposited with lithology of fine-medium sandstone. In $E_{2\text{-}3}y^2$, three types of oil shale bearing rock were further identified as $O_{y\text{-}1}$, $O_{y\text{-}2}$ and $O_{y\text{-}3}$ from the bottom to the top. Swamp microfacies were deposited at $O_{y\text{-}1}$ with lithology of carbonaceous shale, lignite and oil shale. Deep-lake microfacies of brown or hazel colored oil shale were deposited at the middle part of the $E_{2\text{-}3}y^2$ member ($O_{y\text{-}2}$). Shallow-lake subfacies developed at the top part of the $E_{2\text{-}3}y^2$ ($O_{y\text{-}3}$) with grey-black mudstone interbedded with siltstone and sandstone.

Based on the sedimentary environment and lithological characteristics, the Youganwo formation can be interpreted as a 3rd order sequence (Fig. 5). The differences of lithological characteristics in different tracts are revealed through a comprehensive study of borehole core and profile observation. There were significant differences in lithology and thickness in different stages of system tracts with basin evolution [18]. According to the facies architecture and sequence stratigraphic framework of the Youganwo formation shown in Fig. 5, a low-stand systems tract (LST) existed during $E_{2\text{-}3}y^1$. The oil shale bearing strata of $E_{2\text{-}3}y^2$ comprise a transgressive systems tract (TST), a high-stand systems tract (HST) and a regressive systems tract, which corresponds to the sub-members of $O_{y\text{-}1}$, $O_{y\text{-}2}$ and $O_{y\text{-}3}$. The HST and RST constitute the major part of the oil shale layers in the $E_{2\text{-}3}y^2$ member, followed by the TST with major lithology of both oil shale and mudstone. Differences in sedimentary environments and petrological characteristics well verifies the division of sequence in the Youganwo formation.

3.2. *Organic geochemical characteristics*

OM abundance, kerogen type and thermal maturity are the key parameters for evaluating oil shale quality [19]. TOC data is an important index to evaluate the OM abundance, which affects the OM enrichment degree [20]. The TOC content of the tested samples ranged within 3.73–28.88 wt%, with an average value of 13.89 wt% (Table 1). A TOC value of 10 wt% is considered as the dividing limit of organic-rich shales and oil shales [21]. These TOC values indicate that the samples were organic-rich and favourable for the preservation of OM.

T_{max}, °C value is an evaluation parameter for OM maturity in oil shale [19]. The Rock-Eval T_{max} of oil shale samples ranged within 395–436 °C. Relatively low T_{max} values indicate that the Youganwo Formation oil shale is immature, which corresponds to the low maturity oil forming stage in oil and gas evolution. As shown in Table 1, the vitrinite reflectance values (Ro/%) of the tested samples ranged within 0.40–0.65%, with an average value of 0.48%. The vitrinite reflectance has its own shortcomings compared with new

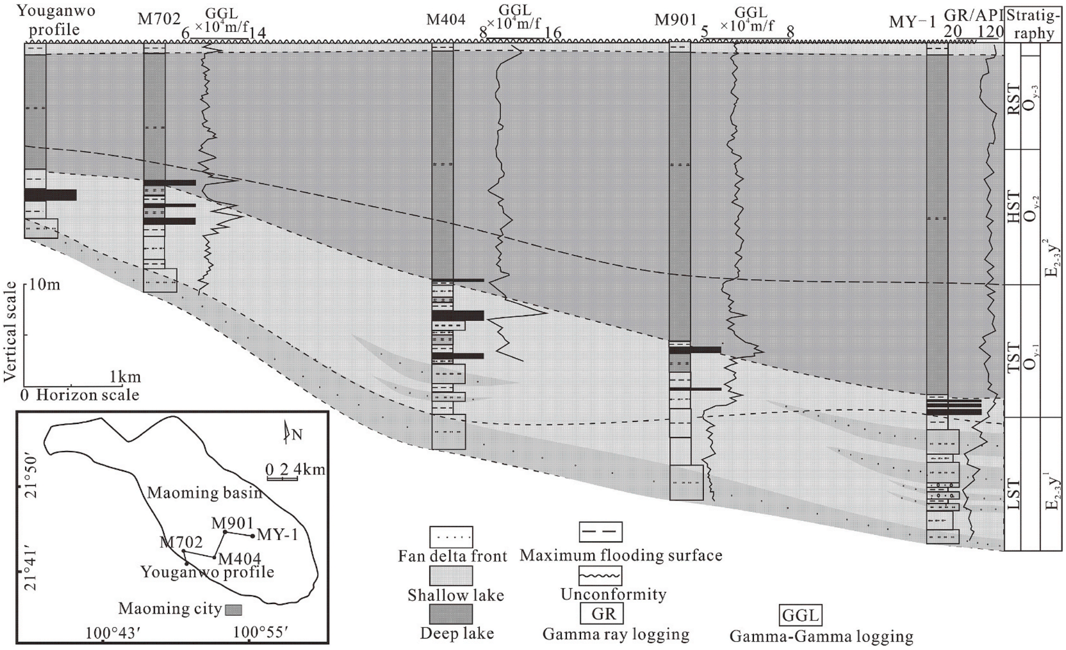

Fig. 5. Lithostratigraphic columnar sections and sedimentary environments revealed by the M − 1 well.

Table 1
Organic matter characteristics of Youganwo Formation oil shale samples.

Samples[a]	TOC/(%)	HI/(mg/g TOC)	T_{max} (°C)	S_1/(mg HC/g rock)	S_2/(mg HC/g rock)	S_1+S_2/(mg HC/g rock)	Ro/(%)
S-1	13.42						0.40
S-2	9.86	610	395	0.83	59.32	60.16	0.65
S-3	7.85						0.64
S-4	9.94	557	415	1.24	55.36	56.6	0.44
S-5	20.8						0.40
S-6	13.6						0.58
S-7	7.16	1016	422	0.84	71.97	72.81	0.44
S-8	16.4						0.51
M-1	11.11	763	435	0.88	84.72	85.6	0.496
M-2	6.68	798	434	0.83	79.6	80.43	0.446
M-3	13.34	719	434	0.64	95.97	96.61	0.450
M-4	9.28	842	424	1.31	110.53	111.84	
M-5	10.74	760	434	0.44	81.58	82.02	0.454
M-6	13.97	825	434	0.74	115.31	116.05	
M-7	12.77	789	435	0.53	100.75	101.28	0.445
M-8	23.06	1034	428	2.05	275.76	277.81	
M-9	23.14	754	429	1.75	167.19	168.94	0.465
M-10	22.14	768	430	1.85	153.93	155.78	0.459
M-11	18.42	940	429	2.21	219.75	221.96	
M-12	14.6	783	422	2.11	113.54	115.65	0.463
M-13	16.26	764	434	1.04	124.28	125.32	0.469
M-14	18.88	785	435	1.28	128.91	130.19	
M-15	11.3	835	434	1.14	135.01	136.15	0.465
M-16	10.06	695	424	1.13	120.03	121.16	0.479
M-17	22.18	754	429	1.75	167.19	168.94	0.500
M-18	3.73	564	425	0.72	44.17	44.89	
M-19	28.88	813	436	1.8	234.79	236.59	0.497
M-20	9.03	598	427	0.33	53.97	54.3	
M-21	4.24	615	429	0.12	26.08	26.2	
M-1#	12.57	701	426	0.83	87.32	88.15	
M-2#	12.05	604	430	0.84	71.97	72.81	

HI stands for hydrogen index, which ranged from 557 to 1034 mg/g TOC. The total generation potential parameter of the ($S_1 + S_2$) values of outcrop samples (S-2, S-4 and S-7) were 60.16, 56.6 and 72.81 mg HC/g rock, respectively. The ($S_1 + S_2$) values of M − 1 well samples ranged within 26.20–277.81 mg/g. The TOC and ($S_1 + S_2$) values indicate that oil shale in the Youganwo Formation has good oil-generating potential. However, the wide range of TOC values also indicates the presence of strong vertical heterogeneity characteristics of OM abundance in Youganwo oil shale [22].

methods such as Raman Spectroscopy [23]. Also, there were differences between bitumen in pores and depositional organic matter when measuring maturity for organic matter in oil shale. The maturity of the tested samples was in agreement with the previous report of Cao et al. (2016) [14].

Youganwo formation oil shales have good oil-generating potential, with an average TOC value of 13.89 wt%, and maturity of the marginally mature phase. However, it has a marked heterogeneity in OM abundance over short distances, especially in the early stage of $E_{2\text{-}3}y^2$ (Fig. 6). At a depth ranging within 856–844 m, which corresponded to $O_{y\text{-}1}$, the TOC values changed from 3.37 to 28.88 wt%. The highest TOC values were found in $O_{y\text{-}2}$ oil shale, which was located at depths between 844 and 832 m. All tested samples in $O_{y\text{-}2}$ had a TOC value higher than 10 wt%. The TOC values of $O_{y\text{-}3}$ oil shale between 832 m and 814.43 m were relatively lower than those of $O_{y\text{-}2}$ oil shale, ranging between 6.68 and 13.97 wt%. This phenomenon can be explained by the variation of depositional environmental factors such as the seasonal different organic input into these sediments [8]. OM characteristics are the major controlling factors of the heterogeneous physical properties and function as the basement for pore-scale characterization of shale reservoirs [24].

3.3. *Paleoclimate*

Spore and pollen are plant reproductive organs that are easy to preserve, which are found in the stratum. The type and abundance of spore and pollen are important indicators of paleoclimate. The sporo-pollen test results of Yu (1983) and Guo (2007) have revealed that angiosperm pollen is the predominant pollen type in Youganwo Formation oil shale, followed by fern pollen and gymnosperms pollen [9,25]. In the present study, fern, gymnosperm, angiosperm and algae plants were identified using outcrop samples (Table 2). Fern pollens were the predominant pollen types, including *Crassoretitriletes, Osmundacidites* and *Polypodiaceaesporites*. Furthermore, the sporo-pollens in sample S-7 were all *Pediastrun*, indicating that the formation of high-quality oil shale may be correlated to the flourishing of ferns and accumulation of algae in lake sediments with a relatively humid climate. The development of gymnosperm and angiosperm pollen reflects a warm and humid subtropical-temperate environment. The palynological assemblage generally represents a subtropical-temperate zone paleoclimate.

The Sr/Cu ratio is another indicator of climate. The TOC contents, Sr/Cu ratios, Sr/Ba ratios, V/Cr ratios, Ce-anomaly values and V/(V + Ni) ratios are presented in Fig. 7. If the Sr/Cu ratio ranges between 1.3 and 5.0, the paleoclimate will be classified as warm-humid climate, and if the ratio is higher than 5.0, it indicates a hot-arid climate [26]. The Sr/Cu ratio of the Youganwo Formation ranged from

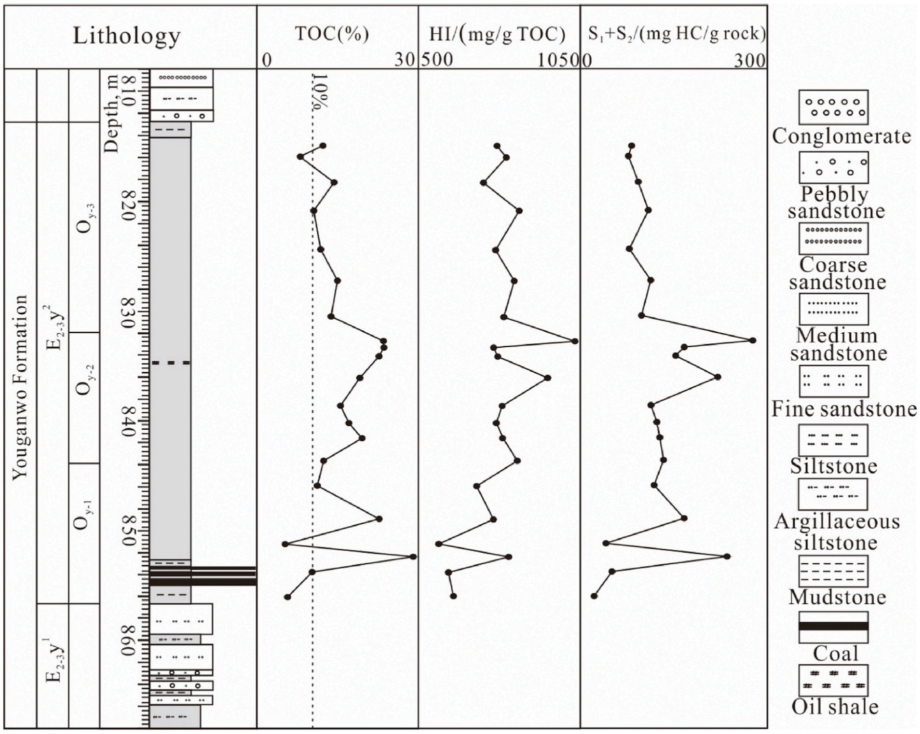

Fig. 6. Depth profiles of TOC, HI and ($S_1 + S_2$) for the M − 1 well.

Table 2
Sporo-pollen test results of the Youganwo outcrop samples.

Sporo-pollen		S-1	S-3	S-4	S-5	S-6	S-7	S-8
Fern	*Crassoretitriletes*	3		8				2
	Osmundacidites	3	77		2			1
	Polypodiaceaesporites	30	19	39	9	2	3	25
Gymnosperm	*Pinuspollenites*	5	7	3			4	2
Angiosperm	*Caryapollenites simplex*	1						
	Ulmipollenitses	4						
	U.undulosus	1						
	Carpinipites					1		
	Engelhardtioidites	6	1					
	Tiliaepollenites			1				
	Liquidambarpollenites						1	
	Rutaceoipollis					1		
	Quercoidites		1					
Algae	*Pediastrun*						20	

2.66 to 5.34, suggesting a transitional climate between hot-arid and warm-humid climates. However, paleoclimate should be classified as a relatively warm and humid climate, because most of the Sr/Cu ratios were lower than 5.0. Merely four samples in $E_{2\text{-}3}y^2$ belonged to a hot-arid climate. One sample was at the transition layer from the $O_{y\text{-}2}$ oil shale to $O_{y\text{-}3}$, while the other three were at the bottom of $O_{y\text{-}1}$.

3.4. Paleo-redox condition

The paleo-redox condition of oil shale was determined using V/Cr ratios, the Cerium anomaly index and V/(V + Ni) ratios. The V/Cr ratios indicate the oxic conditions of oil shale because the vanadium element preserved preferentially in anoxic conditions, while the chromium element's preservation is unaffected by redox conditions [27]. Generally speaking, V/Cr ratios >4.25 suggest anoxic conditions, ratios ranging between 2.00 and 4.25 point to dysoxic conditions, and ratios <2 point to oxic conditions. The V/Cr ratios of the Youganwo Formation in the M − 1 well ranged within 1.222–2.116, with an average value of 1.687, signifying a weak oxidizing to dysoxic condition (Fig. 7). The Sr/Cu ratio also showed a tendency towards a more anoxic condition from $O_{y\text{-}1}$ to $O_{y\text{-}3}$.

Cerium anomaly (Ce anomaly), which is widely used as an indicator for redox conditions of bottom-water, is defined as $\log^{[2Cen/(Lan}$

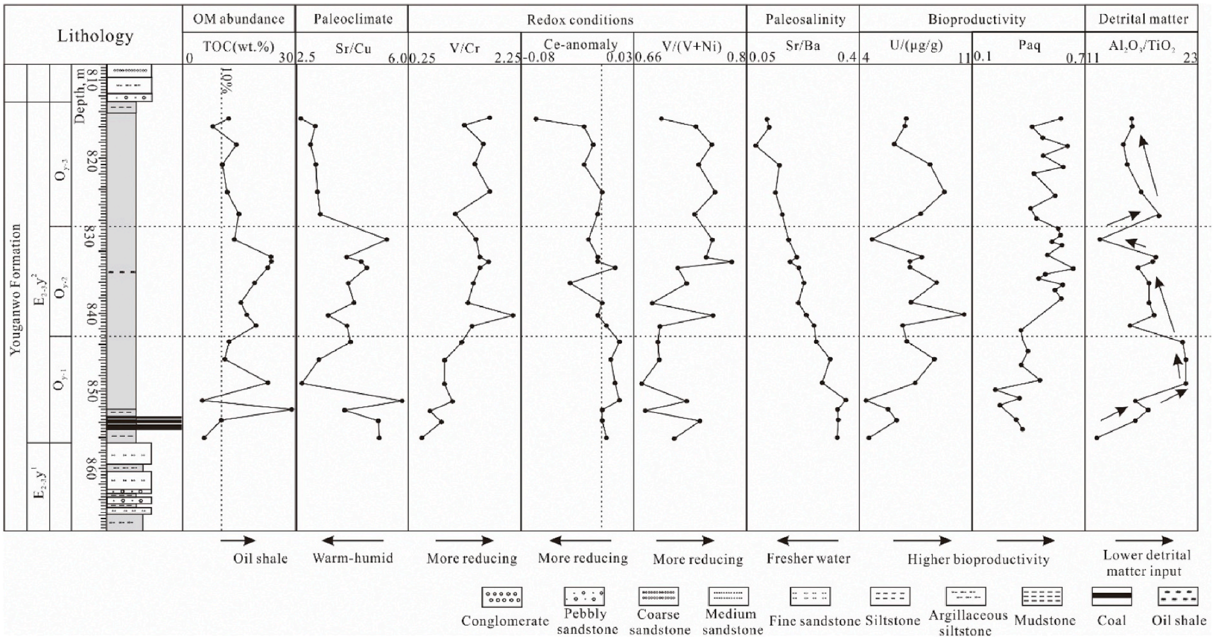

Fig. 7. TOC contents, Sr/Cu ratios, Sr/Ba ratios, V/Cr ratios, Ce-anomaly values, V/(V + Ni) ratios, Sr/Ba ratios, U values and Al_2O_3/TiO_2 ratios of Youganwo Formation oil shale in the M − 1 well.

+ Prn)] (the subscript “n” symbolizes the concentration normalized to chondrite). Ce anomaly values of less than −0.10 indicate reducing conditions, while values greater than −0.10 indicate oxidizing conditions. The cerium anomaly in the Youganwo Formation of the M − 1 well (−0.0655 to 0.017 μg/g; avg., −0.0045 μg/g) generally exhibited oxic conditions [28]. In addition, there was a decreasing trend of Ce anomaly from $O_{y\text{-}1}$ to $O_{y\text{-}3}$, indicating that the reduced condition of water was enhanced.

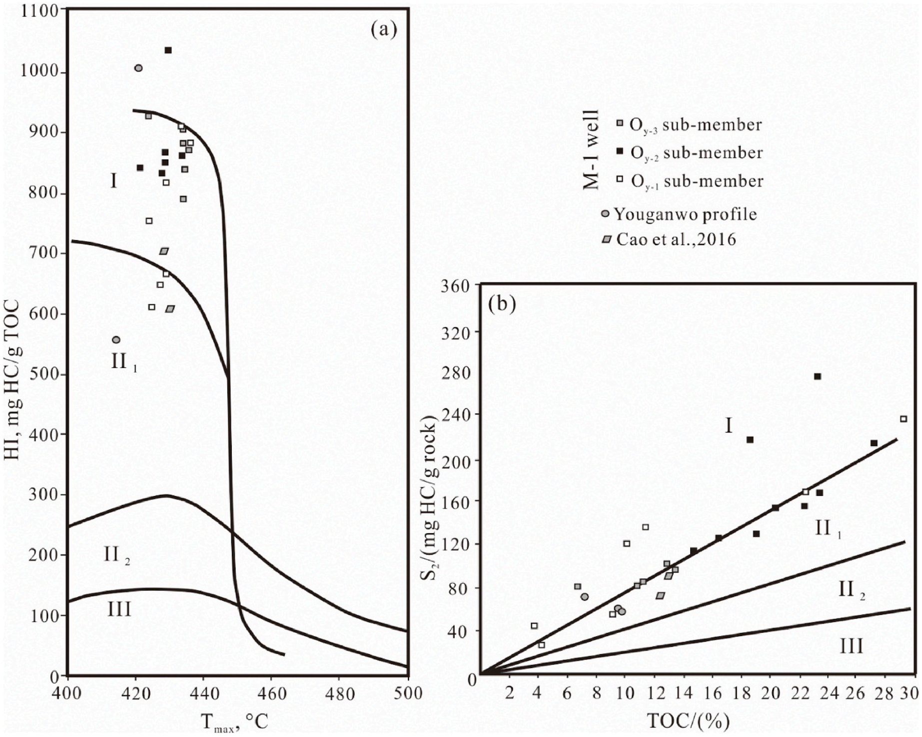

Fig. 8. Plots of HI *vs.* T_{max} and S_2 *vs.* TOC outlining the OM type of oil shale from the Youganwo Formation (Partial data from Ref. [13]).

The V/(V + Ni) ratio is a common indicator to distinguish the redox sedimentary environment. Generally, a high V/(V + Ni) ratio (>0.60) reflects a reducing environment, while a low V/(V + Ni) ratio (<0.46) represents oxic conditions [29]. The V/(V + Ni) ratios of the M − 1 well (0.669–0.781; avg., 0.722) were relatively high (>0.60), reflecting a reducing environment.

From the elemental analysis, it is notable that the Youfanwo Formation was deposited in a weak oxidizing to dysoxic condition. Moreover, the degree of oxidation decreased in turn from the O_{y-1} sub-member to the O_{y-3} sub-member.

3.5. *Paleosalinity*

The Sr/Ba ratio has been widely used as an indicator of lake water paleosalinity [27]. Generally speaking, a low Sr/Ba ratio (<0.5) indicates that the oil shale was formed in fresh water sediments, while a high ratio (>1) reflects saline water. A moderate Sr/Ba ratio (0.5–1.0) in lacustrine sedimentary environments indicates brackish water, with a ratio of 1.0 referring to an arid climate [20]. The Sr/Ba ratio in Youganwo Formation oil shale of the M − 1 well (0.169–0.368; avg., 0.272) exhibited a paleosedimentary environment of fresh water, with a decreasing paleosalinity trend from the bottom to the top (Fig. 7). High-quality oil shale in the O_{y-1} sub-member was deposited with relatively higher Sr/Ba ratios. The Youganwo Formation's early tertiary transgressive events may have contributed to relatively salty lake water, and the water continued to get fresher after the transgression events [9].

3.6. *Origin of the organic matter and bioproductivity*

The kerogen type of OM in oil shale can be subdivided as follows: aquatic type (type I), mixed type (type II), and terrigenous type (type III). The mixed kerogen type can be further divided as follows: Type II_1, more aquatic OM content; Type II_2, more terrigenous OM content [26] (Jia et al., 2013b). Qin (1982) has reported that oil shale from the Youganwo Formation has high H/C ratios and low O/C ratios [30]. The H/C ratios and O/C ratios from Qin (1982), and organic geochemistry data from Cao (2015) indicate that the kerogen of the Youganwo Formation oil shale belongs to Type I and type II_1 [14,21,30]. Plots of HI *vs.* T_{max} (Fig. 8a) and S_2 *vs.* TOC (Fig. 8b) of the oil shale samples comprised both Type I and Type II_1 kerogens. Most of the samples were distributed in the Type I kerogen region in plots with relatively higher HI values and S_2 values (Fig. 8). This suggests that the primary producer of OM in Youganwo Formation oil shale included both aquatic organisms and terrigenous plants. However, the major one was aquatic organisms.

Uranium element (U) is weakly influenced by the grain size effect in reducing environments. Thus, U content is used as an indicator of the bioproductivity of lacustrine oil shales [31,32]. The U values of oil shale samples (4.39–10.50 μg/g; average: 7.10 μg/g) reflected the relatively abundant biological productivity of oil shale. However, the U value distribution from top to bottom greatly changed, indicating that bioproductivity was not stable due to the increase in nutrient input from bottom to top in the Youganwo Formation (Fig. 7).

3.7. *Sediment provenance and terrigenous detrital matter input*

The Al_2O_3/TiO_2 ratio can be used as an indicator of terrigenous detrital matter input in the study of oil shale deposited in a lacustrine environment [32,33]. A higher Al_2O_3/TiO_2 ratio indicates a decrease in detrital matter input [34]. The Al_2O_3/TiO_2 ratios of tested samples ranged from 29.97 to 38, implying a constant sediment source region from intermediate to felsic igneous rocks. Generally speaking, the Al_2O_3/TiO_2 ratios showed three vertical cycles (Fig. 7). From the bottom to the O_{y-1} sub-member, it increased and reached the highest at the end part of O_{y-1}, indicating that the detrital matter input decreased to the lowest as the water level became higher. The Al_2O_3/TiO_2 ratios then exhibited a decreasing trend from O_{y-2} to O_{y-3}, indicating the gradual growth of detrital matter input. From the previous studies of Zhou et al. (2016), the diverse sources of oil shale mainly came from the felsic source region of the upper crust, including felsic igneous rocks, sedimentary rocks and granite [35].

Terrestrial matter input promotes the bloom of aquatic organisms and increases bioproductivity by bringing terrestrial OM and nutrients to the water. However, the over input of terrestrial matter may cause a dilution effect and reduce OM abundance. During the deposition period of Youganwo Formation oil shale, the high-quality oil shale was deposited with a moderate detrital matter input (O_{y-2}), which was favourable for OM preservation.

4. Discussion

4.1. *Multiple influencing factors of oil shale organic matter enrichment*

Oil content (ω) is the most commonly used parameter in evaluating oil shale quality, and an oil content >3.5% is the standard for good industrial oil shale quality [18]. There is a positive correlation between oil content and hydrocarbon source rock evaluation parameters, including the hydrocarbon generation potential parameter of ($S_1 + S_2$) values and TOC values [2,18,26]. This phenomenon indicates that the quality of oil shale is highly controlled by the sedimentary OM enrichment mechanism. Other evaluation parameters, such as ash and sulfur content, are also influenced by the sedimentary environment [36]. Several sedimentary factors influence the OM enrichment of oil shale, including sedimentation rate, biological productivity, paleo-redox condition, paleoclimate, changes in water level, detrital matter input, the origin of OM, redox condition and water stratification [2,18,26,37–40]. Although the OM enrichment in oil shale has multiple influencing factors, it can be synthetically summarized that bioproductivity, preservation and dilution are the major conditions that control OM enrichment [32].

4.1.1. Factors influencing bioproductivity

The paleoclimate conditions of oil shale may influence bioproductivity by controlling the supply of mineral nutrients, and growth of phytoplankton and regional terrigenous plants [37]. As indicated by the geochemical investigation and sporo-pollen test, oil shale in the Youganwo Formation was deposited in a relatively warm and humid climate in the subtropical-temperate zone. A warm and humid paleoclimate was beneficial for the high bioproductivity of water. However, it is notable that the paleoclimate of the $O_{y\text{-}2}$ sub-member with higher TOC values was relatively less warm and humid than that of the $O_{y\text{-}1}$ and $O_{y\text{-}3}$ sub-members. Consequently, paleoclimate was not the dominant influencing factor of OM enrichment in Youganwo Formation oil shales.

OM type $I\text{-}II_1$ was greatly influenced by the input of aquatic organisms. During the sedimentary period of the Youganwo Formation, the lake level continued to increase from $E_{2\text{-}3}y^1$ to $E_{2\text{-}3}y^2$, and the sedimentary environment transformed from limnetic facies to deep-lake facies. This may lead to the blooms of aquatic organisms which greatly increased the bioproductivity of water. The Paq (Paq=(*n*-C23+*n*-C25)/(*n*-C23+*n*-C25+*n*-C29+*n*-C31)) distribution reported by Cao (2015) proves that the $O_{y\text{-}2}$ samples had higher Paq values (0.45–0.65) than $O_{y\text{-}1}$ samples (02.-0.45) and $O_{y\text{-}3}$ samples (0.45–0.65) [14]. It shows that the $O_{y\text{-}2}$ oil shale was deposited in an environment with higher submersible plants and planktonic algae input (Fig. 7). Furthermore, the bioproductivity is also influenced by the origin of OM [26]. However, the samples exhibited a positive correlation between TOC content and HI index (Fig. 9), indicating that OM enrichment was influenced by the origin. The values of TOC content rose as aquatic organism input increased.

4.1.2. Factors influencing preservation

Factors including paleosalinity, water stratification and redox condition have been proven to have controlling effects on the preservation conditions of OM enrichment [30,41].

According to the Sr/Ba ratio, Youganwo Formation oil shale was deposited in fresh water, and the paleosalinity decreased in turn from $O_{y\text{-}1}$ to $O_{y\text{-}3}$, which was consistent with the results reported by Cao (2015) [14]. Accordingly, water salinity stratification was classified as a weak-to-moderately strong stratification, and the degree of stratification decreased from the bottom to the top. The results from the V/(V + Ni), V/Cr ratios and Ce anomaly all demonstrate a weak oxidizing to dysoxic condition. Redox conditions prevailed during the deposition period of $O_{y\text{-}1}$, while $O_{y\text{-}2}$ and $O_{y\text{-}3}$ were deposited in reducing conditions with low water energy.

Strong water stratification and reducing environment are beneficial for organic matter preservation. However, although salinity stratification was stronger, $O_{y\text{-}1}$ had lower TOC content than $O_{y\text{-}2}$. This was because the OM enrichment of the Youganwo formation is more influenced by strong anaerobic stratification [14], rather than by weak salinity stratification. The Sr/Ba *vs.* V/Cr diagram reflects the relationship between oxic conditions and water salinity. In this diagram, the reducibility was relatively stable and the redox conditions showed an inverse correlation with the fluctuant salinity of water (Fig. 10). This indicates that with the rise of water level and the expansion of lake area, fresher water destroyed the salinity stratification. High-quality oil shales mainly accumulated in $O_{y\text{-}2}$ and $O_{y\text{-}3}$ with stronger anaerobic stratification as OM were better preserved in reducing environment and more OM in $O_{y\text{-}1}$ were decomposed.

4.1.3. Factors influencing dilution

The dilution of oil shale is influenced by detrital matter input and nutrient supply [42]. This process can be supported by evidence from the Al_2O_3/TiO_2 ratios, which indicates that the reduction of detrital minerals input and the weakening of dilution contributed to the enrichment of OM. As implied by the Al_2O_3/TiO_2 ratios, the content of Al_2O_3 and TiO_2 showed a positive correlation, indicating that Youganwo Formation oil shale had a constant sediment source region (Fig. 11). High-quality oil shale in the Youganwo Formation was deposited with the subsidence of the basin base and expansion of the lake to its biggest area [9]. It can be expected that during the deposition period of Youganwo Formation oil shale, the detrital matter input was controlled by changes in water base level and the accommodating space in system tracks.

$O_{y\text{-}1}$ was formed during the major lacustrine transgression with increasing lake level and decreasing detrital minerals input. From $O_{y\text{-}1}$ to $O_{y\text{-}2}$, the lake reached its maximum flooding surface with minimum detrital minerals input. From $O_{y\text{-}2}$ to $O_{y\text{-}3}$, the detrital minerals input showed a slightly increasing trend after the highest water level. The data show that the detrital input was low during the deposition period of $O_{y\text{-}2}$ and relatively higher during the early stage of $O_{y\text{-}1}$ and late stage of $O_{y\text{-}3}$. This implies that the relatively low

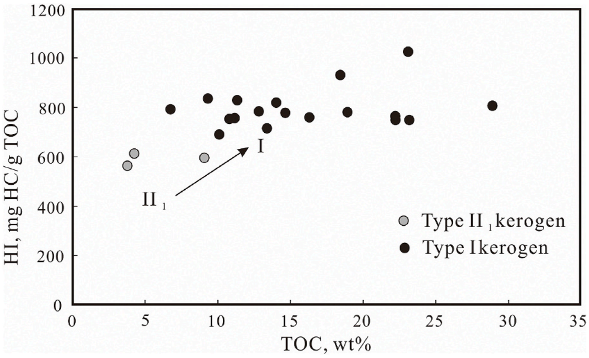

Fig. 9. Diagram of TOC *vs.* HI of Youganwo Formation oil shale.

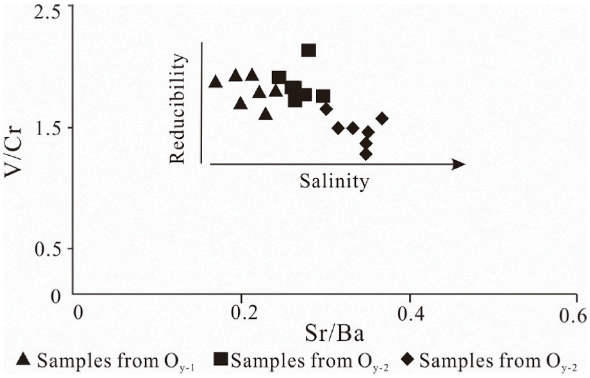

Fig. 10. Diagram of the Sr/Ba *vs.* V/Cr ratios of Youganwo Formation oil shales.

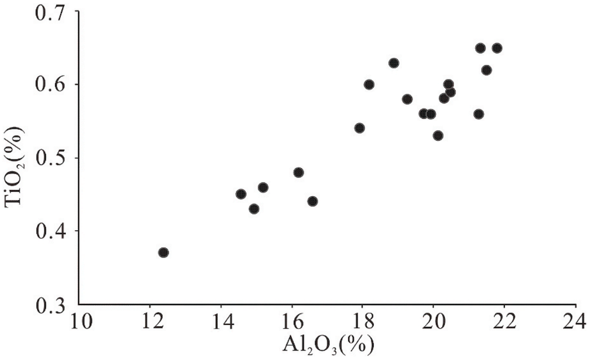

Fig. 11. The diagram of Al_2O_3 content *vs.* TiO_2 content of Youganwo Formation oil shales.

detrital input minimized the effect of OM dilution. Conversely, the effect of dilution in O_{y-1} and O_{y-3} was more evident. This can provide explanation to the fact that although with better preservation conditions, the oil shale quality of O_{y-3} is not as good as O_{y-2}.

4.2. Key influencing factors of OM enrichment

Factors that control the OM enrichment of the Youganwo Formation including bioproductivity, preservation, and dilution are studied and discussed in this section. The findings show that the higher bioproductivity, better preservation conditions, and weaker dilution together had a cumulative effect on the better quality of oil shales in O_{y-2}. However, the key influencing factors of every sub-member have not been revealed. In order to identify them as well as enhance the understanding of OM enrichment and preservation during deposition, a correlation analysis was conducted between TOC values and the indicators of paleoclimate, salinity, water stratification, oxic conditions, bioproductivity and detrital matter input (Figs. 12 and 13). The results reveal that OM enrichment controlling factors in the O_{y-1} sub-member were different from those in the O_{y-2} and O_{y-3} sub-members.

Oil shales in O_{y-1} were mainly deposited in a transitional sedimentary environment of shallow lakes and marshes (Sample M − 17, M − 18, M − 19, M − 20, M − 21), and merely two samples were from semi-deep lakes (Sample M − 15, M − 16). Samples from shallow lakes and marshes had unstable TOC values that ranged from 3.73 wt% to 28.88 wt%, while samples from semi-deep lakes had TOC values of more than 10%. In general, a reducing environment is beneficial for the accumulation of OM. However, no significant correlation was found among TOC values and the redox indicators of Ce anomaly values (Fig. 12a), V/(V + Ni) ratios (Fig. 12b) and V/Cr ratios (Fig. 12c) for the O_{y-1} samples (Fig. 12d). The impact of bioproductivity, water sanity and paleoclimate on TOC distribution was weak, according to the diagram of TOC values *vs.* U content, Sr/Ba and Sr/Cu ratios (Fig. 12e–f). An Al_2O_3/TiO_2 *vs.* TOC diagram was used to investigate the influence of the dilution conditions on OM enrichment (Fig. 12g). The Al_2O_3/TiO_2 ratios showed a weak positive correlation with TOC values, reflecting that the abundance in OM was influenced by dilution conditions to a certain extent.

The Ce anomaly values (Fig. 13a), V/(V + Ni) ratios (Fig. 13b) and V/Cr ratios (Fig. 13c) exhibited a relatively weak correlation with TOC values, indicating that the impact of paleoredox conditions on OM abundance was relatively weak during the deposition of O_{y-2} and O_{y-3} oil shale. However, the Sr/Ba ratios, Sr/Cu ratios, U content and Al_2O_3/TiO_2 ratios showed positive correlations with TOC values (Fig. 13d-g). From O_{y-2} to O_{y-3}, the lake water became fresher and the climate became more warm-humid, with more detrital matter input. In general, a more warm-humid climate is beneficial for the high productivity of water, and fresher water reflects the weakening of salinity stratification. Thus, the formation of high-quality oil shale in O_{y-2} significantly benefited from better dilution conditions and higher bioproductivity, and was influenced by the paleosalinity and paleoclimate (Fig. 13). It is noteworthy that the

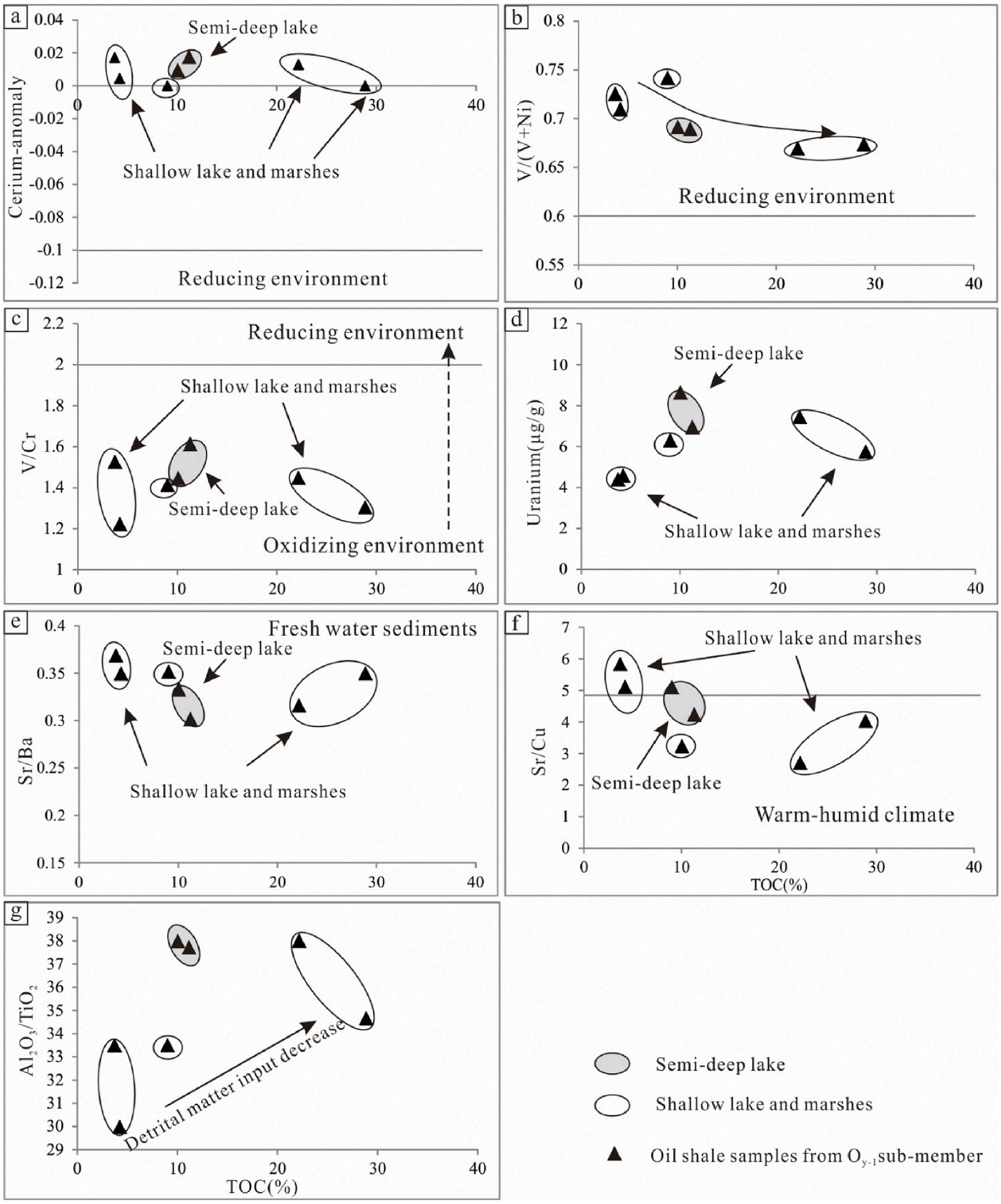

Fig. 12. Correlations between TOC values and the indicators of paleoclimate, salinity, water stratification, redox condition, bioproductivity and detrital matter input for the O_{y-1} sub-member.

preservation condition was also important since it prevented OM from aerobic decomposition. The anoxic condition contributed to the formation of good-quality oil shales in O_{y-3}, regardless of its strong dilution effect.

4.3. Organic matter enrichment mechanism

Oil shales that were deposited in various systems tracts exhibit noticeably different OM characteristics. As a result, the organic enrichment mechanisms in different system tracts varied as well. The sequence stratigraphic work established based on the data from borehole core and profile observation, logging and experimental test, and the sub-intervals divided based on the sequence stratigraphic work have also been verified by the interval differences of organic geochemistry and element geochemistry (Figs. 5 and 7). In the established sequence stratigraphic work, base level, the distribution and relationship of lithology and organic matter characteristics in different system tracts are shown in Fig. 14, which reveals the organic matter enrichment rule in the sequence stratigraphic work.

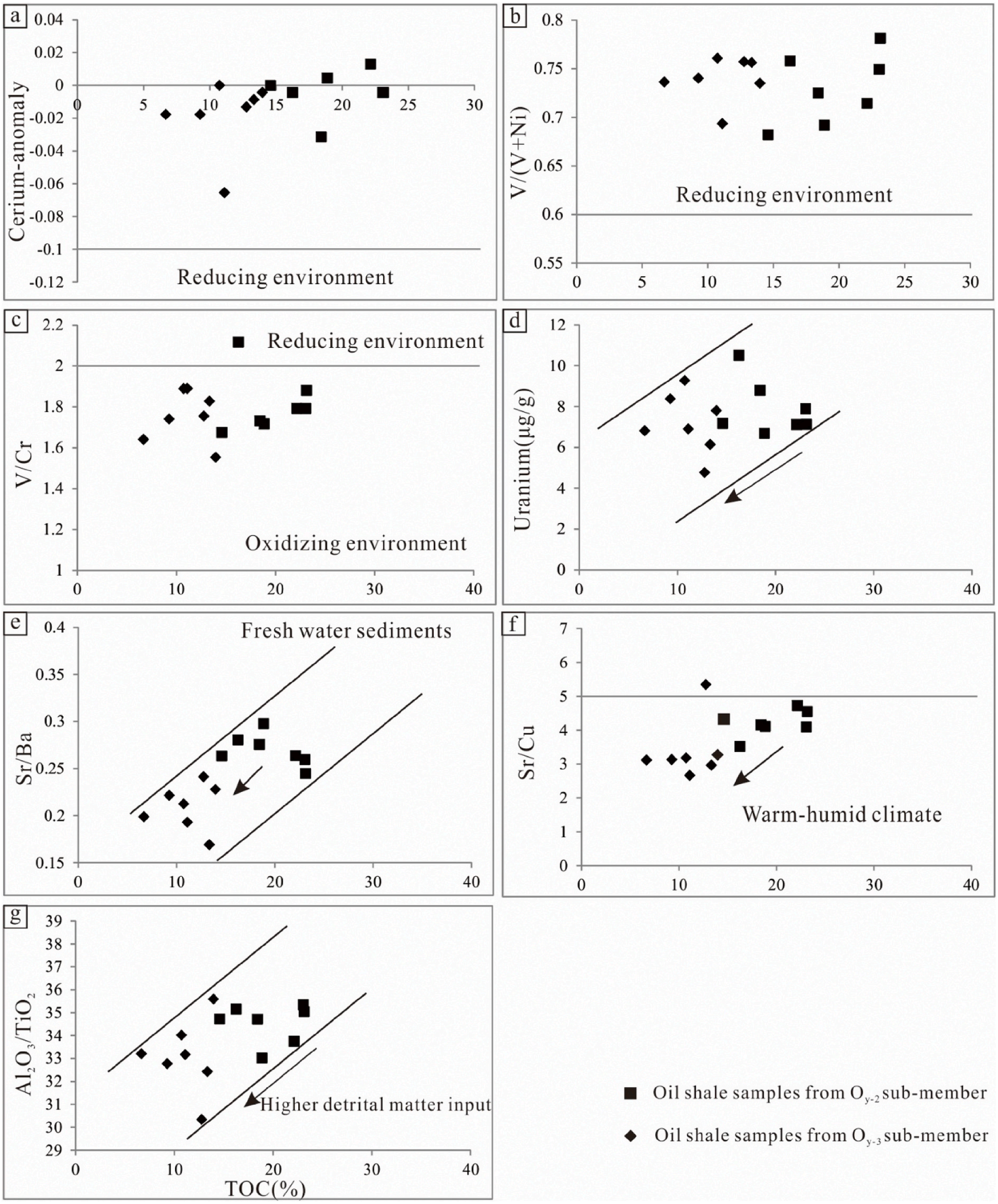

Fig. 13. Correlations between TOC values and the indicators of paleoclimate, salinity, water stratification, redox condition, bioproductivity and detrital matter input for the O_{y-2} and O_{y-3} sub-members.

High-quality lacustrine oil shales were usually formed during the time interval of HST, with a large lake area and minimum detrital minerals input [2]. As shown in Fig. 14, the high-quality oil shale of in O_{y-2} was developed in the HST. The moderate-quality oil shale of the Youganwo Formation was deposited during the TST and RST, which corresponded to the O_{y-1} and O_{y-3} sub-member. Note that the sequence applied in the figure is divided by the aforementioned basic geological work (Fig. 5). The TOC values of O_{y-1}, O_{y-2} and O_{y-3}

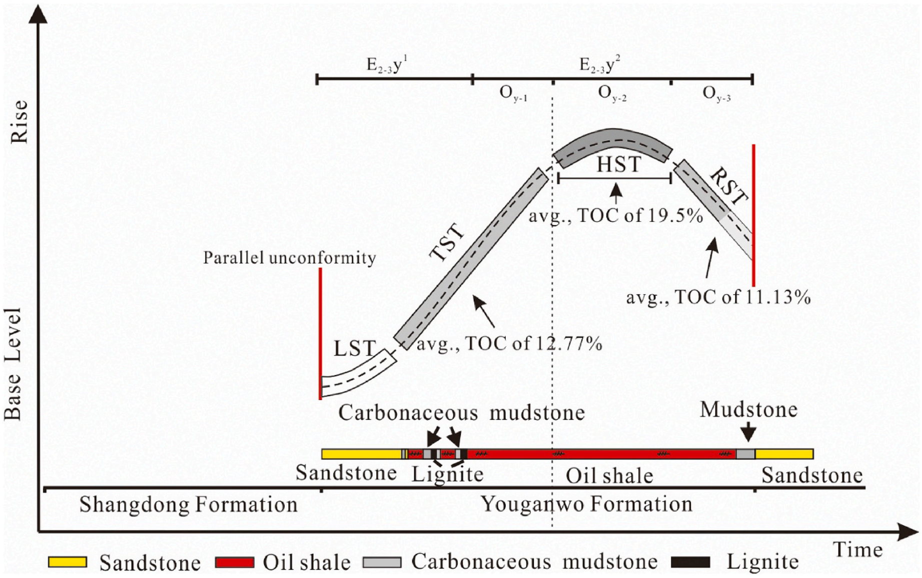

Fig. 14. Oil shale deposited in different system tracts of the Youganwo Formation.

layers in Fig. 14 are used to make comparison and reveal the impact of sequence and sedimentary environment evolution on the enrichment of organic matter.

As shown in Fig. 14, LST represents the early stage of the deposition of the Youganwo Formation. The lake base level was relatively low, and abundant detrital minerals from the delta subfacies were transported by rivers and deposited in the basin. During the deposition of TST and HST, the base level increased and reached the highest level and largest lake area. This significantly increased the accommodation space, and improved the preservation conditions of OM enrichment by water redox stratification. As the base level increased, the climate became warm and humid, which improved the bioprodutivity, and lead to the highest input of higher submersible plants and planktonic algae in the water. The effects of dilution were relatively weak when the lake reached its highest base level. High-quality oil shale was deposited under better bioproductivity, large lake area, minimum detrital minerals input and good preservation conditions during the HST. During the RST, although the lake level remained relatively stable, it slightly decreased when compared with the water level of HST. The lithology of mudstone interbedded with siltstone and sandstone of the RST also indicates that the water base level decreased. Furthermore, the detrital minerals input showed an increasing trend, resulting in stronger OM dilution and moderate quality oil shale at the RST. Another parallel unconformity between the Youganwo Formation and Huangniuling Formation was the change in sedimentary environment from a lacustrine to a delta environment, which ended the deposition of oil shale.

It is notable that some oil shale samples with higher TOC values were distributed within the O_{y-1} sub-member (TST) (Fig. 15). It resulted from the significant contribution of terrestrial OM. On the contrary, the O_{y-2} sub-member was deposited in a relatively stable deep lake environment. The difference in TOC was not as remarkable as the O_{y-1}.

The deposition model and OM enrichment mechanism of oil shale during the Youganwo Formation were established (Fig. 16). The OM enrichment mechanism was complex with an unstable water level during TST (Fig. 16a). Compared with the O_{y-1} and O_{y-3} oil shale,

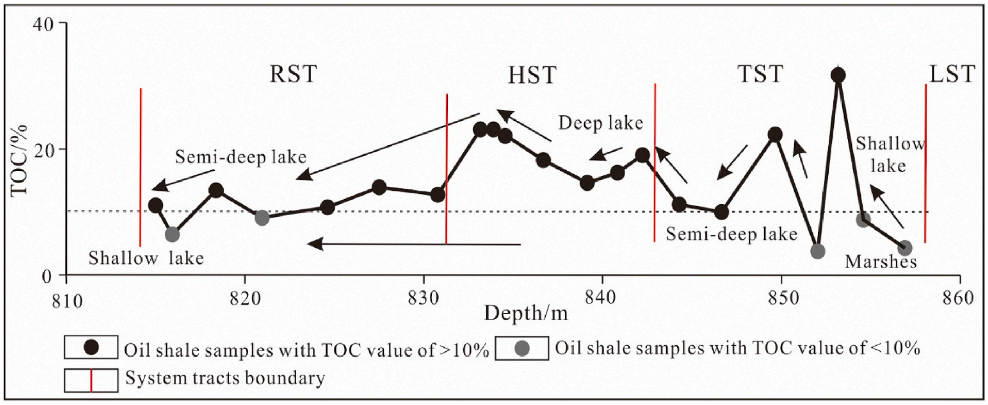

Fig. 15. Sedimentary environment, systems tracts and corresponding TOC values during the deposition of Youganwo Formation oil shale.

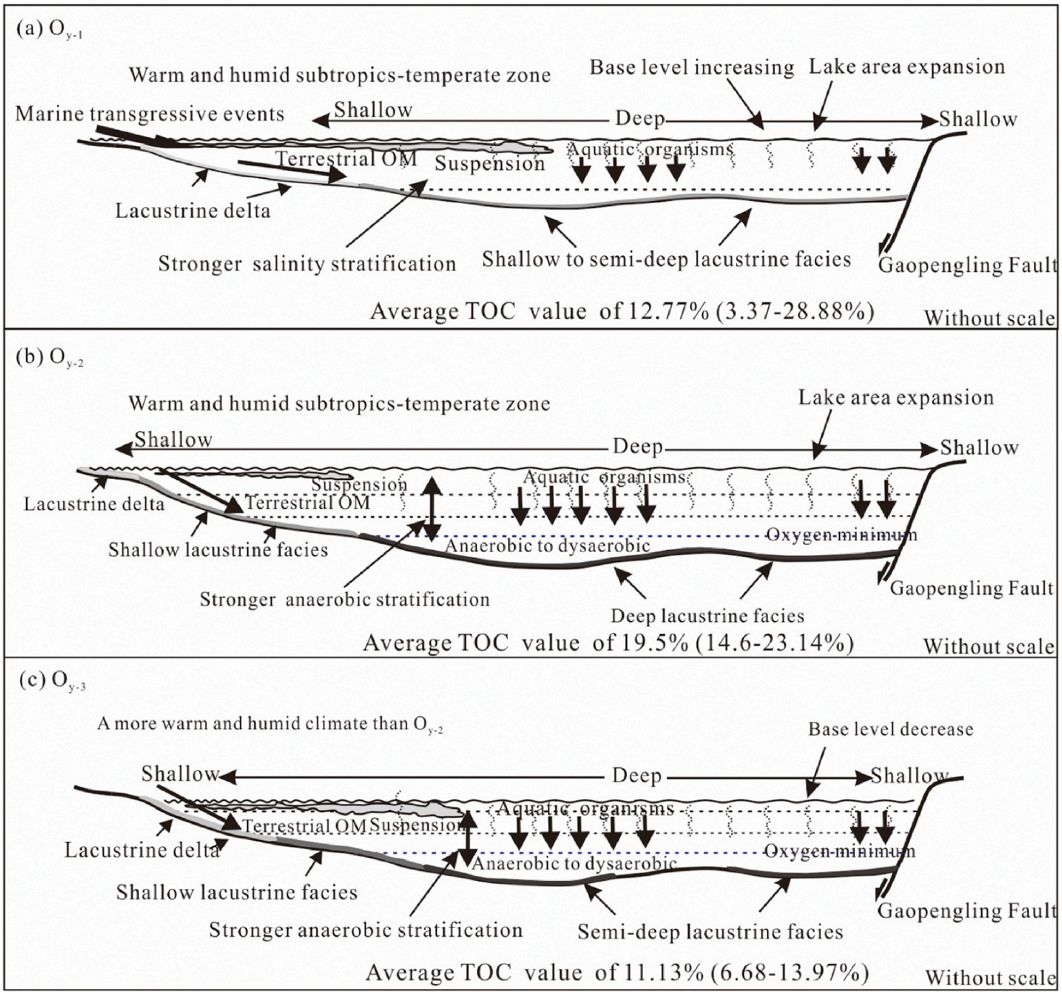

Fig. 16. The deposition model and organic matter enrichment mechanism of oil shale during the Youganwo Formation in the Maoming Basin.

the O_{y-2} oil shale was deposited under a more stable lacustrine environment with higher productivity, and better dilution and preservation conditions within the system tracks of HST (Fig. 16b). During the HST, the climate was warm and humid, which improved the bioprodutivity of water. The relatively strong anaerobic stratification and reducing bottom water conditions led to good preservation conditions. The dilution conditions of the relatively high bioproductivity of water and low input of detrital matter were favourable for the preservation of OM during the HST. After the HST, the worse dilution condition and weaker salinity stratification contributed to the low TOC values (Fig. 16c). Thus, stable and high-quality oil shale was deposited during HST within the Youganwo Formation in the Maoming basin.

5. Conclusion

A geochemical study was conducted to reconstruct the depositional paleoenvironment condition and investigate the OM enrichment mechanisms in Youganwo Formation oil shales. The TOC content of oil shale ranged within 3.73%–28.88%, the ($S_1 + S_2$) values ranged within 26.2 mg/g-277.81 mg/g, and the vitrinite reflectance values (Ro/%) ranged within 0.40%–0.65%, showing good oil-generating potential and a marginally immature maturity phase. The OM belonged to type I and II_1 kerogens. The oil shales deposited during $E_{2-3}y^2$ were subdivided into O_{y-1}, O_{y-2} and O_{y-3} sub-members.

The bulk and elemental geochemical analysis have revealed the multiple influencing factors of OM accumulation during the deposition of oil shales based on the comprehensive study of bioproductivity, preservation and dilution which are reflected by paleoclimate, redox condition, water salinity, water stratification, bioproductivity and detrital matter input. However, with the evolution of the lacustrine sedimentary environment, the influencing factors of OM enrichment changed from O_{y-1} to O_{y-3}.

(1) Youganwo Formation oil shale was mainly deposited in semi-deep to deep-lake environments during $E_{2-3}y^2$, with relatively warm and humid paleoclimate in the subtropical-temperate zone.

(2) From $O_{y\text{-}1}$ to $O_{y\text{-}3}$, the redox condition of water showed a tendency towards a more reducing environment, according to V/Cr ratios, Ce anomaly values and V/(V + Ni) ratios, which reflect the enhancement of water redox stratification.
(3) The predominant primary producer of OM was aquatic organisms. The U values reflect that oil shale had a relatively abundant, but unstable, biological productivity.
(4) The Sr/Ba ratios exhibit a decreasing trend from $O_{y\text{-}1}$ to $O_{y\text{-}3}$, reflecting that the paleosalinity of water became lower and weakened the salinity stratification.
(5) The analysis of trace elements and rare earth elements indicates that sediment provenance was continental crust originated, with a constant sediment source region from intermediate to felsic igneous rocks. The detrital matter input was enhanced during $O_{y\text{-}1}$ and decreased from $O_{y\text{-}2}$ to $O_{y\text{-}3}$.

During $E_{2\text{-}3}y^2$, multiple controlling factors influenced the OM enrichment in system tracts. The dilution condition was proven to be a constant influencing factor of OM enrichment from TST, HST, to RST. However, the OM abundance and variation in HST and RST were also influenced by productivity, paleosalinity and paleoclimate. High-quality oil shales were deposited during the HST, with increasing accommodation space, improved preservation conditions, higher water bioprodutivity and minimum detrital minerals input in $O_{y\text{-}2}$. From HST to RST ($O_{y\text{-}2}$ to $O_{y\text{-}3}$), higher detrital matter input and fresher water led to lower TOC values, even with better climate and more reducing water conditions. The combined effect of bioproductivity, preservation and dilution controlled the OM enrichment and influenced the quality of oil shale.

In this study, the organic matter enrichment mechanism of Youganwo Formation oil shale in the Maoming Basin has been comprehensively analyzed and discussed from the aspects of sedimentary environment, organic geochemistry, elemental geochemistry and stratigraphic classification. The differences between different sub-members and their main influencing factors have been clarified, which can provide scientific guidance for further geological exploration and theory research of oil shale resources in the Maoming Basin. Additionally, the investigation of the organic matter enrichment mechanism of Youganwo Formation oil shale in the Maoming Basin can also provide a reference for oil shale endowment study in other basins. In the future, significant global climatic and geological events during the Youganwo Formation oil shale will be incorporated into the analysis. For example, the study of the response mechanism of terrestrial sedimentary organic matter in Eocene-Oligocene Transition (EOT) climate change can facilitate the understanding of the controlling factors of the organic matter enrichment mechanism in a larger geological context. And the obtained geological laws will have better applicability in providing references for geological exploration in other regions and basins.

Author contribution statement

Di-Fei Zhao: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Wrote the paper.

Ying-Hai Guo; Geoff Wang: Conceived and designed the experiments; Contributed reagents, materials, analysis tools or data.

Xue-Qing Zhou; Jia-Ming Zhang: Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data.

Yuan-Yuan Zhou: Performed the experiments; Contributed reagents, materials, analysis tools or data.

Guang-Ying Ren: Analyzed and interpreted the data.

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Data availability statement

Data included in article/supp. material/referenced in article.

Declaration of interest's statement

The authors declare no conflict of interest.

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