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Original Paper

Evolutions of sedimentary facies and palaeoenvironment and their controls on the development of source rocks in continental margin basins: A case study from the Qiongdongnan Basin, South China Sea

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A B S T R A C T

Hydrocarbon resources in the Qiongdongnan Basin have become an important exploration target in China. However, the development of high-quality source rocks in this basin, especially in its deep-water areas, are still not fully understood. In this study, evolutions of sedimentary facies and palaeoenvironment and their influences on the development of source rocks in diverse tectonic regions of the Qiongdongnan Basin were investigated. The results show that during the Oligocene and to Miocene periods, the sedimentary environment of this basin progressively varied from a semi-closed gulf to an open marine environment, which resulted in significant differences in palaeoenvironmental conditions of the water column for various tectonic regions of the basin. In shallow-water areas, the palaeoproductivity and reducibility successively decrease, and the hydrodynamic intensity gradually increases for the water columns of the Yacheng, Lingshui, and Sanya-Meishan strata. In deep-water areas, the water column of the Yacheng and Lingshui strata has a higher palaeoproductivity and a weaker hydrodynamic intensity than that of the Sanya-Meishan strata, while the reducibility gradually increases for the water columns of the Yacheng, Lingshui, and Sanya-Meishan strata. In general, the palaeoenvironmental conditions of the water column are the most favorable to the development of the Yacheng organic-rich source rocks. Meanwhile, the Miocene marine source rocks in the deep-water areas of the Qiongdongnan Basin may also have a certain hydrocarbon potential. The differences in the development models of source rocks in various tectonic regions of continental margin basins should be fully evaluated in the exploration and development of hydrocarbons.

1. Introduction

The Qiongdongnan Basin located on the northern continental margin of the South China Sea has abundant natural gas and certain amounts of other hydrocarbon resources, including light oils, condensates, and gas hydrates, and it is an important offshore exploration target area in China (Gong and Li, 1997, 2004; Huang et al., 2016; Wei et al., 2021; Wang et al., 2022). Previous explorations in the Qiongdongnan Basin have mainly concentrated on shallow-water areas with water depths less than 300 m in the northwest of the basin, while deep-water areas with water depths larger than 300 m, which account for more than 70% of the basin, are poorly explored. In recent years, explorations of the Qiongdongnan Basin have progressively expanded from shallow-water areas to deep-water areas, and many giant commercial gas fields, including

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LS25, LS18, and LS17 in the Central Canyon and YL8 in the Songnan Uplift, have been discovered (Wang and He, 2003; Zhang et al., 2007, 2019; Zhu, 2007; Wang et al., 2011; Huang et al., 2012, 2017; Xie, 2014; Yang et al., 2019; Guo et al., 2021; You et al., 2021; Zhu et al., 2021). This significant progress in deep-water areas exhibits great hydrocarbon potential for exploration. However, the Qiongdongnan Basin is a multiple petroleum system with several sets of organic-rich source rocks. The development and distribution of the main source rocks are obviously diverse in different tectonic regions, which results in complex enrichment and distribution of hydrocarbon resources in the basin (Wang and He, 2003; He et al., 2006; Xie and Gao, 2020). For example, a variety of hydrocarbon resources, including biogenetic gases, oil-type and coal-type thermogenic gases, light oils, condensates, and gas hydrates, concomitantly occur in this basin, and the sources and maturities of these hydrocarbons vary significantly (Huang et al., 2016; Ding et al., 2018; Zhang et al., 2019; Liang et al., 2020; Wei et al., 2021). Therefore, determinations of the main source rocks in different tectonic regions are important for the next exploration and development of the Qiongdongnan Basin.

In the Cenozoic, with the increasing expansion of transgression, the sedimentary environment of the Qiongdongnan Basin transformed from lacustrine via marine-terrestrial transitional to marine environments from the Eocene to the Miocene. In addition, the palaeogeographical features of the basin are complex due to its location on the edge of the continental shelf, and the sedimentary facies changes obviously in different tectonic regions (Zhu et al., 2008; Zhang et al., 2021), which largely controls the organic provenances of sedimentary strata. In the Qiongdongnan Basin, the organic sources of water column were contributed from both aquatic and terrestrial organisms, while the contributions of the two types of organic sources were diverse for separate tectonic regions. The regions close to the continent have more contributions from terrestrial organisms, while the regions far away from the continent have more contributions from aquatic organisms (Li and Zhang, 2017; Wu et al., 2018).

In the Qiongdongnan Basin, early explorations in shallow-water areas have well documented that the sources of the YC13 giant gas field were the Yacheng Formation marine-terrestrial transitional source rocks (Zhou et al., 2003; Hu et al., 2005; Xiao et al., 2006; Xie and Tong, 2011). Based on this recognition, however, little significant progress has been achieved during subsequent explorations in deep-water areas (Xie, 2014; Zhang et al., 2017, 2019). With the increases in the exploration extent of the Qiongdongnan Basin, increasing geological evidence indicates that the distribution of high-quality source rocks in deep-water areas may be significantly different from that in shallow-water areas (Liang et al., 2019; Zhang et al., 2020). For example, hydrocarbon resources from the Eocene source rocks may have little accumulation potential in deep-water areas because the hydrocarbon generation periods of source rocks were much earlier than the formation periods of effective reservoirs, according to the geological conditions in the adjacent western Pearl River Mouth Basin (Cheng et al., 2013b). However, the Miocene marine source rocks probably have hydrocarbon potential in deep-water areas because some source rocks have accessed the main stage of oil generation resulting from their deep burial depth (Gao et al., 2018; Zhu et al., 2018).

In the petroliferous basins of the South China Sea, the palaeoenvironmental conditions, including palaeoproductivity, redox conditions, and hydrodynamic intensity, significantly impact the development of source rocks (Li, 2015; Yu et al., 2016; Li and Zhang, 2017; Wu et al., 2018; He, 2020; Xu et al., 2021). For example, the palaeoenvironmental conditions were better for the development of the Eocene Liushagang source rocks in the Beibuwan Basin than the contemporaneous Wenchang source rocks in the Pearl River Mouth Basin (Zhu, 2007; Zhao et al., 2021), which resulted in the former source rocks having larger hydrocarbon generation yields than the latter source rocks. The palaeoproductivity of water column largely affects the accumulation of organic matter, and water columns with high palaeoproductivity are beneficial for enriching organic matter and forming organic-rich source rocks (Kryc et al., 2003; Shen et al., 2011; Zhao et al., 2021; Ding et al., 2022). The redox conditions and hydrodynamic intensity of water column have remarkable effects on the preservation of organic matter. An anoxic environment benefits the preservation of organic matter, while an oxygen-rich environment disadvantages the preservation of organic matter (Li et al., 2021). Water columns with a weak hydrodynamic intensity benefit the accumulation of organic matter, while water columns with a robust hydrodynamic intensity disadvantage the enrichment of organic matter (Canfield, 1994; Katz, 2005; Schieber et al., 2007; Macquaker et al., 2010; Zonneveld et al., 2010; Bohacs et al., 2014; Ou et al., 2022). The provenance, enrichment, and preservation of organic matter in water column are diverse in different sedimentary facies and palaeoenvironmental conditions, and thus, the sedimentary facies and palaeoenvironment largely determine the development and distribution of organic-rich source rocks in the Qiongdongnan Basin (Li and Zhang, 2017; Wu et al., 2018; He, 2020; Zhao et al., 2021).

Because of the current lack of sufficient geological information and typical samples, there are few comprehensive studies concerning the controls of the sedimentary facies and palaeoenvironmental conditions on the development and distribution of source rocks in different tectonic regions and strata of the Qiongdongnan Basin, especially in deep-water areas, which controls the development of hydrocarbon resources in this basin. Based on the latest exploration data and geological source rock samples, the evolution of palaeoenvironment and sedimentary facies and their influences on the development of the main source rocks in different tectonic regions of the Qiongdongnan Basin were investigated in this study. This study intends to provide new geological evidence to reveal the distribution of high-quality source rocks in different tectonic regions of the Qiongdongnan Basin, which has practical significance for the next exploration and development of this basin.

2. Geological background

The Qiongdongnan Basin is a Cenozoic faulted basin in the northern continental shelf of the South China Sea, and it covers 8×10^4 km^2 and exhibits a NE–SW distribution (Fig. 1). This basin is tectonically separated by several depressions and uplifts and is generally divided into the Northern Depression, Middle Uplift, and Central Depression blocks from north to south (Fig. 1). The Northern Depression contains the Yabei, Songdong, and Songxi sags; the Middle Uplift is subdivided into the Yacheng, Songtao, Lingshui, and Yanan uplifts; and the Central Depression includes the Yanan, Ledong, Lingshui, and Baodao sags (Fig. 1). The Qiongdongnan Basin underwent rift and the post-rift tectonic stages (Gong and Li, 1997; 2004; Zhu et al., 2008). During the rift stage, independent depressions were formed due to the activities of the controlling faults in this basin. The Eocene Lingtou Formation lacustrine strata, the early Oligocene Yacheng Formation marine-terrestrial transitional strata, and the late Oligocene Lingshui Formation marine strata were successively deposited at this stage (Fig. 2). At the post-rift stage, the independence of various depressions became weak, and a continental slope-shelf system gradually formed in this basin. The Miocene Sanya, Meishan, and Huangliu Formation neritic and bathyal strata and the Pliocene and Quaternary extremely thick abyssal strata were deposited at this stage (Fig. 2). The Qiongdongnan Basin is a typical multiple petroleum system with several sets of source-reservoir-cap assemblages, and the petroleum geological

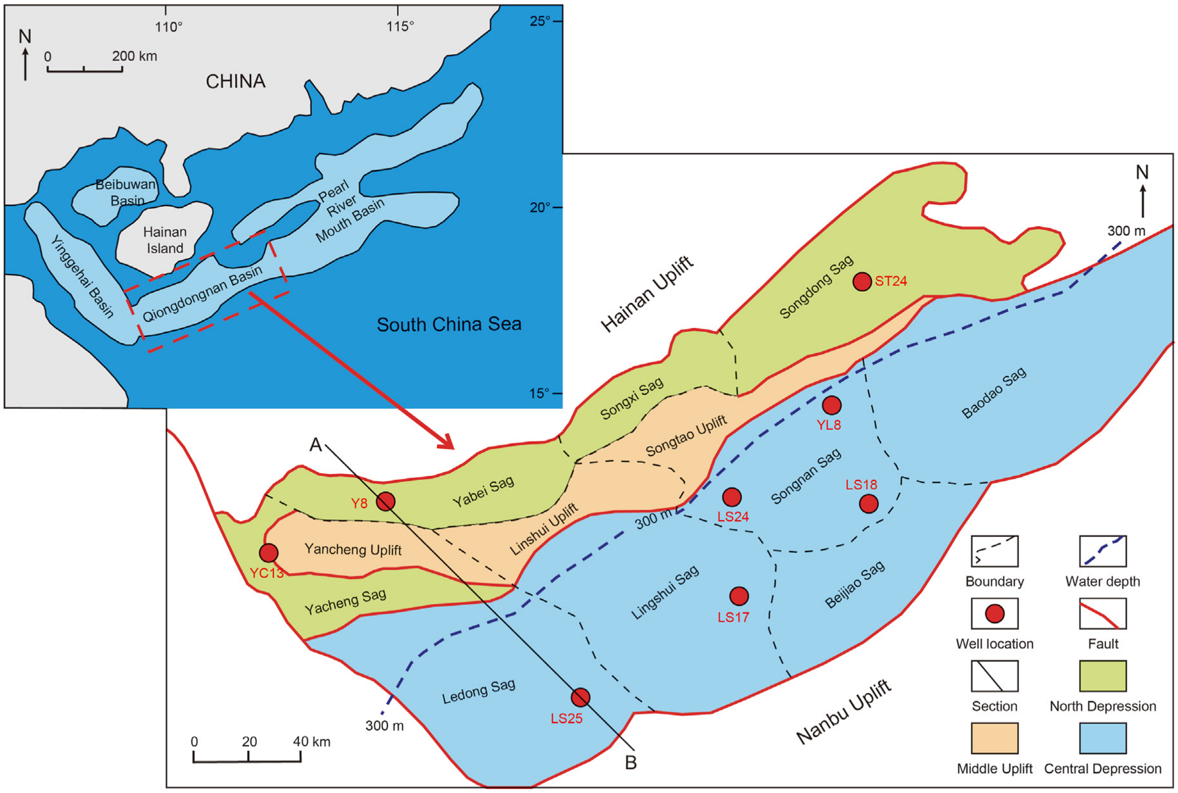

Fig. 1. Schematic map showing the location and structural divisions of the Qiongdongnan Basin (modified after Huang et al., 2016).

conditions of this basin indicate that it has great hydrocarbon potential (Gong and Li, 2004; Zhu, 2007; Xie et al., 2020; Zhang et al., 2021).

3. Samples and experiments

3.1. Samples

In this study, a total of 85 source rock samples were derived from the Yacheng, Lingshui, and Meishan-Sanya Formations in different tectonic regions of the Qiongdongnan Basin, including 23 samples collected from wells Y8 and ST24 in the Northern Depression, 20 samples collected from well YC13 in the Middle Uplift, and 42 samples collected from well LS4 in the Central Depression (Fig. 1).

3.2. Experiments

3.2.1. Rock-eval and TOC analysis

The mudstone samples were ground to less than 80 mesh (i.e., < 180 μm), and the mudstone powder samples were used to analyze their pyrolytic parameters by a Rock-Eval VI instrument. The carbonate compositions of the powder samples were removed by treatment with diluted hydrochloric acids. After the samples were dried, the TOC content was analyzed by an elemental analyzer instrument (Eario El Cube).

3.2.2. Vitrinite reflectance measurement

A microphotometer (3Y-DMR) coupled with a microscope (Leica) was used to measure the vitrinite reflectance (R_o, %) of the source rock samples because these samples generally contain vitrinite macerals. The excitation light from a high-pressure mercury vapor lamp was limited to 420–490 nm by an excitation filter. The vitrinite reflectance values were measured under a 50/1.0 oil immersion objective. A standard yttrium aluminum garnet sample was used to calibrate the instrument before vitrinite reflectance measurements. More than 30 individual vitrinite particles in each sample were measured, and their average value was used in this study.

3.2.3. Analysis of major and trace elements

The mudstone samples with a grain size of less than 180 μm (200 mesh) were successively digested by HNO_3, $HClO_4$, and HF in Teflon cups. The digested samples were dried at 200 °C for 12 h. After these samples were weighed, they were mixed with $Li_2B_4O_7$ flux at a ratio of 1:10 in a platinum crucible. Then, the mixtures were melted at a temperature of 1050 °C, and the molten sample was removed into a platinum mold and cooled to form a melt sheet. The major and trace elements were analyzed by an X-ray fluorescence spectrometer (Philips PW2404 XRF) and an inductively coupled plasma mass spectrometer (Thermo Scientific Element XR ICP–MS). The measurement was performed twice for each sample with an analytical precision of 3% and the averaged value was used.

3.2.4. Determination of sedimentary facies

In this study, diagrams of sedimentary facies are established by the following procedures. First, the thicknesses of various strata are obtained through stratigraphic interpretations based on seismic data. In combination with the regional tectonic background, the paleogeomorphology is restored to determine the overall sedimentary environment and water depth conditions. Then, the sedimentary subfacies are determined by analyzing the seismic reflection characteristics in detail. Finally, the sedimentary microfacies are identified by lithologic, paleontological, and geochemical integrated analysis of typical core samples in different strata and structural regions.

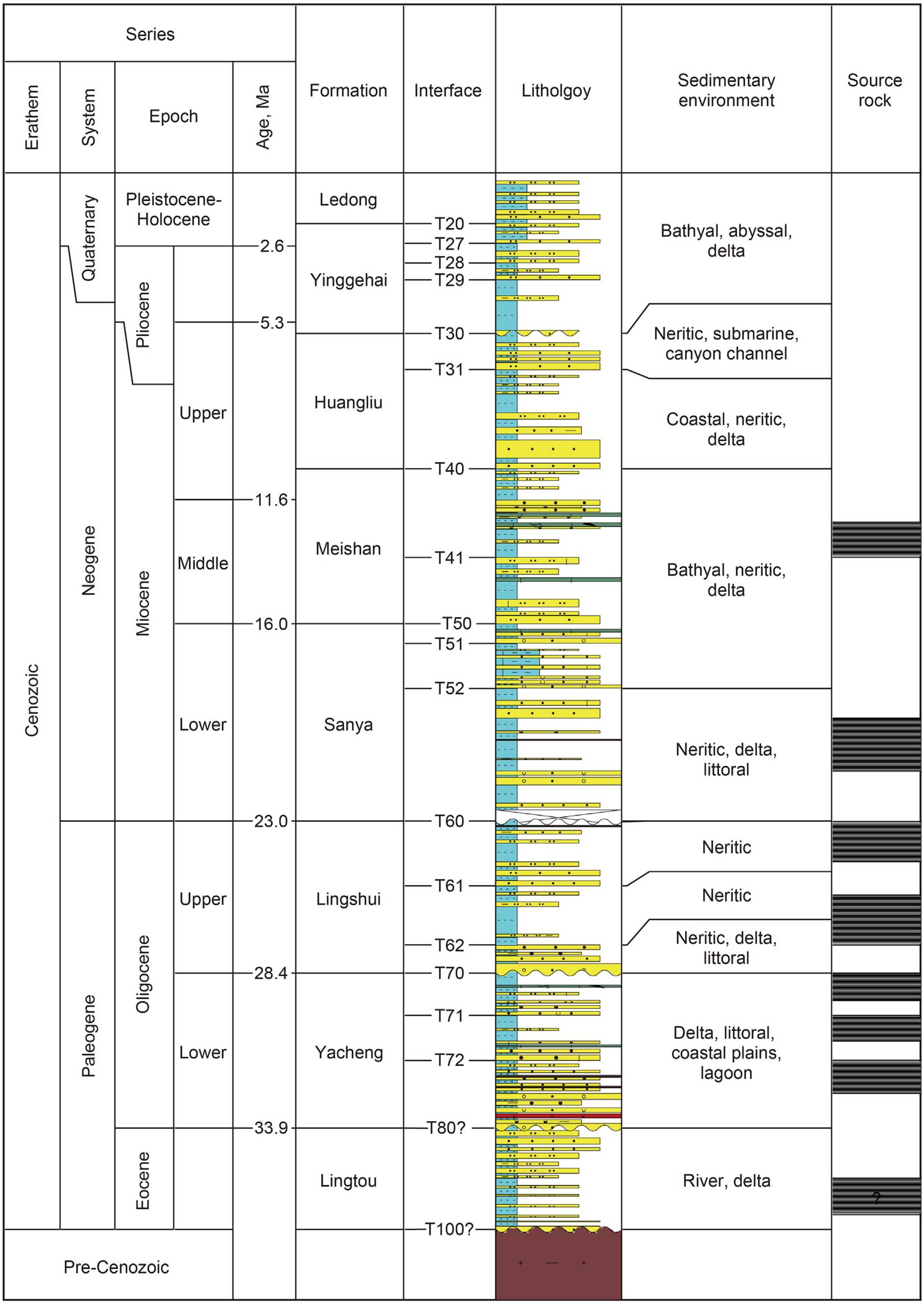

Fig. 2. Stratigraphic column of the Qiongdongnan Basin (modified after Zhu et al., 2021).

4. Results and discussion

4.1. Evolutions of sedimentary strata

The sedimentary environments of the Qiongdongnan Basin gradually transformed from lacustrine via marine-terrestrial transitional to marine environments from the Eocene to the Miocene, and several sets of source rocks were developed during these stages. Because of the diverse tectonic framework and paleogeography, the sedimentary facies of different tectonic regions varied markedly, which influenced the development of organic-rich source rocks in this basin (Figs. 3 and 4).

4.1.1. The early Oligocene Yacheng strata

In the early Oligocene, the revolving extrusion of the Indosinian plate caused the sinistral strike-slip movement of the Honghe Fault system. The western Qiongdongnan Basin split along the No. 1 fault, and thus, the closer to the tail of the strike-slip fault, the greater the extent of regional extension (Fig. 4a). This tectonic movement resulted in the Qiongdongnan Basin exhibiting a trumpet-shaped morphology toward the southeast, with the extents of extension in the west and the north being larger than those in the east and the south, respectively (Gong and Li, 2004; Zhu, 2007; Zhang et al., 2021). The divisions among various depressions were weakened, and the scope of uplifts were smaller compared to the Eocene depositional periods. The early Oligocene strata presented flat terrain in different tectonic regions, and their sedimentary thickness progressively increased from north to south, with thicknesses of 300–2000 m, 500–2000 m, and 500–3000 m for the Northern Depression, Middle Uplift, and Central Depression blocks, respectively (Fig. 3). The early Oligocene strata in the Northern Depression were mainly littoral and coastal plains, and the central sedimentary areas of the sags in this block were mainly lagoon facies. This set of strata was mainly coastal plains and delta facies in the Middle Uplift, and it was primarily littoral and neritic facies in the Central Depression and its sedimentary central areas, respectively (Fig. 4a).

4.1.2. The late Oligocene Lingshui strata

Although the tectonic framework of the Qiongdongan Basin in the late Oligocene was similar to that in the early Oligocene, the intensity of tectonic activity progressively weakened during this period (Gong and Li, 2004; Zhang et al., 2021). Therefore, most of the faults in this basin terminated during the sedimentary period of the Upper Oligocene strata, and divisions among various depressions were further weakened (Fig. 3). The upper Oligocene strata had a greater sedimentary thickness than the Lower Oligocene strata, and the sedimentary thickness progressively increased from north to south, with thicknesses of 1000–2000 m, 500–2500 m, and 2000–4000 m for the Northern Depression, Middle Uplift, and Central Depression blocks, respectively (Fig. 3). The early Oligocene strata were mainly neritic and delta facies in the Northern Depression, neritic and littoral facies in the Middle Uplift, and neritic facies in the Central Depression (Fig. 4b).

4.1.3. The Miocene strata

In the Miocene, the Qiongdongnan Basin entered the post-rift stage and experienced very weak tectonic movements. During this period, a continental shelf-slope system gradually formed along the northern edges of the South China Sea, and the transgression scope progressively expanded in this basin, resulting in a passive continental margin basin (Gong and Li, 1997). The boundaries of various depressions in this basin disappeared, and the depressions were well connected. The sedimentary thickness of the Miocene strata progressively increased from north to south. It was 500–1000 m in the Northern Depression and Middle Uplift and 1500–5000 m in the Central Depression (Fig. 3). The Miocene strata were mainly littoral and delta facies in the Northern Depression, neritic and littoral facies in the Middle Uplift, and abyssal facies in the Central Depression. Meanwhile, submarine fans were also developed in some regions of the Central Depression (Fig. 4c).

4.2. Palaeoenvironmental conditions of water column

The palaeoclimate can affect the provenances and enrichments of organisms in source rocks by influencing vegetation growth and the scale of sea runoff (Hautevelle et al., 2006; Ghosh and Sarkar, 2010; Clift et al., 2015). In the Qiongdongnan Basin, however, the palaeoclimate slightly transformed from humid to half-humid from the Oligocene to the Miocene (Ding et al., 2022), which had little influence on the development of source rocks. Palaeoenvironmental conditions significantly affect the provenance, enrichment, and preservation of organic matter in water columns, which subsequently determine the development of organic-rich source rocks (Sageman et al., 2003; Hao et al., 2011; L'ezin et al., 2013; Alalade, 2016; Yan et al., 2019; Cai et al., 2022). Based on elemental geochemical analysis of source rocks from various tectonic regions of the Qiongdongnan Basin, the differences in palaeoenvironmental conditions during the development of source rocks in diverse strata and tectonic regions are investigated in this study, and the Al/Ti ratio, V/(V + Ni) ratio, and Zr/Rb ratio are applied to

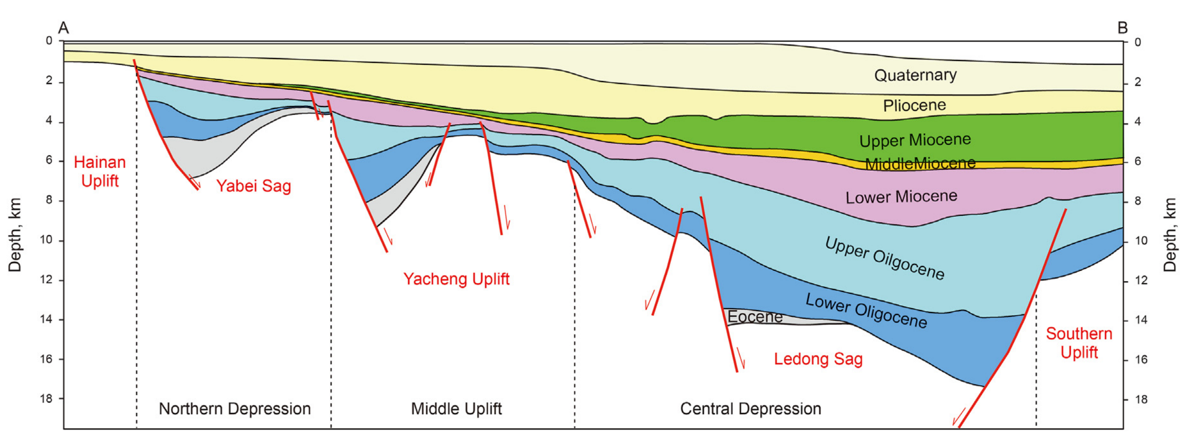

Fig. 3. Geologic section crossing the Northern Depression, Middle Uplift and Central Depression blocks, showing the differences in the development of sedimentary strata in various tectonic regions of the Qiongdongnan Basin. The location of section (A–B) is shown in Fig. 1.

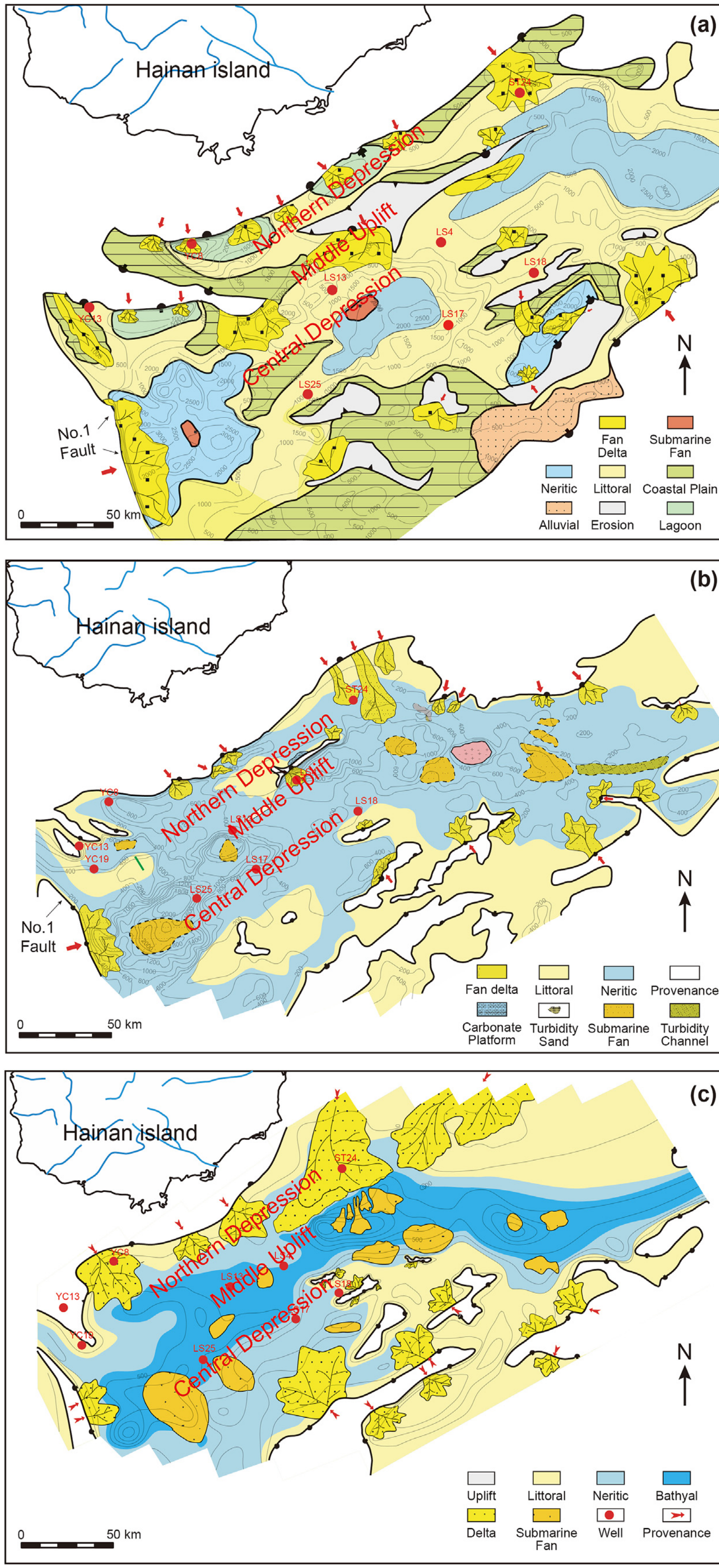

Fig. 4. Distributions of the early Oligocene marine-terrestrial transitional: (a) the late Oligocene semi-close marine, (b) the Miocene marine, and (c) sedimentary strata in different tectonic regions of the Qiongdongnan Basin.

characterize the palaeoproductivity, redox conditions, and hydrodynamic intensity of water column in the Qiongdongnan Basin, respectively (Table 1; Figs. 5 and 6).

4.2.1. Palaeoproductivity

During the diagenetic stage, the accumulation of organic matter in source rocks was largely influenced by the palaeoproductivity of water column (Pirrung et al., 2008; Shen et al., 2011; Zhao et al., 2021). The Al/Ti ratio is an effective index for characterizing the palaeoproductivity of the sedimentary water column (Yarincik et al., 2000; Kryc et al., 2003; Ren et al., 2005; Xiong and Xiao, 2011; Li and Zhang, 2017). In the Qiongdongnan Basin, the Al/Ti values of source rocks are positively correlated with the TOC contents (Fig. 6a), which indicates that the water column with a greater Al/Ti ratio has a higher palaeoproductivity and is more favorable for the enrichment of organic matter (Murray and Leinen, 1996). However, it is worth noting that the positive correlation between the Al/Ti ratio and TOC content exhibits a significant positive correlation for the Lingshui source rocks in the Northern Depression and the Yacheng and Lingshui source rocks in the Middle Uplift, a weak positive correlation for the Yacheng source rocks in the Northern Depression and the Yacheng and Lingshui source rocks in the Central Depression, and no obvious correlations for the Meishan-Sanya source rocks in the whole basin (Fig. 6a). The difference in the correlation may be mainly because the source rocks in various strata and tectonic areas have diverse types of organic provenances.

The average Al/Ti values of the Yacheng, Lingshui, and Sanya-Meishan source rocks in the Qiongdongnan Basin successively decreased from 40.42 via 22.85 to 21.14 in the Northern Depression block and from 43.28 via 32.30 to 20.16 in the Middle Uplift block (Table 1 and Fig. 5a), which indicates that the palaeoproductivity of water column progressively decreased from the early Oligocene through the late Oligocene to the Miocene for the two blocks. In the Central Depression, the average Al/Ti values of the Yacheng and Lingshui source rocks are 72.76 and 87.73, respectively, which are obviously greater than the 20.87 of the Sanya-Meishan source rocks (Table 1 and Fig. 5a). In addition, the average Al/Ti values of the Yacheng and Lingshui source rocks successively increased for the Northern Depression, the Middle Uplift, and the Central Depression blocks because the contributions of aquatic organisms progressively increased for the Northern Depression, Middle Uplift, and Central Depression blocks with the increasing extent of transgression from the north to the south of the Qiongdongnan Basin (Li and Zhang, 2017). However, the average Al/Ti ratio is similar for the Sanya-Meishan source rocks in the three tectonic blocks of the basin (Table 1 and Fig. 5a), which is mainly because the bulk Qiongdongnan Basin entered an open marine environment and the differences in sedimentary environment varied slightly for various tectonic regions during the Miocene (Fig. 5a). In general, the water column had high palaeoproductivity for the Yacheng source rocks in both shallow and deep water areas and for the Lingshui source rocks in deep-water areas (Fig. 5a).

4.2.2. Redox conditions

The redox conditions of water column significantly affect enrichments in organic matter in source rocks. Anoxic environments are conducive to enrichments of organic matter, while oxygen-rich environments are adverse to enrichments of organic matter. The sensitive trace elements V and Ni are generally applied to illustrate the redox conditions of water column in the Qiongdongnan Basin. The water column progressively turns to a more reductive environment with increasing V/(V + Ni) values. This indicates an oxygen-poor environment that is weakly stratified when the V/(V + Ni) values are less than 0.60, and it shows a strongly reductive environment that is significantly stratified when the V/(V + Ni) values are larger than 0.84 (Hatch and Leventhal, 1994; Morford and Emerson, 1999; Piper and Perkins, 2004; Tribovillard et al., 2006). In the Qiongdongnan Basin, the V/(V + Ni) values of source rocks exhibit a significant positive correlation with the TOC contents (Table 1 and Fig. 6b), which indicates the controls of redox conditions on the enrichment of organic matter.

The average V/(V + Ni) values of the Yacheng, Lingshui, and Sanya-Meishan source rocks slightly decrease from 0.85 via 0.77 to 0.75 in the Northern Depression and significantly decrease from 0.96 via 0.82 to 0.44 in the Middle Uplift (Table 1 and Fig. 5b), which indicates that the water column progressively changed to increasing oxygen-rich sedimentary environments from the early Oligocene through the late Oligocene to the Miocene in the two tectonic blocks. However, the average V/(V + Ni) value increases from 0.62 via 0.69 to 0.77 for the Yacheng, Lingshui, and Sanya-Meishan source rocks in the Central Depression (Table 1 and Fig. 5b), which indicates that the water column progressively turns to a more reductive environment in this block from the early Oligocene through the late Oligocene to the Miocene. The redox conditions of water column shows different evolutions for the shallow and deep water areas during the same period (Fig. 5b), which is probably attributed to the differences in sedimentary facies. In general, the redox conditions of water column are more favorable for the development of the Yacheng organic-rich source rocks in shallow-water areas and the Sanya-Meishan organic-rich source rocks in deep-water areas (Fig. 5b).

4.2.3. Hydrodynamic intensity

The hydrodynamic intensity of water column significantly affects the preservation of organic matter during the development of source rocks, and the weaker the hydrodynamic intensity is, the more advantageous the preservation of organic matter (Schieber et al., 2007; Bohacs et al., 2014; Makeen et al., 2015; Ou et al., 2022). The hydrodynamic intensity of water column is generally characterized by the granularity of sedimentary strata. Fine-grained materials, such as clays, generally indicate a calm water column, while coarse-grained sandstones commonly reflect a turbulent water column. The element Zr is a typical terrestrial inert element and is prone to enrichment in coarse-grained sedimentary rocks. Sedimentary strata with a high Zr content generally indicate a high-energy depositional environment with strong hydrodynamic intensity. The element Rb is commonly in a silicate state and is prone to enrichment in fine-grained particles. Sedimentary strata with a high Rb content generally indicate a low-energy depositional environment with a weak hydrodynamic intensity (Dypvik and Harris, 2001). Therefore, the Zr/Rb ratio is an effective index to reflect the hydrodynamic intensity of water column. A water column with a high Zr/Rb ratio generally indicates a robust hydrodynamic intensity, which is adverse to the preservation of its organic matter. In the Qiongdongnan Basin, the Zr/Rb ratio of the source rocks exhibits an apparent negative correlation with the TOC content (Table 1 and Fig. 6c), which clearly shows that the preservations of organic matter in source rocks are significantly influenced by the hydrodynamic intensity.

The average Zr/Rb values of the Yacheng, Lingshui, and Sanya-Meishan source rocks in the Qiongdongnan Basin successively increased from 0.57 via 0.99 to 1.06 in the Northern Depression and from 0.50 via 0.83 to 1.82 in the Middle Uplift (Table 1 and Fig. 5c), which indicates that the hydrodynamic intensity of water column during the development of source rocks gradually increased from the early Oligocene through the late Oligocene to the Miocene. The changes in hydrodynamic intensity mainly occurred because the sedimentary environments of the Qiongdongnan Basin progressively transformed from a semi-closed gulf environment to an

Table 1
Geological and geochemical characteristics and palaeoenvironmental index of source rocks in different tectonic regions and strata of the Qiongdongnan Basin.

Sample information					Palaeoenvironmental index[a]			Source rocks				
Tectonic regions	Epoch	Formation	Well	Burial depth, mbsl	Al/Ti	Zr/Rb	V/(V + Ni)	TOC, %	R_o, %	HI, mg/g TOC	T_{max}, °C	S1+S2, mg/g
Northern Depression	Oligocene	Yacheng	Y8	3617.4	26.88	0.58	0.76	0.54	0.68	–	–	0.00
				3716.5	27.04	0.70	0.77	0.51	0.73	–	–	0.00
				3793.2	32.25	0.62	0.86	0.51	0.75	–	–	0.00
				3819.1	40.91	0.50	0.89	0.51	0.81	–	–	0.00
				3988.3	68.16	0.27	0.88	0.71	0.8	–	449	0.94
				4142.2	47.28	0.72	0.92	0.8	0.78	–	–	0.01
		Lingshui	ST24	2768.0	27.58	1.05	0.78	2.4	0.54	112	432	3.17
				2822.0	20.76	1.12	0.85	1.38	0.58	45	434	0.67
				2928.0	19.55	0.74	0.89	0.74	0.62	241	434	2.75
				3050.0	21.31	1.16	0.79	0.95	0.61	120	436	1.28
				3182.0	19.47	–	0.83	0.57	0.63	237	435	2.17
				3296.0	19.26	1.42	0.80	0.66	0.62	270	435	2.17
				3364.0	25.90	1.11	0.67	0.43	0.63	263	332	2.67
				3458.0	17.77	–	0.81	0.8	0.71	230	310	2.67
				3548.0	21.88	0.68	0.79	0.71	0.72	38	431	0.31
				3634.0	31.95	0.83	0.65	0.48	0.82	144	433	0.84
				3686.0	25.86	0.75	0.64	0.39	0.88	231	358	2.30
	Miocene	Sanya-Meishan		2175.0	23.20	0.69	0.79	0.462	0.51	49	436	0.72
				2235.0	25.25	0.68	0.80	0.301	0.53	375	430	0.46
				2325.0	19.76	0.87	0.81	0.56	0.5	371	432	1.21
				2408.0	20.22	2.07	0.59	0.497	0.51	297	433	0.63
				2468.0	17.57	1.24	0.75	0.336	0.52	253	433	1.27
				2648.0	20.83	0.79	0.78	0.273	0.54	42	428	1.28
Middle Uplift	Oligocene	Yacheng	Y13	4027.4	39.24	0.33	0.97	4.8	1.14	72	459	3.55
				4042.7	48.72	0.42	0.97	5.01	1.25	125	459	7.66
				4061.0	51.18	0.41	0.97	3.84	1.25	194	458	8.81
				4082.3	44.78	0.38	0.97	4.38	1.13	124	461	6.58
				4094.5	54.20	0.42	0.97	4.38	1.23	91	459	4.74
				4128.0	31.95	0.65	0.94	1.96	1.19	72	459	2.01
				4146.3	32.88	0.85	0.91	3.59	1.24	100	460	4.46
		Lingshui		3643.3	19.20	1.86	0.69	0.21	0.77	19	440	0.10
				3649.4	27.27	0.50	0.93	0.33	0.78	33	464	0.17
				3676.8	21.15	1.58	0.58	0.32	1.09	31	459	0.19
				3682.9	21.13	1.15	0.64	0.48	0.97	75	435	0.69
				3692.1	22.87	0.68	0.81	0.42	0.89	46	438	0.32
				3859.8	45.22	0.33	0.96	1.11	0.97	65	452	0.98
				3881.1	51.04	0.25	0.96	5.25	1	201	449	11.10
				3908.5	50.56	0.29	0.97	3.27	1.06	69	450	3.55
	Miocene	Sanya-Meishan		3429.9	20.49	1.60	0.08	0.33	0.63	89	–	0.04
				3509.1	24.14	1.51	0.34	0.32	0.51	55	429	0.17
				3527.4	20.58	1.30	0.54	0.37	0.77	78	429	0.35
				3606.7	17.83	2.43	0.65	0.34	0.78	242	429	0.22
				3612.8	17.78	2.27	0.59	0.34	1.09	82	447	0.15
Central Depression	Oligocene	Yacheng	LS4	4738.0	84.12	0.24	0.42	1.11	1.03	39	435	1.61
				4766.0	162.32	0.14	0.84	1.25	1.04	54	432	1.56
				4800.0	41.20	1.02	0.51	1.35	0.98	169	431	1.83
				4826.0	41.89	1.12	0.58	1.51	1.05	58	434	1.26
				4854.0	31.03	0.42	0.63	1.11	1.11	110	432	1.27
				4882.0	101.57	0.61	0.72	1.3	1.12	103	437	1.41
				4928.0	47.20	0.61	0.66	0.94	1.12	132	432	1.34
		Lingshui		4444.0	45.38	0.56	0.72	0.52	0.52	163	436	2.76
				4456.0	55.47	0.61	0.81	0.69	0.53	253	436	1.79
				4470.0	74.12	0.35	0.83	0.73	0.52	161	430	1.44
				4502.0	109.25	0.25	0.68	0.82	0.57	119	428	1.22
				4526.0	107.54	0.18	0.69	0.67	0.59	94	433	1.20
				4550.0	64.18	0.24	0.61	0.75	0.59	130	434	1.46
				4580.0	131.22	0.17	0.54	0.76	0.55	25	430	1.21
				4598.0	98.53	0.20	0.54	0.77	0.59	42	432	1.43
				4624.0	106.09	0.22	0.66	1.07	0.6	40	–	1.25
				4654.0	95.82	0.26	0.71	1.06	0.7	97	–	1.27
				4680.0	79.50	0.89	0.90	1.1	0.74	60	437	1.61
				4708.0	85.67	0.28	0.67	0.79	0.91	84	432	1.56
	Miocene	Sanya		3330.0	18.98	0.99	0.75	0.56	0.59	304	428	1.80
				3630.0	18.74	1.03	0.78	0.59	0.59	390	431	2.43
				3685.0	19.50	0.90	0.77	0.61	0.55	330	431	2.18
				3795.0	18.60	0.86	0.68	0.82	0.59	177	428	1.62
				3805.0	20.24	0.95	0.80	0.74	0.6	385	430	3.06

(continued on next page)

Table 1 (*continued*)

Sample information					Palaeoenvironmental index[a]			Source rocks				
Tectonic regions	Epoch	Formation	Well	Burial depth, mbsl	Al/Ti	Zr/Rb	V/(V + Ni)	TOC, %	R_o, %	HI, mg/g TOC	T_{max}, °C	S1+S2, mg/g
				3870.0	20.20	0.91	0.78	0.53	0.61	491	429	2.80
				3960.0	20.12	0.96	0.78	0.69	0.64	425	430	3.13
				3980.0	21.22	0.94	0.77	0.75	0.65	265	431	2.08
				4105.0	20.89	0.90	0.81	0.59	0.72	573	430	3.57
				4130.0	21.05	0.92	0.77	0.82	0.67	184	432	1.63
				4160.0	20.61	0.98	0.79	0.68	0.75	515	431	3.76
				4195.0	20.06	0.94	0.79	0.87	0.78	211	427	1.98
				4255.0	20.35	1.02	0.78	0.79	0.43	332	426	2.99
				4282.0	22.74	0.88	0.79	0.61	0.43	366	430	2.45
				4355.0	25.79	0.76	0.78	0.52	0.85	550	427	3.19
				4390.0	29.13	0.80	0.77	0.73	1.03	233	428	1.82
				4410.0	26.58	0.59	0.79	0.67	1.04	366	438	2.76
		Meishan		2810.0	19.10	1.00	0.75	0.49	0.54	210	417	1.10
				3000.0	19.18	1.13	0.80	0.59	0.57	327	423	2.03
				3050.0	19.91	1.13	0.81	0.62	0.6	302	424	2.05
				3095.0	17.08	0.98	0.76	0.51	0.6	461	421	2.54
				3205.0	20.61	1.39	0.79	0.5	0.64	316	425	1.71
				3280.0	19.30	1.11	0.75	0.57	0.64	333	425	2.05

[a] The original data are provided in appendix A.

open-marine environment with the increase in transgression (Figs. 3 and 4). However, the average Zr/Rb values are 0.59, 0.35, and 0.96 for the Yacheng, Lingshui, and Sanya-Meishan source rocks in the Central Depression, respectively, and exhibit no obvious tendency (Table 1 and Fig. 5c). In the Qiongdongnan Basin, the hydrodynamic intensity of the Sanya-Meishan source rocks is greater than those of the other two sets of source rocks (Table 1 and Fig. 5c), probably due to the strong wave effects of the open marine environment.

4.3. Development models of source rocks

According to the sedimentary facies and palaeoenvironment of the Qiongdongnan Basin, the development models of the main source rocks, including the Yacheng marine-terrestrial transitional, the Lingshui semi-closed marine, and the Miocene open-marine source rocks, were determined in diverse tectonic regions of the basin. These source rocks presented remarkably diverse development characteristics in different tectonic regions (Fig. 7).

In the early Oligocene, the development of the Yacheng source rock was primarily developed in littoral, coastal plains and delta facies in the Qiongdongnan Basin, and the organic matter of water column was mainly sourced from terrestrial organisms (Fig. 4a). Because this set of strata was primarily coastal plain facies in shallow-water areas and neritic facies in deep-water areas (Fig. 4a), aquatic organisms have more contributions to the Yacheng source rocks in deep-water areas than in shallow-water areas. The water column had a high palaeoproductivity and a strongly reductive environment during this period, which was beneficial for the enrichment of organic matter. Meanwhile, the hydrodynamic intensity of water column was weak during this period, which benefited the preservation of organic matter (Fig. 7a). Therefore, in the Qiongdongnan Basin, the early Oligocene Yacheng organic-rich source rocks were well developed in both shallow and deep water areas.

In the late Oligocene, the development of the Lingshui source rock was primarily in neritic and littoral facies in the Qiongdongnan Basin, and their organic matter was primarily sourced from terrestrial and aquatic organisms of water column (Fig. 7b). In shallow-water areas, compared to the early Oligocene water column, the late Oligocene water column had a lower palaeoproductivity, a weaker reductive environment, and a stronger hydrodynamic intensity, which was disadvantageous to the accumulation and enrichment of organic matter (Fig. 7b). In deep-water areas, however, the late Oligocene water column had a higher palaeoproductivity, a more strongly reductive environment, and a weaker hydrodynamic intensity than the early Oligocene water column, which was advantageous to the enrichment of organic matter (Fig. 7b). In general, the late Oligocene Lingshui organic-rich source rocks were fairly well developed in the deep-water areas of the Qiongdongnan Basin.

In the Miocene, the development of the Meishan-Sanya source rocks was largely in bathyal, neritic, and delta facies in the Qiongdongnan Basin (Fig. 7c). Although the organic matter of water column in the Qiongdongnan Basin was also sourced from terrestrial and aquatic organisms, the contributions of aquatic sources significantly increased during this period. In shallow-water areas, compared to the Oligocene water column, the Miocene water column had a lower palaeoproductivity, a weaker reductive environment, and a stronger hydrodynamic intensity, which was significantly adverse to the accumulation and enrichment of organic matter (Fig. 7c). In the deep-water areas, the palaeoproductivity and hydrodynamic intensity of the Meishan-Sanya water column are also more disadvantageous than those of the Oligocene water column, but the reductive environment is more reductive for the Meishan-Sanya column in these areas (Fig. 7c). In general, the Miocene source rocks are poorly developed in the Qiongdongnan Basin.

The seismic sections of the Eocene strata in the Qiongdongnan Basin exhibit continuous, low-frequency and strong reflections, which are similar to those of the Eocene Wenchang Formation lacustrine strata in the adjacent western Pearl River Mouth Basin (Zhang et al., 2017; Zhu et al., 2021). Therefore, a set of high-quality Eocene lacustrine source rocks may have developed in this basin. The sandstone extracts and the concomitant condensate oils in gas fields in some regions of the Qiogndongnan Basin, such as the Northern Depression, generally contain the diagnostic biomarkers of C_{30}-4 methylsteranes for the Eocene deep lacustrine source rocks (Huang et al., 2003; Cheng et al., 2013a, 2015), which further indicates the presence of Eocene source rocks in this basin. However, the development model of this formation source rock was not investigated in this study because few typical Eocene source rock samples have been drilled in this basin.

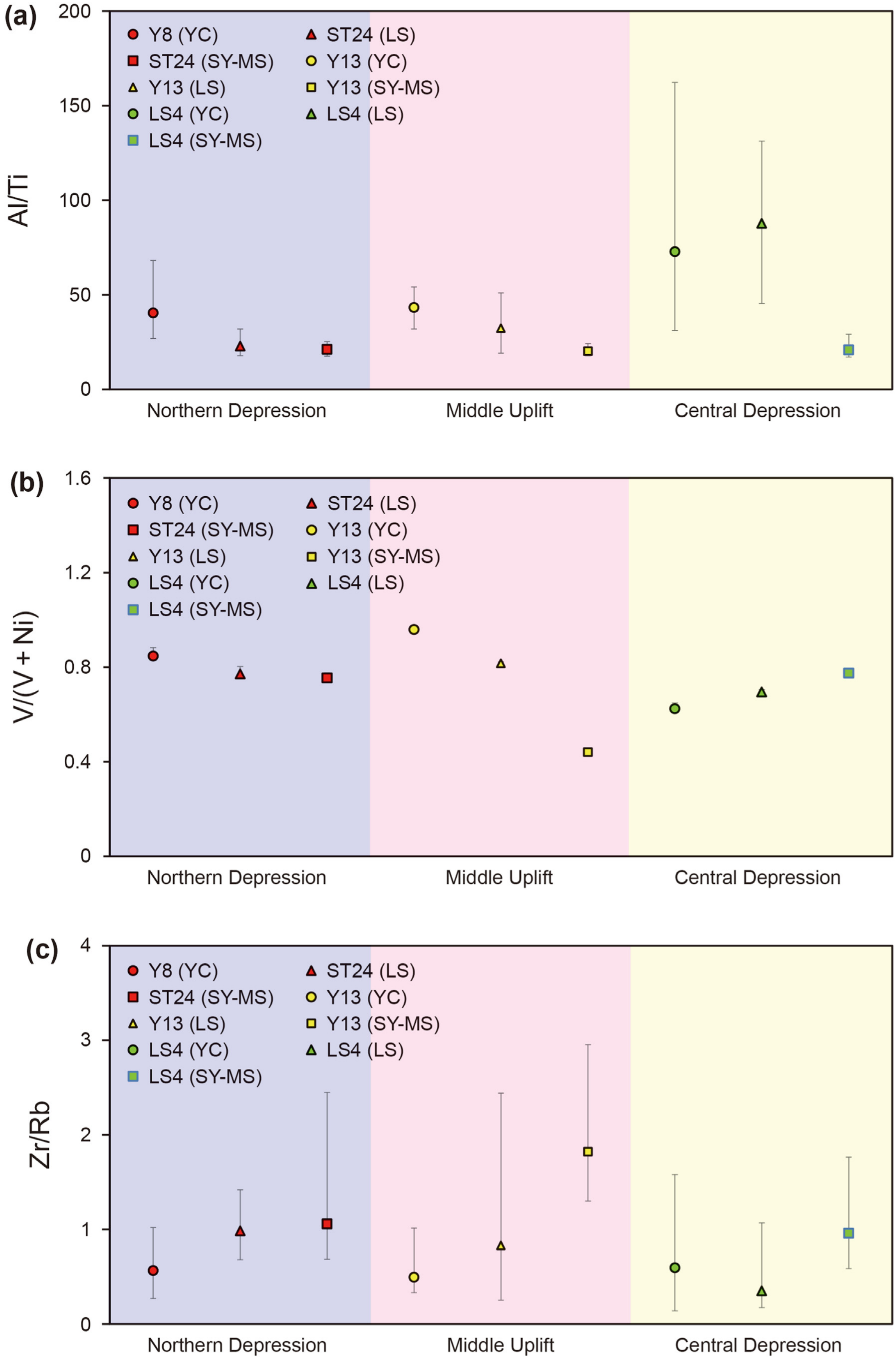

Fig. 5. The Al/Ti (a), V/(V + Ni) (b), and Zr/Rb (c) values for the source rocks in different strata and tectonic regions of the Qiongdongnan Basin.

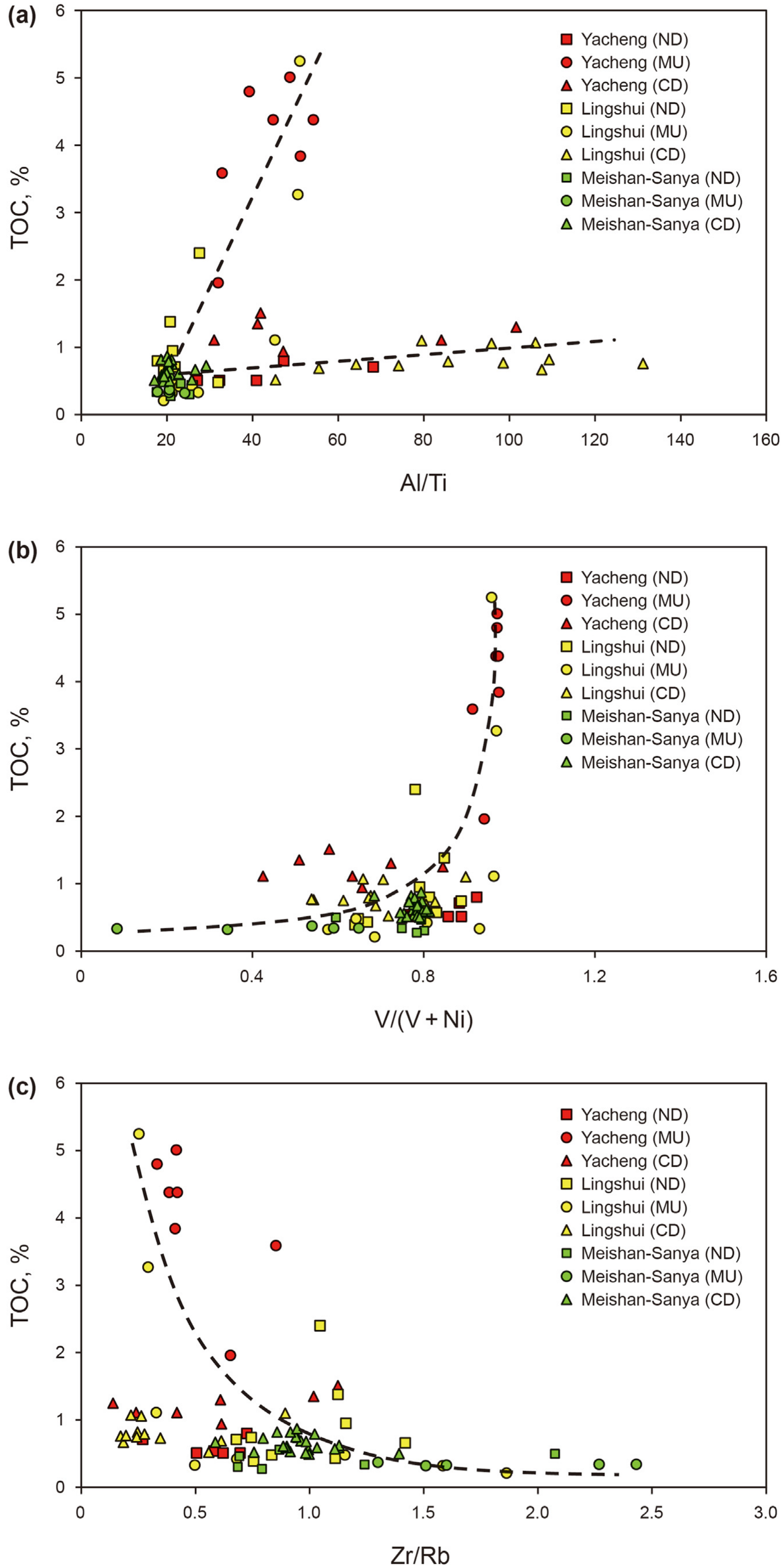

Fig. 6. Correlation of the TOC contents with the Al/Ti (a), V/(V + Ni) (b), and Zr/Rb (c) values for the source rocks in different strata and tectonic regions of the Qiongdongnan Basin.

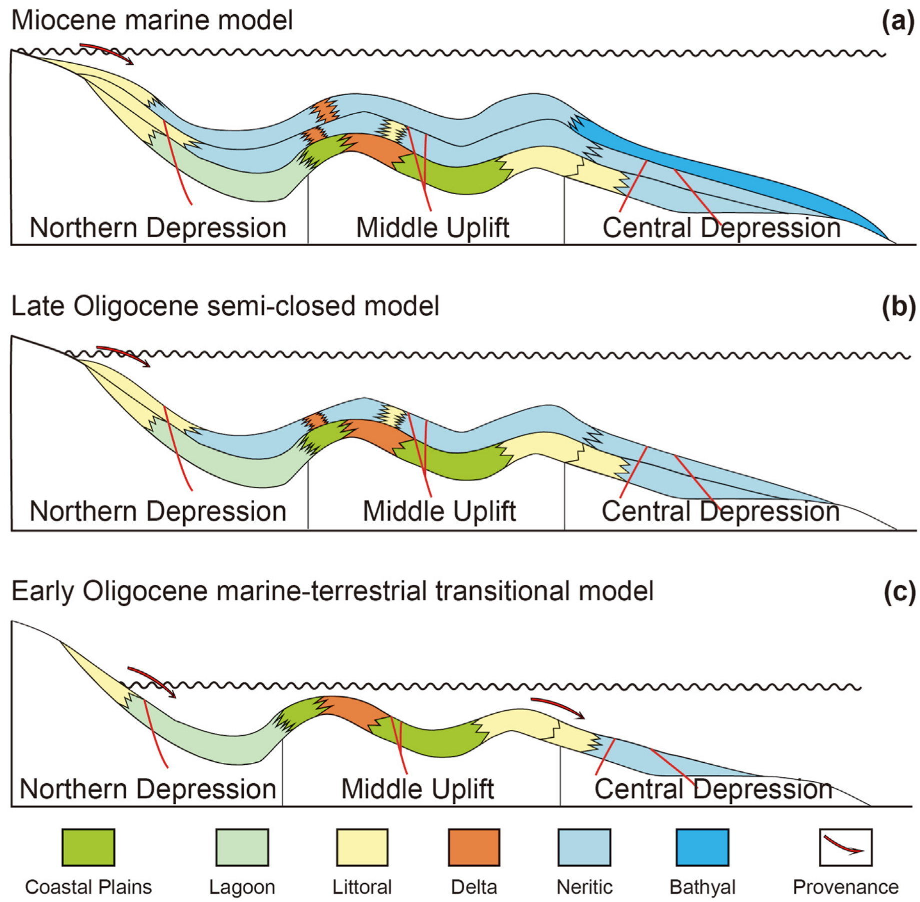

Fig. 7. Schematic maps showing the development models of the main source rocks in different tectonic regions of the Qiongdongnan Basin. (a) Early Oligocene marine-terrestrial transitional model; (b) late Oligocene semi-closed marine model; (c) Miocene marine model.

4.4. Controls on the development of source rocks

The palaeoenvironmental conditions were diverse in different periods and tectonic regions in the Qiongdongnan Basin, which largely affected the provenance, enrichment, and preservation of organic matter during the development of source rocks and further controlled the geological and geochemical properties and the hydrocarbon potential of source rocks. The difference in the development of the main source rocks in various blocks of this basin was revealed in this study (Table 1 and Figs. 8 and 9).

In the early Oligocene, the Yacheng source rocks were generally developed in littoral and coastal plain facies in the Qiongdongnan Basin, and they had moderate thicknesses with coal and carbonaceous mudstone intervals. The organic matter was primarily contributed by terrestrial organisms to the Yacheng source rocks, and thus, the Yacheng source rocks generally have type II_b-III kerogen and low HI values (Table 1, Fig. 8). The TOC content of this set of source rocks successively decreases in the Northern Depression, Middle Uplift, and Central Depression blocks, with average TOC contents of 3.99%, 1.22%, and 0.60%, respectively (Fig. 9a). The thermal maturity of this set of source rocks successively decreases in the Northern Depression, Middle Uplift, and Central Depression blocks, with average R_o values of 1.20%, 1.06%, and 0.76%, respectively (Fig. 9b). Because effective reservoirs developed in the deep-water areas before the Yacheng source rocks accessed the main gas generation stages, most of the gas resources from the Yacheng source rocks were reserved. In general, the Yacheng source rocks have large hydrocarbon generation potential in the Qiongdongnan Basin and are the main source rocks of the YC13 giant gas field (Hu et al., 2005; Xie and Tong, 2011).

The late Oligocene Lingshui source rocks were thick and basically developed in neritic and littoral facies. The organic matter was contributed by terrestrial and aquatic organisms to the Lingshui source rocks. Similar to the Yacheng source rocks, the Lingshui source rocks also have type II_b-III kerogen and low HI values (Table 1, Fig. 8). The average TOC contents of the Lingshui source rocks are 1.42%, 0.86, and 0.81% in the Northern Depression, Middle Uplift, and Central Depression blocks, respectively, and the Lingshui source rocks in the Middle Uplift exhibit a larger range of TOC contents than those in the other two blocks (Fig. 9a). The thermal maturity of the Lingshui samples is lower than that of the Yacheng samples, and it successively decreases in the Middle Uplift, Northern Depression, and Central Depression blocks, with average R_o values of 0.94%, 0.67%, and 0.62%, respectively (Fig. 9b). In general, the Lingshui source rocks exhibit a large hydrocarbon potential in the Middle Uplift.

The Miocene Sanya-Meishan source rocks had thick thicknesses and were mainly developed in bathyal and neritic facies. The organic matter of the Sanya-Meishan source rocks is mainly contributed by aquatic organisms, and thus, the Sanya-Meishan source rocks generally have type II_a-II_b kerogen and high HI values (Table 1, Fig. 8). The TOC content and thermal maturity of the Sanya-Meishan marine source rocks are smaller than those of the Yacheng and Lingshui marine-terrestrial transitional source rocks. The average TOC contents of the Sanya-Meishan source rocks are 0.40%, 0.34%, and 0.45% in the Northern Depression, Middle Uplift,

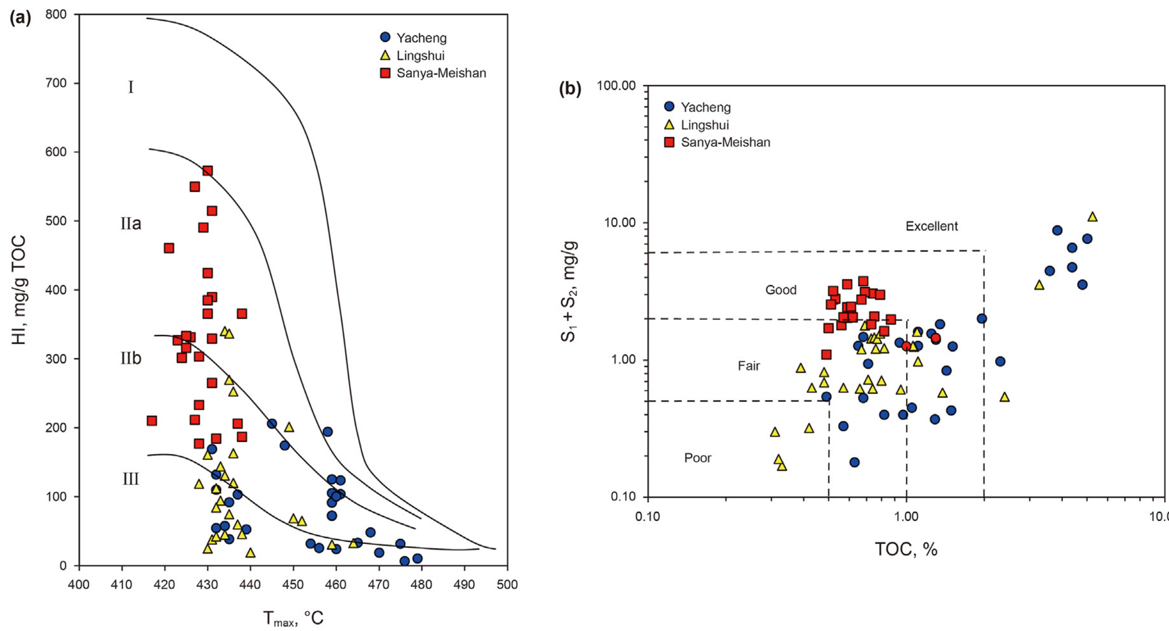

Fig. 8. Modified Van Krevelen diagram showing the relationship between the hydrogen index (HI) and the maximum pyrolysis temperature (T_{max}) (a) and the correlation between TOC content and the total content of free and pyrolysis hydrocarbons (S1+S2) (b) for typical source rocks in different strata and tectonic regions of the Qiongdongnan Basin. (The classification of kerogen types (a) and source rocks (b) are cited from Uspenskiy et al., 1980 and Zhu et al., 2021, respectively).

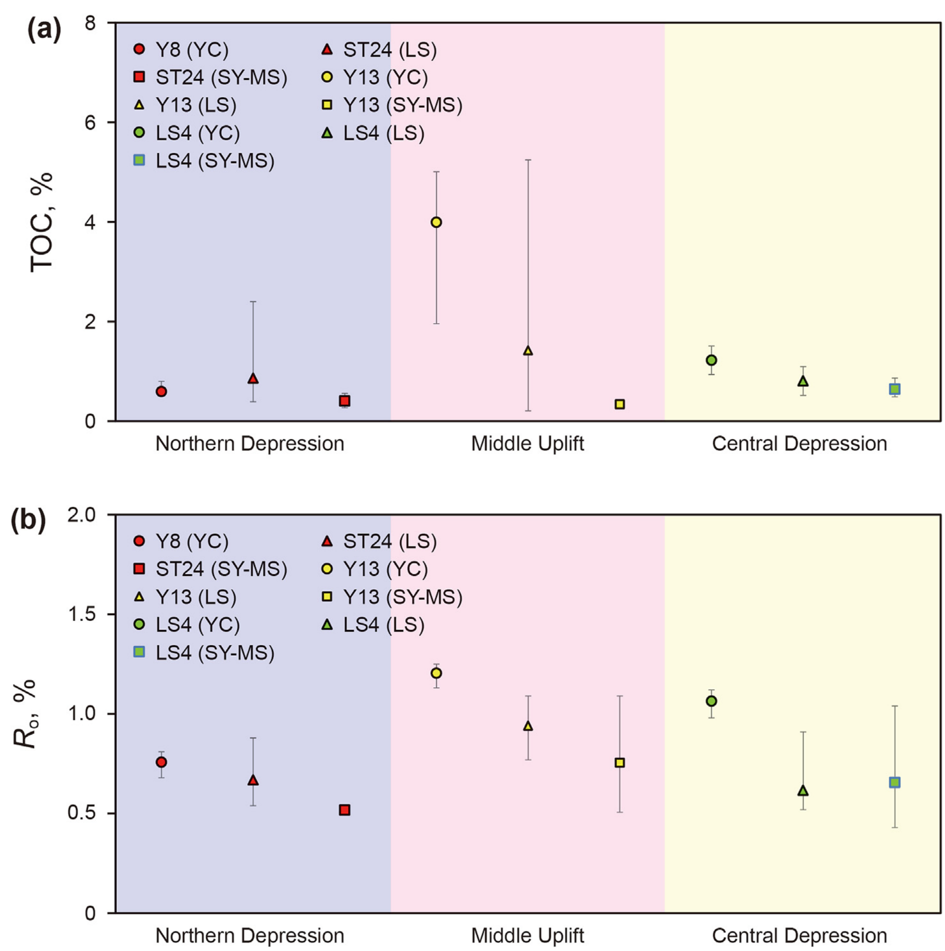

Fig. 9. The TOC contents and R_o values of source rocks in different strata and tectonic regions of the Qiongdongnan Basin.

and Central Depression blocks, respectively (Fig. 9a), and the average R_o values of the Sanya-Meishan source rocks are 0.94%, 0.67%, and 0.62% in the Northern Depression, Middle Uplift, and Central Depression blocks, respectively (Fig. 9b). It is worth noting that all the Miocene source rock samples were collected in slope areas of the Central Depression. In contrast, the Miocene source rocks with higher TOC contents may have mainly developed in sedimentary central areas of the Central Depression, and these marine source rocks have accessed a mature stage due to their deep burial depth. Therefore, the Miocene source rocks may have high hydrocarbon potential in the deep-water areas of the Qiongdongnan Basin.

In the Qiongdongnan Basin, the development of source rocks was significantly affected by the sedimentary facies and palaeoenvironmental conditions, and the differences in geological and geochemical characteristics of the source rocks with similar sedimentary facies are mainly controlled by the sedimentary environment. In general, the Yacheng source rocks are the main source rocks in both shallow and deep water areas of the Qiongdongnan Basin. In addition, the Eocene lacustrine source rocks in the North depression and the Miocene marine source rocks in the Central depression areas have certain hydrocarbon generation potential.

5. Conclusions

Based on the latest exploration data and geochemical analyses of typical source rock samples, the evolution of sedimentary facies and palaeoenvironment in different tectonic regions of the Qiongdongnan Basin were integrated and analyzed in this study, to reveal the controls on the development of source rocks. Several conclusions are obtained in this study:

(1) In the Qiongdongnan Basin, the sedimentary environment progressively varied from a semi-closed gulf to an open-marine environment from the early Oligocene to the Miocene, which resulted in significant differences in the palaeoenvironmental conditions of water columns for various tectonic regions of the basin.
(2) In the shallow-water areas, the palaeoproductivity and reducibility successively decreased, and the hydrodynamic intensity gradually increased for the water columns of the Yacheng, Lingshui, and Sanya-Meishan source rocks. In the deep-water areas, however, the water column of the Yacheng and Lingshui source rocks has a higher palaeoproductivity and a weaker hydrodynamic intensity than that of the Sanya-Meishan source rocks, and the reducibility gradually increases for the water columns of the Yacheng, Lingshui, and Sanya-Meishan source rocks.
(3) In the Qiongdongnan Basin, the palaeoenvironmental conditions are most favorable to the development of early Oligocene organic-rich source rocks. Meanwhile, the Miocene marine source rocks also have a certain hydrocarbon potential in deep-water areas.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.petsci.2023.04.019.

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