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Application of C_{30} tetracyclic polyprenoids as effective biomarker in oil-to source rock correlation in the Zhu Ⅲ depression, Zhujiangkou Basin, northern South China

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Abstract

The northern South China Sea, including the Zhujiangkou Basin and the Beibuwan Basin, developed high-quality lacustrine source rocks during the Eocene rifting period. These source rocks are vital for hydrocarbon generation in the northern South China Sea. The Zhu Ⅰ depression in the Zhujiangkou Basin and the Beibuwan Basin typically exhibit high abundance of C_{30} 4-methyl steranes. However, shales in the Eocene Wenchang Formation in the Zhu Ⅲ depression of the Zhujiangkou Basin contains lower quantities of high-quality lacustrine source rocks with 4-methyl steranes, which often co-elute with some pentacyclic triterpanes in gas chromatography-mass spectrometry (GC-MS). Therefore, the single 4-methylsterane parameter based on GC-MS cannot accurately distinguish organic source in the deep to semi-deep water lacustrine source rocks of the Wenchang Formation from other source rocks, thus impeding the recognition of their contributions to petroleum reservoirs. In this study, GC-MS of aliphatic hydrocarbons, palynofacies and algal identification, as well as stable carbon isotope data of organic matter were used to identify the algal species and construct the paleoclimate during deposition of the Wenchang Formation source rocks in the Zhu Ⅲ depression of the Zhujiangkou Basin. It is suggested that during the Wenchang Formation period, freshwater green algae prevailed in the lake, which is likely account for the relatively low content of 4-methyl steranes in the high-quality lacustrine source rocks. Controlled by the algal species, it is proposed that the content of C_{30} tetracyclic polyprenoids (TPP) can better indicate the quality of the Wenchang source rocks than C_{30} 4-methyl steranes. Consequently, a relationship between the TPP index and the quality of the lacustrine source rocks in the Wenchang Formation of the Zhu Ⅲ depression was established. A higher TPP index indicates higher organic matter abundance and hydrogen index of the lacustrine source rocks. When applied to the origin analysis of oils in the Zhu Ⅲ depression, it is believed that the organic-rich deep lacustrine source rocks in the Wenchang Formation made great contribution to the transitional zone crude oils in the Wenchang A and Wenchang B depressions.

Key words: lacustrine source rocks, C_{30} tetracyclic polyprenoids, green algae, Zhujiangkou Basin.

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1 Introduction

High-quality source rocks are the dominant hydrocarbon sources in the exploration practice of large oil and gas fields. Type and abundance of organic matter determine the oil and gas generating potential of source rocks. Biomarkers have biological information and are therefore used to study the organic matter origins in source rocks. Numerous biomarkers are generally related to algal input, but few of them are specific in nature. Botryococcane is a specific indicator of the fresh water algal *Botryococcus braunii*, yet is rarely observed in oils outside Southeast Asia and therefore is limited in application (Seifert and Moldowan, 1981). Other parameters that are indicative of lacustrine depositional environments include elevated 4-methyl steranes from dinoflagellate (Summons et al., 1987; Goodwin et al., 1988; Murray et al., 1994) and β-carotane (Jiang and Fowler, 1986; Fu et al., 1990). Elevated 4-methyl steranes are considered as a typically biomarker of high-quality lacustrine source rocks in China offshore areas, especially in the Beibuwan Basin and the Zhu Ⅰ depression in the Zhujiangkou Basin. Hence, the 4-methyl sterane parameter has been widely used in oil-source correlations in Chinese lacustrine sedimentary basins (Fu, 2018). However, the abundance of C_{30} 4-methylstanes in the lacustrine source rocks of the Wenchang Formation in the study area is generally low (Bao et al., 2007; Fu et al., 2011; Ding et al., 2015; Li et al., 2022). Meanwhile, C_{30} 4α-methyl-24-ethyl-5α(H),14β(H),17β(H) cholestane (20R) co-eluted with C_{29}Ts in some GC-MS experimental conditions (Fu et al., 2012). Hence, the credibility of the C_{30} 4-methylstanes ratios calculated by the peak areas are compromised. Exploring the reasons for the low abundance of 4-methylsteranes in the high-quality lacustrine source rocks of the Wenchang Formation in the Zhu Ⅲ depression, and seeking characteristic biomarkers that can better indicate these high-quality lacustrine source rocks of the Wenchang Formation, can help identify the contribution of the Wenchang formations. It also has great theoretical and practical significance for the oil and gas exploration in the northern South China Sea.

In this study, in addition to the bulk organic geochemical study on the Wenchang Formation mudstones in the Zhu Ⅲ de-

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pression, more specific algae biomarkers than previous studies were tentatively analyzed to indicate the dominant algae in paleo lakes during the corresponding geological interval. Correlations between the organic matter content in mudstones and algal contributions, specific biomarker abundance, were conducted to establish methods of the Wenchang source rock evaluation and oil identification in Zhu Ⅲ depression. This work also will be beneficial for understanding the development environment differences of the Eocene lacustrine source rocks and proposing the proper biomarkers in the oil-source correlations among basins/depressions in the northern South China Sea.

2 Geological setting

The Zhujiangkou Basin is an important petroliferous basin in China. It is located in the northern continental margin of the South China Sea and includes the Zhu Ⅰ depression, the Zhu Ⅱ depression, the Zhu Ⅲ depression and the Zhu Ⅳ depression (Fig. 1). The Zhu Ⅲ depression, located in the western part of the Zhujiangkou Basin, can be further divided into nine secondary structure units including the Yangjiang depression, the Qionghai depression, the Wenchang A depression, the Wenchang B depression and the Wenchang C depression (Yang et al., 2019). Among these five depressions, the Wenchang A and the Wenchang B are the main hydrocarbon generation. Oil and gas exploration in the Zhu Ⅲ depression is mainly concentrated in the Wenchang A depression, Wenchang B depression and their surrounding high areas of structure (Quan, 2018). Hydrocarbons in the Wenchang A depression is dominated by natural gas and condensate, while petroleum discovery in the Wenchang B depression is dominated by crude oils (Fig. 1).

Significant tectonic episodes have resulted in the deposition of different types of source rocks in the Zhu Ⅲ depression including (1) the Paleocene to Early Oligocene rifting period, during which formed several deep water lakes and deposited lacustrine source rocks of the Wenchang Formation, (2) the Oligocene to Early Miocene transitive period, during which caused the shrinking of the lakes and the deposition of fluvial marsh facies source rocks occured the Enping Formation and (3) the Miocene up to present depression period. As a product of the rift-sinking heyday, the Wenchang Formation mainly develops a (fan) delta-lake system, with the (fan) delta mainly coming from the Shenhu uplift, and the lakes mainly dominated by shallow-lakes and semi-deep lakes, which formed the main source rocks in the study area. The study area experienced a large-scale lake-phase deposition durings the deposition of the third and second members of the Wenchang Formation. The number and size of fan deltas decreased, and the semi-deep lake-phase deposition developed in the deep parts of the Wenchang B and C depressions. The Wenchang A depression also experienced an expansion of the middle-depth lake area and thicker stratigraphy. The thick-layered lacustrine mudstones that were developed during this period were the most significant hydrocarbon source rocks in the study area (Xu et al., 2023). Well W19-1Q-1 in the Wenchang B depression was drilled to reveal the lacustrine shales of the second member of the Wenchang Formation with a thickness of 100 m. Source rocks of the Wenchang Formation are mainly algae-dominated mudstones and are charatcerized by low abundance of 4-methyl sterane, trace amount of oleanane and C_{30} rearranged hopane, and the absence of bicadinane (Fu et al., 2012). Source rocks of the Oligocene Enping Formation are dominated by terrigenous higher plant inputs and are characterized by high abundance of bicadinane relative to C_{30} $\alpha\beta$ hopane (You et al., 2020). This biomarker

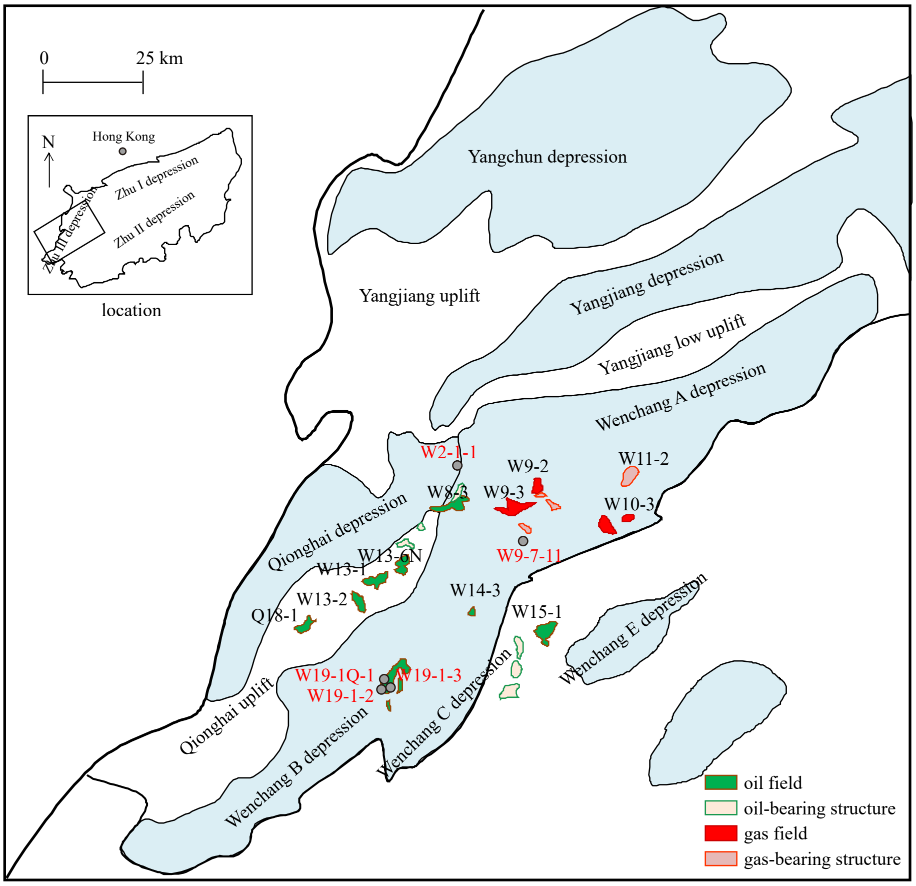

Fig. 1. Tectonic units of the Zhu Ⅲ depression and the oil/gas distributions.

characteristic allows the Oligocene Enping Formation to be easily distinguished from the Eocene lacustrine Wenchang Formation.

3 Samples and analytical methods

In this study, total organic carbon (TOC) measurement, Rock-Eval pyrolysis and GC-MS of aliphatic hydrocarbons were conducted on 19 cuttings samples of lacustrine mudstones from the Wenchang Formation, with 12 from the mid-deep lacustrine source rocks obtained from W19-1-3 and W19-1Q-1 wells, and 7 from the shallow-marine lacustrine source rocks obtained from W2-1-1 and W9-7-1 wells (Table 1). Spore-pollen and algal analysis were carried out on 32 mudstones/shales (Table 2). Stable carbon isotope data of organic matter in shales in Well W19-1-3 were cited from Sun et al. (2009). Stable carbon isotope data of Well W19-1M-1 were measured by Petroleum Geology Research and Laboratory Center, Reasearch Institute of Petroleum Exploration & Development (RIPED). Additionally, aliphatic GC-MS analysis was performed on 35 oil samples from the Zhu Ⅲ depression. Locations of each well were shown in Fig. 1.

3.1 *Rock-Eval pyrolysis and total organic carbon measurement*

For Rock-Eval pyrolysis and TOC measurement, the cutting samples were powdered to 200 mesh. In order to calculate the amounts of free hydrocarbons (S_1), potential hydrocarbons (S_2), and the temperature of maximal generation (T_{max}), Rock-Eval pyrolysis was carried out using a Rock-Eval Ⅳ. After pre-treating the samples with 5% diluted hydrochloric acid for 12 h and thoroughly rinsing with deionized water 50 times to eliminate any remaining hydrochloric acid, the TOC was measured using a Leco CS-344 analyzer.

3.2 *Spore and pollen analyses*

The pollen extraction method was used to treat the samples for spore and pollen analysis (Faegri and Iversen, 1989; Tweddle and Edwards, 2010). This method uses 10% hydrochloric acid to remove carbonates and 40% hydrofluoric acid to remove silicates. The residual inorganic portion was employed to separate the pollen and spores using a heavy liquid (HBr, KI, Zn) with a specific gravity of 2.0 (d = 2). Nylon sieves with a mesh size of 10 microns were used to sieve the palynological residues in an ultrasonic bath. Sporopollen were discovered and counted using a binocular microscope with a 400x magnification (Leica DM4000B). The number of particles of each sporopollen taxon is recorded if the number less than 60, and the number of particles is counted as a percentage in relation to the total amount of spores and pollen if the number lager than 60.

3.3 *Solvent extraction, fractionation and gas chromatography-mass spectrometry*

The cuttings samples were pulverized to 200 mesh and then solvent extracted with dichloromethane in a Soxhlet device for 72 h after washing the sample surfaces with deionized water and ethanol to eliminate any surface contamination. By adding excess n-hexane, the asphaltenes in the extractable organic matter were precipitated, leaving the soluble maltene fractions. By using column chromatography on silica gel, the maltenes were divided into aliphatic and aromatic hydrocarbons (elution with n-hexane) (elution with n-hexane: dichloromethane,1:2, v:v). Gas chromatography-mass spectrometry (GC-MS) was used to analyze the aliphatic fractions of the shale extracts using an Agilent 6 890 GC connected to an Agilent 5 973 mass selective detector (ionisation source operated at an electron impact energy of 70 eV). A fused silica capillary column (30 m, 0.25 mm, film thicknesses of 0.25 m) made of HP-5MS was employed. The oven was set to begin at 20℃(hold for 1 min), increase to 100℃ at 20℃/min, and then increase from 100℃ to 310℃ at 3℃/min with a final hold of 18 min. The carrier gas, helium, flowed at a rate of 1.0 mL/min.

4 Results and discussion

4.1 *Development of the Wenchang source rocks*

4.1.1 *Geochemical characteristics*

The Wenchang Formation organic-rich shales have been thought to be the major source rocks in the Zhu Ⅲ depression

Table 1. Rock-Eval pyrolysis data and biomarker parameters of the Wenchang cutting samples in the Zhu Ⅲ depression

Well name	Depth/ m	Lithology	TOC/ %	(S_1+S_2)/ (mg·g^{-1})	HI/ (mg·g^{-1})	T_{max}	Pr/ Ph	Pr/ nC_{17}	Ph/ nC_{18}	4MS/ $C_{29}S$	TPP/ C_{27}dia	OL/ $C_{30}H$	(W+T)/ $C_{30}H$	C_{21}/ (C_{19}-C_{26})	SS/ (SS + RR)	bb/ (aa + bb)
W19-1-3	1 700	mudstone	4.5	21.44	469	432	0.92	0.77	1.00	/	10.86	/	/	0.31	/	/
W19-1-3	1 956	mudstone	2.38	5.81	298	436	0.70	0.73	0.42	/	5.70	/	/	0.30	/	/
W19-1-3	2 102	mudstone	3.74	17.32	463	432	1.09	2.45	1.63	/	7.43	/	/	0.38	/	/
W19-1-3	2 342	sandy mudstone	3.46	14.32	408	435	1.16	1.45	1.28	/	6.74	/	/	0.32	/	/
W19-1-3	2 699	mudstone	2.98	11.53	379	432	0.95	0.79	0.54	/	7.59	/	/	0.18	/	/
W19-1-3	2 759	mudstone	2.34	6.65	280	435	1.00	1.36	0.84	/	4.17	/	/	0.29	/	/
W19-1Q-1	3 292	shale	2.4	12.56	487	443	1.83	0.52	0.30	0.41	6.85	0.14	0.02	0.26	0.52	0.48
W19-1Q-1	3 314	shale	3.03	16.55	514	442	1.52	0.76	0.53	0.38	8.09	0.11	0.01	0.25	0.50	0.43
W19-1Q-1	3 326	shale	2.79	15.77	554	443	1.48	0.61	0.45	0.47	6.87	0.12	0.01	0.25	0.50	0.48
W19-1Q-1	3 344	shale	3.83	20.77	513	444	1.61	0.71	0.47	0.41	8.56	0.09	0.01	0.25	0.49	0.50
W19-1Q-1	3 376	shale	2.86	12.56	404	442	1.60	0.48	0.28	0.43	7.60	0.14	0.02	0.21	0.48	0.53
W19-1Q-1	3 400	shale	3.4	18.05	485	443	1.60	0.55	0.34	1.14	7.38	0.11	0.01	0.23	0.50	0.47
W2-1-1	3 512	sandy mudstone	0.36	1.21	219	445	2.57	0.26	0.09	0.00	0.79	0.74	3.02	0.13	0.57	0.67
W2-1-1	3 514	mudstone	1.09	0.16	133	448	0.69	0.21	0.27	0.14	1.12	0.08	1.63	0.18	0.39	0.40
W2-1-1	3 517	mudstone	0.64	0.82	100	447	/	/	/	0.37	1.18	0.63	5.08	0.19	0.41	0.40
W9-7-1	4 278	mudstone	0.62	1.24	98	433	1.39	1.08	0.59	0.25	1.97	0.07	0.16	0.17	0.28	0.36
W9-7-1	4 288	mudstone	0.48	1.27	127	435	1.51	1.10	0.61	0.17	1.52	0.07	0.59	0.15	0.25	0.33
W9-7-1	4 292	mudstone	0.50	0.58	50	443	1.49	0.68	0.40	0.26	1.77	0.10	0.45	0.17	0.30	0.37
W9-7-1	4 308	mudstone	0.75	3.41	283	425	1.91	1.58	0.68	0.09	1.67	0.04	0.49	0.17	0.11	0.32

Note: / represents no data.

(Quan et al., 2019). It is therefore of great significance to decipher the heterogeneity of the Wenchang Formation for petroleum reserve sessment and for oil-source correlation (Keym et al., 2006; Curiale, 2008). All the 19 cutting samples display wide variations in Rock-Eval hydrogen indices but relatively narrow range of T_{max} from 430℃ to 450℃ (Fig. 2a), suggesting thermal maturity from early to late oil generation (Peters, 1986). Therefore, the wide variations in hydrogen indices were thought to be not much influenced by maturity changes in shales in the Wenchang Formation.

The deep-lake facies source rocks developed in the Wenchang Formation were confirmed by Well W19-1Q-1. Almost 120 m thick continuous shale samples were drilled and obtained in the Wenchang Formation from 3 284 m to 3 403 m in Well 19-1Q-1. Samples from Well W19-1Q-1 have TOC ranging from 2.4% to 3.8%, $S_1 + S_2$ in the range of 12.56 mg/g to 20.77 mg/g rock, and HI of 404 mg/g to 554 mg/g TOC. The Wenchang Formation drilled by Well W19-1-3 was sand-mud interbedded and was supposed to be a transition zone between the delta front and deep-lake facies. Samples from this well have lower range of TOC, HI

Table 2. Statistical table of the dominant fossil spore and pollen of the mudstone samples from the Zhu Ⅲ depression, Zhujiangkou Basin

No.	Well name	Depth/m	Lithology	(Spore+Pollen)/%	Algae		
					Green algae/%	Actitarchs/%	Non-marine dinoflagellates/%
1	W19-1Q-1	2 912	mudstone	89.40	1.90	8.70	0.00
2	W19-1Q-1	2 962	sandy mudstone	82.20	0.00	17.80	0.00
3	W19-1Q-1	2 980	silty mudstone	92.20	2.90	4.90	0.00
4	W19-1Q-1	3 024	mudstone	86.90	7.50	5.60	0.00
5	W19-1Q-1	3 062	mudstone	91.30	1.00	7.70	0.00
6	W19-1Q-1	3 084	mudstone	89.50	0.00	10.50	0.00
7	W19-1Q-1	3 090	mudstone	38.10	35.20	26.70	0.00
8	W19-1Q-1	3 104	mudstone	66.33	5.00	28.67	0.00
9	W19-1Q-1	3 228	mudstone	69.40	10.20	20.40	0.00
10	W19-1Q-1	3 250	shale	41.76	37.86	20.38	0.00
11	W19-1Q-1	3 276	shale	45.85	28.43	25.73	0.00
12	W19-1Q-1	3 300	shale	58.80	27.50	13.70	0.00
13	W19-1Q-1	3 326	shale	43.90	41.10	15.00	0.00
14	W19-1Q-1	3 346	shale	35.66	49.45	13.89	1.00
15	W19-1Q-1	3 366	shale	48.05	35.64	16.32	0.00
16	W19-1Q-1	3 402	shale	36.54	51.95	11.51	0.00
17	W19-1-3	1 832	sandy mudstone	50.48	48.57	0.95	0.00
18	W19-1-3	1 931	mudstone	99.03	0.97	0.00	0.00
19	W19-1-3	1 940	mudstone	60.97	39.03	0.00	0.00
20	W19-1-3	1 997	sandy mudstone	93.16	6.84	0.00	0.00
21	W19-1-3	2 051	mudstone	86.00	12.00	2.00	0.00
22	W19-1-3	2 069	mudstone	97.00	1.00	2.00	0.00
23	W19-1-3	2 099	mudstone	98.00	2.00	0.00	0.00
24	W19-1-3	2 207	sandy mudstone	84.62	14.42	0.96	0.00
25	W19-1-3	2 261	mudstone	94.95	4.04	1.01	0.00
26	W19-1-3	2 408	mudstone	99.19	0.81	0.00	0.00
27	W19-1-3	2 438	mudstone	99.20	0.80	0.00	0.00
28	W19-1-3	2 504	sandy mudstone	98.30	1.70	0.00	0.00
29	W19-1-3	2 531	mudstone	98.60	0.70	0.70	0.00
30	W19-1-3	2 564	sandy mudstone	95.36	4.64	0.00	0.00
31	W19-1-3	2 618	sandy mudstone	98.40	1.60	0.00	0.00
32	W19-1-3	2 654	sandy mudstone	98.13	1.87	0.00	0.00

Note: The number of particles are >60 and calculated in percent.

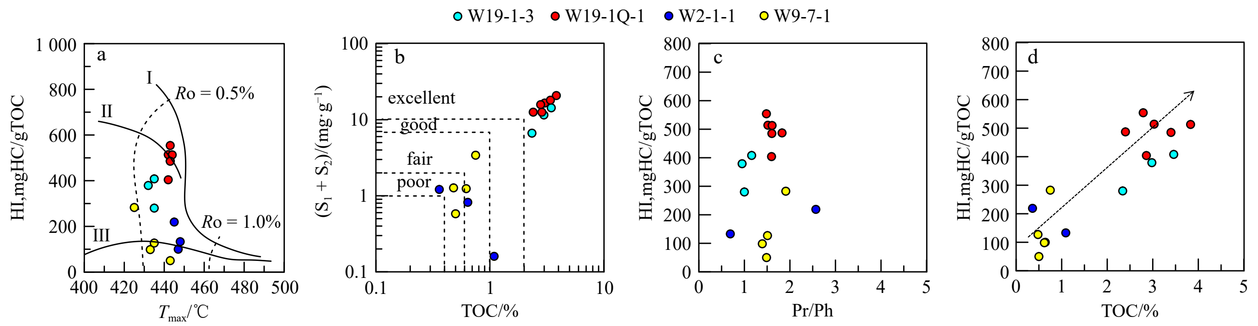

Fig. 2. Hydrocarbon potentials and organic matter types of the Wenchang source rocks deposited in the Zhu Ⅲ depression.

and $S_1 + S_2$ than the samples from Well W19-1Q-1 but are still in excellent quality area with Well W19-1Q-1. Lake-margin sediments were confirmed by Wells W2-1-1 and W9-7-1. Pyrolysis data of mudstones from Well W2-1-1 and W9-7-1 are characterized by TOC in the range of 0.36%–1.09%, $S_1 + S_2$ of 0.16–1.27 mg/g rock, and HI of 50–283 mg/g TOC which are in poor-fair source rock quality area (Fig. 2b).

It is worth noting that there is a weak correlation between hydrogen indices and Pristane/Phytane (Pr/Ph). Meanwhile, the TOC values increase as hydrogen indices increase (Figs 2c and d). Pr/Ph are effctive sedimentary environmental parameters. The low Pr/Ph ratio (<2) suggests that anoxic conditions prevailed in the bottom water. Among the 19 cutting samples, except for only one sandy mudstone sample from Well WC2-1 has high Pr/Ph ratio (> 2). The low and relatively stable ratios of Pr/Ph with wide range of hydrogen indices indicates that the preservation conditions are not the key factor controlling the good-quality source rock development. Source rocks with high hydrogen indices were derived mainly from algal organic matter. During the Wenchang period in the Zhu III depression, the positive correlation between TOC and hydrogen indices suggests that the algal blooms controlled the formation of good-quality source rocks.

4.1.2 *Biological sources*

The results of spore-pollen and algal identification shows the planktonic algal changes in Well W19-1M-1 and W19-1-3 (Table 2, Fig. 3). In Well W19-1M-1, significant changes in algal content occurred above and below the depth of 3 090 m. The content of planktonic algae is rather low in the Wenchang Formation with buried depths lower than 3 084 m, ranging from 1% to 26.8%, while the content of planktonic algae was significantly higher with depth deeper than 3 090 m, ranging from 30.6% to 64.4%. The contents of various components of planktonic algae show that the Wenchang Formation with depth deeper than 3 090 m are predominantly freshwater green algae. The Wenchang fine sediments drilled by Well W19-1-3 are sandy mudstones and mudstones. Although the content of planktonic algae varied significantly, green algae were always the dominant algal species genus with content of 0.7% to 48.6%. The results of spore-pollen and algal identification show that during the Wenchang period, the paleo-lake was relatively homogeneous in terms of algal speices mainly include , freshwater green algae *Pediatrum boryanum* and *Pediatrum simplex*. This understanding is consistent with the previous belief that the dominant algal species during the deposition period of the Wenchang Formation in the Zhu III depression was the green algae, specifically the genus Pediastrum (Xia and Wu, 1996; Zhu et al., 1997).

Unlike the Wenchang Formation in the Zhu III depression, during deposition of the Liushagang in the Beibuwan Basin, the dominant algae species in the lakes are non-marine dinoflagel-

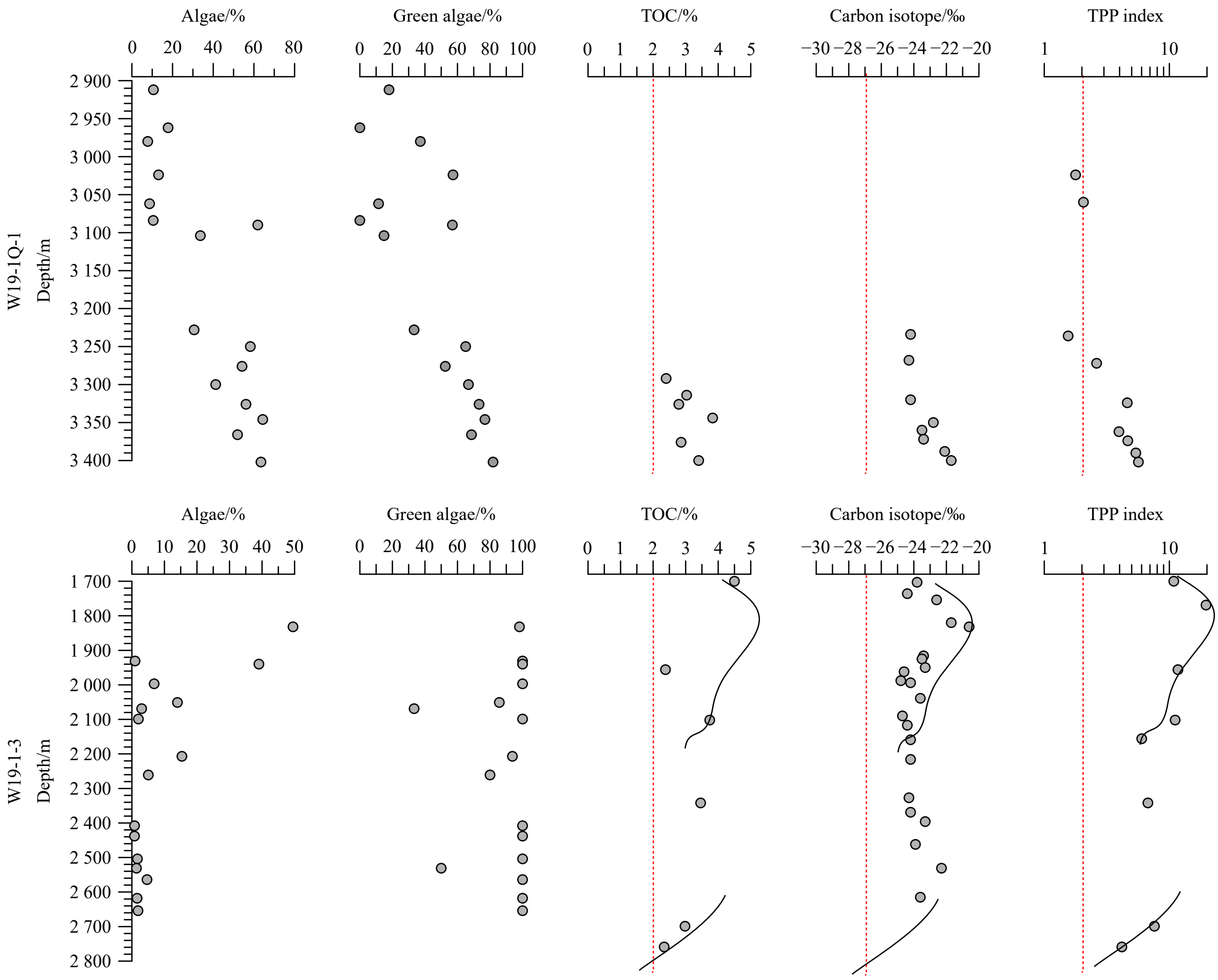

Fig. 3. Geochemical profiles of well W19-1Q-1 and W19-1-3. Algae/%: algae/(algae+pollen pore), green algae/%: green algae/algae, TPP index: [18α(H), 21R-TPP × 2]/(C_{27} S rearranged sterane + C_{27} R rearranged sterane).

late and green algae (Chang et al., 2023). The high abundance of C_{30} 4-methylsteranes in the Liushagang Formation in the Beibuwan Basin is associated with the input of dinoflagellate (Fu, 2018). In fact, shale samples from the Liushagang Formation are characterized by high abundance of TPP aliphatic hydrocarbons because of the concurrent green algal contribution. This difference in planktonic algae composition likely explains the much lower abundance of C_{30} 4-methylsteranes in the high-quality lacustrine source rocks of the Wenchang Formation in the Zhu III depression compared to the high 4-methylsteranes abundance in the Liushagang Formation in the Beibuwan Basin.

4.1.3 *Development model of the Wenchang source rocks*

The development of lacustrine source rocks is commonly influenced by the ancient productivity and the preservation conditions of organic matter. During the sedimentation period of the Wenchang Formation in the Zhu III depression, the spore-pollen assemblages are characterized by the dominance of pentagonal and oak pollen (Zhu et al., 1997), indicating a typical tropical climate. Tropical climates were thought to be beneficial for high primary productivity in lake basins (Katz, 1995). The anomalous distribution of stable carbon isotopes in the high-quality lacustrine source rocks of the mid-deep lacustrine facies in the Wenchang Formation also reflects the high productivity in the lake basin during the sedimentation period of the Wenchang Formation. The stable carbon isotope ratios of the shallow-marine lacustrine source rocks in the Wenchang Formation of the Zhu III depression ranges from –27.8‰ to –25.8‰, showing a characteristic of “normal” carbon isotope composition for an ecological environment. However, the mid-deep lacustrine source rocks and the corresponding crude oil in the Wenchang Formation both exhibit anomalously heavy carbon isotope compositions, ranging from –25.3‰ to –20.6‰, which was thought to be associated with algal blooming (Sun et al., 2009; Huang, 1993).

There are two reasons for the “anomalously positive” stable carbon isotope composition in the lacustrine sapropelic source rocks. First, the high ancient productivity of the lake leads to the predominance of aquatic biota, which contributes to the enrichment of more positive stable carbon isotopes in the sedimentary organic matter. Second, in a saline depositional environment, the high content of inorganic salts causes the carbon isotope composition of the hydrocarbon source rocks, primarily derived from aquatic biota, to become heavier.

Previous research based on studies of foraminifera content, carbon and oxygen isotopes of authigenic carbonates, and trace elements have revealed that the mineralization degree of water during the Wenchang Formation period in the Zhu III depression was low, below 200 mg/l, and the pH value was < 8, indicating a typical freshwater lake environment (Zhu, 2009). This is incongruent with the saline water conditions for “anomalously positive” carbon isotope composition of the algal organic matter. Furthermore, the Wenchang source rock intervals with anomalously positive carbon isotope composition in Well W19-1Q-1 and Well W19-1-3 show clear positive correlations among the stable carbon isotope ratios, the content of green algae and TOC values (Fig. 3). At the depth interval of 2 900–3 080 m in Well W19-1Q-1, the content of algae in the samples mostly accounts for approximately 10% of the total content of algae and spores. Meanwhile, the TOC is consistently below 1%, with the majority being below 0.5%. At the depth of 3 090 m, the content of algae in the samples significantly increases, accounting for about 60% of the total content of algae and spores. The content of green algae also increases, exceeding 50%. Simultaneously, the TOC also significantly increases to 3.5%. In the depth interval of 3 100–3 230 m, the content of algae in the samples decreases, ranging from 30% to 40%, with green algae accounting for 20%–30% of the total algae content. The TOC also decreases to below 1%. From a depth of 3 250 m and deeper, with increasing burial depth, the content of algae in the samples increases, and the TOC also gradually increases. As the green algae content and TOC values increases, the stable carbon isotope composition of organic matter becomes more postive (Fig. 3). This synchronous variation among algae content, together with TOC value and carbon isotope variations indicate the occurrence of green algae bloom in the mid-deep lacustrine facies of the Wenchang Formation, suggesting a high primary productivity in the lake basin.

In the shallow source rock interval in Well W19-1-3, the percentage of algal inputs are relatively high. This indicates that planktonic algae is an important source of organic matter in the source rocks intervals. In the deeper interval, possibly due to the sample inhomogeneity or fossil preservation, the contents of algae inputs were low but TOC values were still high. Nevertheless, there is also a good positive correlation between carbon isotope and TOC of organic matter in all the intervals, indicating that the positive carbon isotopes were triggered by the algae blooming.

Tropical lakes experience small seasonal temperature differences and are less prone to seasonal overturning. They have weaker water circulation and relatively quiet water bodies. After the bloom of algae in tropical lakes, when they die, they accumulate and decompose at the lake bottom. This process consumes a large amount of oxygen in lake water. This increased anaerobic respiration of microorganisms leads to a reducing environment in the water, enabling the preservation of algae. As a result, high-quality hydrocarbon source rocks are formed and deposited. Therefore, the enrichment of organic matter in the Wenchang Formation source rocks is controlled by high productivity of the ancient water, and the proliferation of algae determines the formation of high-quality hydrocarbon source rocks in the Wenchang Formation in the Zhu III depression.

4.2 *Geochemical implications of C_{30} tetracyclic polyprenoids*

4.2.1 *Identification of C_{30} TPP*

In this study, C_{30} tetracyclic polyprenoids (TPP) was detected in varying abundance in all thirty-four lacustrine source rock samples from the Wenchang Formation in the Zhu III depression (Figs 4 and 5). C_{30} TPP is a diagnostic indicator for freshwater or slightly brackish water algae input, especially for some green algae (Holba et al., 2000; Peters et al., 2005). In sedimentary layers where green algae blooming in the Sub-Andean Basin of South America, high contents of C_{30} TPP have been observed, suggesting that green algae are likely the biological source of C_{30} TPP

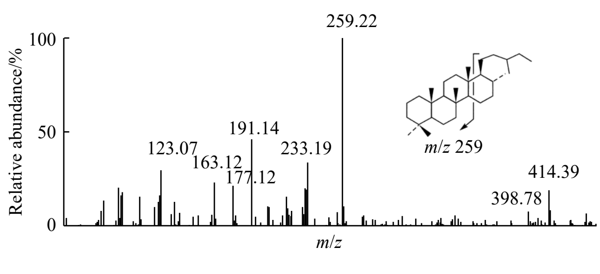

Fig. 4. Mass spectra of C_{30} tetracyclic polyprenoids (TPP) detected in the Wenchang semi-deep lacustrine source rocks in the Zhu III depression.

(Holba et al., 2003).

The C_{30} TPP, commonly elute as a doublet of peaks of the 21R and 21S isomers with nearly equal intensity when analyzed under common experimental conditions of *m*/*z* 259 mass chromatogram. In the lacustrine Wenchang Formation source rocks in the Zhu Ⅲ depression, the detected C_{30} TPP also consists of two distinct isomeric compounds. The later peak co-elutes with propylcholestane and exhibits a significantly higher abundance than the earlier peak. Therefore, when quantifying the abundance of the compounds, the area of the earlier peak [18*α*(H), 21R-TPP] is multiplied by 2 to represent the TPP content (Holba et al., 2000). The ratio between a C_{30} 20R TPP relative to 27-norcholestanes and C_{30} 20R TPP/C_{27} diacholestanes (TPP index) are useful indicators for assessing nonmarine freshwater algal input (Schiefelbein et al., 1999; Mello et al., 2013). Previous research has shown that the TPP index is highly specific to the algal source of organic matter and is an effective indicator for distinguishing the lacustrine sources from marine organic matter. For example, it has been successfully used in identifying lacustrine source oils on both sides of the South Atlantic (Silva et al., 2011; Casilli et al., 2014; Laakia et al., 2017). In the mid-deep lacustrine facies of the Wenchang Formation, the abundance of TPP is generally higher, with TPP index values ranging from 5.7 to 10.8. In contrast, the shallow-marine lacustrine source rocks exhibit significantly lower TPP abundance, with TPP index values ranging from 0.5 to 4.5 (Fig. 5). This suggests that there is a significantly higher input of algae in the mid-deep lacustrine source rocks compared to the shallow-marine lacustrine source rocks, which is consistent with higher hydrogen index of the mid-deep lacustrine source rocks.

4.2.2 *Application of TPP index in the Wenchang source rocks*

The geochemical profiles of wells W19-1Q-1 in the Zhu Ⅲ depression show a good positive correlation between the TPP index of the lacustrine source rocks from the Wenchang Formation and the content of freshwater green algae in total algae (Fig. 3). This indicates that freshwater green algae are likely the biological source of TPP. Hence, due to the blooming of green algae in the lake during the Wenchang period in the Zhu Ⅲ Depression, the high-quality lacustrine source rocks of the Wenchang Formation are characterized by high TPP index.

It was aforementioned that the dominant species of planktonic algae in the lake basin of the Wenchang Formation mainly come from freshwater green algae. Therefore, the TPP abundance can serve as an indicator of the quality of the lacustrine source rocks from the Wenchang Formation. In this study, there is a strong positive correlation between the TPP abundance and TOC and hydrogen index of the lacustrine source rocks from the Wenchang Formation in the Zhu Ⅲ depression (Fig. 6). High-quality hydrocarbon source rocks, which have high hydrogen content and abundant organic matter, exhibit high TPP index values. On the other hand, medium-quality hydrocarbon source rocks with lower hydrogen content and TOC have lower TPP in-

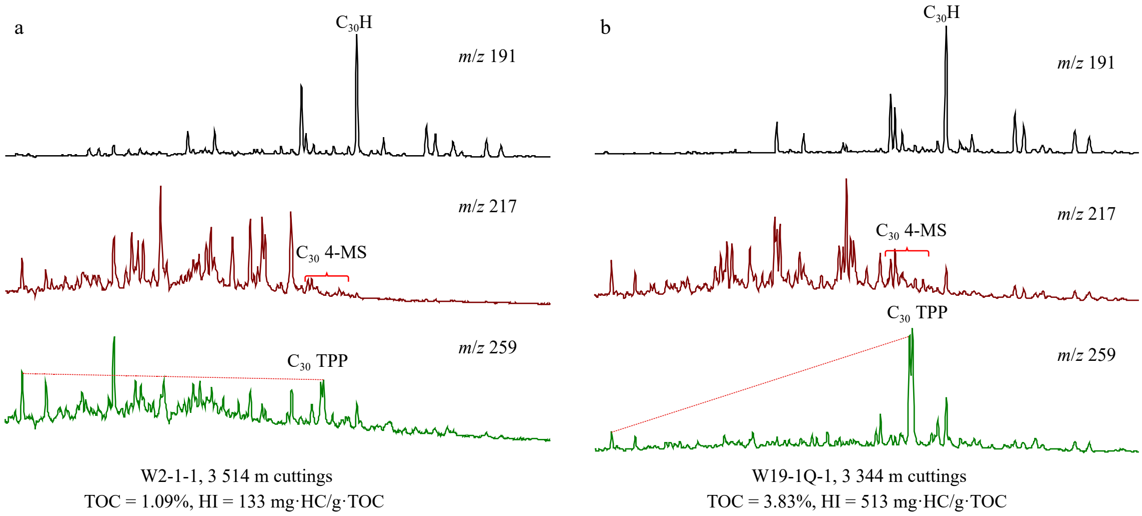

Fig. 5. Typical biomarker distributions of the Wenchang source rocks with different qualities in Zhu Ⅲ depression.

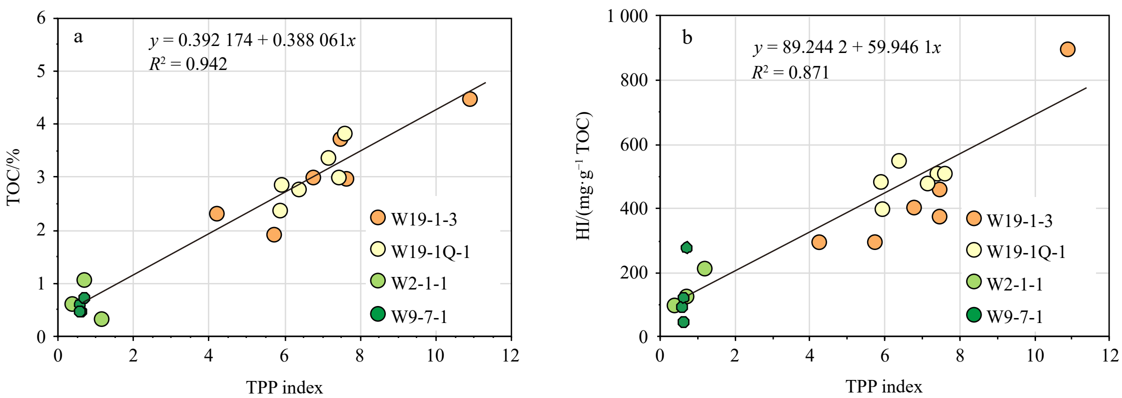

Fig. 6. Correlations between TPP indices and TOC, hydrogen index (HI) of the Wenchang source rocks in the Zhu Ⅲ depression.

dex values (Fig. 6). The correlation coefficients between the TPP index and the organic matter richness (TOC) and hydrogen index (HI) of the source rocks are 0.94 and 0.87, respectively. The good correlation between the TPP index and TOC reveals that good hydrocarbon source rocks (TOC > 1%) typically have TPP index > 2, and high-quality hydrocarbon source rocks (TOC > 2%) commonly have TPP index values >4. When the TPP index is >4.5, the HI is >400 mg/g, which aligns with the understanding that the high-quality hydrocarbon source rocks in the Wenchang Formation of the Zhu Ⅲ depression are rich in hydrogen and are predominantly sourced from planktonic algae in these ancient lakes.

Biomarker parameters are generally influenced by maturity and/or redox conditions, which were thought to exert limited control on the TPP index in this study. For the middle-deep lacustrine source rocks revealed by well W19-1Q-1, the burial depth ranges within 100 m, and the maturity differences are not significant. The calculated isomerization parameters 20S/(20S + 20R)C_{29} sterane and $\beta\beta/(\alpha\alpha+\beta\beta)$ C_{29} sterane are distributed in the range of 0.48–0.52 and 0.43–0.53, respectively (Figs 7a and b), indicating peak hydrocarbon generation stage. The corresponding TPP index values are distributed in the range of 5.7–7.5, which is significantly higher than the TPP index of the lacustrine source rocks from the shallow lake facies revealed by wells W2-1-1 and W9-7-1. Shales in wells W2-1-1 and W9-7-1 have similar TPP index distributions with the shallow lake facies of the Wenchang Formation, ranging from 0.34 to 1.16, which is significantly lower than that of the middle-deep lacustrine source rocks in Well W19-1Q-1. However, thermal maturity differences of shales in the shallow lake facies in these two wells are considerable. The 20S/(20S + 20R) C_{29} sterane isomerization parameter of shales in shallow lake facies from Well W9-7-1 ranges from 0.25 to 0.30, and the $\beta\beta/(\alpha\alpha+\beta\beta)C_{29}$ sterane isomerization parameter ranges from 0.33 to 0.37, while the shallow lake facies from Well W2-1-1 has 20S/(20S + 20R)C_{29} sterane and $\beta\beta/(\alpha\alpha+\beta\beta)C_{29}$ sterane isomerization parameters ranging from 0.36 to 0.57 and 0.4 to 0.67, indicating a higher thermal maturity level than the shallow lake facie shales from Well W9-7-1. There is large difference in maturity of the shallow lake facies between wells W2-1-1 and W9-7-1, but with similar TPP index, suggesting that the TPP index is not significantly influenced by thermal maturity. In addition, the similarity in maturity range between Well W2-1-1 and Well W19-1Q-1with difference in TPP index values, reflects the significant influence of sedimentary environments on the TPP index.

The cross plotting Pr/Ph and TPP index was used to assess the influence of redox condition of organic matter on TPP index. A total of 18 out of 19 cutting samples have low Pr/Ph ratios (<2.0), indicating the relatively stable redox conditions. However, these samples have wide range of TPP indices from less than 1.0 to around 11.0. Hence, the lack of positive or negative correlation between Pr/Ph and TPP indices suggests that the influence of redox conditions of organic matter on TPP contents is limited. Therefore, the TPP index can effectively used to indicate the variations of algal inputs in the Wenchang Formation in the Zhu Ⅲ depression.

4.2.3 *Application of TPP ratios in oil-source correlation*

By conducting aliphatic hydrocarbon chromatography analysis, TPP compound identification, and integration calculations on 35 crude oil samples from the Zhu Ⅲ depression, combined with the analysis of bicadinane content, the source of the crude oil was re-evaluated. The results showed that the TPP index of crude oils from the Wenchang B depression and the Qionghai uplift were all larger than 2, with the majority larger than 3, while the bicadinane content was extremely low. This indicates that these crude oils were mainly derived from lacustrine source rocks of the Wenchang Formation. On the other hand, crude oils from the Wenchang A depression had TPP index values all <2, indicating a distant relationship with the lacustrine source rocks of the Wenchang Formation. These crude oils also contained medium to high abundance of bicadinane, indicating that they mainly originated from the fluvial-lacustrine source rocks of the Enping Formation (Fig. 8). This further solidified the understanding of the petroleum system in the area. However, there were slight differences in the recognition of the petroleum source between the relatively high structures encountered in the Wenchang A and Wenchang B depressions.

In the two oil samples from the W8-3 structure, the shallower reservoir sample exhibits a high TPP index greater than 2, indicating a close relationship and significant contribution from the middle to deep lacustrine source rocks. Additionally, the deepest reservoir sample shows a low TPP index with abundant bicadinanes, suggesting a closer affinity with the Enping Formation source rocks. Among the three DST oil samples from the W9-1 structure, the deepest sample has a remarkably high TPP index of 5.0 and almost no bicadinane, indicating its origin from the mid to deep lacustrine source rocks of the Wenchang Formation. The two relatively shallower samples show moderate TPP index values along with higher bicadinane content, implying a mixed origin from the middle to deep lacustrine source rocks of the Wenchang Formation and the river-lacustrine source rocks of the Enping Formation. In the case of the W9-2 structure, the two oil samples show significant differences. The deep DST sample almost lacks TPP and contains high abundance of bicadinane, pointing to its origin from the Enping Formation. On the other hand, the shallow RFT oil sample has a high TPP index and low bicadinane abundance, suggesting a mixed origin from the middle to deep lacustrine source rocks of the Wenchang Forma-

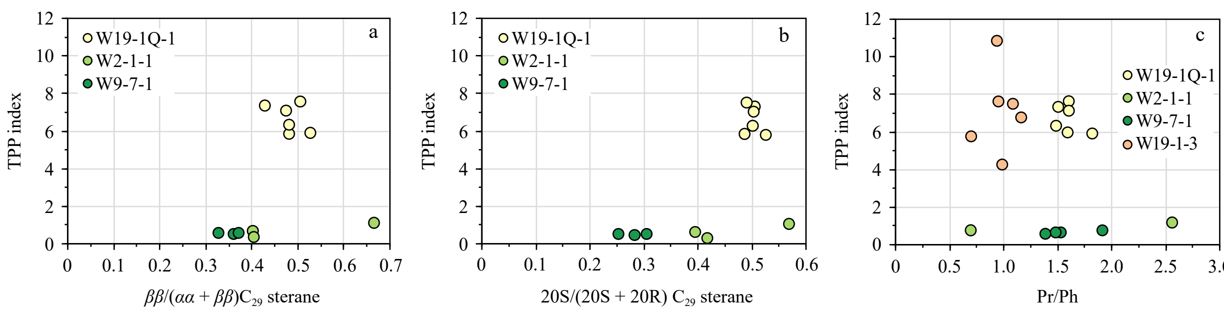

Fig. 7. Impact of thermal maturity and redox condition of source rocks on the TPP index.

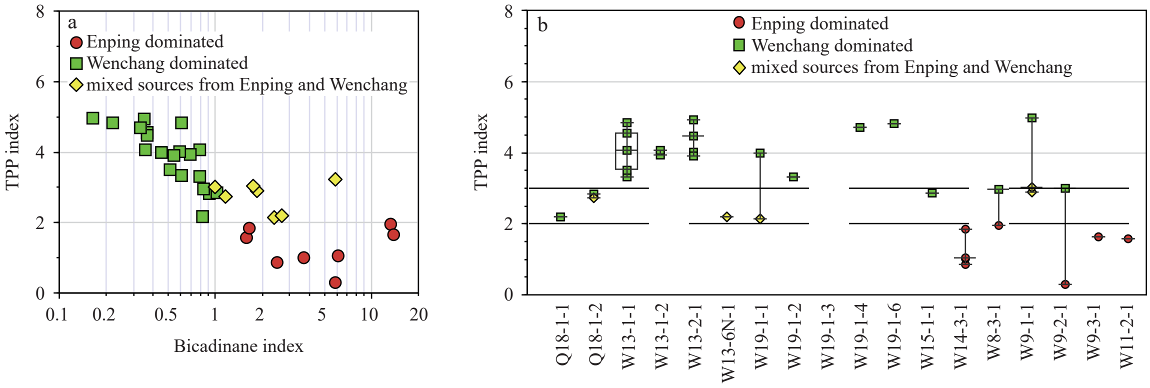

Fig. 8. Analysis of the oil origins in the Zhu III depression. Bicadinane index indicates the ratio of (W-bicadinane+T-bicadinane)/ C_{30} Hopane on $m/z = 412$.

tion and the river-lacustrine source rocks of the Enping Formation (Fig. 8). This is consistent with previous studies of a two-stage charging model based on fluid inclusion analysis in the same hydrocarbon reservoirs (Gan et al., 2007).

From a geological perspective, the conclusions drawn from the oil-source analysis result are feasible. During the deposition of the main reservoirs, the Zhuhai Formation and the Zhujiang Formation in the Zhu III Sag, both the Wenchang Formation and the Enping Formation were already in a thermally mature stage, which were effective hydrocarbon source rocks. Fluid inclusion studies have indicated that the Zhu III Sag experienced multiple stages of oil and gas charging (Fu et al., 2011). The differences in the timing of trap formation can result in variations in the sources of oil and gas accumulation. For instance, in the WC10-3 structure, three layers of oils from the Zhuhai Formation were identified, with one layer exhibiting characteristics of the middle to deep lacustrine source rocks of the Wenchang Formation. This finding indicates the presence of middle to deep lacustrine source rocks in the Wenchang A Sag. When the timing of trap formation aligns well with the expulsion timing of the middle to deep lacustrine source rocks of the Wenchang Formation. Therefore, it is possible for the reservoir to capture the oil sourced from these rocks (Li and Li, 2013). This understanding further supports the feasibility of using the TPP index for oil-source correlation.

5 Conclusions

(1) In the lacustrine Wenchang Formation in the Zhu III Sag, the green algae, particularly Prediadum, are the dominant algal species. This explains the relatively low abundance of C_{30} 4-methyl steranes in the Wenchang Formation source rocks. The TPP index shows a positive correlation with the input of green algae, making it a more effective indicator for evaluating the quality of Wenchang Formation source rocks and oil-source analyse in the Zhu III Sag.

(2) The deep lacustrine source rocks in the Wenchang Formation in the Zhu III Sag contain high abundances of C_{30} tetracyclic polyprenoids, with TPP index values ranging from 5.7 to 10.8. In contrast, the shallow lacustrine source rocks of the Wenchang Formation have lower TPP index ranging from 0.5 to 4.5. There is a good positive correlation between the TPP index and the quality of the Wenchang Formation lacustrine source rocks.

(3) Based on the distribution of TPP index in the crude oils, the contribution of the deep lake facies source rocks in the Wenchang Formation can be identified in the transition zone between the Wenchang A depression and the Wenchang B depression, particularly in oil and gas reservoirs including W8-3, W9-1, and W9-2. High TPP index values (larger than 2) of oil samples suggest the origin from the deep lake facies source rocks in the Wenchang Formation. This identification method is beneficial for better understanding the oil and gas sources in this region and provide valuable information for further petroleum exploration and development in study area.

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