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ORIGINAL ARTICLE

Organic geochemical characteristics of Eocene crude oils from Zhanhua Depression, Bohai Bay Basin, China

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Abstract Geochemical studies of crude oil and source rock play an important role in future exploration in Zhanhua Depression. In this study, thirty-one oil samples collected from Shahejie Formation in Zhanhua Depression, Bohai Bay Basin, NE China have been geochemically analyzed and their organic geochemical characteristics have been applied to differentiate groups of oils. These oil samples can be classified into two families based on multiple biomarker proxies and stable carbon isotopic values. Family I is characterized by a low ratio of pristane over phytane (Pr/Ph < 0.7), a relatively high ratio of phytane over n-C_{18} (Ph/n-C_{18}), varying ratios of gammacerane over C_{30} hopane (Ga/C_{30}H) and C_{22}/C_{21} tricyclic terpane, and a low ratio of C_{19}/C_{23} tricyclic terpane. Family II is marked by a relatively high Pr/Ph ratio (0.7–1.6), relative low ratios of Ph/n-C_{18} and C_{22}/C_{21} tricyclic terpane, and avarying ratio of C_{19}/C_{23} tricyclic terpane. Both families I and II within these crude oils can be subdivided into two families based on different values of stable carbon isotopic composition of individual n-alkanes. Moreover, the potential source rocks of oil samples in Family I and Family II were likely derived from the upper Es_4 member and Es_3 member, respectively, based on the correlation of organic geochemical characteristics of the oils and source rocks. The results of oil–source rock correlation provide insight into the process from oil generation to migration and to final accumulation, providing a better understanding of factors controlling oil–gas distribution for prediction of sweet spots.

Keywords Zhanhua depression · Bohai Bay Basin · Biomarkers · Stable carbon isotopic compositions · Crude oil

1 Introduction

A key problem in the oil–source study is to distinguish the effects of source on oil compositions, including organic matter input, depositional environment, and thermal maturity (Peters et al. 2005). Biomarkers are widely used in identifying groups of genetically related oils and correlating oils with source rocks (Moldowan et al. 1985; Peters et al. 2005). Their distributions and parameters can be used to interpret the depositional conditions and describe the source rock of migrated oil of uncertain origin (Seifert and Moldowan 1978, 1981). Variations in specific biomarker parameters are controlled by source input, such as n-C_{15}, n-C_{17}, n-C_{19} versus n-C_{25}, n-C_{27}, n-C_{29} (Bourbonniere and Meyers 1996) and the abundance of 4-methylsteranes (Bird et al. 1971; Wolff et al. 1986; Volkman et al. 1990); others are dominated by maturity, such as $22S/(22S + 22R)$ homohopane isomerization (Seifert and Moldowan 1980) and moretane/hopanes (Mackenzie et al. 1980; Seifert and Moldowan 1980); some others are affected by both source and maturity, such as diasteranes/steranes (Rubinstein et al. 1975; Sieskind et al. 1979; Mello et al. 1988).

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While carbon isotope analysis of bulk fractions has been frequently used in oil–source rock correlation (e.g., Rullkötter et al. 1986; Chung et al. 1997; Cai et al. 2015), compound-specific carbon isotope analysis of individual hydrocarbons has shown its advantage of accurate data and confidence in the interpretation of oil–oil and oil–source rock correlations (Freeman et al. 1990; Hayes et al. 1990; Bjorøy et al. 1991, 1994; Bakel et al. 1993; Boreham et al. 1995; Odden et al. 2002).

The Zhanhua Depression is located in the northeastern Jiyang sub-basin, Bohai Bay Basin, eastern China (Fig. 1). Intensive petroleum exploration suggests that the petroleum reserves were more than 19.1 × 10^8 t (Li et al. 2017; Yuan et al. 2019). Shengli oil field is one of the primary oil–gas fields in China. As a highly explored area, the Zhanhua Depression area has stepped into the exploration stage of subtle reservoirs. However, the origin of oil and gas has remained as the focus. In previous geochemical studies of oil–source correlations in Zhanhua (e.g., Shi et al. 2005; Wang 2011; Song and Men 2019), compound-specific isotopic data was rarely reported compared to the biomarker. This study investigates both biomarkers and stable carbon isotopic compositions of *n*-alkanes from oils in Zhanhua Depression. In addition, most geochemical studies of oils from Zhanhua Depression (e.g., Gong et al. 2005; Liu et al. 2006a, b; Zhang et al. 2006) have limited oil samples from single well or source rock of a single member. This study investigates the geochemical characteristics of oils in Es_{1-4} members from multiple wells in Zhanhua Depression. Two families of oils are classified and further subdivided on the basis of biomarker and carbon isotopic proxies. The original strata and the distribution of oils and gas from different source rocks are interpreted through fine oil–source rock correlation on the basis of the geochemical characteristic of crude oil and source rock. This study provides effective methods on the division of oil–gas systems and the prediction of oil–gas migration. A better understanding of these controlling factors, with the synthesis of local geological information (e.g., structure and fault), assist in the promotion of oil production in existed wells and contributes new evidence and supplements to future exploration of oils in Zhanhua Depression. This comprehensive study combined biomarkers and compound-specific carbon isotopic data and provided insight into possible origins of the two oil families, in order to provide helpful recognitions and progress of oil–source rock correlation, which provides new evidence and supplements to the previous study of Zhanhua Depression.

2 Samples and methods

2.1 Geological settings

Zhanhua Depression is one of the seven individually developed depressions in the Bohai Bay Basin (Fig. 1b; Zhang et al. 2005). Chenjiazhuang Uplift is in the South and Yihezhuang Uplift is in the north of Zhanhua Depression (Fig. 1c; Liu et al. 2006a, b; Li et al. 2017). It

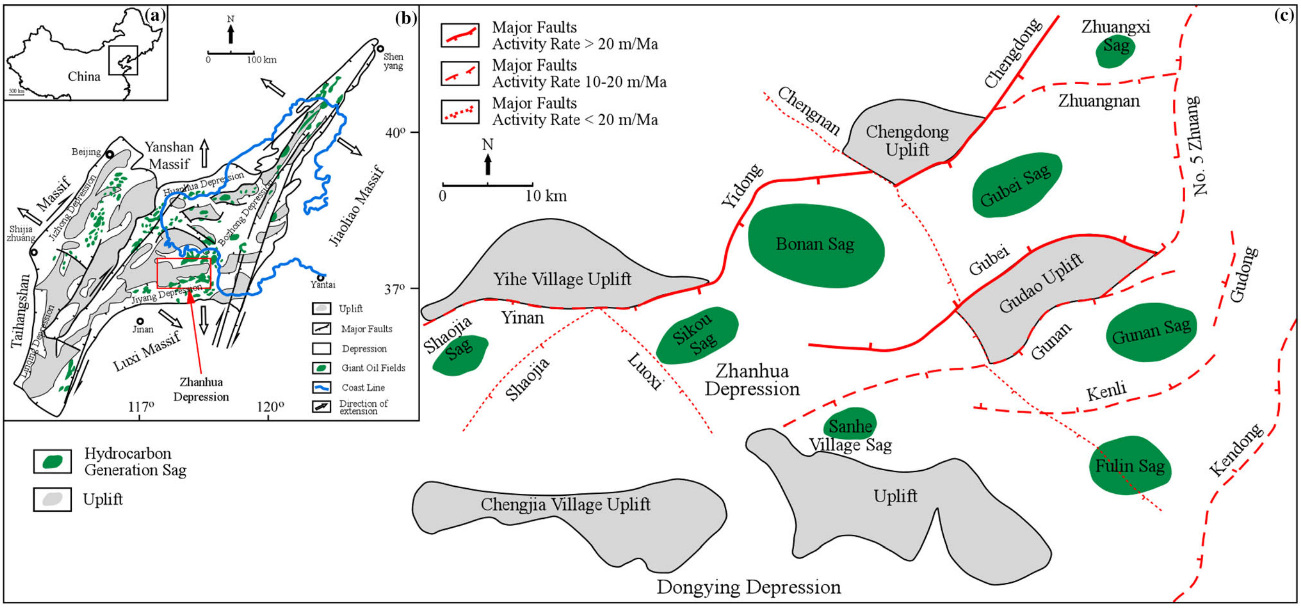


Fig. 1 Location of the study area: Maps show the elementary structural features of Zhanhua Depression (Modified from Ma 2017): Location of the Bohai Bay Basin in China (a), subbasins of the Bohai Bay Basin (b), location map showing the distribution of hydrocarbon generation sags in Zhanhua depression (c)

forms a typical half-graben rift basin whose north side is faulted, and south side overlapped, covering an area of 2800 km^2 (Ma 2017).

The lacustrine deposits in Zhanhua Depression comprise three Paleogene units: Kongdian (Ek), Shahejie (Es), and Dongying (Ed) Formations (Liu 2017). The Shahejie Formationis subdivided into four members: Es_1, Es_2, Es_3, and Es_4 in descending order (Fig. 2). The potential source rocks are considered located in the upper Es_4, Es_3, and Es_1 members (Shi et al. 2005; Song and Men 2019). The lower Es_4 comprises mudstone with anhydrate and salt while the upper Es_4 contains laminated oil shale and lime mudstone with carbonate, interpreted as saline lake deposits during lake contraction (Liu 2017; Song and Men 2019). The lower Es_3 has thick laminated shale and calcareous shale with intercalated siltstone while the middle–upper Es_3 is composed of thick massive siltstone and sandy siltstone, suggesting a transition from salt-brackish lake to the fresh lake during lake expansion (Zhu 2002; Ma et al. 2016; Liu 2017). Es_2 has mudstone with intercalated sandstone and

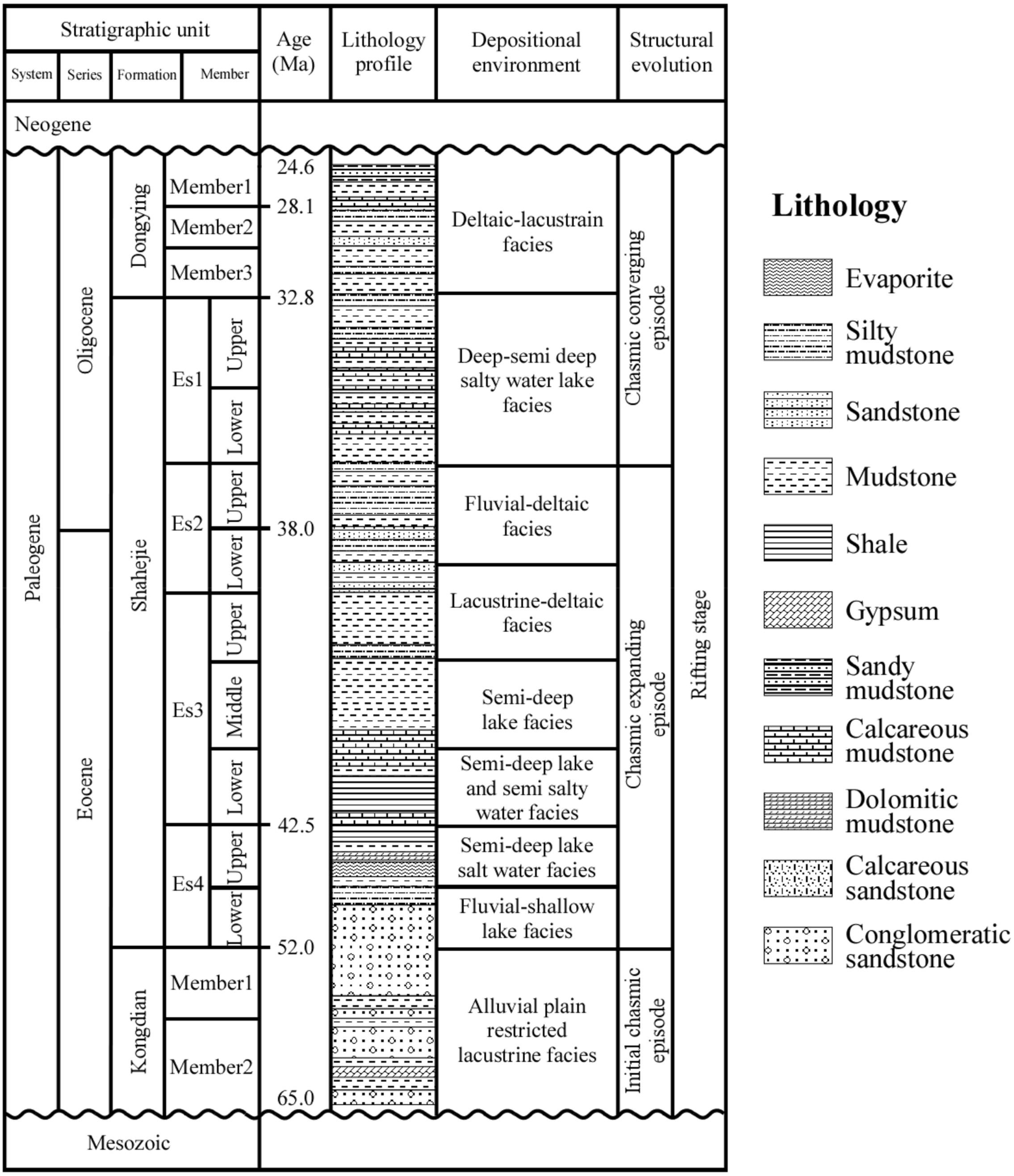


Fig. 2 Tertiary stratigraphy of the Zhanhua Depression (Modified after from Jiu et al. 2013)

conglomerate, representing fluvial-deltaic deposits during lake contraction (Allen et al. 1997; Zhu 2002). Es_1 contains thick laminated shale, mudstone, sandstone, and thin biogenic carbonates, interpreted as profundal deposits during lake expansion (Zhu 2002; Song and Men 2019).

2.2 Samples

Forty-eight crude oil samples in Shahejie Formation have been collected from reservoirs in multiple drilling wells in Zhanhua Depression, Bohai Bay Basin. Highly biodegraded oils have been excluded because the secondary alteration process may have caused variations on type and abundance of biomarkers in these oils, result in misinterpretation of the oil–source relationship (Peters et al. 2007; 2016). Thus, the remaining thirty-one oil samples without biodegradation have been selected and analyzed in this study.

2.3 Methods

The oils were dissolved in *n*-hexane and ultrasonicated, the soluble fraction was further separated into saturated, aromatic, and polar fractions on a silica/alumina column using hexane, hexane/dichloromethane (3:2), and methanol, respectively. The saturated fraction was further treated with urea adduction to yield the branched and linear chain alkane fractions, and each fraction was conducted on GC-IRMS for compound-specific isotope analysis.

GC/MS analysis was performed on a Trace Ultra GC coupled to a Thermo DSQ-II mass spectrometer operated at an ionization energy of 70 eV with a full scan ranging from *m*/*z* 50–600. An HP-5 fused silica capillary column (30 m × 0.25 mm i.d. × 0.25 μm film thickness) was used. The GC oven temperature was set initially at 80 °C and held for 2 min, then increased to 295 °C at a heating rate of 3 °C/min and held for 20 min. High purity nitrogen was used as the carrier gas with a constant flow of 1.2 mL/min.

GC-IRMS analysis was performed on the saturated hydrocarbon fractions for compound-specific carbon isotopic measurements, using an Isoprime IRMS instrument interfaced to an HP6890 GC instrument via a combustion interface. A DB-5 column (30 m × 0.32 mm i.d. × 0.25 μm film thickness) was used for chromatographic separation. The GC oven was increased from 80 to 300 °C at a heating rate of 4 °C/min and held for 30 min. Each sample was analyzed in triplicate, and the average value for the three runs was regarded as the final $\delta^{13}C$ value of the sample. Isotope ratios are determined based on delta notation (‰) relative to the Vienna Pee Dee Belemnite (V-PDB) standard. The precision of replicated samples is better than ± 0.08 ‰ for the compound-specific carbon isotopic measurements.

3 Results and discussion

3.1 Molecular geochemistry

3.1.1 n-alkanes and acyclic isoprenoids

The *n*-alkane distribution of oils collected in Es_1 and Es_3-members are quite similar (Fig. 3a; Table 1). *n*-alkanes in most oil samples have a predominance of medium molecular weight compounds (*n*-C_{19-25}) and arrange from *n*–C_{13} to *n*-C_{33}, with only a few of them, contain *n*-C_{34-37}, suggesting weak biodegradation. Oils from Es_2 and Es_4 members are generally dominated by both low (*n*-C_{18-20}) and high (*n*-C_{28-30}) molecular weight compounds (Fig. 3b; Table 1), suggesting a major source contribution from both algae and terrestrial higher plants into organic matter (Peters et al. 2005).

The Pr/Ph ratios of the oil samples range between 0.25 and 1.59. The oils from Es_1 and Es_3 members are commonly marked by a weak dominance of pristane over phytane, with an average Pr/Ph ratio of 1.23, suggesting the source rock of the oils were likely deposited in sub-oxic conditions (Didyk et al. 1978). Oils from Es_2 and Es_4 members have a pronounced predominance of phytane over pristane, with an average Pr/Ph ratio of 0.39, indicating the oils were derived from source rocks deposited in reducing conditions. According to the ternary diagram of Pr/Ph–Pr/*n*-C_{17}–Ph/*n*-C_{18} (Fig. 4a), source rocks of oils from Es_2 and Es_4 were likely deposited in brackish-saline water environment and almost source rocks of oils from Es_1 and Es_3 members likely formed in freshwater. This is consistent with the high phytane/*n*-C_{18} ratios of oils from Es_1 and Es_3 compared with that of oils from Es_2 and Es_4 members. A crossplot of Pr/*n*C_{17} versus Ph/*n*C_{18} can be used to indicate thermal maturity as well as the depositional environment and the organic matter type of the source rock (Peters et al. 1999; Hanson et al. 2001). The Pr/*n*C_{17} versus Ph/*n*C_{18} results suggest the source rocks of oils from Es_2 and Es_4 members mainly developed in algal and reducing environment, while that of oils from Es_1 and Es_3 members developed in a transitional environment with mixed organic matter of algae and terrestrial higher plants (Fig. 4b).

3.1.2 Terpenoids

Terpanes are identified using GC/MS by monitoring the ions *m*/*z* 191 (Fig. 3c, d).The C_{19}–C_{20} tricyclic terpanes (TTs) have been considered originated mainly from

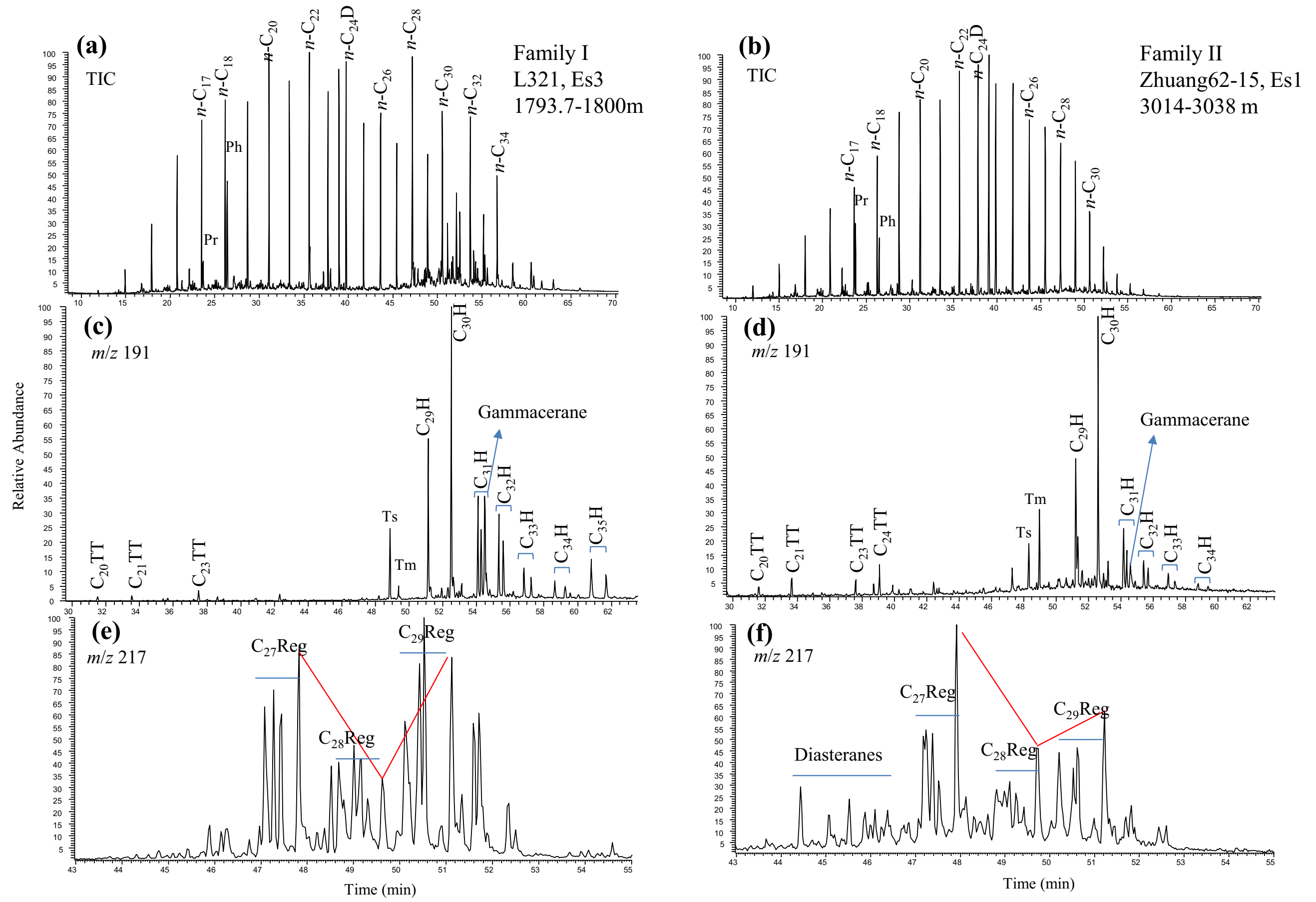


Fig. 3 Representative mass chromatograms of total ion chromatogram (TIC), hopane (*m/z* 191) and sterane (*m/z* 217) series of saturated hydrocarbon fraction for crude oils in the Zhanhua Depression

diterpenids (Peters et al. 2005), which are produced by vascular plants (Peters et al. 2005). Marine and saline water conditions promote the generation of C_{23} TTs (Neto et al. 1981; Tao et al. 2015; Chen et al. 2017), whereas C_{23} TTs are dominated in freshwater (Zhu 1997). Therefore, carbon number distributions of the TTs are considered a useful indicator of organic matter input and environmental conditions (Peters et al. 2005; Xiao et al. 2019). C_{19}/C_{23} TTs ratio ranges from 0.09 to 0.45 in all oil samples, suggesting varying organic matter input and environmental conditions.

Hopanes, derived from membranes of aerobic bacteria and photosynthetic cyanobacteria (Ourisson and Albrecht 1992), have been widely used as proxies for organic matter input, depositional environments, and thermal maturity level (Peters et al. 2005).C_{27} 18α-trisnorneohopane (Ts) is thermally more stable than the C_{27} 17α-trisnorhopane (Tm) during catagenesis (Seifert and Moldowan 1978). The investigated oils have a wide range of Ts/(Ts + Tm) ratios ranging from 0.09 to 0.47, suggesting the presence of multiple groups of crude oils with different maturities (Moldowan et al. 1985) or diverse source depositional environments (Peters et al. 2005).

The distribution of the homohopanes (C_{31}–C_{35}) has been used to evaluate redox conditions based on homohopanes index (Peters et al. 2005). High C_{35}/C_{34} homohopanes ratios of oil samples from Es_4 range from 1.15 to 2.61 while the low C_{35}/C_{34} homohopanes ratios (0.42–0.90) in Es_{1-3} are lower than 1 except two samples in Es_3 suggesting source rocks of the oils in Es_4 were deposited in highly reducing conditions and that in Es_{1-3} were deposited in suboxic conditions (Moldowan et al. 1985; Peters and Moldowan 1991). This is consistent with interpretation from low Pr/Ph ratios of oil samples.

High abundance of gammacerane is typically related to highly reducing and hypersaline conditions during the deposition of organic matter (Damsté et al. 1995; Peters et al. 2005; Summons et al. 2008; Albaghdady 2013). The average ratios of gammacerane to C_{30} hopane (Ga/C_{30}H) of oil samples from Es_2(0.34) and Es_4(0.21) are higher than that of Es_1(0.16) and Es_3(0.12), suggesting source rocks of oils from Es_2 and Es_4 were likely deposited in the brackish-saline lake and that from Es_1 and Es_3 were deposited in fresh-water lake conditions.

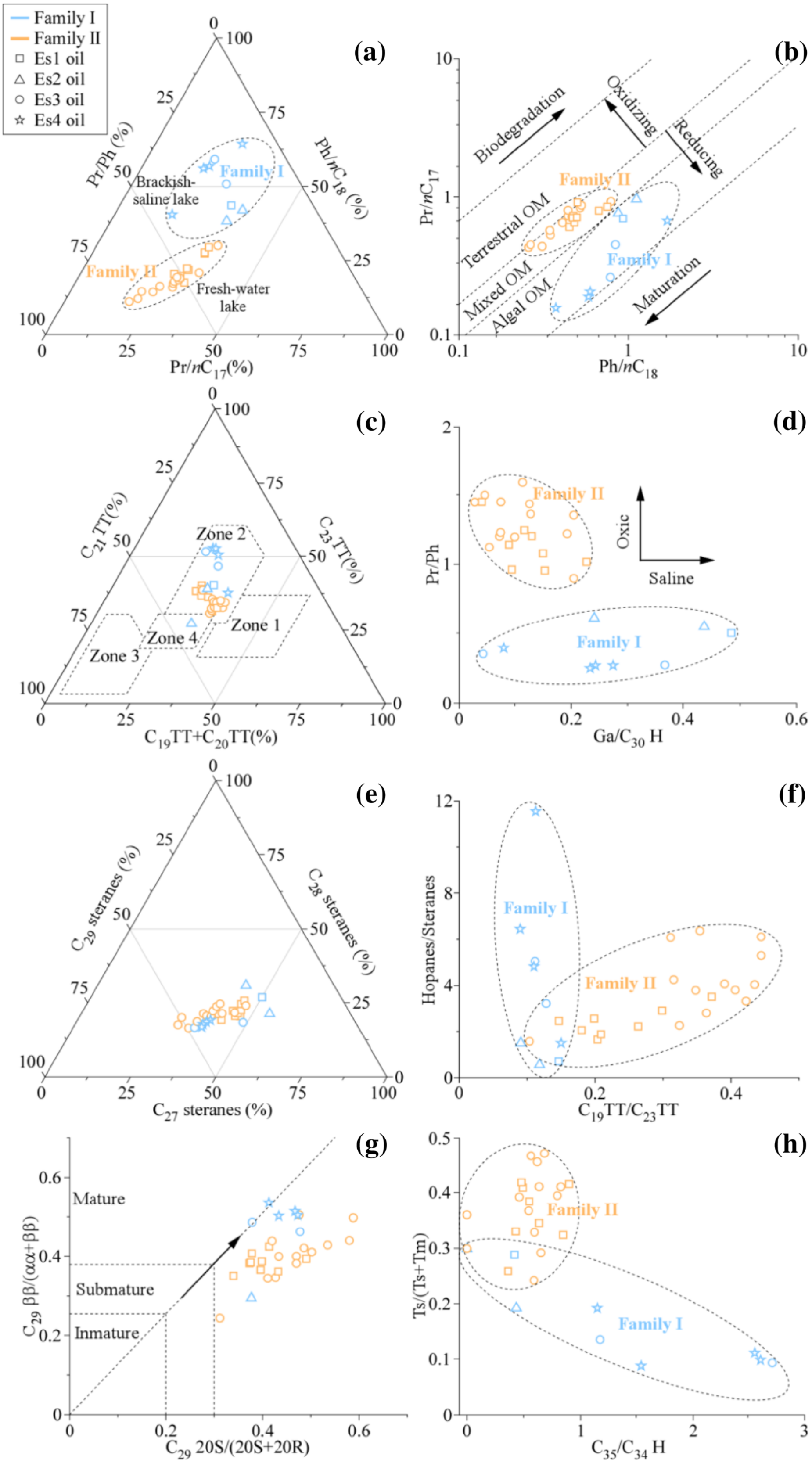
Table 1 Sample information and geochemical parameters

Sample	Interval	Depth (m)	Family	*n*-alkane and isoprenoids				Terpanes (*m*/*z* 191)					H/S	Steranes and diasteranes (*m*/*z* 217)					
				CPI	Pr/Ph	Pr/*n*-C_{17}	Ph/*n*-C_{18}	C_{19}/C_{23} TT	Ts/(Ts + Tm)	C_{30}M/C_{30}H	G/C_{30}H	C_{35}H/C_{34}H		C_{27}S (ααα)%	C_{28}S (ααα)%	C_{29}S (ααα)%	C_{29} 20S/(20S + 20R)	C_{29} αββ/(αββ + ααα)	C_{27} dia/(reg + dia)
Z391	Es1	2724.2–2756.4	I	1.05	0.51	0.70	0.93	0.15	0.29	0.09	0.48	0.42	0.69	50.10	26.82	23.08	0.47	0.51	0.00
Z399	Es2	2851–2861.6	I	1.05	0.63	0.77	0.87	0.12	0.19	0.09	0.24	0.43	0.56	55.20	21.40	23.40	0.38	0.49	0.00
Z351	Es2	3155–3160	I	1.05	0.56	0.97	1.11	0.09	0.30	0.09	0.44	–	1.49	43.47	30.97	25.56	0.38	0.29	0.00
L321	Es3	1793.7–1800	I	0.96	0.28	0.26	0.78	0.13	0.09	0.06	0.37	2.71	3.20	35.70	16.62	47.68	0.48	0.46	0.00
L354	Es3	2654–2657	I	1.00	0.35	0.45	0.84	0.11	0.14	0.06	0.05	1.18	5.04	48.92	18.44	32.64	0.38	0.49	0.00
L9–6–71	Es4	2138.5–2153	I	0.97	0.25	0.21	0.59	0.09	0.11	0.06	0.23	2.56	6.45	37.93	18.62	43.45	0.47	0.50	0.00
L9–5–12	Es4	2154.9–2171.5	I	0.96	0.27	0.19	0.58	0.11	0.10	0.05	0.28	2.61	4.82	37.31	17.84	44.85	0.43	0.50	0.00
L358	Es4	2663.9–2691.2	I	0.99	0.27	0.67	1.69	0.15	0.09	0.05	0.24	1.54	1.48	38.87	19.19	41.94	0.41	0.54	0.00
L7	Es4	3039.4–3048.4	I	0.99	0.39	0.16	0.37	0.11	0.19	0.06	0.08	1.15	11.56	37.47	16.86	45.67	0.47	0.51	0.00
Z303	Es1	2472–2473	II	1.03	1.08	0.71	0.50	0.21	0.32	0.09	0.15	0.85	1.86	43.87	22.23	33.90	0.34	0.35	0.23
Z47	Es1	2753.0–2757.5	II	1.04	1.02	0.65	0.48	0.15	0.33	0.09	0.23	0.43	2.44	45.76	21.40	32.84	0.38	0.38	0.00
Z62–15	Es1	3014–3038	II	1.05	1.21	0.70	0.47	0.26	0.42	0.09	0.13	0.90	2.20	47.02	20.37	32.61	0.37	0.38	0.28
Z134	Es1	3015–3024	II	1.04	1.45	0.91	0.50	0.37	0.41	0.09	0.04	0.49	3.49	42.08	19.16	38.76	0.49	0.39	0.29
Z42	Es1	3048–3061	II	1.05	1.25	0.73	0.47	0.20	0.38	0.09	0.12	0.55	2.54	46.15	22.46	31.39	0.38	0.41	0.26
Z351	Es1	3056.8–3042.7	II	1.04	0.96	0.85	0.75	0.20	0.26	0.12	0.10	0.36	1.63	45.38	20.51	34.11	0.40	0.37	0.22
Z300	Es1	3118–3122	II	1.04	0.96	0.79	0.67	0.18	0.35	0.08	0.15	0.63	2.04	45.16	24.54	30.30	0.40	0.39	0.24
Z390	Es1	3118–3122	II	1.04	0.96	0.85	0.75	0.20	0.26	0.12	0.10	0.36	1.63	45.38	20.51	34.11	0.40	0.37	0.22
Z11–1	Es1	–	II	1.05	1.14	0.60	0.45	0.30	0.42	0.07	0.09	0.48	2.89	40.21	21.16	38.63	0.41	0.42	0.26
Z202–3	Es3	2583–2600.4	II	1.04	1.36	0.44	0.31	0.39	0.47	0.11	0.21	0.56	4.05	38.23	22.26	39.51	0.41	0.34	0.23
Z306	Es3	2603–2613	II	1.05	1.20	0.66	0.45	0.33	0.41	0.09	0.10	0.63	2.24	44.84	21.70	33.46	0.43	0.40	0.25
Z8–1	Es3	2836–2851	II	1.03	1.23	0.72	0.46	0.42	0.30	0.11	0.08	–	3.31	39.09	24.72	36.19	0.48	0.50	0.28
Z43	Es3	3004–3006.5	II	1.03	1.59	0.43	0.26	0.36	0.41	0.13	0.11	0.83	6.36	33.94	16.52	49.54	0.47	0.40	0.26
Z242	Es3	3158–3179	II	1.02	1.44	0.45	0.27	0.45	0.46	0.11	0.13	0.62	6.10	30.30	17.61	52.09	0.54	0.43	0.26
Z66	Es3	3193–3259	II	1.03	0.90	0.92	0.79	0.10	0.24	0.09	0.21	0.59	1.55	46.93	24.08	28.99	0.31	0.24	0.00
Z42	Es3	3196.5–3199	II	1.02	1.45	0.57	0.34	0.31	0.37	0.12	0.08	0.54	6.08	35.38	18.72	45.90	0.49	0.42	0.26
Z421	Es3	3209–3218	II	1.04	1.37	0.73	0.44	0.36	0.36	0.10	0.13	–	2.78	38.55	20.54	40.91	0.58	0.44	0.35
Z110	Es3	3215.2–3276	II	1.06	1.45	0.83	0.52	0.32	0.29	0.09	0.03	0.65	4.24	41.33	21.42	37.25	0.47	0.38	0.26
Z78	Es3	3252–3262	II	1.03	1.12	0.87	0.53	0.35	0.33	0.09	0.06	0.59	3.77	37.15	20.62	42.23	0.42	0.44	0.29
Z242	Es3	3509.4–3063	II	1.03	1.20	0.65	0.41	0.44	0.40	0.09	0.07	0.80	4.02	30.18	20.08	49.74	0.59	0.50	0.31
Z125	Es3	–	II	1.03	1.50	0.80	0.44	0.45	0.39	0.08	0.05	0.46	5.29	35.91	21.35	42.74	0.50	0.41	0.30
Z2011	Es3	–	II	1.03	1.23	0.53	0.35	0.41	0.47	0.10	0.19	0.68	3.79	38.51	23.78	37.71	0.43	0.35	0.24

CPI = 1/2 $[(\sum(nC_{25} \sim nC_{33}))/(\sum(nC_{24} \sim nC_{32})) + (\sum(nC_{25} \sim nC_{33}))/(\sum(nC_{26} \sim nC_{34}))]$

CPI, carbon preference index; Pr/Ph, pristane/phytane; TT, tricyclic terpane; Ts, C_{27} 18α-trisnorneohopane; Tm, C_{27} 17α-trisnorhopane; H, hopanes; G, gammacerane; M, moretane; S, steranes

Fig. 4 Cross plots of geochemical parameters from oils discovered from different members of the Shahejie Formation in the Zhanhua Depression: Ternary plot showing the relationship between pristane/phytane, pristane/$n\mathrm{C}_{17}$ and phytane/$n\mathrm{C}_{18}$ (**a**); Pristane/$n\mathrm{C}_{17}$ ratio versus phytane/$n\mathrm{C}_{18}$ ratio (**b**); Ternary plot showing the relationship between tricyclic terpanes $\mathrm{C}_{19}+\mathrm{C}_{20}$, C_{21} and C_{23}, Zone 1: marine and saline source rocks and oils; Zone 2: fresh water source rocks and oils; Zone 3: swamp source rocks and oils; Zone 4: fluvial/delta source rocks and oils (Zone 1–4 after Xiao et al. 2019) (**c**); Pristane/phytane ratio versus gammacerane/C_{30} hopane (**d**); Ternary plot showing the relative distribution of C_{27}–C_{28}–C_{29} steranes in the studied oils (**e**); Tricyclic terpane $\mathrm{C}_{19}/\mathrm{C}_{23}$ ratio versus Hopanes/Steranes (**f**); C_{29} sterane 20S/(20S + 20R) ratio versus C29 sterane $\alpha\beta\beta/(\alpha\alpha\alpha+\alpha\beta\beta)$ (**g**); Ts/(Ts + Tm) ratio versus $\mathrm{C}_{35}/\mathrm{C}_{34}$ homohopane ratio (**h**)



The ratio of C_{30} moretane over C_{30} hopane ($C_{30}M/C_{30}H$) is less than 0.15 in all samples. The C_{30} 17β, 21α(H)-moretanes are thermally less stable than the C_{30} 17α, 21β(H)-hopanes and therefore the $C_{30}M/C_{30}H$ value decreases with increasing thermal maturity (Peters et al. 2005). Low $C_{30}M/C_{30}H$ values of all the oil samples suggest that they are in moderate-high maturity. The ratio of the 22*S*/(22*S* + 22*R*) for C_{32} 17α-hopane (C_{32}22*S*/(22*S* + 22*R*) Hopane) is in the range of 0.48–0.65, indicating the oils have reached equilibrium and are thermally mature (Mackenzie et al. 1984; Wang et al. 2018).

3.1.3 Steroids

The distributions of diasteranes and steranes are characterized by the *m*/*z* 217 in chromatograms (Fig. 3e, f). The proportions of regular $C_{27–29}$ steranes (%) have been used to indicate types of photosynthetic eukaryotes. C_{27} steranes are mainly derived from algae, diatom, and zooplankton in lacustrine deposits, C_{28} steranes are believed to be derived from lacustrine algae, while C_{29} steranes typically constitute the sterols of vascular plants (Volkman 2003; Peters et al. 2005; Albaghdady 2013). The distribution of C_{27}, C_{28}, and C_{29}ααα(20*R*) steranes is plotted on a ternary diagram (Fig. 4a). The dominance of both C_{27} and C_{29} steranes suggests a mixture of organic matter input from algae and terrestrial higher plants into the source rocks.

Diasteranes are present in oil samples from Es_1 and Es_3, but not detected in the Es_2 and Es_4 members. The ratios of C_{27}dia/(dia + reg sterane), defined as C_{27} diasterane/(diasterane + regular sterane), range between 0.22 and 0.35 in the samples contain diasteranes. Regular steranes convert to diasteranes with thermal maturity or acidic catalysis in oxic and clay-rich conditions (Rubinstein et al. 1975). The values of C_{27}dia/(dia + reg sterane) in oil samples from Es_1 and Es_3 may suggest moderate thermal maturity or suboxic and clay-poor conditions.

The hopane/sterane ratio ranges between 0.56 and 11.56, with an average value of 3.51 in all the oil samples. The hopane/sterane ratio is calculated as the sum of $C_{27–35}$ 17α-hopanes homologs over the sum of $C_{27–29}$ααα(20S + 20R) and αββ(20S + 20R). The hopane/sterane ratio is generally used as an indicator of prokaryotic inputs versus eukaryotic inputs of organic matter into the source rocks (Ourisson and Albrecht 1992). The high value of hopane/sterane in all the oil samples suggests large input of bacterial organic matter or bacterial modification of organic matter.

Isomerization at C-20 in C_{29} 5α, 14α, 17α-steranes causes an increase of 20S/(20S + 20R) ratio with increasing thermal maturity while isomerization at C-14 and C-17 in the C_{29} 20S and 20R regular steranes causes an increase of ββ/(ββ + αα) ratio with increasing thermal maturity (Seifert and Moldowan 1986).The ratios of C_{29} 20*S*/(20*R* + 20*S*) and C_{29}αββ/(ααα + αββ) steranes of all the oils are in the range of 0.34–0.59 and 0.61–0.85, indicating that the analyzed oils are thermally mature.

3.2 Oil family classification

The oils in this study are classified into two families and each of them is further subdivided into two groups based on biomarker and compound-specific carbon isotopic compositions. Families I and II have 9 and 22 oil samples, respectively. The Family I oils are mainly from Es_2 and Es_4 members of Shahejie Formation while Family II oils occur in the Es_1 and Es_3 reservoirs at diverse depths in oilfields in the Zhanhua Depression.

Family I is characterized by a low Pr/Ph value of 0.25–0.63 and a relatively high $Ga/C_{30}H$ ratio of 0.05–0.48 (Fig. 4d). The Ph/*n*-C_{18} ratio is higher than the Pr/*n*-C_{17} ratio in the Family I oils (Fig. 4a). The ratio of hopanes/steranes in the Family I has a wide range between 0.56 and 11.56, whereas C_{19}/C_{23} tricyclic terpane has a narrow range between 0.09 and 0.15 (Fig. 4f). Most samples in the Family I have a relatively higher abundance of C_{35} homohopanes than that of C_{34} homohopanes, with C_{35}/C_{34}homohopanes ratio of majority of samples larger than 1.0 (Fig. 4h). In addition, Ts/(Ts + Tm) values in the Family I are less than 0.3 (Fig. 4h) and no diasteranes have been detected in the Family I oils. Family II is marked by a high Pr/Ph value in the range of 0.9–1.59 and a low $Ga/C_{30}H$ ratio of less than 0.23 (Fig. 4d). The Ph/*n*-C_{18} ratio is lower than the Pr/*n*-C_{17} ratio in Family II (Fig. 4a, b). Moreover, the ratio of hopanes/steranes in Family II ranges from 1.55 to 6.36, whereas C_{19}/C_{23} tricyclic terpane has a wide range from 0.10 to 0.45 (Fig. 4f). Additionally, C_{35}/C_{34} homohopanes ratios of all oils in Family II are lower than 1 and Ts/(Ts + Tm) ratios of all oils in Family II are all higher than 0.2 (Fig. 4e). Diasteranes have been only detected in Family II; the ratios of C_{27} diasterane/(diasterane + regular) steranes are in the range of 0.22–0.31 (Table 1). Finally, C_{29} sterane 20*S*/(20*S* + 20*R*) ratios of oils in both Family I and II are higher than 0.3 and C_{29} sterane αββ/(ααα + αββ) ratios of oils in both Family I and II are higher than 0.25 (Fig. 4f).

3.3 Stable carbon isotopic compositions of individual n-alkanes

The long-chain *n*-alkanes are more stable than the bulk components of organic matters, so the $\delta^{13}C$ value of oils is very conservative parameters to reflect the original organic facies (Liu et al. 2006a, b). The stable carbon isotopic value of *n*-alkanes (*n*-$C_{13–35}$) in the 31 crude oils from Es_1 to Es_4 members ranges from − 23.87 to − 33.46 ‰

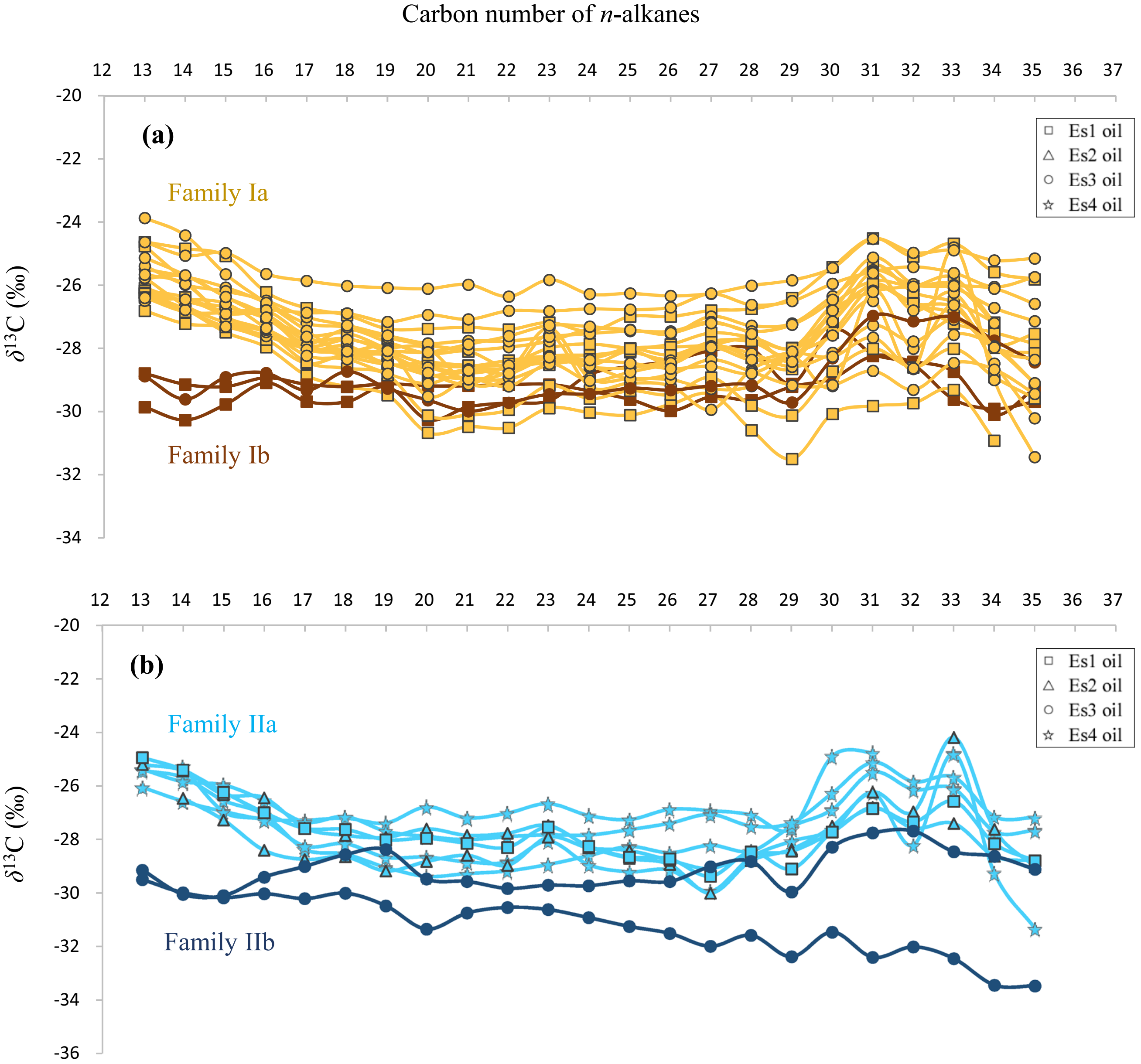


Fig. 5 The δ^{13}C values of individual *n*-alkanes for the crude oils from the Shahejie Formation in the Zhanhua Depression: Family I (**a**) and Family II (**b**)

(Fig. 5). The average δ^{13}C value of *n*-alkanesin Family I is − 28.2 ‰, slightly lower than that of Family II, which is − 27.8 ‰. Both Family I and Family II oils can be divided into two subfamilies a and b based on distribution patterns of carbon isotopic value of the *n*-alkanes. The δ^{13}C has an average value of − 29.3 ‰ for *n*-$C_{13–18}$ and shows a "U" pattern from *n*-C_{13} to *n*-C_{35} in Family Ia, whereas the δ^{13}C value has an average value of − 26.8 ‰ for *n*-$C_{13–18}$ and shows a flat pattern from *n*-C_{13} to *n*-C $_{35}$ in Family Ib (Fig. 5a).The *n*-$C_{13–18}$ show the lightest ^{13}C values in Family Ib, ranging from − 28.73 to − 30.27 ‰, which results in more negative ^{13}C values of Family Ib oils. In Family IIa, the overall pattern of the isotopic profile appears to be a "U" pattern, with an average δ^{13}C value of − 27.6 ‰ for *n*-$C_{13–35}$, slightly heavier than average δ^{13}C value of − 30.2 ‰ in Family IIb oils (Fig. 5b).The ^{13}C value of *n*-$C_{13–35}$ in Family IIb oils displays approximately a flat pattern, with an average δ^{13}C value of − 30.17 ‰. Both carbon isotope ratios of the *n*-alkanes of the oils from Family Ia and IIa are heavier than that of oils from Family Ib and IIb, indicating an increasing terrestrial organic matter input for Family Ia and IIa oils. Plank tonic algae has more negative ^{13}C values than that of terrestrial plants because concentrations of dissolved CO_2 are high in fluvial and lacustrine environments, and therefore more negative ^{13}C values may suggest a predominance of algal biomass, whereas more positive $\delta^{13}C$ value may suggest the dominance of input higher terrestrial plants into organic matter (Hollander and Mckenzie 1991; Mayer and Schwark 1999; Sun et al. 2000).

3.4 Inferred source-rocks for the oil families

Previous studies have proposed that the Es_1, lower Es_3, and upper Es_4 are the main source rocks of oils in the Zhanhua Depression (Zhu 2002; Wang 2011; Song and Men 2019; Table 2). They have different hydrocarbon generation potential due to various types of organic matter (Wang et al. 2005, 2010). The upper Es_4 deposits exhibit a prominent advantage of phytane over pristane and have a relatively high abundance of gammacerane and C_{35} homohopanes, with a "V" pattern of C_{27}, C_{28}, and C_{29}-ααα20R steranes. They were likely deposited in a brackish to a saline lacustrine setting (Table 2; Sun et al. 2015; Li et al. 2016; Song and Men 2019). However, the lower Es_3 deposits have a high Pr/Ph ratio and a low Ga/C_{30} hopanes ratio, with an "L" pattern of C_{27}, C_{28} and, C_{29}ααα20R steranes. They were likely deposited in a freshwater lacustrine setting (Table 2; Sun et al. 2015; Song and Men 2019). The Es_1 deposits have a low ratio of Pr/Ph and Ga/C_{30}H, with the relative abundance of C_{28}ααα20R steranes more than 30 %. They were likely deposited in a saline lacustrine setting (Table 2; Sun et al. 2015; Song and Men 2019). In addition, organic matter of Es_3 and Es_4 is mainly mature, while the organic matter of the Es_1 is still in the stage of immature (Song and Men 2019).

In this study, low Pr/Ph and relatively high Ga/C_{30}H ratiosin oils of Family I suggest the source rocks of the oils were probably derived from anoxic saline water conditions. High Ph/n-C_{18} and low Pr/n-C_{17} ratios of Family I in the ternary diagram of Pr/Ph, Ph/n-C_{18}, and Pr/n-C_{17} (Fig. 4a) suggest source rocks of Family I oils were likely originated from a brackish-saline lake. High Ph/n-C_{18} and low Pr/n-C_{17} ratios of Family I oils in cross plot of Pr/nC_{17} versus Ph/nC_{18} suggest source rock of Family I oils were likely developed in algal reducing environments (Fig. 4b). An extremely high ratio of hopane/sterane of Family I oils suggests strong bacterial input or bacterial modification of organic matter. A low ratio of C_{19}/C_{23} tricyclic terpane in Family I oils suggests a little contribution from terrestrial organic matter (Hanson et al. 2001). A high C_{35}/C_{34} hopane ratio in Family I suggests the source rocks of Family I may have been derived from highly reducing conditions. High ratios of C_{29} sterane 20S/(20S + 20R) and C_{29} sterane αββ/(ααα + αββ) in Family I oils suggest the oils are thermally mature. The low Ts/(Ts + Tm) value in the Family I may suggest reducing depositional environments (Moldowan et al. 1986), considering all oils are thermally mature. Overall, geochemical characteristics of the Family I oils match that of source rocks in upper Es_4 perfectly and the interpretation of the depositional conditions, organic matter input, and maturity level are in consistent for both the oils and the source rocks.

Relatively high Pr/Ph and low Ga/C_{30}H ratiosin Family II oils suggest the source rocks of Family II were probably derived from suboxic freshwater conditions. Low Ph/n-C_{18} and high Pr/n-C_{17} ratios of Family II in the ternary diagram of Pr/Ph, Ph/n-C_{18}, and Pr/n-C_{17} (Fig. 4a) suggest the source rocks of Family II oils were likely originated from a freshwater lake. Low Ph/n-C_{18} and high Pr/n-C_{17} ratios of Family II oils in cross plot of Pr/nC_{17} versus Ph/nC_{18} suggest source rock of Family II oils were likely developed in a transitional environment with mixed organic matter of algae and terrestrial higher plants (Fig. 4b). A high ratio of hopane/sterane suggests an important bacterial input or bacterial modification of organic matter. A high ratio of C_{19}/C_{23} tricyclic terpane in the Family II oils suggests an important contribution from terrestrial organic matter (Hanson et al. 2001; Volk et al. 2005). A low C_{35}/C_{34} hopane ratio in the Family II suggests the source rocks of Family II may have been derived from suboxic conditions. High ratios of C_{29} sterane 20S/(20S + 20R) and C_{29} sterane αββ/(ααα + αββ) ratio in Family II oils suggest the oils are thermally mature. The high Ts/(Ts + Tm) value in

Table 2 Diagnostic characteristics of the three main source rocks in the Zhanhua Depression

Parameters	Upper Es_4	Lower Es_3	Es_1	References
TOC	0.93 %–4.0 %	3.9 %–19 %	2.8 %–7.5 %	Song and Men (2019)
Type of kerogen	I, II_1	I, II_1	I	Song and Men (2019)
Ro	0.76 %–1.3 %	0.64 %–0.9 %	0.43 %–0.60 %	Song and Men (2019)
Pr/Ph	0.3–0.68	0.61–1.67	0.38–0.8	Song and Men (2019)
G/C_{30}H	0.2–0.56	0.04–0.18	0.53–1.6	Sun et al. (2015), Song and Men (2019)
Ts/Tm	< 1	> 1	< 0.8	Sun et al. (2015)
C_{29}20S/(20S + 20R)	> 0.42	> 0.35	< 0.32	Sun et al. (2015)

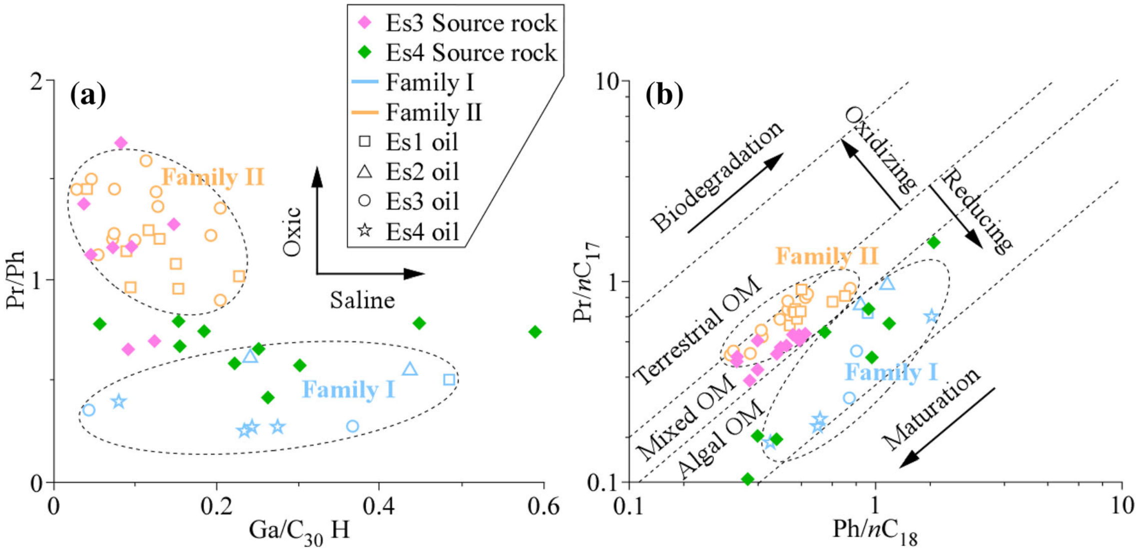


Fig. 6 Geochemical correlations for the oils and source rock samples: **a** Pristane/phytane ratio versus gammacerane/C_{30} hopane; **b** Pristane/nC_{17} ratio versus phytane/nC_{18} ratio. (Source rocks data originated from Liu et al. 2006a, b; Wang et al. 2010)

the Family II may suggest suboxic depositional environments (Moldowan et al. 1986), considering all Family II oils are thermally mature. Above all, geochemical characteristics and their interpretation of the Family II oils are inconsistent with that of source rocks in lower Es_3.

Based on the results and discussions above, we infer that Family I and II oils may have been derived from source rocks in upper Es_4 and lower Es_3, respectively. The results also supported by comparing the data with source rocks in lower Es3 and upper Es4 respectively (Fig. 6, source rocks data originated from Liu et al. 2006a, b; Wang et al. 2010).

The stable carbon isotopic values of the two families are very similar to each other because they shared similar kerogen types of Type I and Type II_1 for the source rocks in Es_4 and Es_1, respectively (Table 2). The stable carbon isotopic value of kerogen Type I is heavier than that of type II (Wang and Chen 2004).Thus, Family Ia and Family IIa oils were mainly controlled by the dominance of sapropelic matter input, while Family Ib and Family IIb oils were mainly originated from the humic substance. In summary, the subdivision of group a and b in Family I and II oils helps to better understand the process and mechanism of oil generation in a single family, and further assist in oil exploration.

4 Conclusion

Thirty-one crude oil samples in Shahejie Formation from in Zhanhua Depression, Bohai Bay Basin have been classified into two families on the basis of biomarkers and isotopic compositions. Family I oils are characterized by a low Pr/Ph value (< 0.7), relatively high Ga/C_{30}H and tricyclic terpane C_{22}/C_{21} ratio, and low C_{19}/C_{23} ratio. Family II oils are marked by a relatively high Pr/Ph value (0.7–1.6), low ratio of Ga/C_{30} H and C_{22}/C_{21} tricyclic terpane, and a wide range of C_{19}/C_{23} ratio. The Family I oils are largely collected from Es_2 and Es_4 members and were likely originated from the upper Es_4 source rocks, while the Family II oils are largely collected from Es_1 and Es_3 members and were likely derived from the lower Es_3 source rocks. Both the Families I and II oils can be further subdivided into two groups by compound-specific carbon isotopic compositions of *n*-alkanes.

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Compliance with ethical standards

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

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