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Original Paper

New alkylbenzene parameters to identify organic matter sources for source rocks of light oils and condensates from the Tarim Basin and Beibuwan Basin

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ABSTRACT

As an important component of light hydrocarbon compounds, alkylbenzene compounds lack indicators to indicate the source of organic matter of light oils and condensates. Forty-one oil samples from the Tarim Basin and Beibuwan Basin were analyzed by comprehensive two-dimensional gas chromatography time-of-flight mass spectrometry (GC × GC-TOFMS). The concentration distributions of thirteen light hydrocarbon compounds with organic matter source and sedimentary environment indication were studied. There is no significant difference in the concentrations of 1-methylpropylbenzene (MPB) in all studied oils. However, the concentrations of 2-MPB in the Tarim swamp oils are higher than that in the Beibuwan lacustrine oils and Tarim marine oils. Based on the significant concentration difference of 1- and 2-MPB in all studied oils, 1-/2-MPB (MPBr) was proposed as an indicator to identify the source of organic matter in crude oils. The MPBr values greater than 1.5 indicate that the crude oil mainly comes from lower aquatic organisms, bacteria, and algae. The MPBr values greater than 1.0 and less than 1.5 indicate that crude oil was derived from the combined contributions of lower aquatic organisms, bacteria and algae, and terrestrial higher plants. The MPBr values less than 1.0 suggest that the crude oil was mainly derived from terrigenous higher plants. The MPBr values in crude oils basically are not or slightly affected by depositional environment and secondary alteration. The MPBr values can be used to infer the organic matter origin in sediments, especially for the lack of biomarkers of light oils and condensates.

1. Introduction

Light hydrocarbons (LHs) usually account for about 30% of crude oil composition, and can account for 90% in light oils and condensates (Hunt et al., 1980). Therefore, LHs can represent the geochemical information of the whole light oils and condensates. Alkylbenzenes (monoaromatic hydrocarbons) compounds are important components of light hydrocarbon compounds (2–6 wt.%) (Hartgers et al., 1994a) and have been widely detected in various sediments, including crude oil, kerogen, coal, source rock extracts and bitumen (Gallegos, 1981; Ostroukhov et al., 1983; Summons and Powell, 1987; Hartgers et al., 1994a; Jia et al., 2007, 2008; Lis et al., 2008; Zhang et al., 2014a; Cheng et al., 2015a). Alkylbenzenes are widely used in petroleum geochemistry studies to identify crude oil groups from the same source rock, thermal maturity, and secondary alteration of crude oils (water washing, biodegradation and evaporative fractionation) (Volkman et al., 1984; Connan, 1984; Thompson, 1987, 1988; Sinninghe Damsté et al., 1993; Hartgers et al., 1994b, 1994c; Hill et al., 2004; Lis et al., 2008).

Alkylbenzenes are mainly produced by α, β and δ cleavage of the side chain of the aromatic moiety connected to the macromolecular structure (Hartgers et al., 1994a). Specific aromatic moieties usually have a specific biological origin. 1,3-/1,4-dimethylbenzene (DMB) and 1,2,3-trimethylbenzene (TMB) may originate from nonaromatic carotenoids, such as β,β-carotene (Hartgers et al., 1994a). 1,2,4-TMB may originate from plastoquinones, which is widely distributed in algae and cyanobacteria (Hartgers et al., 1994a). 1-methyl-4-isopropylbenzene (1-M-4-iPB) has a related to α-terpineol and

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limonene, which exist in terrestrial and marine depositional environments (Hartgers et al., 1994a; Yassaa et al., 2008). 1,2,3,4-tetramethylbenzene (TeMB) and 1,2,3,5-TeMB are derived from the aromatic carotenoids with specific substitution patterns linked to kerogen (Requejo et al., 1992; Hartgers et al., 1994b). In addition, 1,2,3,4-TeMB has an algal origin (Hoefs et al., 1995; Pedentchouk et al., 2004).

Studies on the biological origin of alkylbenzenes have been limited to possible source prediction based on similar structure and carbon isotope composition. The application of biological sources of alkylbenzenes is limited to the indication of sedimentary environments using isotopic signatures (Jia et al., 2007, 2008). The ratios of isopropyltoluene isomers have been proposed to indicate the source of organic matter in the sediments (Meng et al., 2022). Indications of the application of alkylbenzene compounds to the sources of organic matter in sediments are lacking and need to be further explored. In addition, because alkylbenzene compounds are usually analyzed by gas chromatography (GC) or GC-mass spectrometry (GC-MS), the co-elution of alkylbenzene compounds has seriously hindered the study of the potential geochemical significance of alkylbenzene compounds. However, the comprehensive two-dimensional gas chromatography time-of-flight mass spectrometry (GC × GC-TOFMS) has a good ability to separate alkylbenzene compounds and provides a good means to study the potential geochemical significance of alkylbenzene compounds (Meng et al., 2022, 2023).

In this study, a total of forty-one oil samples from different organic matter sources were analyzed by GC × GC-TOFMS, and the distribution characteristics of light hydrocarbons with indicative geochemical characteristics are analyzed. Combining with the established geochemical parameters of sedimentary environment and organic matter sources, the parameters of alkylbenzenes with organic matter sources were studied.

2. Samples and methods

2.1. Samples

A total of forty-one oil samples were collected from the Tarim Basin and Beibuwan Basin in China, including 26 oil samples from the Tarim Basin, 15 oil samples from the Beibuwan Basin. Sample detailed information is shown in Table 1.

2.1.1. Tarim marine and swamp oils

A total of twenty-six oil samples were collected from the Tarim Basin, including fifteen oil samples from the Ordovician reservoirs in the Halahatang depression of the Tabei uplift, six oil samples from the Luntai sub-uplift and five oil samples from the Kuqa depression (Fig. 1(a)). The API gravities of all studied oils indicates that these oils are light oils and condensates (Chang et al., 2013; Song et al., 2015). The light oils and condensates from the Halahatang depression and the Luntai sub-uplift were derived from the Carmbrian-Ordovician marine carbonate source rocks deposited under anoxic and reducing conditions (Chang et al., 2013). The light oils and condensates from the Kuqa depression originated from the thick Jurassic limnetic coal measures deposited in sub-oxic conditions (Liang et al., 2003; Fan et al., 2009; Zhang et al., 2011a; Zhu et al., 2012). All marine and swamp oils from the Tarim Basin are in mature to high-mature phase (Wang et al., 2008; Chang et al., 2013; Song et al., 2015).

2.1.2. Beibuwan lacustrine oils

A total of fifteen light oil and condensate samples were collected from the Fushan depression, Beibuwan Basin, South China Sea (Fig. 1(b)), including nine oil samples from the Huachang Oilfields, two oil samples from the Bohou-Chaoyang Oilfields, two oil samples from the Yong'an Oilfields, one oil sample from the Jinfeng Oilfeilds, and one oil sample from the Huadong Oilfileds. These oils are characterized by low density, low concentrations of steranes and terpenes biomarkers, and low content of asphaltenes and non-hydrocarbon components (Li et al., 2008a). The light oil and condensate samples mainly comes from organic-rich black mudstone and shale of the Liushagang Formation (E_2l), with type II and III kerogens (Li et al., 2008b). The source rocks of Liushagang formation (E_2l) are characterized by oxidizing sedimentary environment with dominant terrigenous organic matter input (Li et al., 2007, 2009). According to the methylphenanthrene index, the thermal maturity of crude oil is relatively high, and the equivalent R_o values is 0.9%–1.1%, which reflects the evolution stage of these oils from mature to high maturity (Li et al., 2008a).

2.2. Methods

The GC of the light oils and condensates was analyzed using an Agilent 6890A gas chromatography. The GC column is a fused silica (HP-PONA, 50 m × 0.2 mm × 0.5 μm). The injector temperature was 300 °C. The initial oven temperature was 35 °C, maintained for 5 min, then heated up to 70 °C at 3 °C/min, and to 300 °C at 4.5 °C/min, held for 35 min.

Saturated and aromatic hydrocarbons can be separated from all studied oils by column chromatography. The GC-MS of saturated and aromatic hydrocarbons were analyzed using an Agilent 6890A GC-5979i MS. The GC column is an HP-5MS (60 m × 0.25 μm × 0.25 μm). The programmed temperature of GC oven of saturated fraction was as follows: 50 °C (held for 1 min) to 100 °C at 20 °C/min, and then to 310 °C at 3 °C/min (maintain for 10 min); for aromatic fraction was as follows: 80 °C (held for 1 min) to 300 °C at 3 °C/min (maintain for 20 min).

The GC × GC-TOFMS of the light oils and condensates was analyzed using Agilent 7890 GC-Pegasus 4D TOFMS. The GC was equipped with two opposite phase columns, including non-polar capillary column (HP-PONA, 50 m × 0.20 mm × 0.50 μm) and polar capillary column (Rxi-17, 1.7 m × 0.1 mm × 0.1 μm) in the first and second dimensions, respectively. The programmed temperature of GC oven of the first dimensional column was as follows: 35 °C (held for 5 min) to 295 °C at 2 °C/min (maintain for 30 min); for the second dimensional column was as follows: 40 °C (held for 5 min) to 280 °C at 2 °C/min (maintain for 30 min). The modulation period was 6 s with 1.2 s hot pulse time. The detector voltage is 1350 V. The ion source voltage is 70 eV. Helium was used as carrier gas in the above experiments. All experimental analyses were performed in the National Key Laboratory of Petroleum Resources and Engineering.

3. Results

3.1. Identification of light hydrocarbon compounds in light oils and condensates

Thirteen light hydrocarbon compounds commonly used to determine organic matter source and sedimentary environment were detected and identified in light oil and condensate by comparing mass spectra, retention indices and retention times with published literature (Hartgers et al., 1992; Wang et al., 2014; Cheng et al., 2015a, 2015b; Meng et al., 2022, 2023). Fig. 2 shows thirteen light hydrocarbon compounds between *n*-nonane (nC_9) and *n*-dodecane (nC_{12}). Thirteen light hydrocarbon compounds include *n*-propylcyclohexane (*n*-PCHx), 2,6-dimethyloctane (2,6-DMO), 2-methyl-3-ethylheptane (2-M-3-EH), 1,1,2,3-tetramethylcyclohexane (1,1,2,3-TeMCHx), 3-methylnonane (3-MN), 2-methylpropylbenzene

Table 1
Basic geological and geochemical information for light oils and condensates.

No.	Name	Location	Source age	Depositional environment of source rock	References
1	H1-01	Beibuwan	E	Lacustrine	Li et al. (2007, 2009)
2	H1-02	Beibuwan	E	Lacustrine	As above
3	H2	Beibuwan	E	Lacustrine	As above
4	H2-3	Beibuwan	E	Lacustrine	As above
5	H3	Beibuwan	E	Lacustrine	As above
6	H3-4	Beibuwan	E	Lacustrine	As above
7	H5-01	Beibuwan	E	Lacustrine	As above
8	H6	Beibuwan	E	Lacustrine	As above
9	HX4-01	Beibuwan	E	Lacustrine	As above
10	FCl-01	Beibuwan	E	Lacustrine	As above
11	FCl-02	Beibuwan	E	Lacustrine	As above
12	Y-1-1	Beibuwan	E	Lacustrine	As above
13	Y1-02	Beibuwan	E	Lacustrine	As above
14	JFM-1	Beibuwan	E	Lacustrine	As above
15	HD-1-1	Beibuwan	E	Lacustrine	As above
16	RP14-5X	Tarim	Є-O	Marine	Zhang and Huang (2005)
17	RP8-1	Tarim	Є-O	Marine	As above
18	RP8001	Tarim	Є-O	Marine	As above
19	XK501C	Tarim	Є-O	Marine	As above
20	XK8-1	Tarim	Є-O	Marine	As above
21	JY103C	Tarim	Є-O	Marine	As above
22	JY5H	Tarim	Є-O	Marine	As above
23	YM1	Tarim	Є-O	Marine	As above
24	YM2	Tarim	Є-O	Marine	As above
25	YM3	Tarim	Є-O	Marine	As above
26	YM4	Tarim	Є-O	Marine	As above
27	YM5	Tarim	Є-O	Marine	As above
28	YM102	Tarim	Є-O	Marine	As above
29	YJ1X	Tarim	Є-O	Marine	As above
30	YJ2X	Tarim	Є-O	Marine	As above
31	S3	Tarim	J	Swamp	Cheng et al. (2015b)
32	S3-1	Tarim	J	Swamp	As above
33	YL2-2	Tarim	J	Swamp	As above
34	YL2-1	Tarim	J	Swamp	As above
35	YD2-2	Tarim	J	Swamp	As above
36	YD2-3	Tarim	J	Swamp	As above
37	DW105	Tarim	J	Swamp	Zhang et al. (2011a)
38	DB2	Tarim	J	Swamp	As above
39	DB202	Tarim	J	Swamp	As above
40	KU1	Tarim	J	Swamp	Fan et al. (2009)
41	DN201	Tarim	J	Swamp	Zhu et al. (2012)

Notes: Є-O: Cambrian-Ordovician; J: Jurassic; E: Eocene.

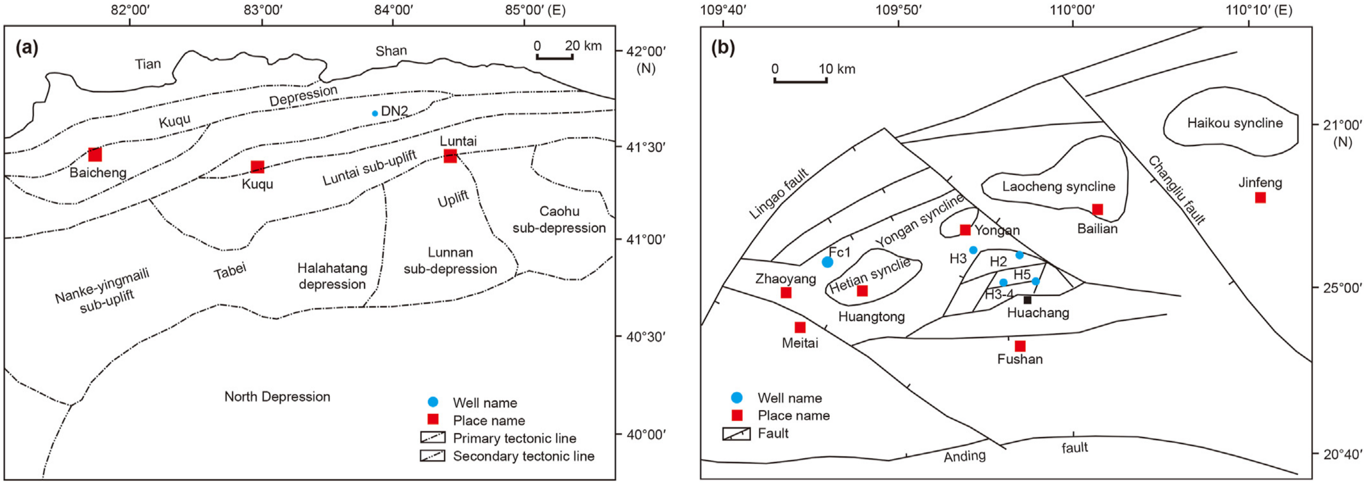

Fig. 1. The regional tectonic characters of **(a)** the northern Tarim Basin and **(b)** the Fushan depression of the Beibuwan Basin.

(MPB), 1-MPB, 1-methyl-3-isopropylbenzene (1-M-3-iPB), 1-M-4-iPB, 1-methyl-2-isopropylbenzene (1-M-2-iPB), 1,2,4,5-TeMB, 1,2,3,5-TeMB and 1,2,3,4-TeMB. The first dimension (1D) and second dimension (2D) retention time, the retention index of first dimension (1D RI), molecular mass (MW) and quantitative quality (QM) of thirteen light hydrocarbon compounds are shown in Table 2.

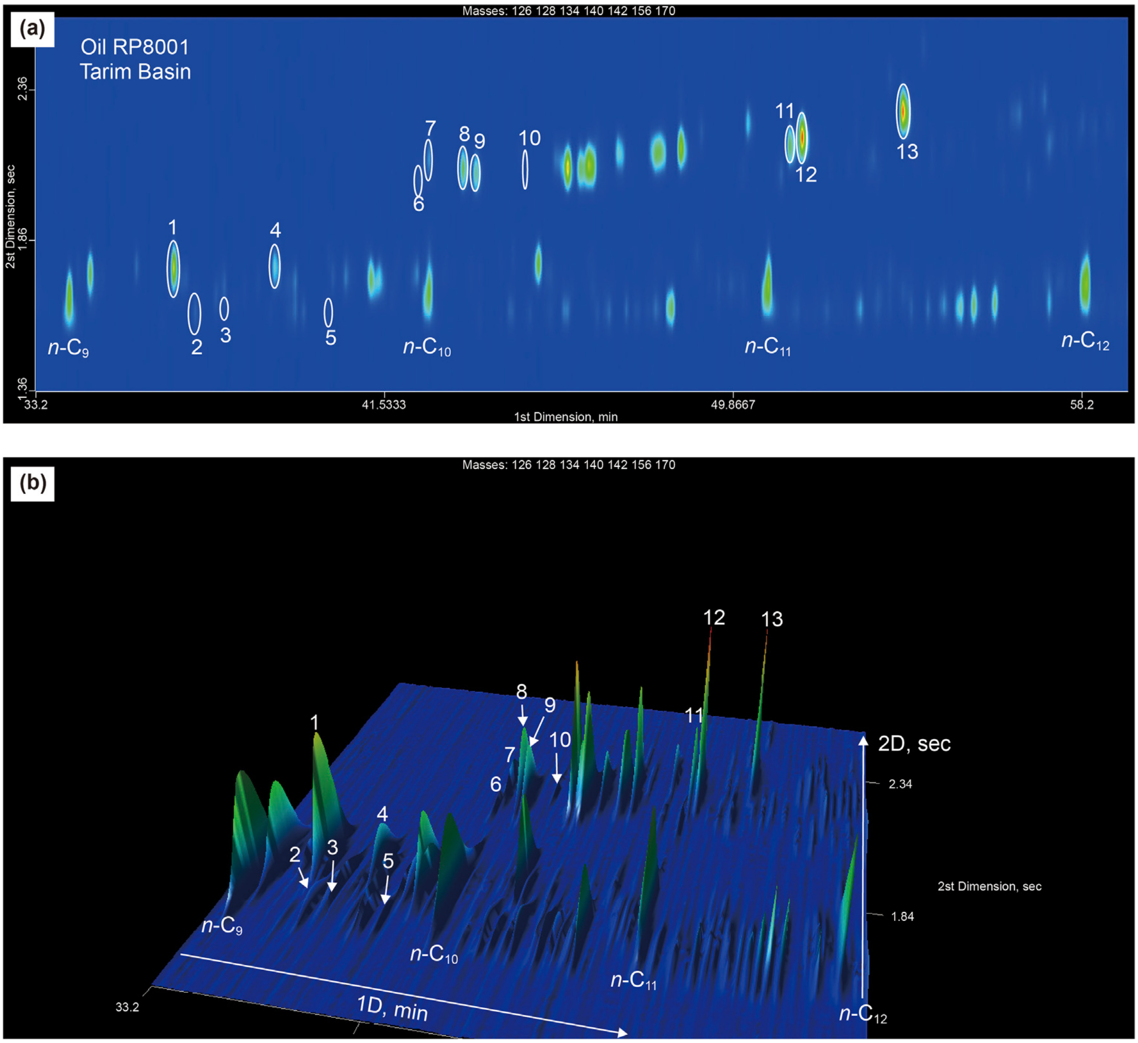

Fig. 2. Summed mass chromatogram of oil RP8001 from the Tarim Basin exhibits the distribution of thirteen light hydrocarbon compounds in GC × GC-TOFMS. The names of the compounds corresponding to peak numbers are listed in Table 2.

Table 2
Thirteen light hydrocarbon compounds were detected and identified in light oil and condensate.

No.	Compound name	1D	1D RI	2D	MW	QM	Abbreviation	References
1	*n*-propylcyclohexane	36.5	929.1	1.77	126	83	*n*-PCHx	Cheng et al. (2015a, 2015b)
2	2,6-dimethyloctane	37.0	934.9	1.62	142	57	2,6-DMO	As above
3	2-methyl-3-ethylheptane	37.7	943.0	1.64	142	57	2-M-3-EH	As above
4	1,1,2,3-Tetramethylcyclohexane	38.9	957.0	1.77	140	69	1,1,2,3-TeMCHx	As above
5	3-methylnonane	40.2	972.1	1.62	142	57	3-M-N	As above
6	2-Methylpropylbenzene	42.4	997.7	2.06	134	91	2-MPB	As above
7	1-Methylpropylbenzene	42.6	1000.0	2.13	134	105	1-MPB	As above
8	1-Methyl-3-isopropylbenzene	43.4	1009.9	2.10	134	119	1-M-3-iPB	As above
9	1-Methyl-4-isopropylbenzene	43.7	1013.6	2.08	134	119	1-M-4-iPB	As above
10	1-Methyl-2-isopropylbenzene	44.8	1027.2	2.15	134	119	1-M-2-iPB	As above
11	1,2,4,5-Tetramethylbenzene	51.2	1106.6	2.18	134	119	1,2,4,5-TeMB	As above
12	1,2,3,5-Tetramethylbenzene	51.5	1110.5	2.20	134	119	1,2,3,5-TeMB	As above
13	1,2,3,4-Tetramethylbenzene	53.9	1142.1	2.29	134	119	1,2,3,4-TeMB	As above

Notes: No: Peak number refers to Fig. 2; 1D: first dimension retention time (min); 1D RI: first dimension retention index; 2D: second dimension retention time (s); MW: molecular weight; QM: quantitative mass.

3.2. Distribution of light hydrocarbon compounds in light oils and condensates

The quantification of the concentration of light hydrocarbon compounds was based on the known quantity of the internal standard (1-hexene). The quantitative results of thirteen light hydrocarbon compounds concentrations are shown in Table 3. According to the distribution characteristics of the concentration of light hydrocarbon compounds, thirteen light hydrocarbon compounds can be divided into four groups.

Table 3
Concentration distribution of thirteen light hydrocarbon compounds.

No.	Name	Concentration, mg/g (in whole oil)												
		a	b	c	d	e	f	g	h	i	j	k	l	m
1	H1-01	2.29	3.88	1.06	0.64	3.61	0.49	0.59	1.73	0.93	0.15	2.72	2.87	1.67
2	H1-02	2.96	3.43	1.21	0.73	3.95	0.51	0.63	1.73	1.03	0.16	2.89	3.09	1.80
3	H2	0.73	2.26	0.94	0.40	2.82	0.24	0.39	1.25	0.58	0.12	2.94	4.34	2.61
4	H2-3	0.24	0.92	0.39	0.16	0.60	0.15	0.19	0.56	0.42	0.07	2.33	3.37	2.05
5	H3	2.51	3.62	1.48	0.64	3.58	0.42	0.61	1.34	0.94	0.14	2.20	3.25	1.92
6	H3-4	1.62	1.70	1.88	0.69	1.36	0.07	0.08	0.31	0.42	0.03	0.49	0.84	0.96
7	H5-01	3.26	2.49	0.58	0.16	2.74	0.41	0.53	0.00	0.91	0.00	3.57	5.14	2.14
8	H6	2.29	2.98	1.38	0.66	1.95	0.27	0.34	1.03	0.91	0.10	1.49	2.72	2.15
9	HX4-01	2.60	3.43	0.63	0.56	3.49	0.36	0.41	1.34	0.79	0.08	2.13	3.15	1.55
10	FCl-01	2.50	3.12	2.23	1.01	1.90	0.17	0.38	0.98	0.90	0.09	1.07	1.60	2.04
11	FCl-02	1.64	2.39	0.93	0.37	1.96	0.15	0.28	0.90	0.36	0.05	1.61	2.65	1.44
12	Y-1-1	2.77	5.22	3.15	1.35	1.63	0.37	0.41	0.33	0.37	0.13	1.94	1.20	1.31
13	Y1-02	2.65	2.43	1.56	0.65	2.50	0.29	0.45	1.39	0.91	0.13	1.46	1.94	1.61
14	JFM-1	4.42	4.77	3.05	1.55	3.26	0.59	0.63	2.22	1.50	0.20	2.47	3.62	2.66
15	HD-1-1	2.66	2.84	1.11	0.44	3.96	0.02	0.20	0.37	0.04	0.05	2.97	3.40	1.21
16	RP14-5X	1.78	5.01	4.50	2.21	3.13	0.36	0.64	0.59	0.72	0.08	0.61	1.08	1.26
17	RP8-1	2.01	5.74	3.80	1.76	3.18	0.32	0.54	0.74	0.57	0.07	0.53	1.01	1.30
18	RP8001	0.72	5.78	4.59	1.84	3.33	0.35	0.55	0.76	0.57	0.08	0.53	1.00	1.29
19	XK501C	1.37	3.63	3.88	1.11	2.05	0.18	0.39	0.39	0.42	0.06	0.35	0.76	1.21
20	XK8-1	1.89	4.91	4.70	1.92	2.85	0.24	0.45	0.58	0.57	0.07	0.50	0.97	0.92
21	JY103C	2.90	7.22	6.48	2.61	3.58	0.38	0.96	1.14	0.94	0.12	0.80	1.81	2.58
22	JY5H	1.66	5.56	4.98	2.06	2.67	0.26	0.62	0.80	0.71	0.06	0.40	1.15	2.30
23	YM1	2.35	6.77	4.56	2.51	3.50	0.33	0.58	0.84	0.73	0.09	0.60	1.19	1.17
24	YM2	2.26	6.34	5.70	2.56	3.49	0.32	0.81	1.06	0.87	0.09	0.59	1.41	2.42
25	YM3	2.53	6.31	4.86	2.21	3.50	0.40	0.66	1.02	0.80	0.10	0.67	1.31	1.28
26	YM4	0.74	6.09	4.29	2.44	3.37	0.33	0.71	1.04	0.79	0.09	0.71	1.45	2.07
27	YM5	2.06	6.27	3.80	2.11	3.61	0.31	0.48	0.63	0.77	0.08	0.63	1.16	1.12
28	YM102	1.52	5.82	4.38	2.21	3.01	0.21	0.57	0.93	0.74	0.07	0.51	1.32	1.91
29	YJ1X	1.88	5.04	3.85	1.83	2.97	0.32	0.69	0.87	0.46	0.07	0.58	1.19	1.67
30	YJ2X	2.44	5.99	5.55	1.79	3.31	0.35	0.60	0.93	0.39	0.09	0.63	1.17	1.08
31	S3	0.08	0.24	0.12	0.05	0.36	0.04	0.05	0.06	0.05	0.02	0.59	0.95	0.44
32	S3-1	2.29	3.96	1.61	1.17	3.09	0.93	0.70	0.86	0.41	0.29	1.24	1.74	0.83
33	YL2-2	3.34	3.53	1.21	1.16	3.69	1.31	0.18	0.37	0.09	0.30	0.39	0.54	0.29
34	YL2-1	1.50	2.80	0.47	0.65	2.65	0.39	0.08	0.07	0.03	0.12	0.26	0.32	0.20
35	YD2-2	1.45	2.36	1.37	0.62	2.74	0.47	0.27	0.36	0.61	0.19	0.61	0.77	0.47
36	YD2-3	1.73	3.50	1.46	0.83	3.35	1.03	0.32	0.28	0.64	0.34	0.76	0.90	0.44
37	DW105	1.77	2.51	1.38	0.64	2.49	0.83	0.57	0.61	0.38	0.34	1.01	1.26	0.79
38	DB2	2.20	4.04	1.81	0.92	3.96	1.47	0.82	0.75	0.37	0.65	1.11	1.08	0.89
39	DB202	1.41	2.48	0.87	0.44	3.74	0.99	0.48	0.63	0.19	0.39	0.71	0.88	0.64
40	KU1	1.39	1.80	0.99	0.81	2.21	0.68	0.20	0.17	0.07	0.16	0.46	0.72	0.58
41	DN201	1.77	2.96	0.56	0.70	3.84	0.88	0.17	0.11	0.11	0.31	0.23	0.77	0.40

Notes: a: *n*-PCHx; b: 2,6-DMO; c: 2-M-3-EH; d: 1,1,2,3-TeMCHx; e: 3-M-N; f: 2-MPB; g: 1-MPB; h: 1-M-3-iPB; i: 1-M-4-iPB; j: 1-M-2-iPB; k: 1,2,4,5-TeMB; l: 1,2,3,5-TeMB; m: 1,2,3,4-TeMB.

(1) In Fig. 3(a) and (b), the concentrations of 1,1,2,3-TeMCHx and 2-M-3-EH are high in the Tarim marine oils (1.11–2.61 (average 2.08) and 3.80–6.48 (average 4.66) mg/g whole oil, respectively), but there is no significant difference in the concentrations 1,1,2,3-TeMCHx and 2-M-3-EH between the Tarim Basin swamp oils (0.05–1.17 (average 0.73) and 0.12–1.81 (average 1.08) mg/g whole oil, respectively) and the Beibuwan lacustrine oils (0.16–1.55 (average 0.67) and 0.39–3.15 (average 1.44) mg/g whole oil, respectively), showing a relatively lower concentration (Table 3).

(2) In Fig. 3(c) and (d), the concentrations of 1,2,4,5-TeMB and 1,2,3,5-TeMB are relatively high in the Beibuwan lacustrine oils (0.49–3.57 (average 2.15) and 0.84–5.14 (average 2.88) mg/g whole oil, respectively), but there is no obvious difference in the concentrations of 1,2,4,5-TeMB and 1,2,3,5-TeMB in marine oils (0.35–0.80 (average 0.58) and 0.76–1.81 (average 1.20) mg/g whole oil, respectively) and swamp oils (0.23–1.24 (average 0.67) and 0.32–1.74 (average 0.90) mg/g whole oil, respectively) in the Tarim Basin, and the concentrations of 1,2,4,5-TeMB and 1,2,3,5-TeMB are relatively lower (Table 3).

(3) In Fig. 3(e) and (f), the concentrations of 2-MPB and 1-M-2-iPB in the Tarim swamp oils (0.04–1.47 (average 0.82) and 0.02–0.65 (average 0.28) mg/g whole oil, respectively) are higher than those in the Beibuwan lacustrine oils (0.02–0.59 (average 0.30) and 0.00–0.20 (average 0.10) mg/g whole oil, respectively) and the Tarim marine oils (0.18–0.40 (average 0.31) and 0.06–0.12 (average 0.08) mg/g whole oil, respectively), and there is no significant difference between the concentrations of 2-MPB and 1-M-2-iPB in the Beibuwan lacustrine oils and Tarim marine oils (Table 3).

(4) There was no significant difference in the concentrations of the remaining seven light hydrocarbon compounds (*n*-PCHx, 2,6-DMO, 3-M-N, 1-MPB, 1-M-3-iPB, 1-M-4-iPB, 1,2,3,4-TeMB) in the light oils and condensates (Table 3).

4. Discussion

4.1. Sources of light hydrocarbon compounds

The concentration difference of light hydrocarbons in light oils and condensates is directly related to the sedimentary environment and organic matter source of crude oils. Therefore, the concentration difference of light hydrocarbons in light oils and condensates deserve more attention. 2-M-3-EH is derived from limonene through the saturation of double bonds in the side chain and the breaking of double bonds in the ring (Mair et al., 1966). 1,1,2,3-TeMCHx may originate from the breakdown of carotenoids (Yabuta et al., 2002). Carotenoids may also be esterified with acids and then trapped in kerogen, where they undergo diagenesis to form 1,1,2,3-TeMCHx through reduction and cleavage (Wang et al., 2014). In addition, the concentrations of 1,1,2,3-TeMCHx and 2-M-3-EH in the Tarim marine oils formed in reduction environment are higher than those in the Beibuwan lacustrine oils and Tarim swamp oils formed in sub-oxidized/oxidized depositional environment (Table 3). The above studies indicate that 1,1,2,3-TeMCHx and 2-M-3-EH may be more likely to form in anoxic and reductive deposition environments (Wang et al., 2014; Cheng et al., 2015b).

1,2,3,5-TeMB is likely to be derived from macromolecularly-bound aromatic carotenoids, because their aromatic moieties possess specific substitution patterns relative to the isoprenoid chain, which yield upon β-cleavage 1,2,3,5-TeMB (Hartgers et al., 1994a). In Fig. 3(c) and (d), the concentration distribution of 1,2,4,5- and 1,2,3,5-TeMB in all studied oils is consistent, indicating that 1,2,4,5- and 1,2,3,5-TeMB have similar source (Table 3). For the sources of the Tarim marine oils and swamp oils, the organic matter input is obviously different, and the concentrations of 1,2,4,5- and 1,2,3,5-TeMB are similar (Fig. 3(c) and (d)). Therefore, the origin of organic matter may not be the main controlling factor for the formation of 1,2,4,5- and 1,2,3,5-TeMB. The Beibuwan Basin oils formed in strong oxidizing environment has higher 1,2,4,5- and 1,2,3,5-TeMB content ((Fig. 3(c) and (d)), which indicates that 1,2,4,5- and 1,2,3,5-TeMB may be inclined to form in strong oxidizing sedimentary environment. In addition, previous studies have shown that the 1,2,3,5-TeMB moieties could have been bound preferentially via oxygen linkages (Cheng et al., 2015a).

For the sources of the Beibuwan Basin oils and Tarim marine oils, the sedimentary environment is obviously different, and the concentrations of 2-MPB and 1-M-2-iPB are similar (Fig. 3(e) and (f)). Therefore, the sedimentary environment may not be the main controlling factor for the formation of 2-MPB and 1-M-2-iPB. The Tarim swamp oils from the terrigenous higher plants has higher 2-MPB and 1-M-2-iPB content (Fig. 3(e) and (f)), which indicates that 2-MPB and 1-M-2-iPB may be inclined to come from terrigenous higher plants. In addition, monoterpenoids are widely present in the secretory tissues of terrestrial higher plants, especially in the essential oils of terrestrial higher plants (Tissot and Welte, 1984). Therefore, 2-MPB and 1-M-2-iPB may derive mainly from similar skeletal monocyclic terpenes during diagenesis (Meng et al., 2022).

Although the types of light hydrocarbon compounds are generally present in crude oils from different sedimentary environments and organic matter sources, the concentrations of different compounds are different. Therefore, different combinations of light hydrocarbon compounds can indicate the organic matter source and sedimentary environment of crude oils. In this study, based on the concentration difference of 1- and 2-MPB in all studied oils (Table 3), 1-/2-MPB (MPBr) is proposed to distinguish the organic matter source of crude oils. MPBr values has a good correlation with other established geochemical parameters.

4.2. MPBr as an indicator of organic matter source

4.2.1. Effect of depositional environment on MPBr values

Pristane/phytane (Pr/Ph) is widely used to determine the sedimentary environment of organic matter (Didyk et al., 1978; Wang et al., 2022; You et al., 2023). Among alkanes, the ratios of 2-M-3-EH/3-MN (MT1), 1,1,2,3-TeMCHx/*n*-PCHx (MT2), 2,6-DMO/2-M-3-EH (MTR) have been proposed to indicate the redox conditions during the deposition of ancient sediments (Wang et al., 2014; Cheng et al., 2015a, 2015b). In Fig. 4, based on Pr/Ph, MT1, MT2, MTR and MPBr values, all studied light oils can be divided into three groups. The Tarim marine oils have the lowest Pr/Ph values, the relatively high MTR, MT1, MT2 and the highest MPBr values (1.57–2.75, average 2.02) (Table 4), indicating that the Tarim marine oils was formed in reductive and anoxic sedimentary environments. The Beibuwan lacustrine oils have the highest Pr/Ph values, lower MTR, MT1, MT2 and MPBr values (1.08–2.32, average 1.40) (Table 4), suggesting that the Beibuwan lacustrine oils was formed in oxidized sedimentary environment. The Tarim swamp oils have higher Pr/Ph values, lower MTR, MT1, MT2 and the lowest MPBr values (0.14–1.25, average 0.50) (Table 4), indicating that the Tarim swamp oils were formed in an oxic/dysoxic sedimentary environment.

In the alkylbenzene compounds, 1,2,3,5-/1,2,3,4-TeMB (TeMBr) and 1,2,4,5-/1,2,3,4-TeMB ratio have been proposed to distinguish the sedimentary environment of crude oils (Cheng et al., 2015a; Meng et al., 2023). In Fig. 5, all crude oils can be divided into three groups based on TeMBr, 1,2,4,5-/1,2,3,4-TeMB ratio combined with MPBr values. The Tarim marine oils have relatively low TeMBr, 1,2,4,5-/1,2,3,4-TeMB ratio, and the highest MPBr values (1.57–2.75, average 2.02) (Table 4), which imply their source rocks formed in reducing conditions. The Beibuwan lacustrine oils have higher TeMBr, 1,2,4,5-/1,2,3,4-TeMB ratio, and MPBr values (1.08–2.32, average 1.40) (Table 4), which indicate their source rocks formed in oxidization conditions. The Tarim swamp oils, with higher TeMBr, 1,2,4,5-/1,2,3,4-TeMB ratio, and the lowest MPBr values (0.14–1.25, average 0.50) (Table 4), was derived from source rocks formed in an oxic/dysoxic conditions.

The indicator parameters of sedimentary environment show that the Beibuwan lacustrine oils and the Tarim swamp oils for their source rocks were formed in oxidized sedimentary environment, while the Tarim marine oils for their source rocks was formed in reduced sedimentary environment. Therefore, the influence of sedimentary environment on MPBr value distribution difference in all studied oils cannot be completely ruled out. However, the Beibuwan Basin oils formed in strong oxidation sedimentary environment (Pr/Ph: 2.93–4.97) has relatively high MPBr value compared with the Tarim swamp oils formed in weak oxidation environment (Pr/Ph: 1.68–2.72), while the Tarim marine oils formed in strong reduction sedimentary environment (Pr/Ph: 0.82–1.05) has the highest MPBr value. In other words, there is no proportional relationship between the degree of redox in the sedimentary environment and the MPBr value. In addition, the MPBr values of the Beibuwan Basin oils and Tarim swamp oils, both formed in oxic/dysoxic sedimentary environment, are obviously different. Based on the above discussion, the difference in the distribution of MPBr values may not be mainly controlled by the depositional environment.

4.2.2. Influence of organic matter sources on MPBr values

The ratios of (1-M-3-iPB+1-M-4-iPB)/1-M-2-iPB (iPTr), 1-M-3-iPB/1-M-2-iPB (iPTr1) and 1-M-4-iPB/1-M-2-iPB (iPTr2) have been proposed to indicate the organic matter origin of crude oils (Meng et al., 2022). The Tarim marine oils, with the highest iPTr, iPTr1, iPTr2 and MPBr values (1.57–2.75, average 2.02) (Table 4), was derived from the lower aquatic organisms, bacteria, algae. The Beibuwan lacustrine oils have higher iPTr, iPTr1, iPTr2 and MPBr values (1.08–2.32, average 1.40) (Table 4), suggesting these oils was derived from the contribution of the lower aquatic organisms, bacteria, algae, and terrestrial higher plants. The Tarim swamp oils, with the lowest iPTr, iPTr1, iPTr2 and MPBr values (0.14–1.25, average 0.50) (Table 4), was derived from terrestrial higher plants.

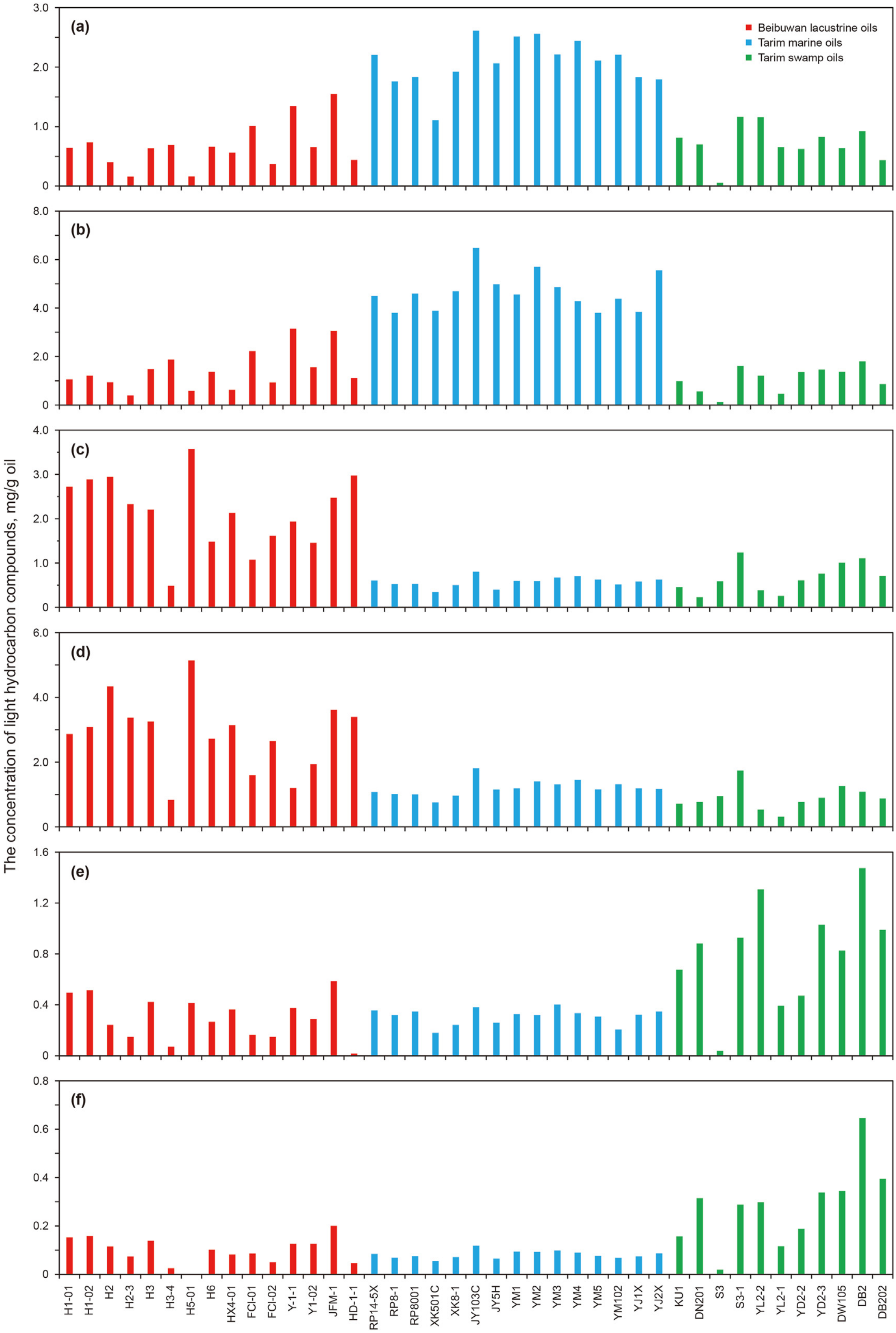

Fig. 3. The concentrations of six light hydrocarbon compounds are distributed in light oils and condensates of the Tarim Basin and Beibuwan Basin. **(a)** 1,1,2,3-TeMCHx; **(b)** 2-M-3-EH; **(c)** 1,2,4,5-TeMB; **(d)** 1,2,3,5-TeMB; **(e)** 2-MPB; **(f)** 1-M-2-iPB.

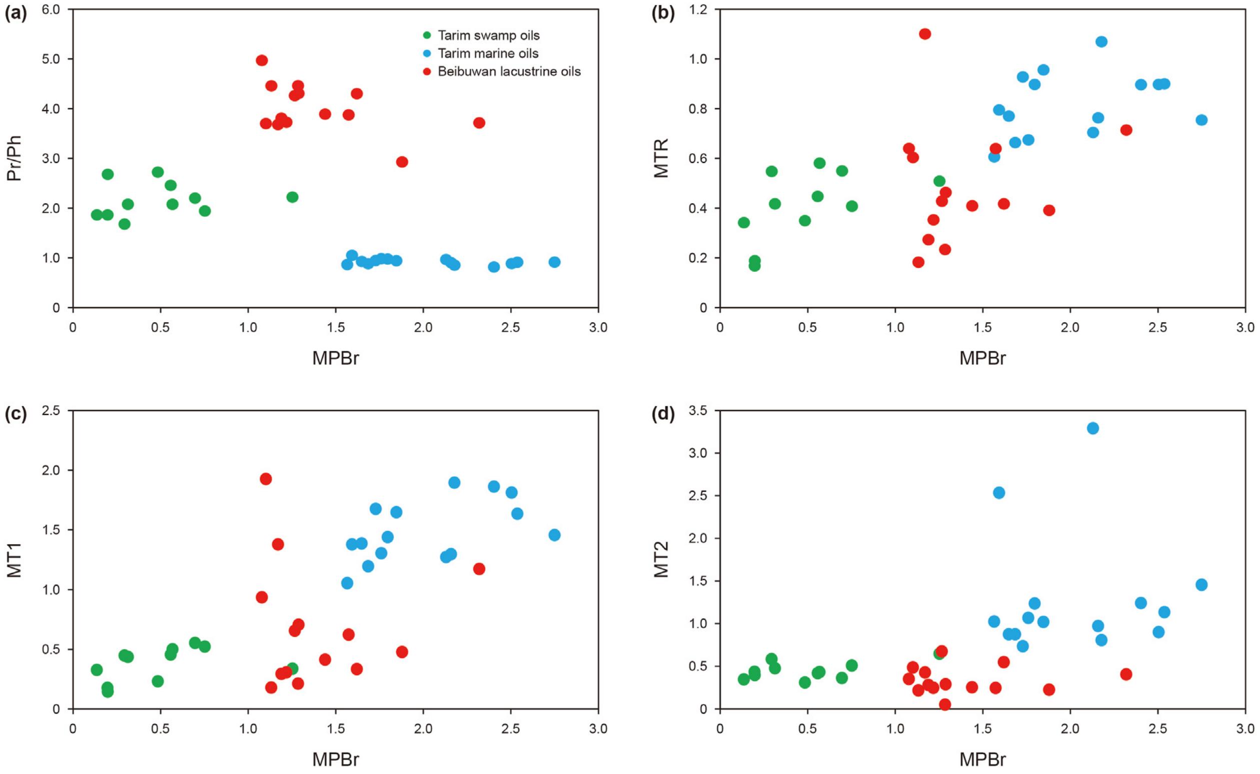

Fig. 4. Cross-plots of MPBr vs. Pr/Ph **(a)**, MTR **(b)**, MT1 **(c)** and MT2 **(d)** in all studied oils.

Therefore, all studied oils can be divided into three groups according to the difference of organic matter sources (Fig. 6).

The C_{15} bicyclic sesquiterpanes, including 2,2,4a,7,8-pentamethyl-decalin, 1,2,2,5,5-pentamethyl-trans-decalin, 1,1,2,5,5-pentamethyl-trans-decalin, were probably formed by degradation of oleanoids (Nytoft et al., 2009). Cadinane is considered as a reliable biomarker of terrigenous organic matter input (van Aarssen et al., 1992). In Fig. 7, the ratios of C_{15} bicyclic sesquiterpanes combined with MPBr values can well classify all studied oils into three groups. The Tarim marine oils have relatively low the ratios of C_{15} bicyclic sesquiterpanes, and the highest MPBr values (1.57–2.75, average 2.02) (Table 4). The Beibuwan lacustrine oils have higher the ratios of C_{15} bicyclic sesquiterpanes and MPBr values (1.08–2.32, average 1.40) (Table 4). The Tarim swamp oils have higher the ratios of C_{15} bicyclic sesquiterpanes and the lowest MPBr values (0.14–1.25, average 0.50) (Table 4).

The dominant *n*-heptane (nC_7) and methylcyclohexane ($MCyC_6$) are usually associated with the input of marine and terrigenous organic matter, respectively (ten Haven, 1996; Odden et al., 1998). Cycloalkanes (CyC_{5-7}) predominance is mainly distributed in terrigenous sedimentary organic matter, while marine sedimentary organic matter usually has *n*-alkanes (nC_{5-7}) predominance (Song et al., 2019). In Fig. 8, all studied oils can be divided into three groups by the ratios of $nC_7/MCyC_6$ and CyC_{5-7}/nC_{5-7} linked to MPBr values. The Tarim marine oils have relatively high $nC_7/MCyC_6$, low CyC_{5-7}/nC_{5-7} values, and the highest MPBr values (1.57–2.75, average 2.02) (Table 4). The Beibuwan lacustrine oils have lower $nC_7/MCyC_6$, higher CyC_{5-7}/nC_{5-7} values and MPBr values (1.08–2.32, average 1.40) (Table 4). The Tarim swamp oils have lower $nC_7/MCyC_6$, higher CyC_{5-7}/nC_{5-7} values and the lowest MPBr values (0.14–1.25, average 0.50) (Table 4).

The C_{21} tricyclic terpene (TT) and C_{23}TT predominance usually occur in marine and saline lacustrine source rocks (Aquino Neto et al., 1983; Tao et al., 2015; Xiao et al., 2019). The dominance of C_{19}TT and C_{20}TT are usually related to the contribution of terrigenous organic matter (Peters et al., 2005). In Fig. 9, all studied oils can be divided into three groups by (C_{19}TT + C_{20}TT)/C_{23}TT and MPBr values. The Tarim marine oils have the lowest (C_{19}TT + C_{20}TT)/C_{23}TT and the highest MPBr values (1.57–2.75, average 2.02) (Table 4). The Beibuwan lacustrine oils have lower (C_{19}TT + C_{20}TT)/C_{23}TT and higher MPBr values (1.08–2.32, average 1.40) (Table 4). The Tarim swamp oils have the highest (C_{19}TT + C_{20}TT)/C_{23}TT and the lowest MPBr values (0.14–1.25, average 0.50) (Table 4).

The anomalies in Fig. 7(b) are mainly due to the fact that the Tarim marine oils is light oil and condensate, which is in the stage of over-mature evolution, leading to the decomposition of macromolecular biomarkers to a certain extent, and even the extreme shortage of biomarkers in some crude oils (Table 4). Therefore, there is an inaccurate problem when the macromolecule biomarker compounds indicate the geochemical information of crude oil. In this study, alkylbenzene compounds can make up for the shortage of macromolecular biomarkers in light oil and condensate.

Based on the above discussion, there is a good correlation among MPBr values and organic matter source parameters in all studied oils. The Tarim marine oils have the highest MPBr values (>1.5), indicating that the crude oil mainly comes from lower aquatic organisms, bacteria, and algae. The Beibuwan lacustrine oils have relatively high MPBr (1.0<MPBr<1.5), indicating that crude oils were derived from the combined contributions of lower aquatic organisms, bacteria and algae, and terrestrial higher plants. The Tarim swamp oils have the lowest MPBr values (<1.0), which indicates that the crude oils were mainly derived from terrigenous higher plants. With MPBr values gradually approaching zero, the input proportion of terrestrial organic matter gradually increased, and the input proportion of lower aquatic organisms, algae and

Table 4
Geochemical indicators of sedimentary environment, organic matter sources, thermal maturity and secondary alteration of light oils and condensates.

No.	Name	R1	R2	R3	R4	R5	R6	R7	R8	R9	R10	R11	R12	R13	R14	R15	R16	R17	R18	R19	R20	R21	R22	R23	R24
1	H1-01	3.81	0.27	0.29	0.28	1.63	1.71	1.19	11.32	6.08	17.40	0.41	0.42	3.66	1.53	0.22	5.42	3.76	1.38	6.73	0.87	5.97	0.86	0.54	0.29
2	H1-02	3.73	0.35	0.31	0.25	1.60	1.71	1.22	10.93	6.52	17.45	0.33	0.16	1.71	1.18	0.40	2.25	3.19	0.80	6.44	0.87	5.64	0.85	0.52	0.28
3	H2	4.30	0.42	0.33	0.55	1.13	1.66	1.62	10.73	4.96	15.69	0.40	0.34	3.11	1.46	0.44	2.28	3.48	0.89	5.80	0.85	5.97	0.86	0.51	0.28
4	H2-3	4.26	0.43	0.66	0.67	1.13	1.64	1.27	7.59	5.61	13.20	0.43	0.41	3.85	1.55	0.53	2.29	4.20	0.94	6.73	0.87	5.99	0.86	0.53	0.28
5	H3	3.89	0.41	0.41	0.25	1.15	1.69	1.44	9.61	6.74	16.35	0.45	0.42	3.58	1.55	0.18	6.87	3.41	0.86	5.52	0.85	5.41	0.84	0.50	0.27
6	H3-4	3.68	1.10	1.38	0.43	0.51	0.87	1.17	12.39	16.75	29.13	0.31	0.28	2.27	1.27	0.73	1.18	2.50	0.06	1.58	0.61	2.88	0.74	0.39	0.21
7	H5-01	4.46	0.23	0.21	0.05	1.67	2.40	1.28	/	/	/	0.62	0.55	4.06	1.28	0.41	2.83	3.15	0.77	4.32	0.81	3.68	0.79	0.47	0.24
8	H6	4.31	0.46	0.71	0.29	0.69	1.26	1.29	10.06	8.92	18.98	0.40	0.27	2.20	1.14	0.43	1.47	3.23	0.60	3.08	0.75	3.56	0.78	0.46	0.24
9	HX4-01	4.46	0.18	0.18	0.22	1.38	2.04	1.13	16.23	9.52	25.74	0.38	0.40	2.99	1.34	0.48	1.95	3.77	0.61	4.56	0.82	4.16	0.81	0.45	0.24
10	FCl-01	3.71	0.71	1.17	0.41	0.53	0.78	2.32	11.38	10.41	21.78	0.42	0.39	2.12	1.21	0.52	1.46	3.40	0.30	2.05	0.67	3.01	0.75	0.45	0.23
11	FCl-02	2.93	0.39	0.48	0.22	1.12	1.84	1.88	18.00	7.29	25.29	0.35	0.23	2.63	1.31	0.26	4.72	2.17	0.61	3.81	0.79	5.13	0.84	0.44	0.23
12	Y-1-1	3.70	0.60	1.93	0.49	1.48	0.92	1.10	2.59	2.89	5.48	0.33	0.26	1.79	1.12	0.44	3.17	4.10	0.26	2.82	0.74	4.30	0.81	0.47	0.24
13	Y1-02	3.87	0.64	0.62	0.25	0.90	1.20	1.57	10.99	7.15	18.14	0.40	0.42	2.16	1.30	0.43	2.20	1.93	0.62	4.13	0.80	5.31	0.84	0.52	0.27
14	JFM-1	4.97	0.64	0.94	0.35	0.93	1.36	1.08	11.09	7.47	18.56	0.32	0.37	2.61	1.34	0.42	2.34	3.55	0.73	3.66	0.79	4.30	0.81	0.50	0.27
15	HD-1-1	3.65	0.39	0.28	0.16	2.45	2.80	/	8.06	0.83	8.89	0.41	0.40	4.19	1.72	0.35	3.13	5.69	2.66	12.05	0.92	7.34	0.88	0.61	0.32
16	RP14-5X	0.98	0.90	1.44	1.24	0.48	0.86	1.80	7.02	8.51	15.53	0.03	0.09	0.45	0.35	1.71	0.64	0.90	0.17	9.72	0.91	9.78	0.91	0.47	0.27
17	RP8-1	0.88	0.66	1.20	0.88	0.41	0.78	1.69	10.74	8.26	19.00	0.07	0.11	0.57	0.40	1.75	0.58	0.81	0.20	7.44	0.88	9.12	0.90	0.46	0.26
18	RP8001	1.05	0.79	1.38	2.53	0.41	0.78	1.59	9.99	7.50	17.49	0.06	0.14	0.54	0.40	1.72	0.73	0.90	0.21	7.90	0.89	9.21	0.90	0.46	0.26
19	XK501C	0.85	1.07	1.90	0.81	0.29	0.63	2.18	7.09	7.48	14.58	0.01	0.08	0.30	0.27	1.64	0.58	0.62	0.19	6.13	0.86	6.44	0.87	0.47	0.27
20	XK8-1	0.94	0.96	1.65	1.02	0.55	1.05	1.85	8.05	7.93	15.99	0.05	0.09	0.54	0.40	1.73	0.56	0.52	0.22	13.98	0.93	12.11	0.92	0.49	0.28
21	JY103C	0.89	0.90	1.81	0.90	0.31	0.70	2.50	9.57	7.91	17.48	0.03	0.09	0.33	0.30	1.75	0.59	0.69	0.14	6.43	0.87	7.20	0.88	0.43	0.25
22	JY5H	0.82	0.90	1.86	1.24	0.17	0.50	2.40	12.32	11.04	23.37	0.03	0.09	0.38	0.31	1.79	0.59	0.69	0.14	5.28	0.84	7.15	0.88	0.41	0.24
23	YM1	0.98	0.67	1.30	1.07	0.51	1.02	1.76	8.89	7.74	16.63	0.06	0.17	0.71	0.47	1.72	0.55	1.59	0.23	15.57	0.94	12.56	0.93	0.51	0.30
24	YM2	0.91	0.90	1.63	1.13	0.25	0.58	2.54	11.37	9.30	20.67	0.01	0.07	0.30	0.26	2.04	0.52	0.72	0.08	5.99	0.86	7.14	0.88	0.42	0.25
25	YM3	0.93	0.77	1.39	0.87	0.53	1.03	1.65	10.29	8.09	18.38	0.07	0.14	0.65	0.47	1.74	0.60	1.36	0.26	15.28	0.94	12.65	0.93	0.50	0.30
26	YM4	0.97	0.70	1.27	3.29	0.34	0.70	2.13	11.54	8.80	20.34	0.05	0.09	0.40	0.31	1.98	0.50	0.77	0.17	8.72	0.90	8.52	0.89	0.48	0.28
27	YM5	0.87	0.61	1.05	1.02	0.56	1.03	1.57	8.29	10.06	18.35	0.06	0.13	0.75	0.50	1.75	0.52	0.81	0.25	16.46	0.94	13.19	0.93	0.51	0.30
28	YM102	0.92	0.75	1.46	1.46	0.27	0.69	2.75	13.56	10.78	24.35	0.02	0.08	0.28	0.25	1.98	0.53	0.71	0.11	8.02	0.89	7.92	0.89	0.46	0.27
29	YJ1X	0.90	0.76	1.30	0.97	0.35	0.71	2.16	11.67	6.11	17.77	0.09	0.10	0.36	0.27	1.86	0.54	0.77	0.26	7.85	0.89	8.78	0.90	0.45	0.26
30	YJ2X	0.95	0.93	1.68	0.73	0.58	1.08	1.73	10.66	4.51	15.17	0.08	0.13	0.61	0.44	1.77	0.51	1.24	0.31	13.82	0.93	12.47	0.93	0.50	0.29
31	S3	2.22	0.51	0.34	0.65	1.34	2.17	1.25	2.81	2.73	5.55	0.68	0.24	3.81	1.54	0.39	3.18	/	3.02	/	/	/	/	/	/
32	S3-1	1.94	0.41	0.52	0.51	1.50	2.10	0.75	2.98	1.42	4.40	0.49	0.11	3.43	1.44	0.19	0.70	/	1.96	28.94	0.97	34.27	0.97	0.73	0.38
33	YL2-2	1.86	0.34	0.33	0.35	1.35	1.88	0.14	1.23	0.30	1.53	0.28	0.31	3.56	1.83	0.35	0.41	/	0.49	15.05	0.94	24.81	0.96	0.53	0.30
34	YL2-1	1.86	0.17	0.18	0.44	1.28	1.56	0.20	0.58	0.25	0.82	/	/	/	/	0.35	0.41	/	0.49	15.05	0.94	24.81	0.96	0.53	0.30
35	YD2-2	2.08	0.58	0.50	0.43	1.31	1.65	0.57	1.89	3.23	5.12	0.31	0.18	2.04	1.18	/	/	3.26	/	9.62	0.91	13.55	0.93	0.51	0.29
36	YD2-3	2.08	0.42	0.44	0.48	1.74	2.05	0.31	0.84	1.90	2.74	0.35	0.21	2.86	1.52	/	1.40	4.73	/	/	/	/	/	/	/
37	DW105	2.20	0.55	0.55	0.36	1.27	1.59	0.70	1.78	1.09	2.87	/	/	/	/	/	/	/	/	/	/	/	/	/	/
38	DB2	2.46	0.45	0.46	0.42	1.25	1.22	0.56	1.17	0.58	1.74	/	/	/	/	0.69	1.50	/	2.32	/	/	/	/	/	/
39	DB202	2.72	0.35	0.23	0.31	1.10	1.37	0.48	1.59	0.48	2.07	/	/	/	/	0.51	1.96	/	2.71	/	/	/	/	/	/
40	KU1	1.68	0.55	0.45	0.58	0.78	1.23	0.29	1.08	0.43	1.51	0.58	0.26	3.46	1.31	0.34	3.06	/	3.55	9.45	0.90	12.09	0.92	0.37	0.21
41	DN201	2.68	0.19	0.15	0.40	0.58	1.93	0.20	0.34	0.36	0.70	0.32	0.26	4.24	1.67	/	/	/	/	/	/	/	/	/	/

Notes: R1: Pr/Ph; R2: MTR; R3: MT1; R4: MT2; R5: 1,2,4,5/1,2,3,4-TeMB; R6: TeMBr; R7: MPBr; R8:iPTr1; R9: iPTr2; R10: iPTr; R11: cadinane/(cadinane+8β(H)-drimane); R12: 2,2,4a,7,8-pentamethyl-decalin/(2,2,4a,7,8-pentamethyl-decalin+8β(H)-drimane); R13: 1,2,2,5,5-pentamethyl-trans-decalin/(1,2,2,5,5-pentamethyl-trans-decalin +8β(H)-drimane); R14 : 1,1,2,5,5-pentamethyl-*trans*-decalin/(1,1,2,5,5-pentamethyl-*trans*-decalin+8β(H)-drimane); R15: nC_7/$MyCC_6$; R16: CyC_{5-7}/nC_{5-7}; R17: ($C_{19}TT$ + $C_{20}TT$)/$C_{23}TT$; R18: Tol/nC_7; R19: MDR; R20: MDR'; R21: EDR; R22: EDR'; R23: F1; R24: F2;/: no data. Geochemical parameters calculation method and compound complete name were listed in Meng et al. (2023). Items R2~R10 are obtained through GC × GC-TOFMS analysis, and other data are obtained through GC-MS analysis.

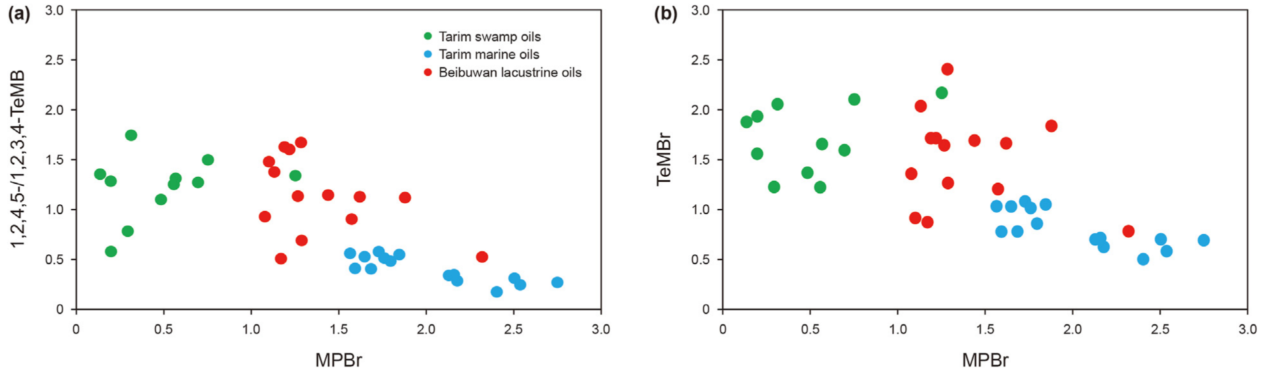

Fig. 5. Cross-plots of MPBr vs. 1,2,4,5-/1,2,3,4-TeMB (**a**) and TeMBr (**b**) in all studied oils.

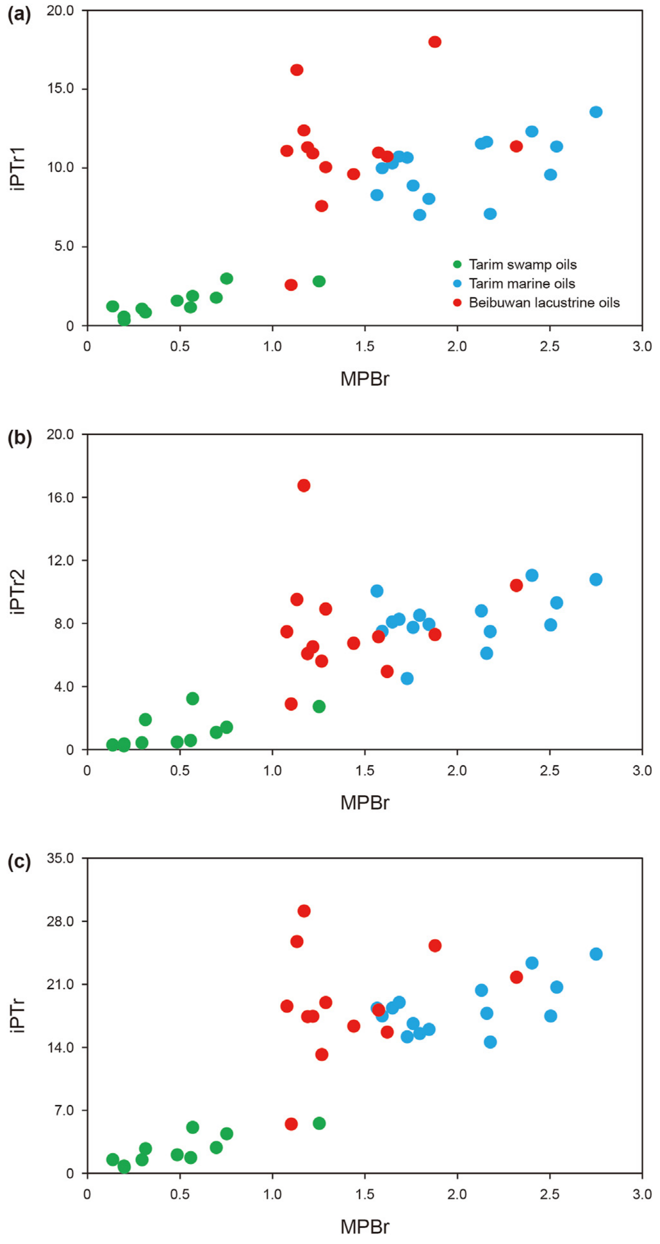

Fig. 6. Cross-plots of MPBr vs. iPTr1 (**a**), iPTr2 (**b**), and iPTr (**c**) in all studied oils.

bacteria gradually decreased.

4.3. Secondary alteration influence on MPBr values

Different physicochemical processes usually lead to significant changes in crude oil composition, including evaporative fractionation, biodegradation, and thermal maturity (Evans et al., 1971; Peters and Fowler, 2002). Therefore, the secondary changes will affect the reasonable and correct use of light hydrocarbon geochemical parameters. It is very important to understand the influence of secondary changes on the identification parameters of light hydrocarbons.

4.3.1. Evaporative fractionation

Evaporative fractionation refers to the conversion of low and medium molecular weight hydrocarbons into vapor phase under suitable physical conditions (Thompson, 1987, 1988). In order to quantitatively evaluate the intensity of evaporative fractionation of crude oil, the F ($n\mathrm{C}_7$/MCyC6) and B (Tol/$n\mathrm{C}_7$) were proposed by Thompson (1988). When evaporative fractionation occurs in crude oils, the B and F values in crude oils will deviate from the F (>1.0) and B (<0.5) values of normal oils (Thompson, 1987). The values of F and B in the Tarim marine oils are 1.64–2.04 (average 1.80) and 0.08–0.31 (average 0.20) respectively (Table 4), indicating that Tarim marine oils has not experienced evaporative fractionation. Previous studies have shown that marine oils from the Tabei uplift in the Tarim Basin has undergone a certain degree of evaporative fractionation (Zhang et al., 2000, 2011b, 2015). Compared with the Tarim marine oils, the Beibuwan lacustrine oils and the Tarim swamp oils have lower F value (0.18–0.73 and 0.19–0.69, respectively) and higher B values (0.06–2.66, 0.49–3.55, respectively). The F values of crude oil depend on the input of organic matter and thermal maturity. The low F values in the Beibuwan lacustrine oils and the Tarim swamp oils may be influenced by the input of terrigenous organic matter. Benzene and toluene compounds are more abundant in terrigenous organic matter than in marine sedimentary organic matter (Leythaeuser et al., 1979a, 1979b). Therefore, the B values in the Beibuwan lacustrine oils and the Tarim swamp oils are higher than the Tarim marine oils. In addition, the determining factors for a compound to enter the vapor phase are structure and molecular weight. The 1-MPB and 2-MPB have the same molecular weight and similar structure, therefore, MPBr values is basically not affected by evaporation fractionation. The correlation analysis between the concentration of 1-MPB and 2-MPB or the MPB ratio and F or B values shows that there is no good correlation between the above parameters, which further indicates that MPBr values is basically not affected by evaporative fractionation.

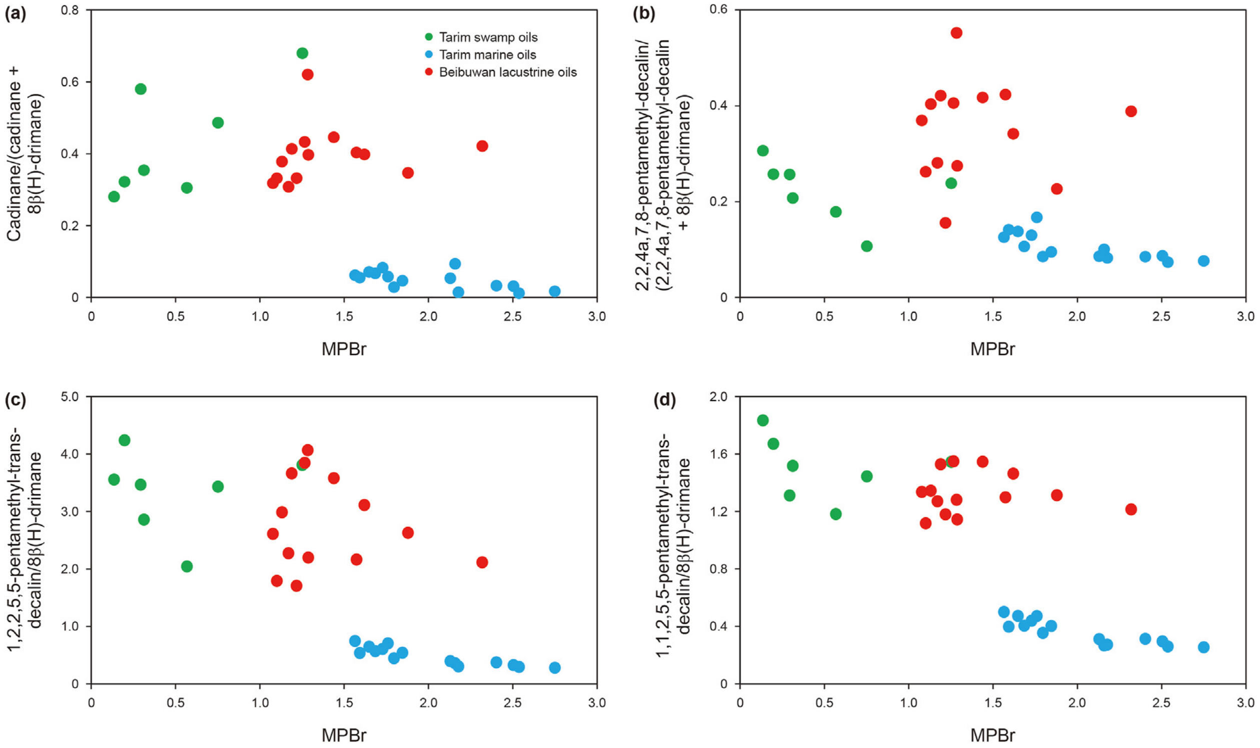

Fig. 7. Cross-plots of MPBr vs. cadinane/(cadinane+8β(H)-drimane) **(a)**, 2,2,4a,7,8-pentamethyl-decalin/(2,2,4a,7,8-pentamethyl-decalin+8β(H)-drimane) **(b)**, 1,2,2,5,5-pentamethyl-*trans*-decalin/(1,2,2,5,5-pentamethyl-*trans*-decalin +8β(H)-drimane) **(c)**, 1,1,2,5,5-pentamethyl-*trans*-decalin/(1,1,2,5,5-pentamethyl-*trans*-decalin +8β(H)-drimane) **(d)** in all studied oils.

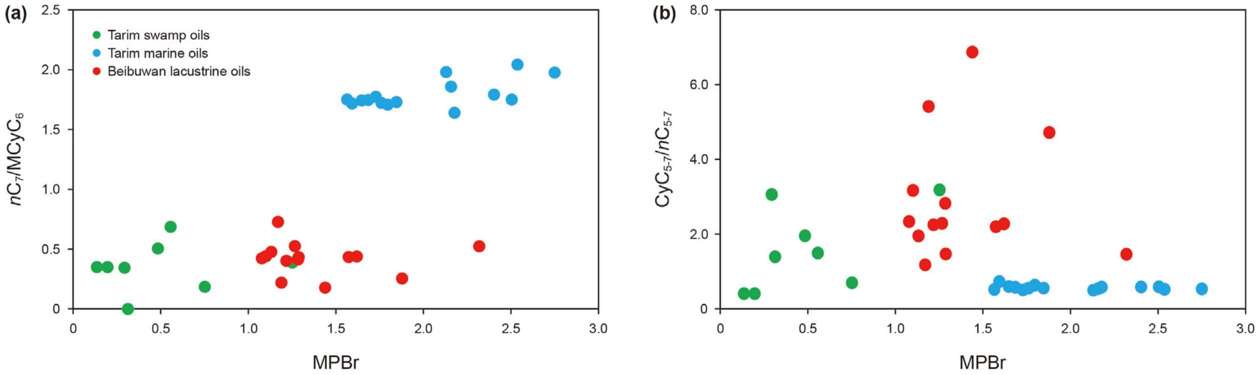

Fig. 8. Cross-plots of MPBr vs. nC_7/$MCyC_6$ **(a)** and $CyC_{5\text{-}7}$/$nC_{5\text{-}7}$ **(b)** in all studied oils.

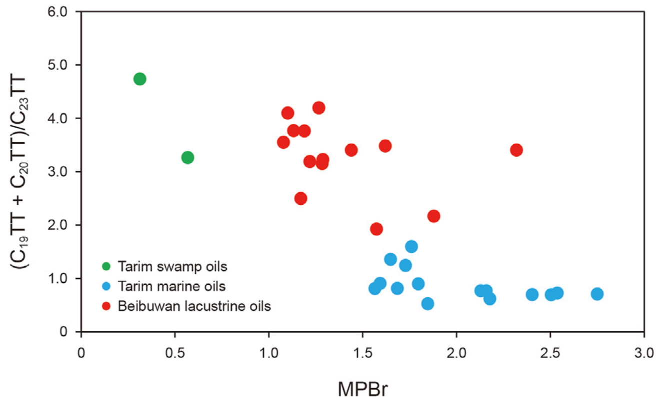

Fig. 9. Cross-plots of MPBr vs. (C_{19}TT + C_{20}TT)/C_{23}TT in all studied oils.

4.3.2. *Biodegradation*

Biodegradation usually occurs in shallow formations, resulting in changes in the oil composition, such as reduced API gravities values and increased viscosity (Peters and Fowler, 2002). In addition, the 25-norhopane is believed to be formed by the biodegradation of crude oils, so the compound is used as an important indicator for the biodegradation of crude oils (Evans et al., 1971; Peters and Fowler, 2002). Although 25-norhopane has been detected in the Tarim marine oils, it is generally considered to be the remain of palaeobiodegraded oils preserved in late charged fresh oils (Chang et al., 2013; Zhang et al., 2014b). No obvious 25-norhopane series compounds were detected in the Beibuwan lacustrine oils and the Tarim swamp oils, suggesting these oils did not undergo biodegradation. The rate of biodegradation of aromatic compounds is inversely proportional to the number of aromatic rings and directly proportional to the number of alkyl substituents.

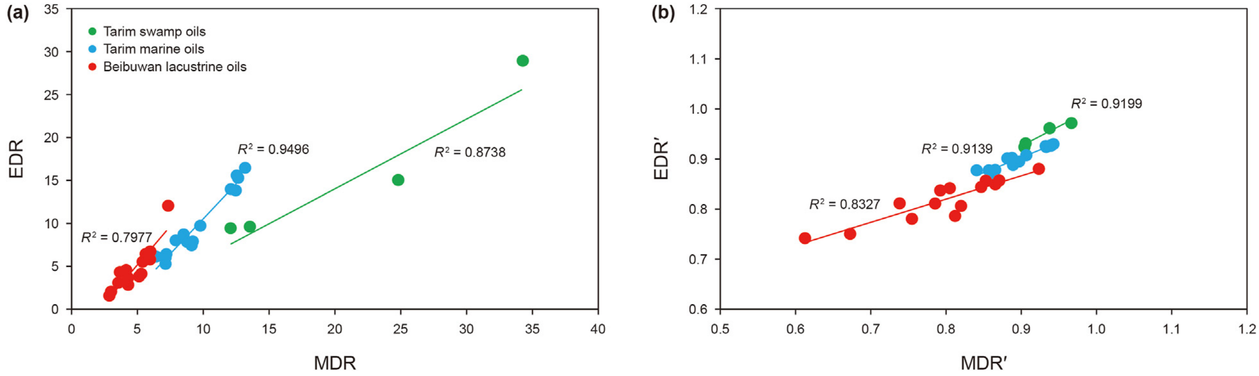

Fig. 10. Cross-plots of MDR vs. EDR (**a**) and MDR' vs. EDR' (**b**) for all studied oils.

Thus, the 1- and 2-MPB have similar sensitivity to biodegradation. The MPBr values may not be affected by biodegradation.

4.3.3. *Thermal maturity*

According to the different thermal stability of dibenzothiophene compounds in different thermal evolution stages, dibenzothiophene parameters (MDR, EDR, MDR' and EDR') have been proposed to characterize the thermal maturity of crude oils (Radke and Willsch, 1994; SantamaríA-Orozco et al., 1998). In Fig. 10, the maturity of the Tarim marine oils is lower than that of the Tarim swamp oils, but higher than that of the Beibuwan lacustrine oils. In addition, the ratios of methylphenanthrene isomers (F1 and F2) are used to indicate the degree of thermal evolution of sedimentary organic matter (Kvalheim et al., 1987). In Fig. 11, all studied oils are generally in mature evolution stage, and some the Beibuwan lacustrine oils and the Tarim swamp oils are in high mature evolution stage. Therefore, the differences in MPBr values in all studied oils that are not affected by thermal maturity cannot be completely excluded. Because the thermal maturity of the Beibuwan lacustrine oils is similar to that of the Tarim marine oils, but lower than that of the Tarim swamp oils, the MPBr values of the Beibuwan lacustrine oils is higher than that of the Tarim swamp oils, but lower than that of the Tarim marine oils. In addition, there was no correlation between the concentrations of 1-MPB and 2-MPB or the MPBr and the above maturity parameters. Therefore, the difference of MPBr values is not controlled by thermal maturity in crude oils.

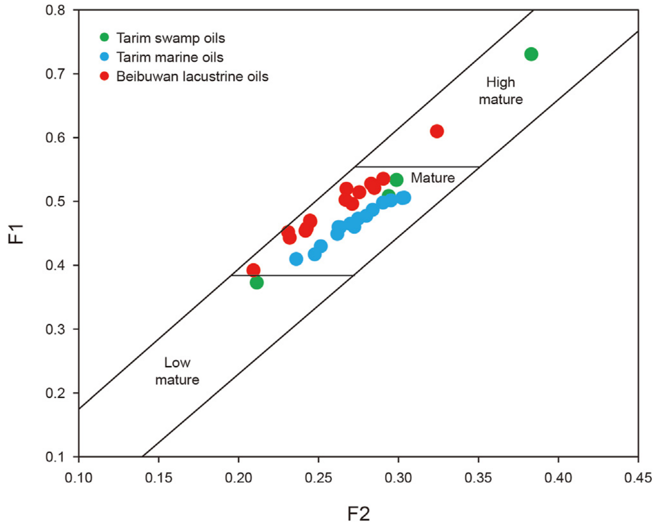

Fig. 11. Cross-plots of F1 vs. F2 in all studied oils.

5. Conclusions

Forty-one oil samples from the Tarim Basin and Beibuwan Basin were analyzed by GC × GC-TOFMS, and the concentrations distribution of thirteen light hydrocarbons with organic matter source and sedimentary environment indication were analyzed. The research results show that 1,1,2,3-TeMCHx and 2-M-3-EH may be more likely to form in anoxic and reductive deposition environments. 1,2,4,5- and 1,2,3,5-TeMB are more likely to form in the oxidized sedimentary environment. The 2-MPB and 1-M-2-iPB are mainly derived from terrigenous higher plants. The concentrations of the remaining seven light hydrocarbons showed no significant difference in all studied oils. Based on the concentration difference of 1- and 2-MPB in all studied oils, MPBr values is proposed to distinguish the organic matter sources of crude oils. The MPBr values greater than 1.5 indicate that the crude oil mainly comes from lower aquatic organisms, bacteria, and algae. The MPBr values less than 1.0 and greater than 1.5 indicate that crude oil was derived from the combined contributions of lower aquatic organisms, bacteria and algae, and terrestrial higher plants. The MPBr values less than 1.0 suggest that the crude oil was mainly derived from terrigenous higher plants. With MPBr values gradually approaching zero, the input proportion of terrestrial organic matter gradually increased, and the input proportion of lower aquatic organisms, algae and bacteria gradually decreased. The MPBr values in crude oils basically are not or slightly affected by depositional environment and secondary alteration (biodegradation, evaporative fractionation and thermal maturity).

CRediT authorship contribution statement

Bing-Kun Meng: Writing – review & editing, Writing – original draft, Software, Resources, Methodology, Investigation, Funding acquisition, Conceptualization. **Dao-Fu Song:** Supervision, Resources, Funding acquisition. **Yuan Chen:** Software, Investigation. **Sheng-Bao Shi:** Software, Methodology.

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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