



## Organic geochemistry of Carboniferous source rocks and their generated oils from the Eastern Junggar Basin, NW China



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### ABSTRACT

Carboniferous source rocks have been gaining increasing attention after the discovery of the Kelameili gasfield in the Eastern Junggar Basin in 2005. Two sets of source rocks within the Lower Carboniferous Dishuiquan Formation ( $C_{1d}$ ) and Upper Carboniferous Batamayineishan Formation ( $C_{2b}$ ), respectively, have been identified in this area. In this paper, clear differences between these two source rocks are demonstrated in molecular and carbon isotopic compositions. The  $C_{1d}$  source rocks, in comparison with the  $C_{2b}$  source rocks, have remarkably lower  $Pr/n-C_{17}$  ratio, higher  $Ts/(Ts + Tm)$  and  $C_{30}$  diahopane/ $(C_{30}$  diahopane +  $C_{30}$  hopane) ratios, lower concentration of  $C_{24}$  tetracyclic terpane relative to  $C_{23}$  and  $C_{26}$  tricyclic terpanes and higher gammacerane/ $C_{31}$  hopane ratio.  $\delta^{13}C$  values of individual  $n$ -alkanes decrease with carbon number for  $C_{1d}$  source rocks while the opposite is true for  $C_{2b}$  source rocks.

Among the 10 oils collected from the studied area, four were generated exclusively from  $C_{1d}$  source rocks while the others were mainly derived from  $C_{1d}$  source rocks but contaminated by a small or trace amount of oil components which were derived from the  $C_{2b}$ , Middle Permian Pingdiqian Formation ( $P_{2p}$ ) and Lower Jurassic source rocks.

The free, adsorbed and inclusion oils from three oil-containing volcanic reservoir rocks within the  $C_{2b}$  formation generally correlate with  $C_{1d}$  source rocks based on molecular and carbon isotopic data. However, the three types of oil from the same reservoir rock vary significantly in molecular and carbon isotopic compositions, reflecting facies and maturity changes of charging oil during the reservoir filling.

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### 1. Introduction

The Junggar Basin is a major oil and gas producing province of China. Numerous studies have demonstrated that the most important source rocks in the basin are within the Permian strata and for most reservoirs, especially in the northwestern area, oils are dominantly derived from three Permian source rocks, which from old to young are: (1) the Lower Permian Jiamuhe Formation, (2) the Lower Permian Fencheng Formation, and (3) the Middle Permian Lower Wuerhe Formation in the northwestern area and its age-equivalent analogs Pingdiqian Formation in the eastern area and the Lucaogou Formation in the southern basin margin (Jiang and Fowler, 1986; Jiang et al., 1988a,b, 1990; Zhou et al., 1989; Graham et al., 1990; Carrol et al., 1992; Yang et al., 1992; Zhang et al., 1993; King et al., 1994; Clayton et al., 1997; Carroll, 1998;

Wang and Kang, 1999, 2001; Pan and Yang, 2000; Pan et al., 2003; Cao et al., 2005, 2006).

The Carboniferous strata widely occur in the Junggar Basin and its neighboring areas and contain marine clastic source rocks which are mostly postmature (Zhao, 1994). Wang (1996) suggested that (1) the Carboniferous source rocks mainly generated oils during the Carboniferous and Permian, which were hardly preserved due to the Hercynian tectonic movements in the basin, and (2) in some areas of the basin, they were possibly within the oil generation window and generated oils during and after the Triassic Period, which can be partly preserved. Several studies have documented the geochemical characteristics of oils that were generated from the Carboniferous source rocks based on limited oil samples from boreholes in the Eastern Junggar Basin with oil shows, and demonstrated that these oils are characterized by heavy carbon isotopes, with whole oil  $\delta^{13}C$  values ranging from  $-25\%$  to  $-23\%$ , and molecular compositions similar to terrigenous organic matter and coal (Yang and Li, 1992; Yang et al., 2002; Chen et al., 2003a). The Carboniferous source rocks have gained increasing attention due

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to the discovery and exploration of the Kelameili gasfield in the Eastern Junggar Basin during 2005–2008, the largest gasfield so far found in this basin with in-place gas reserves of over  $100 \times 10^9 \text{ m}^3$  (Da et al., 2010; Wang et al., 2010). This gasfield contains gaseous hydrocarbons with a dryness ratio ( $C_1/\Sigma C_{1-4}$ ) ranging from 0.88–0.98 and condensates and light oils with densities  $< 0.85 \text{ g/cm}^3$  (Da et al., 2010). Gas reservoirs of this field are mainly within the volcanic reservoir rocks of the Upper Carboniferous Batamayineishan Formation ( $C_{2b}$ ) with minor in the Jurassic and Cretaceous sandstones (Da et al., 2010). It has been suggested that both gaseous and liquid hydrocarbons in this field were mainly derived from the Carboniferous source rocks based on geological setting (Da et al., 2010; Wang et al., 2010). Two Carboniferous source rocks have been identified in the Eastern Junggar Basin, i.e., the Lower Carboniferous Dishuiquan Formation ( $C_{1d}$ ) and Upper Carboniferous Batamayineishan Formation ( $C_{2b}$ ) (Yang and Li, 1992; Yang et al., 2002; Chen et al., 2003a; Da et al., 2010; Wang et al., 2010). So far, the differences in molecular and isotopic compositions between these two sets of Carboniferous source rocks have not been well documented and the origins of oils within the Kelameili gasfield have not been clearly understood. In the present study, we document organic geochemical characteristics of these two sets of Carboniferous source rocks and oils in the Eastern Junggar Basin based on bulk and molecular parameters, and carbon isotopic compositions of individual *n*-alkanes. In addition, as demonstrated by previous studies, analysis of oil-bearing fluid inclusions and sequential extraction of oil-containing reservoir rocks are effective approaches to reveal geochemical signatures of the initial oil charges (Karlsen et al., 1993; Wilhelms et al., 1996; George et al., 1997, 1998, 2004, 2008; Schwark et al., 1997; Jones and Macleod, 2000; Leythaeuser et al., 2000, 2007; Pan and Yang, 2000; Pan et al., 2000, 2003, 2005, 2007; Gong et al., 2007; Yu et al., 2011, 2012). A large set of oil-containing volcanic reservoir rocks from the Kelameili gasfield were also analyzed to evaluate the source variations during the reservoir filling. We hope that the results of the present study could enhance our understanding on the origin of oil and gas components of the Kelameili gasfield in the Eastern Junggar Basin.

## 2. Geological setting

The Junggar Basin is situated in the north of Xinjiang Uygur Autonomous Region of China (Fig. 1), and is a composite stacked basin of the central landmass type, characteristic of the Precambrian crystalline and the lower Paleozoic bi-layer basement (Zhao, 1992a,b). After the basement of the pre-Carboniferous was shaped, the evolutionary history of the Junggar Basin can be divided into four stages in general: (1) Late Carboniferous to Early Permian: foreland marine-residual marine basin; (2) Middle to Late Permian: interior foreland basin; (3) Triassic to Eogene: intra-craton downwarped basin; and (4) Neogene to Quaternary: reactivated collisional foreland basin (Graham et al., 1990; Song Yan, 1995; Chen et al., 2003a). The area studied in this paper lies in the eastern part of the basin (Fig. 1).

The generalized stratigraphy in the studied area is demonstrated in Fig. 2. The Carboniferous strata include the Lower Carboniferous Tamugang Formation ( $C_{1t}$ ) and Dishuiquan Formation ( $C_{1d}$ ), and Upper Carboniferous Batamayineishan Formation ( $C_{2b}$ ), Shijiantan Formation ( $C_{2s}$ ), and Liukeshu Formation ( $C_{2l}$ ) (Zhang et al., 1993; Wang et al., 2010). The Tamugang Formation contains pyroclastic sediments, mudstones and carbonates deposited in a paralic environment (Zhang et al., 1993). The Dishuiquan Formation ( $C_{1d}$ ) mainly contains dark grey mudstones with minor sandstones deposited in a paralic environment with a thickness of about 1000 m (Zhang et al., 1993; Wang et al., 2010). The Batamayineishan Formation ( $C_{2b}$ ) contains grayish green basalts,

andesites, tuffs with minor grey sandstones, dark grey mudstones, carbonaceous mudstones and coals deposited in a terrigenous environment with the largest thickness  $> 4 \text{ km}$  (Zhang et al., 1993; Wang et al., 2010). The Shijiantan Formation ( $C_{2s}$ ) mainly contains marine clastic sediments and bioclastic limestones (Zhang et al., 2007; Wu et al., 2008). The Liukeshu Formation ( $C_{2l}$ ) mainly contains coarse clastic sediments with minor limestones, tuffs and liparites (Zhang et al., 2007; Wu et al., 2008).

The Permian strata include the Lower Permian Jingou Formation ( $P_{1j}$ ), Middle Permian Jiangjunmiao Formation ( $P_{2j}$ ) and Pingdiquan Formation ( $P_{2p}$ ) and Upper Permian Lower Cangfanggou Group ( $P_{3c}$ ) (Zhang et al., 2007). The Jingou Formation ( $P_{1j}$ ) contains grayish-green tuffaceous conglomerates and sandstones, and grey mudstones (Zhang et al., 1993). The Jiangjunmiao Formation ( $P_{2j}$ ) consists of fluvial and deltaic deposits, red mudstones, conglomerates and sandy mudstones. The Pingdiquan Formation consists of conglomerates, sandstones and dark grey mudstones and oil shales deposited in fluvial, delta and lacustrine environments. The Lower Cangfanggou group ( $P_{3c}$ ) contains mainly sandstones and mudstones deposited in fluvial and shallow lacustrine environments (Zhang et al., 1993).

The Triassic and Jurassic strata contain mainly sandstones, mudstones, carbonaceous mudstones and coals deposited in fluvial and shallow lacustrine environments while the Cretaceous, Tertiary and Quaternary strata contain mainly alluvial and fluvial clastic sediments (Zhang et al., 1993).

The major source rocks in the studied area are within the Lower Carboniferous Dishuiquan Formation ( $C_{1d}$ ) and Upper Carboniferous Batamayineishan Formation ( $C_{2b}$ ). The thickness of source rocks within the Dishuiquan Formation ranges from 60–300 m drilled by boreholes and is about 300 m in the Dishuiquan outcrop (Wang et al., 2010). The thickness of source rocks within the Batamayineishan Formation ranges from 23–520 m drilled by boreholes (Wang et al., 2010). The Middle Permian Pingdiquan Formation ( $P_{2p}$ ) contains oil prone source rocks with high quality in the regions south and southeast of the studied area, as documented by Chen et al. (2003a). However, this set of source rocks scarcely occurs and is insignificant in the studied area due to facies variation (Da et al., 2010; Wang et al., 2010). The Lower Jurassic Badaowan and Sangonghe formations and Middle Jurassic Xishanyao Formation contain some source rocks, such as dark grey mudstones, carbonaceous mudstones and coals (e.g., Chen et al., 2003a,b). In the studied area, the maturities of these Jurassic source rocks are very low ( $\%Ro < 0.5$ ) and have not yet reached the oil generation window (Da et al., 2010; Wang et al., 2010).

## 3. Samples and experimental

### 3.1. Samples

Source rocks (69 samples), oil-containing reservoir rocks (38) and oils (10) were collected in the present study (Tables 1 and 2, Fig. 1). For the 69 source rocks, 60 samples, from young to old, DSQ-S1 to DSQ-S60, were collected from the Lower Carboniferous Dishuiquan Formation ( $C_{1d}$ ) from Dishuiquan outcrop. The other nine samples were collected from the Upper Carboniferous Batamayineishan Formation ( $C_{2b}$ ) from boreholes DX8 and Q5.

For the 10 oils, six were from volcanic reservoirs within the Upper Carboniferous Batamayineishan Formation ( $C_{2b}$ ) while the other four oils were collected from the sandstone reservoirs within the Upper Permian, Upper Triassic, Lower Jurassic and Lower Cretaceous strata (Table 2 and Fig. 1). The 38 oil-containing volcanic reservoir rocks were collected from the Upper Carboniferous Batamayineishan Formation ( $C_{2b}$ ) from boreholes D402, D403 and DX18, but only three of them are presented in this paper (Table 2 and Fig. 1).

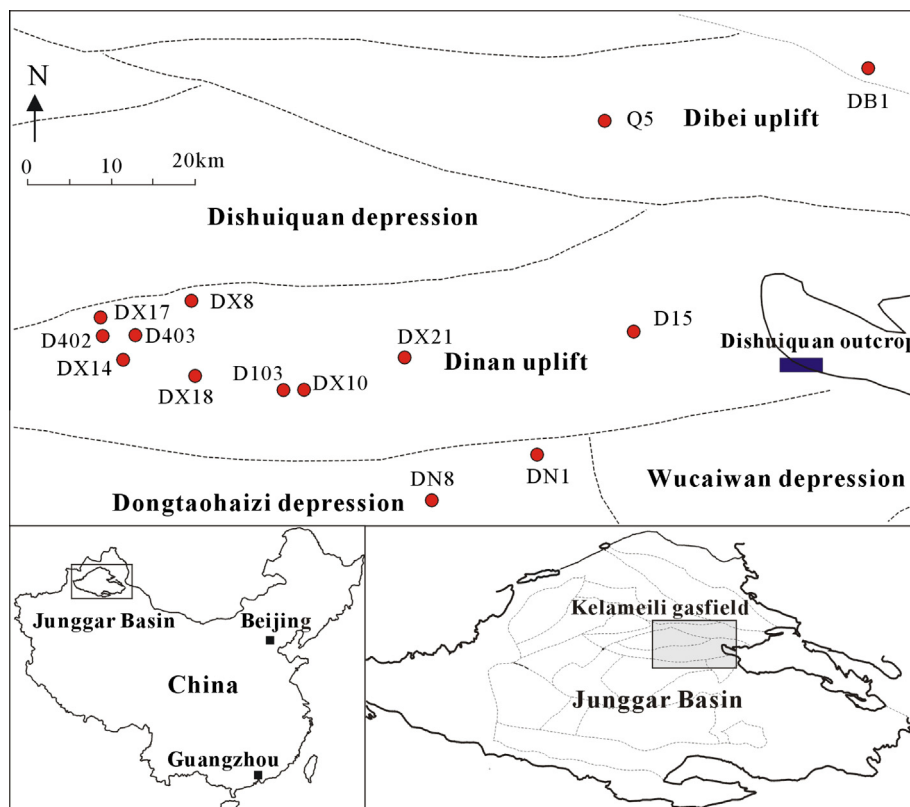


Fig. 1. Location map of Kelameili gasfield and the sampled boreholes in the Junggar Basin.

### 3.2. Total organic carbon (TOC) content, Rock-Eval and bitumen extraction of source rocks

All 69 source rocks were first cleaned and then ground into powder (about 200 mesh). A small aliquot of powder was taken from each sample for measurement of TOC using a Leco-230 C/S analyzer. Another small aliquot of powder was taken for Rock-Eval analysis using an IFP Rock-Eval 6 for nine core samples (from DX8S61 to Q5S69) and OG-200V oil-gas show evaluation instrument for 60 outcrop samples (from DSQ-S1 to DSQ-S60). Rock-Eval analysis for the outcrop samples was performed a year after the core sample analysis. The remaining powdered samples (~100 g each) for eight cores within the Batamayineishan Formation ( $C_{2b}$ ) and 24 outcrop rocks within the Dishuiquan Formation ( $C_{1d}$ ) with relatively higher TOC and HI values were Soxhlet extracted with dichloromethane:methanol (DCM:MeOH 93:7 v:v) for 72 h to obtain bitumen.

### 3.3. Sequential extraction of oil-containing volcanic reservoir rocks

The free, adsorbed and inclusion oils were obtained from oil-containing reservoir rocks by sequential extraction as described previously (Pan and Liu, 2009; Yu et al., 2011). Briefly, each core was crushed and sieved to obtain about 200 g of 0.10–0.30 mm size fraction. The grain sample was divided into two parts. In the tables and figures the free, adsorbed and inclusion oil sample codes carry the suffix “a”, “b” and “c”, respectively.

### 3.4. Oil fractionation

The extracts from the source rocks and the free, adsorbed and inclusion oils from oil-containing volcanic rocks were first diluted with about 1 ml or less DCM and then were deasphalted using a 40× excess of hexane. All the deasphalted samples were

fractionated on a silica:alumina column using hexane, hexane:DCM (2:1 v:v) and methanol as eluants to yield the saturated, aromatic and resin fractions, respectively.

### 3.5. GC, GC-MS and GC-IRMS analyses

Saturated fractions from the crude oils and source and reservoir rocks were first analyzed by gas chromatography (GC) using a HP6890 GC fitted with a 30 m × 0.32 mm i.d. HP-5 column with a film thickness of 0.25 μm and using nitrogen carrier gas. A constant flow mode and the flame ionization detector were employed. The GC oven temperature was held initially at 70 °C for 5 min, ramped from 70–290 °C at 4 °C/min, and then held at 290 °C for 30 min.

After GC analysis, the saturated fractions were further treated with urea adduction to separate *n*-alkanes and iso- and cyclic-alkanes. Gas chromatographic–mass spectrometric (GC–MS) analysis of iso- and cyclic-alkane fractions were carried out using a Thermal Scientific DSQ II quadrupole mass spectrometer interfaced to a Trace GC ULTRA. The GC ULTRA was fitted with a 30 m × 0.25 mm i.d. HP-5MS column with a film thickness of 0.25 μm and using helium carrier gas. A constant flow was used. The mass spectrometer was operated in electron impact (EI) mode at 70 eV. The GC oven temperature was initially held at 80 °C for 2 min, ramped from 80–180 °C at 8 °C/min, from 180–290 °C at 2 °C/min, and then held at 290 °C for 20 min.

Gas chromatography–isotope ratio mass spectrometry (GC–IRMS) analysis on *n*-alkane fractions was performed on a GV isoprime IRMS instrument interfaced to a HP6890 GC via a combustion interface. HP6890 GC was fitted with a 30 m × 0.25 mm i.d. HP-5MS column with a film thickness of 0.25 μm and using helium carrier gas. The initial temperature was 80 °C for 1.5 min, ramped from 80–130 °C at 20 °C/min, and further from 130–290 °C at 4 °C/min, and then held at 290 °C for 15 min. A mixture of *n*-alkane

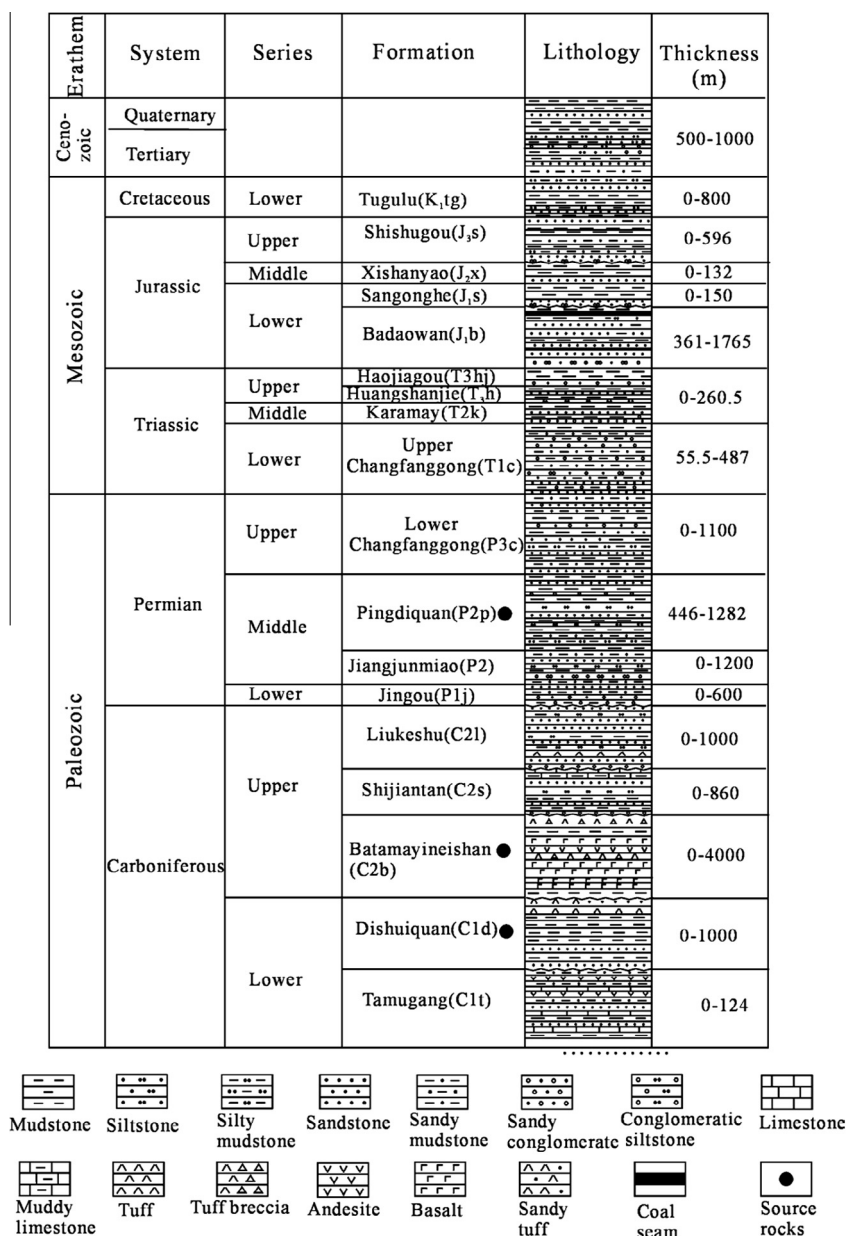


Fig. 2. Stratigraphy of the Eastern Junggar Basin (modified from Zhang et al., 1993).

standards ( $n\text{-C}_{12}$ ,  $n\text{-C}_{14}$ ,  $n\text{-C}_{16}$ ,  $n\text{-C}_{18}$ ,  $n\text{-C}_{20}$ ,  $n\text{-C}_{22}$ ,  $n\text{-C}_{25}$ ,  $n\text{-C}_{28}$ ,  $n\text{-C}_{30}$ ,  $n\text{-C}_{32}$  and  $n\text{-C}_{35}$ , provided by A. Schimmelmann of Indiana University, USA) was measured daily to monitor the GC-IRMS system. Replicate analyses of this mixture show that the standard deviation for each compound is less than 0.3%. Carbon isotopic values are reported relative to the VPDB standard. Each sample was analyzed at least twice. The reproducibility was generally within  $\pm 0.4\%$ . The average for the two or more runs was accepted as the final isotopic result for a sample.

## 4. Results

TOC and Rock-Eval data for the 69 source rocks are shown in Table 1 and Fig. 3. The gross compositions for the 10 oils are shown in Table 3. The selected molecular parameters for 32 source rocks, 10 oils and three selected oil-containing reservoir rocks are shown in Table 4 and Figs. 4 and 5. The gas chromatograms and  $m/z$  191

and  $m/z$  217 mass chromatograms for two selected source rocks, 10 oils and three selected reservoir rocks are shown in Figs. 6–12. Terpene and sterane peak assignments for Fig. 6 and Figs. 8–12 are listed in Table 5. The carbon isotopic compositions of individual  $n$ -alkanes for 32 source rocks, 10 oils and three selected oil-containing reservoir rocks are in Fig. 13.

## 5. Discussion

### 5.1. Differences between two Carboniferous source rocks

#### 5.1.1. Total organic carbon (TOC) contents

For 60 outcrop source rocks from the Lower Carboniferous Dishuiquan Formation ( $C_{1d}$ ), TOC contents range from 0.46–4.85%, mainly within 1.0–1.5%. For the nine core source rocks from the Upper Carboniferous Batamayineishan Formation ( $C_{2b}$ ), TOC contents range from 0.99–13.19% (Table 1). The outcrop samples



**Table 1**  
Total organic carbon content (TOC) and Rock–Eval parameters of the Carboniferous source rocks.

Depth (m)	Strata	TOC%	HI	Tmax	Depth (m)	Strata	TOC%	HI	Tmax		
DSQ-S1	Outcrop	C <sub>1d</sub>	1.53	64.3	471	DSQ-S36	Outcrop	C <sub>1d</sub>	0.94	62.2	492
DSQ-S2	Outcrop	C <sub>1d</sub>	1.32	61.7	501	DSQ-S37	Outcrop	C <sub>1d</sub>	0.94	82.3	497
DSQ-S3	Outcrop	C <sub>1d</sub>	4.85	124.2	474	DSQ-S38	Outcrop	C <sub>1d</sub>	0.80	32.4	493
DSQ-S4	Outcrop	C <sub>1d</sub>	0.74	64.0	478	DSQ-S39	Outcrop	C <sub>1d</sub>	2.56	139.2	473
DSQ-S5	Outcrop	C <sub>1d</sub>	1.24	131.7	491	DSQ-S40	Outcrop	C <sub>1d</sub>	1.08	98.5	476
DSQ-S6	Outcrop	C <sub>1d</sub>	0.73	39.3	500	DSQ-S41	Outcrop	C <sub>1d</sub>	1.13	35.9	484
DSQ-S7	Outcrop	C <sub>1d</sub>	0.82	20.0	501	DSQ-S42	Outcrop	C <sub>1d</sub>	1.18	69.6	478
DSQ-S8	Outcrop	C <sub>1d</sub>	0.76	62.4	498	DSQ-S43	Outcrop	C <sub>1d</sub>	1.33	77.5	491
DSQ-S9	Outcrop	C <sub>1d</sub>	0.46	64.4	484	DSQ-S44	Outcrop	C <sub>1d</sub>	2.91	78.3	475
DSQ-S10	Outcrop	C <sub>1d</sub>	0.85	56.7	491	DSQ-S45	Outcrop	C <sub>1d</sub>	1.55	104.1	476
DSQ-S11	Outcrop	C <sub>1d</sub>	0.96	94.3	480	DSQ-S46	Outcrop	C <sub>1d</sub>	1.42	104.0	476
DSQ-S12	Outcrop	C <sub>1d</sub>	0.99	82.2	482	DSQ-S47	Outcrop	C <sub>1d</sub>	1.33	94.3	483
DSQ-S13	Outcrop	C <sub>1d</sub>	0.91	55.5	498	DSQ-S48	Outcrop	C <sub>1d</sub>	0.98	89.6	472
DSQ-S14	Outcrop	C <sub>1d</sub>	1.81	144.7	475	DSQ-S49	Outcrop	C <sub>1d</sub>	1.19	65.4	500
DSQ-S15	Outcrop	C <sub>1d</sub>	1.60	104.7	470	DSQ-S50	Outcrop	C <sub>1d</sub>	1.08	90.0	476
DSQ-S16	Outcrop	C <sub>1d</sub>	0.73	81.2	481	DSQ-S51	Outcrop	C <sub>1d</sub>	1.29	109.0	474
DSQ-S17	Outcrop	C <sub>1d</sub>	0.78	89.6	490	DSQ-S52	Outcrop	C <sub>1d</sub>	1.32	92.4	473
DSQ-S18	Outcrop	C <sub>1d</sub>	1.26	90.6	489	DSQ-S53	Outcrop	C <sub>1d</sub>	1.70	108.4	467
DSQ-S19	Outcrop	C <sub>1d</sub>	1.30	109.5	490	DSQ-S54	Outcrop	C <sub>1d</sub>	1.14	55.3	511
DSQ-S20	Outcrop	C <sub>1d</sub>	1.10	90.4	481	DSQ-S55	Outcrop	C <sub>1d</sub>	1.47	113.0	471
DSQ-S21	Outcrop	C <sub>1d</sub>	1.54	114.4	486	DSQ-S56	Outcrop	C <sub>1d</sub>	1.45	96.4	474
DSQ-S22	Outcrop	C <sub>1d</sub>	1.30	119.2	490	DSQ-S57	Outcrop	C <sub>1d</sub>	1.47	83.7	473
DSQ-S23	Outcrop	C <sub>1d</sub>	1.25	89.2	481	DSQ-S58	Outcrop	C <sub>1d</sub>	1.68	122.7	464
DSQ-S24	Outcrop	C <sub>1d</sub>	1.10	86.6	488	DSQ-S59	Outcrop	C <sub>1d</sub>	1.46	116.0	474
DSQ-S25	Outcrop	C <sub>1d</sub>	0.80	63.8	486	DSQ-S60	Outcrop	C <sub>1d</sub>	0.98	24.5	457
DSQ-S26	Outcrop	C <sub>1d</sub>	1.11	93.6	492	DX8S61	3407.3	C <sub>2b</sub>	7.45	160.1	448
DSQ-S27	Outcrop	C <sub>1d</sub>	0.92	137.2	483	DX8S62	3410.0	C <sub>2b</sub>	11.00	120.8	451
DSQ-S28	Outcrop	C <sub>1d</sub>	1.16	73.6	510	DX8S63	3511.8	C <sub>2b</sub>	13.19	238.0	446
DSQ-S29	Outcrop	C <sub>1d</sub>	1.08	76.2	480	DX8S64	3605.6	C <sub>2b</sub>	3.47	106.0	453
DSQ-S30	Outcrop	C <sub>1d</sub>	1.10	60.2	468	DX8S65	3605.9	C <sub>2b</sub>	5.34	153.6	468
DSQ-S31	Outcrop	C <sub>1d</sub>	0.95	54.9	484	Q5S66	2140.6	C <sub>2b</sub>	3.80	47.0	453
DSQ-S32	Outcrop	C <sub>1d</sub>	1.02	77.8	477	Q5S67	2572.8	C <sub>2b</sub>	1.45	44.0	459
DSQ-S33	Outcrop	C <sub>1d</sub>	1.24	74.6	489	Q5S68	2573.7	C <sub>2b</sub>	0.99	45.0	461
DSQ-S34	Outcrop	C <sub>1d</sub>	1.00	52.7	502	Q5S69	2871.8	C <sub>2b</sub>	2.38	38.7	476
DSQ-S35	Outcrop	C <sub>1d</sub>	0.78	106.1	448						

HI: in "mg/g TOC"; Tmax is in "°C".

**Table 2**  
Samples of crude oils and oil-containing volcanic rocks from the Eastern Junggar Basin in the present study.

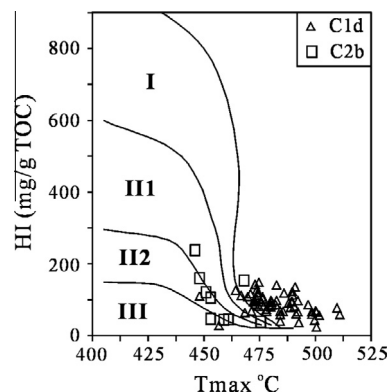
	Depth (m)	Strata
<i>Oils</i>		
D150	1477–1496	K <sub>1tg</sub>
DB10	1679–1689	J <sub>1b</sub>
DX10-10	2695–2698	T <sub>3b</sub>
DN80	3956–3972	P <sub>3c</sub>
DX10-20	3024–3048	C <sub>2b</sub>
DX210	2906–2913	C <sub>2b</sub>
D1030	3050–3062	C <sub>2b</sub>
D4030	3194–3606	C <sub>2b</sub>
DX170	3519–3526	C <sub>2b</sub>
DX140	3523–3550	C <sub>2b</sub>
<i>Oil-containing volcanic rocks</i>		
D402R3	3788.0	C <sub>2b</sub>
D403R20	3676.5	C <sub>2b</sub>
DX18R35	3452.0	C <sub>2b</sub>

were unlikely much influenced by weathering as demonstrated by gas chromatograms of saturated fractions (Fig. 6a). Therefore, C<sub>1d</sub> outcrop source rocks apparently have lower TOC contents than C<sub>2b</sub> core source rocks.

C<sub>1d</sub> source rocks generally have higher Tmax values, and therefore have higher maturities than C<sub>2b</sub> source rocks (Table 1, Fig. 3).

### 5.1.2. Molecular geochemistry

C<sub>1d</sub> outcrop source rocks generally have lower Pr/n-C<sub>17</sub> and Ph/n-C<sub>18</sub> ratios than C<sub>2b</sub> core source rocks (Table 4, Figs. 4a, 6a, and d). C<sub>1d</sub> source rocks contain substantially lower concentrations of



**Fig. 3.** Crossplot of hydrogen index (HI) and Tmax from Rock–Eval analyses of source rock samples included in this study.

terpanes and steranes than C<sub>2b</sub> source rocks either due to differential maturities or differential initial concentrations of these components, or both (Fig. 6b–c and e–f). The major differences in terpane and sterane compositions between these two source rocks can be listed as follows: (1) C<sub>1d</sub> source rocks contain substantially higher concentrations of C<sub>30</sub> diahopane and Ts relative to C<sub>30</sub> hopane and Tm than C<sub>2b</sub> source rocks (Table 4, Figs. 4b, 6b and e), possibly demonstrating efficient acidic catalysis in the C<sub>1d</sub> source rocks (e.g., Moldowan et al., 1991; Farrimond and Telnæs, 1996; Peters et al., 2005). (2) C<sub>2b</sub> source rocks contain unusually high concentrations of C<sub>24</sub> tetracyclic terpane relative to C<sub>23</sub> and C<sub>26</sub> tricyclic terpanes compared with C<sub>1d</sub> source rocks (Table 4, Figs. 4c, 6b and e),

**Table 3**

Gross compositions of the crude oils.

	Sat%	Aro%	Res%	Asp%		Sat%	Aro%	Res%	Asp%
D150	89.21	6.88	1.02	2.89	DX210	89.99	7.49	0.84	1.68
DB10	86.65	7.77	3.29	2.29	D1030	97.32	1.73	0.72	0.23
DX10-10	98.59	0.83	0.29	0.30	D4030	96.68	2.59	0.17	0.56
DN80	88.43	8.21	1.44	1.92	DX170	96.98	2.20	0.47	0.35
DX10-20	97.80	1.36	0.29	0.54	DX140	95.50	2.88	1.15	0.47

demonstrating lower maturity and terrigenous source of organic matter for C<sub>2b</sub> source rocks (Philp and Gilbert, 1986; Chen et al., 2001, 2003a,b; Peters et al., 2005). (3) C<sub>2b</sub> source rocks generally contain higher concentrations of C<sub>19</sub>–C<sub>21</sub> relative to C<sub>23</sub>–C<sub>24</sub> tricyclic terpanes than C<sub>1d</sub> source rocks (Table 4, Figs. 4d, 6b and e), demonstrating higher plant source for C<sub>2b</sub> source rocks (e.g., Chen et al., 2001, 2003a,b; Peters et al., 2005). (4) C<sub>1d</sub> source rocks have substantially higher ratios of C<sub>23</sub> tricyclic terpane/(C<sub>23</sub> tricyclic terpane + C<sub>30</sub> hopane) and C<sub>21</sub>/(C<sub>21</sub> + ΣC<sub>29</sub>) steranes than C<sub>2b</sub> source rocks (Table 4, Figs. 4e, 6b–c and e–f), which can be mainly ascribed to higher maturities of C<sub>1d</sub> source rocks than C<sub>2b</sub> source rocks (e.g., Aquino Neto et al., 1983; Shi et al., 1988; Van Graas, 1990; Farrimond et al., 1999; Pan and Yang, 2000; Yu et al., 2011). (5) C<sub>1d</sub> source rocks contain relatively higher concentrations of C<sub>27</sub> steranes and lower concentrations of C<sub>29</sub> steranes compared with C<sub>2b</sub> source rocks (Table 4, Figs. 5, 6c and f), demonstrating substantial facies difference between these two source rocks. (6) C<sub>1d</sub> source rocks contain higher C<sub>27</sub>–C<sub>29</sub> diasteranes relative to C<sub>27</sub>–C<sub>29</sub> regular steranes than C<sub>2b</sub> source rocks (Table 4, Figs. 4f, 6c and f), possibly demonstrating more efficient acidic catalysis and higher maturity for the C<sub>1d</sub> source rocks compared with C<sub>2b</sub> source rocks (e.g., Peters et al., 2005). (7) C<sub>2b</sub> source rocks contain very low concentrations of gammacerane relative to hopanes compared with C<sub>1d</sub> source rocks. Gammacerane/C<sub>31</sub> hopanes (R + S) ratios range 0.01–0.08 and 0.15–0.34, respectively for C<sub>2b</sub> and C<sub>1d</sub> source rocks (Table 4), demonstrating higher extent of water-column stratification for C<sub>1d</sub> source rocks than C<sub>2b</sub> source rocks (e.g., Peters et al., 2005).

In summary, in terms of terpane and sterane distributions, C<sub>2b</sub> source rocks are similar to Jurassic source rocks and their derived oils (e.g., Chen et al., 2001, 2003a,b). In general, C<sub>2b</sub> source rocks deposited in relative oxic environment and contain dominantly terrigenous organic matter while C<sub>1d</sub> source rocks deposited in relative anoxic environment and contain relatively more marine aquatic organic matter. C<sub>1d</sub> source rocks have substantially higher maturities than C<sub>2b</sub> source rocks.

### 5.1.3. Carbon isotopic compositions of individual *n*-alkanes

For both C<sub>1d</sub> and C<sub>2b</sub> source rocks, the δ<sup>13</sup>C values of individual *n*-alkanes are higher than –30‰ and vary in a wide range. However, they generally decrease with carbon number for C<sub>1d</sub> source rocks while opposite for C<sub>2b</sub> source rocks (Fig. 13a and b). Wang et al. (2010) and Da et al. (2010) reported that δ<sup>13</sup>C values of kerogen for C<sub>1d</sub> and C<sub>2b</sub> source rocks range –27.50‰ to 21.00‰ and –25.96‰ to –21.00‰, respectively. The result of the present study is generally consistent with these previous studies.

## 5.2. Oil sources

### 5.2.1. Oils D1030, D4030, DX170 and DX140 from C<sub>2b</sub> volcanic reservoirs

These four oils are overwhelmingly dominated by saturated fraction, ranging 95.50–97.32% (Table 3). This result can be ascribed to two causes: These oils have high maturities, or (2) these oils were likely generated from gas prone source rocks, and therefore only a small amount of oil components can be expelled

from the source rocks which are overwhelmingly dominated by saturates (e.g., Pepper, 1992).

As to molecular geochemistry, these four oils are characterized by (1) lower Pr/*n*-C<sub>17</sub> and Ph/*n*-C<sub>18</sub> ratios, (2) lower concentrations of terpanes and steranes, (3) higher ratios of C<sub>30</sub> diahopane/(C<sub>30</sub> diahopane + C<sub>30</sub> hopane) and Ts/(Ts + Tm), (4) lower concentration of C<sub>24</sub> tetracyclic terpane relative to C<sub>23</sub> and C<sub>26</sub> tricyclic terpanes, (5) lower ratios of C<sub>20</sub>/(C<sub>20</sub> + C<sub>23</sub>) tricyclic terpanes and C<sub>21</sub>/(C<sub>21</sub> + C<sub>23</sub>) tricyclic terpanes, and (6) higher ratios of C<sub>23</sub> tricyclic terpane/(C<sub>23</sub> tricyclic terpane + C<sub>30</sub> hopane) and C<sub>21</sub>/(C<sub>21</sub> + ΣC<sub>29</sub>) steranes (Table 4, Figs. 4, 7 and 9). These four oils generally correlate with C<sub>1d</sub> source rocks and are substantially different from C<sub>2b</sub> source rocks based on molecular parameters.

δ<sup>13</sup>C values of *n*-alkanes for all the four oils decrease with carbon number, similar to those for C<sub>1d</sub> source rocks and different substantially from those for C<sub>2b</sub> source rocks (Fig. 13). For oils D1030, DX4030 and DX140, δ<sup>13</sup>C values of *n*-alkanes are within the range of those for C<sub>1d</sub> source rocks. However, δ<sup>13</sup>C values of *n*-alkanes for oil DX170 are more negative than those for C<sub>1d</sub> source rocks.

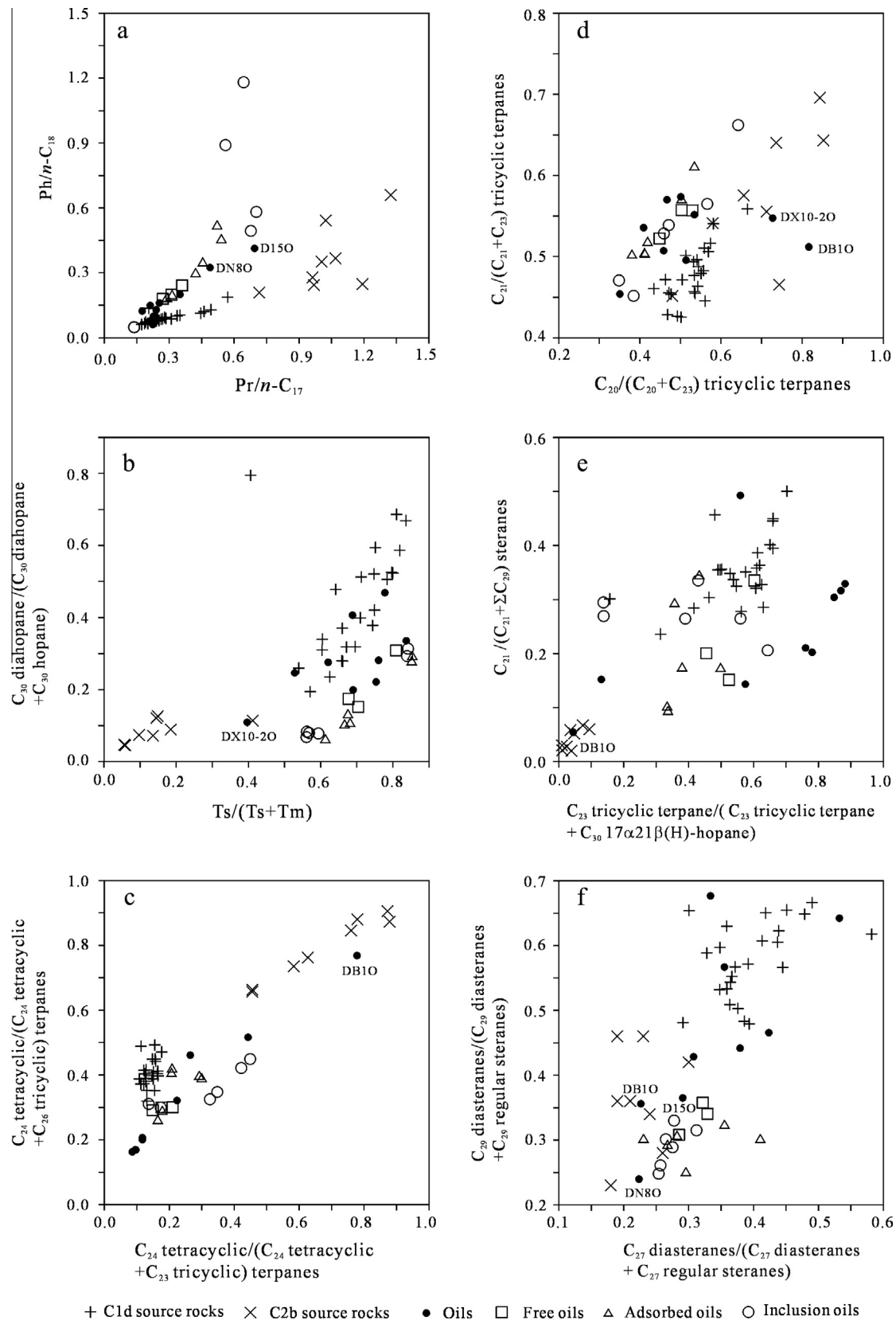
From molecular and isotopic compositions, it can be determined that these four oils were generated from C<sub>1d</sub> source rocks, rather than C<sub>2b</sub> source rocks. However, oil DX170 was generated from C<sub>1d</sub> source rocks with δ<sup>13</sup>C values more negative than those for samples from Dishuiquan outcrop.

### 5.2.2. Oils DX10-20 and DX210 from C<sub>2b</sub> volcanic reservoirs

Oil DX10-20 consists of 97.80% saturated fraction (Table 3). It has lower Pr/*n*-C<sub>17</sub> and Ph/*n*-C<sub>18</sub> ratios and contains very low concentrations of terpanes and steranes, similar to C<sub>1d</sub> source rocks (Table 4, Figs. 7e, 8d and i). On the other hand, this oil is characterized with (1) lower ratios of C<sub>30</sub> diahopane/(C<sub>30</sub> diahopane + C<sub>30</sub> hopane) and Ts/(Ts + Tm), (2) higher concentration of C<sub>24</sub> tetracyclic terpane relative to C<sub>23</sub> and C<sub>26</sub> tricyclic terpanes, and (3) higher ratios of C<sub>20</sub>/(C<sub>20</sub> + C<sub>23</sub>) and C<sub>21</sub>/(C<sub>21</sub> + C<sub>23</sub>) tricyclic terpanes (Table 4, Fig. 8d and i). From these molecular parameters, this oil is similar to C<sub>2b</sub> source rocks. δ<sup>13</sup>C values of *n*-alkanes for this oil generally decrease with carbon number and are within the range of those for C<sub>1d</sub> source rocks (Fig. 13c). These results demonstrate that this oil is dominantly derived from C<sub>1d</sub> source rocks but contaminated by components from C<sub>2b</sub> source rocks. As a result, the major components, i.e. *n*-alkanes and acyclic isoprenoids, are derived from C<sub>1d</sub> source rocks but terpanes and steranes are mainly derived from C<sub>2b</sub> source rocks.

Oil DX210 consists of 90% saturated fraction, lower than the other five oils from C<sub>2b</sub> volcanic reservoirs (Table 3). This oil has lower Pr/*n*-C<sub>17</sub> and Ph/*n*-C<sub>18</sub> ratios (Table 4, Fig. 7e), similar to C<sub>1d</sub> source rocks. However, it contains higher concentrations of terpanes and steranes than the other five oils from C<sub>2b</sub> volcanic reservoirs and extracts from C<sub>1d</sub> source rocks (Figs. 6, 8 and 9). This oil has higher ratios of C<sub>30</sub> diahopane/(C<sub>30</sub> diahopane + C<sub>30</sub> hopane) and Ts/(Ts + Tm), similar to C<sub>1d</sub> source rocks (Table 4, Figs. 4b, 8e and j). On the other hand, it contains higher concentration of C<sub>24</sub> tetracyclic terpanes relative to C<sub>23</sub> and C<sub>26</sub> tricyclic terpanes, close to C<sub>2b</sub> source rocks and different substantially from C<sub>1d</sub> source rocks (Table 4, Figs. 4c, e, 8e and j). δ<sup>13</sup>C values of *n*-alkanes for this





**Fig. 4.** Crossplots of molecular parameters for oils and oil components from source and reservoir rocks. (a) crossplot of the ratios of  $Pr/n-C_{17}$  versus  $Ph/n-C_{18}$ ; (b) crossplot of the ratios of  $Ts/(Ts+Tm)$  versus  $C_{30}$  diahopane / ( $C_{30}$  diahopane +  $C_{30}$  hopane); (c) crossplot of the ratios of  $C_{24}$  tetracyclic / ( $C_{24}$  tetracyclic +  $C_{26}$  tricyclic) terpanes versus  $C_{24}$  tetracyclic / ( $C_{24}$  tetracyclic +  $C_{23}$  tricyclic) terpanes; (d) crossplot of the ratios of  $C_{20}/(C_{20}+C_{23})$  tricyclic terpanes versus  $C_{21}/(C_{21}+C_{23})$  tricyclic terpanes; (e) crossplot of the ratios of  $C_{23}$  tricyclic terpane / ( $C_{23}$  tricyclic terpane +  $C_{30}$   $17\alpha 21\beta(H)$ -hopane) versus  $C_{21}/(C_{21}+\Sigma C_{29})$  steranes; (f) crossplot of the ratios of  $C_{27}$  diasteranes / ( $C_{27}$  diasteranes +  $C_{27}$  regular steranes) versus  $C_{29}$  diasteranes / ( $C_{29}$  diasteranes +  $C_{29}$  regular steranes).



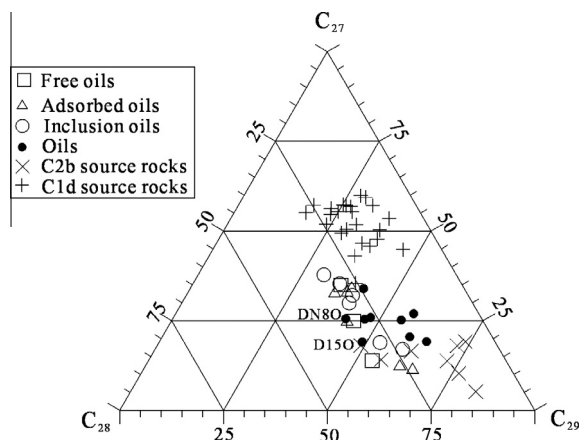


Fig. 5. Ternary diagram of  $C_{27}$ ,  $C_{28}$  and  $C_{29}$  sterane composition for oils and oil components from source and reservoir rocks.

fraction for the two oils are 89.21% and 88.43%, respectively, relatively lower than the six oils from  $C_{2b}$  volcanic reservoirs (Table 3).  $Pr/n-C_{17}$ ,  $Ph/n-C_{18}$  and  $Pr/Ph$  ratios are 0.69, 0.41 and 1.78, respectively for oil D150 and 0.49, 0.32 and 1.90, respectively for oil DN80, substantially different from those of the other eight oils and both  $C_{1d}$  and  $C_{2b}$  source rocks. These two oils have higher  $Ph/n-C_{18}$  ratio and lower  $Pr/Ph$  ratio than  $C_{1d}$  source rocks, lower  $Pr/n-C_{17}$  and  $Pr/Ph$  ratios than  $C_{2b}$  source rocks and higher

$Pr/n-C_{17}$  and  $Ph/n-C_{18}$  ratios than the other eight oils (Table 4). These two oils have higher concentration of gammacerane relative to  $C_{31}$  hopanes than the other seven oils and  $C_{1d}$  and  $C_{2b}$  source rocks (Table 4, Fig. 8a). As to sterane distribution, these two oils are characterized by (1) higher ratios of  $C_{29}/\Sigma C_{27-29}$  steranes and  $C_{28}/\Sigma C_{27-29}$  steranes and lower ratio of  $C_{27}/\Sigma C_{27-29}$  steranes, and (2) lower ratios of  $C_{27}$  diasteranes/( $C_{27}$  diasteranes +  $C_{27}$  regular steranes) and  $C_{29}$  diasteranes/( $C_{29}$  diasteranes +  $C_{29}$  regular steranes), compared with the other seven oils (Table 4, Figs. 4, 8 and 9).  $\delta^{13}C$  values of  $n$ -alkanes for these two oils generally decrease with carbon number but are relatively low among those for the 10 oils (Fig. 13).

The molecular parameters of acyclic isoprenoids, terpanes and steranes for oils D150 and DN80 are similar to oils which were generated from the source rocks within the Middle Permian Pingdiquan Formation ( $P_{2w}$ ), as documented in the previous studies (e.g., Zhou et al., 1989; Yang et al., 1992; Clayton et al., 1997; Pan and Yang, 2000; Chen et al., 2003a; Pan et al., 2003; Cao et al., 2005, 2006, 2012). Chen et al. (2003a) and Pan et al. (2003) documented molecular characteristics of oil DN10 from the interval 2760–2767 m of the Jurassic sandstone reservoir and oil components from oil-containing sandstone DN1-1 at 2764 m, respectively from borehole DN1 located near borehole DN8 (Fig. 1), demonstrating that the oil sample and oil components from the reservoir rock were derived from source rocks within the Permian Pingdiquan Formation ( $P_{2p}$ ). Compared with oil DN10 and oil-containing sandstone DN1-1, oils D150 and DN80 have several fundamental differences

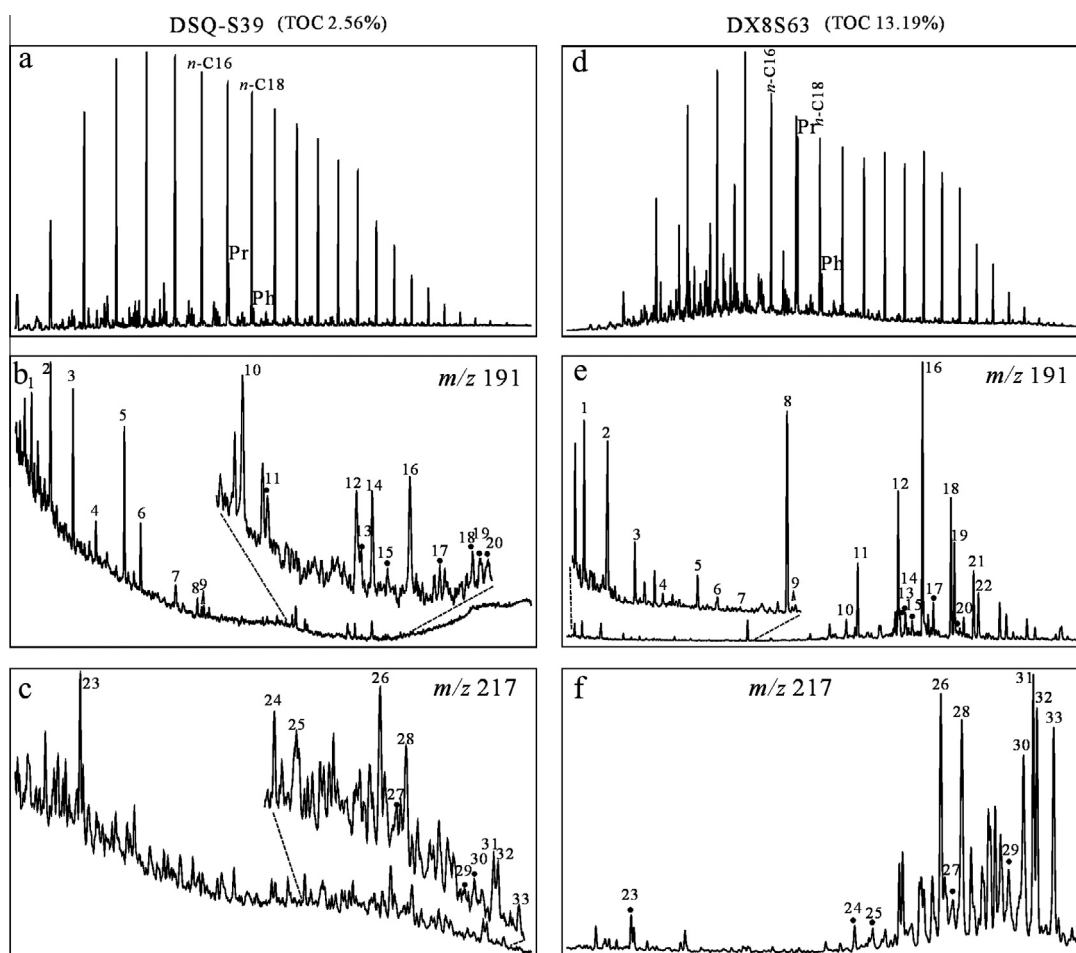


Fig. 6. Gas chromatograms and  $m/z$  191 and  $m/z$  217 mass chromatograms of  $C_{1d}$  source rock DSQ-S39 and  $C_{2b}$  source rock DX8S63; peak identifications are in Table 5.

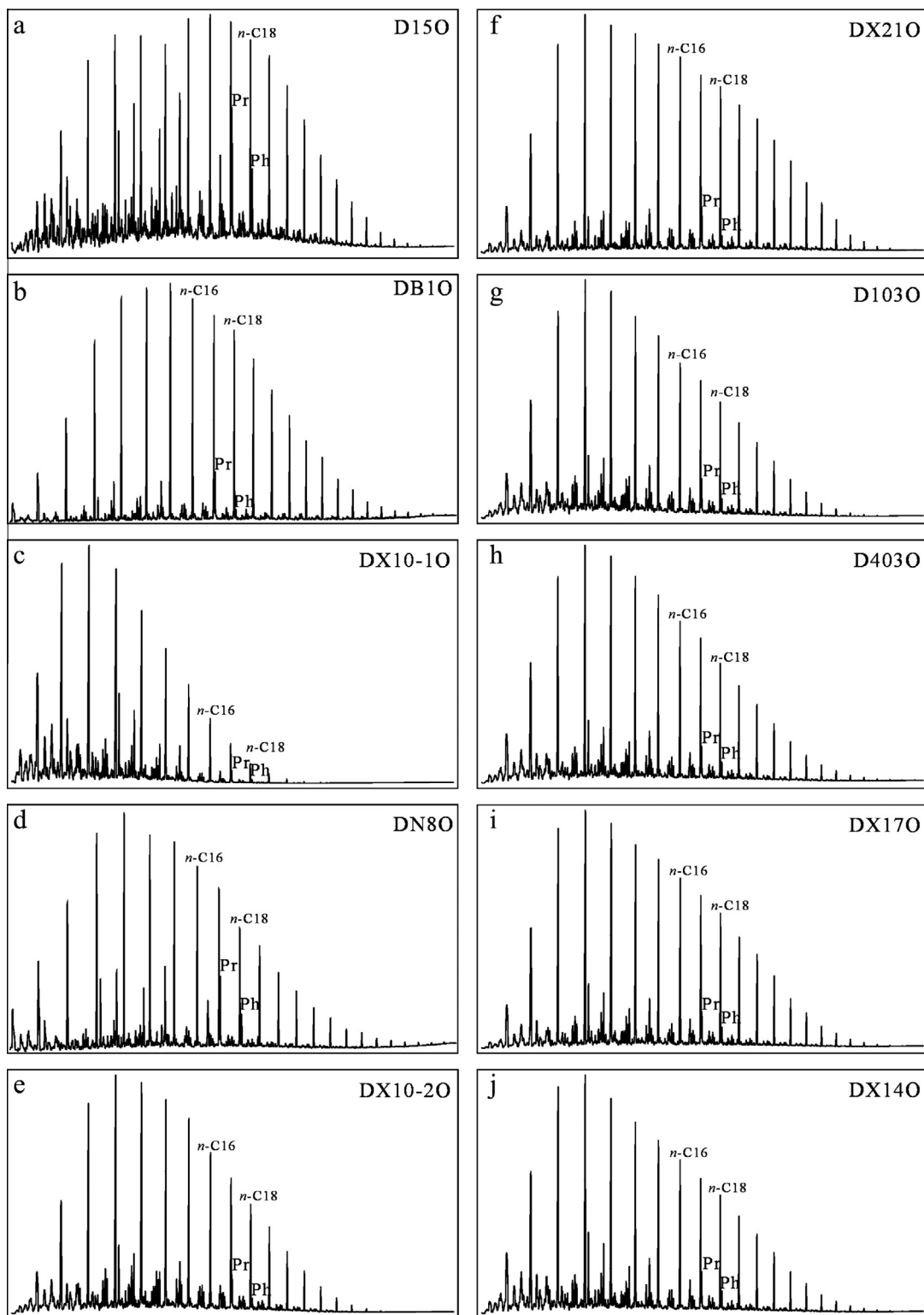


Fig. 7. Gas chromatograms of the 10 oil samples.

in gross and molecular compositions: (1) these two oils contain higher saturated fraction than oil DN10, i.e., 74.98%; (2) these two oils have substantially higher ratios of  $C_{23}$  tricyclic terpane / ( $C_{23}$  tricyclic terpane +  $C_{30}$  hopane) and  $C_{21} / (C_{21} + \Sigma C_{29})$  steranes than oil DN10 and oil-containing sandstone DN1-1; (3) these two oils have substantially higher ratios of  $C_{30}$  diahopane / ( $C_{30}$  diahopane +  $C_{30}$  hopane) and  $Ts / (Ts + Tm)$  than oil DN10 and oil-containing sandstone DN1-1. Points (1) and (2) demonstrate

that oils D150 and DN80 have higher maturities than oil DN10 and oil components from oil-containing sandstone DN1-1. Points (1) to (3) can be interpreted to indicate that D150 and DN80 were mainly derived from  $C_{1d}$  source rocks and mixed by minor oils generated from  $P_{2p}$  source rocks. Da et al. (2010) documented that gaseous hydrocarbons and oils for the Kelameili gasfield were generated from Carboniferous source rocks mainly within the area of this field and accumulated after short distance migration.

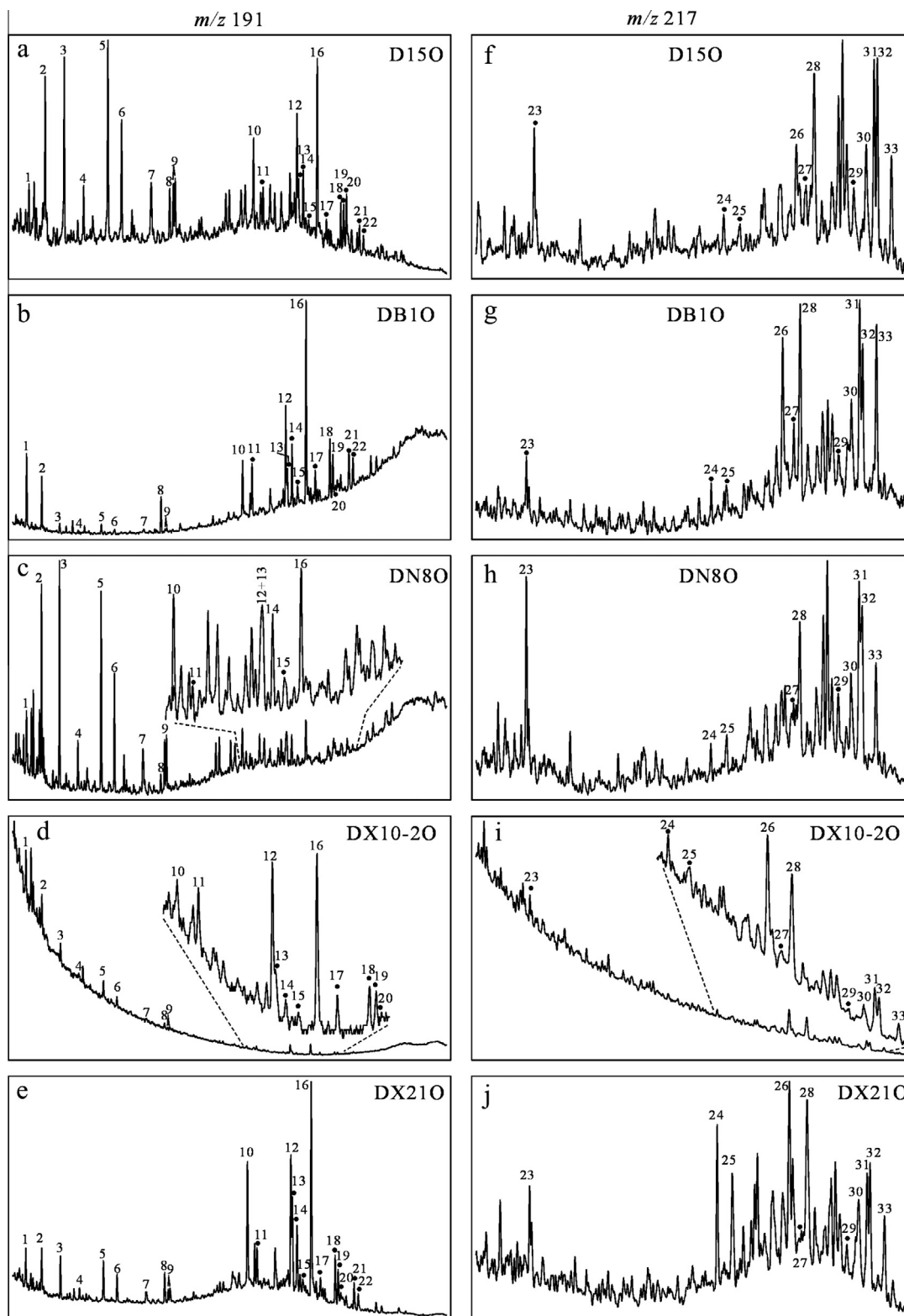


Fig. 8. Mass chromatograms  $m/z$  191 and  $m/z$  217 of oils D150, DB10, DN80, DX10-20 and DX210.

However, oils from Permian source rocks were most likely generated from the central area of the Dongtaohaizi depression, south of the gasfield (Fig. 1, Chen et al., 2003a; Pan et al., 2003).

Oil DX10-10 is a condensate collected from Triassic sandstone gas reservoir. This oil contains 98.59% saturated fraction, highest

among the 10 oils (Table 3).  $Pr/n-C_{17}$ ,  $Ph/n-C_{18}$  and  $Pr/Ph$  ratios for this oil are 0.35, 0.20 and 3.55, respectively, comparable to those for  $C_{1d}$  source rocks (Table 4, Fig. 4a). The concentrations of terpanes and steranes for this oil are below the detection level.  $\delta^{13}C$  values of individual  $n$ -alkanes for this oils decrease with

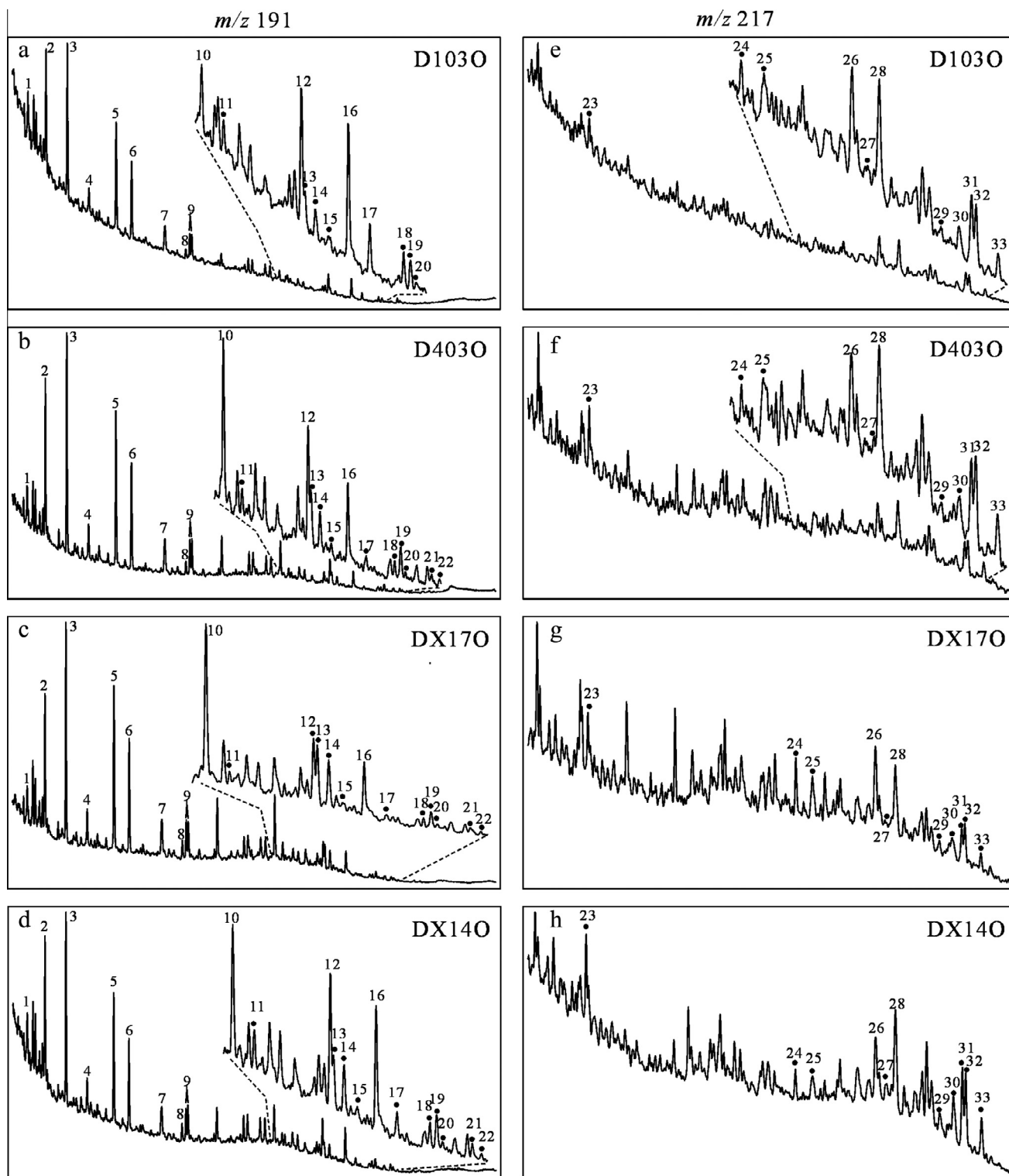
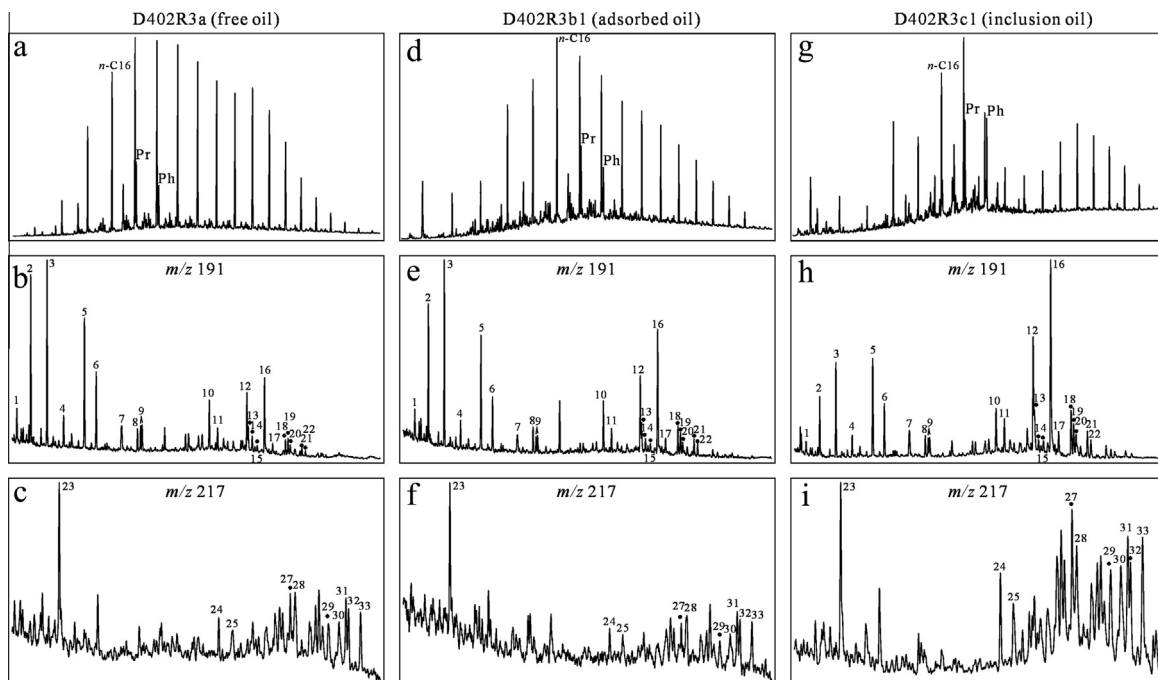


Fig. 9. Mass chromatograms  $m/z$  191 and  $m/z$  217 of oils D1030, D4030, DX170 and DX140.

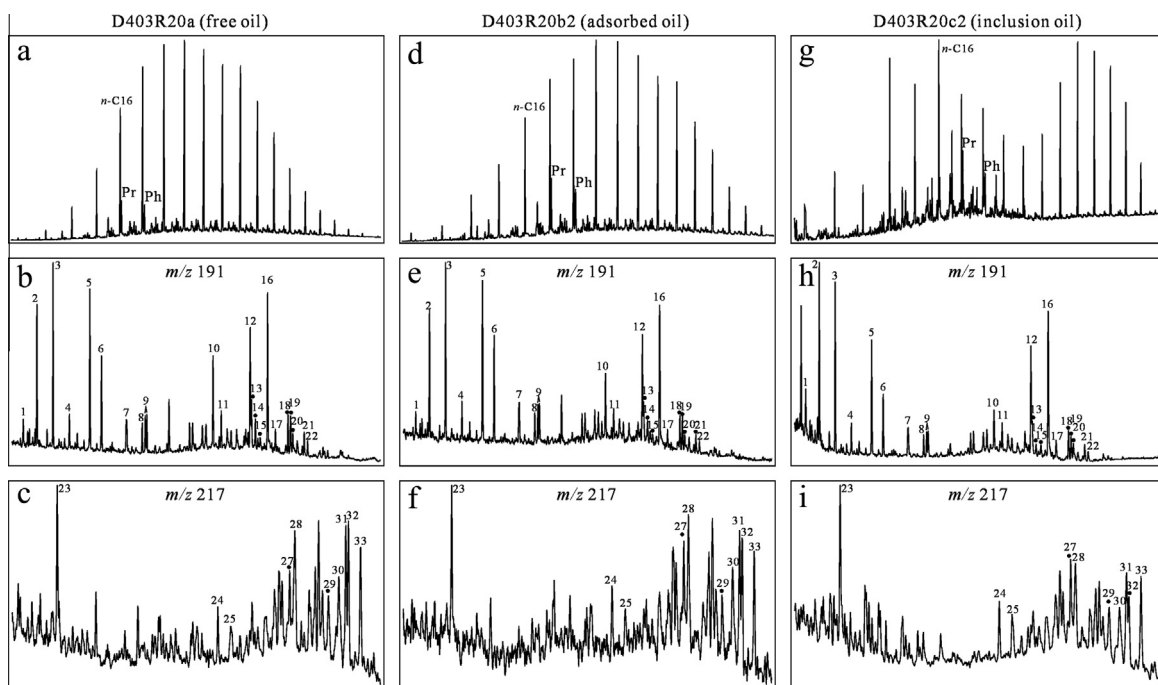
carbon number and are relatively low among those for the 10 oils, similar to oil DX170 (Fig. 13). This oil was most likely generated from  $C_{1d}$  source rocks, similar to oil DX170.

Oil DB10 was collected from the Lower Jurassic sandstone reservoir. It contains 86.65% saturated fraction, lowest among the 10 oils (Table 3). It has lower Pr/ $n$ - $C_{17}$ , Ph/ $n$ - $C_{18}$  ratios and higher Pr/Ph ratio, comparable to those of  $C_{1d}$  source rocks (Table 4, Fig. 7). It contains relatively higher concentrations of terpanes and ster-

anes, compared with oils from  $C_{2b}$  volcanic reservoirs and extracts from  $C_{1d}$  source rocks. Terpane and sterane distributions for this oil are characterized by (1) higher concentrations of  $C_{19}$ – $C_{21}$  tricyclic terpanes relative to  $C_{23}$ – $C_{24}$  tricyclic terpanes, (2) higher concentration of  $C_{24}$  tetracyclic terpane relative to  $C_{23}$  and  $C_{26}$  tricyclic terpanes, (3) lower ratios of  $C_{23}$  tricyclic terpane/( $C_{23}$  tricyclic terpane +  $C_{30}$  hopane) and  $C_{21}/(C_{21} + \Sigma C_{29})$  steranes, (4) higher ratio of  $C_{29}/\Sigma C_{27-29}$  steranes and lower ratios of



**Fig. 10.** Gas chromatograms and  $m/z$  191 and  $m/z$  217 mass chromatograms of reservoir rock D402R3. (a–c) Free oil; (d–f) adsorbed oil; (g–i) inclusion oil; peak identifications are in Table 5.

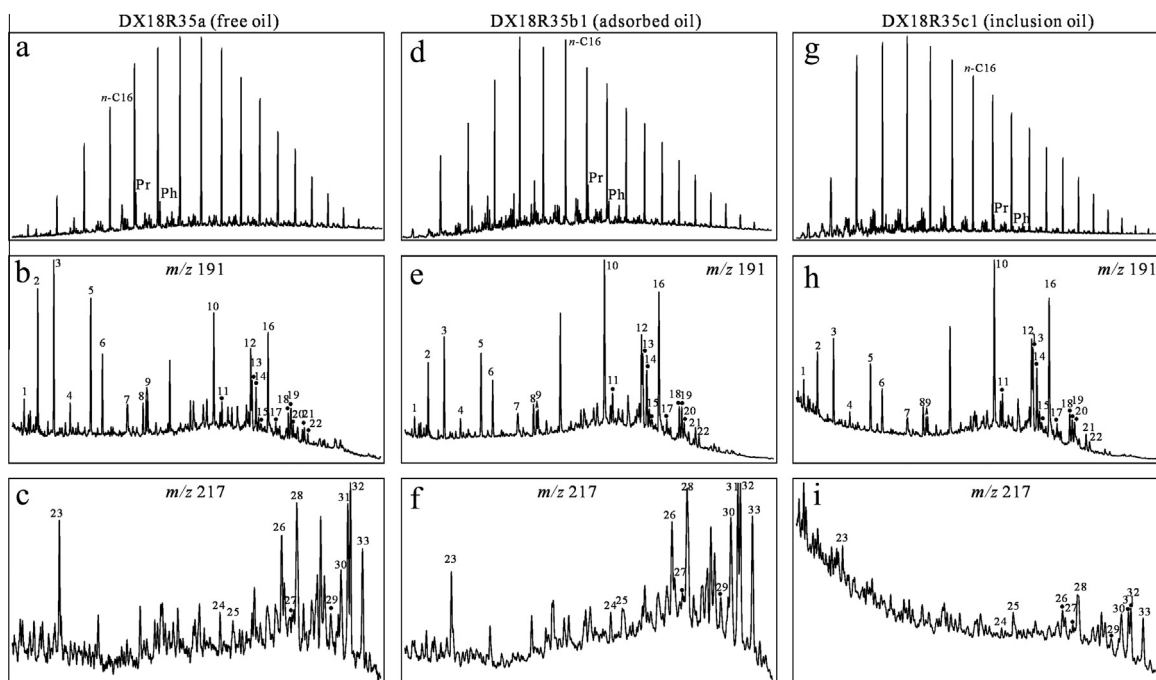


**Fig. 11.** Gas chromatograms and  $m/z$  191 and  $m/z$  217 mass chromatograms of reservoir rock D403R20. (a–c) Free oil; (d–f) adsorbed oil; (g–i) inclusion oil; peak identifications are in Table 5.

$C_{28}/\Sigma C_{27-29}$  steranes and  $C_{27}/\Sigma C_{27-29}$  steranes, and (5) lower ratios of  $C_{27}$  diasteranes/( $C_{27}$  regular steranes) and  $C_{29}$  diasteranes/( $C_{29}$  diasteranes +  $C_{29}$  regular steranes) (Table 4, Fig. 8b and g). These terpane and sterane signatures are similar to those of the Lower Jurassic source rocks and their derived oils (e.g., Chen et al., 2003a), as well as  $C_{2b}$  source rocks. However, this oil has higher ratios of  $C_{30}$  dihopane/( $C_{30}$  dihopane +  $C_{30}$  hopane) and  $Ts/(Ts + Tm)$  (Table 4, Fig. 8b), similar to  $C_{1d}$  source rocks but

substantially different from both  $C_{2b}$  and the Lower Jurassic source rocks (Chen et al., 2003).  $\delta^{13}C$  values of individual  $n$ -alkanes for this oil decrease with carbon number and within the range for  $C_{1d}$  source rocks (Fig. 13c). It can be determined that this oil was mainly derived from  $C_{1d}$  source rocks but contaminated by minor components from  $C_{2b}$  or Lower Jurassic source rocks, or both based on molecular and isotopic composition. A similar phenomenon has been documented by Li et al. (2001).





**Fig. 12.** Gas chromatograms and  $m/z$  191 and  $m/z$  217 mass chromatograms of reservoir rock DX18R35. (a–c) Free oil; (d–f) adsorbed oil; (g–i) inclusion oil; peak identifications are in Table 5.

### 5.3. Origins of oil components obtained by sequential extraction from reservoir rocks

#### 5.3.1. Oil components from reservoir rocks D402R3 and D403R20

The ratios of Pr/ $n$ -C<sub>17</sub>, Ph/ $n$ -C<sub>18</sub> and Pr/Ph for the free, adsorbed and inclusion oils from these two samples are significantly different from those of both C<sub>1d</sub> and C<sub>2b</sub> source rocks. However, these three ratios for the free and adsorbed oils from these two samples and inclusion oils from D403R20 are more close to those of C<sub>1d</sub> source rocks than C<sub>2b</sub> source rocks. The inclusion oils from sample D402R3 have spectacularly higher Ph/ $n$ -C<sub>18</sub> ratio and lower Pr/Ph ratio (Table 4, Figs. 4a, 10 and 11).

The free, adsorbed and inclusion oils from these two reservoir rocks correlate with C<sub>1d</sub> source rocks based on following biomarker parameters, (1) higher Ts/(Ts + Tm) ratio (Fig. 4b), (2) lower concentration of C<sub>24</sub> tetracyclic terpene relative to C<sub>23</sub> and C<sub>26</sub> tricyclic terpenes (Fig. 4c), (3) lower concentrations of C<sub>19</sub>–C<sub>21</sub> tricyclic terpenes relative to C<sub>23</sub>–C<sub>24</sub> tricyclic terpenes (Fig. 4d), (4) higher ratios of C<sub>23</sub> tricyclic terpene/(C<sub>23</sub> tricyclic terpene + C<sub>30</sub> hopane) and C<sub>21</sub>/(C<sub>21</sub> +  $\Sigma$ C<sub>29</sub>) steranes (Fig. 4e), and (5) higher ratio of gammacerane/C<sub>31</sub> hopanes (Table 4). However, oil components from these two reservoir rocks contain higher concentrations of terpenes and steranes, and have lower ratio of C<sub>30</sub> diahopane/(C<sub>30</sub> diahopane + C<sub>30</sub> hopane), contrast to C<sub>1d</sub> source rocks (Fig. 4b). They also have the distribution pattern of C<sub>27</sub>, C<sub>28</sub> and C<sub>29</sub> steranes substantially different from both C<sub>1d</sub> and C<sub>2b</sub> source rocks (Fig. 5).

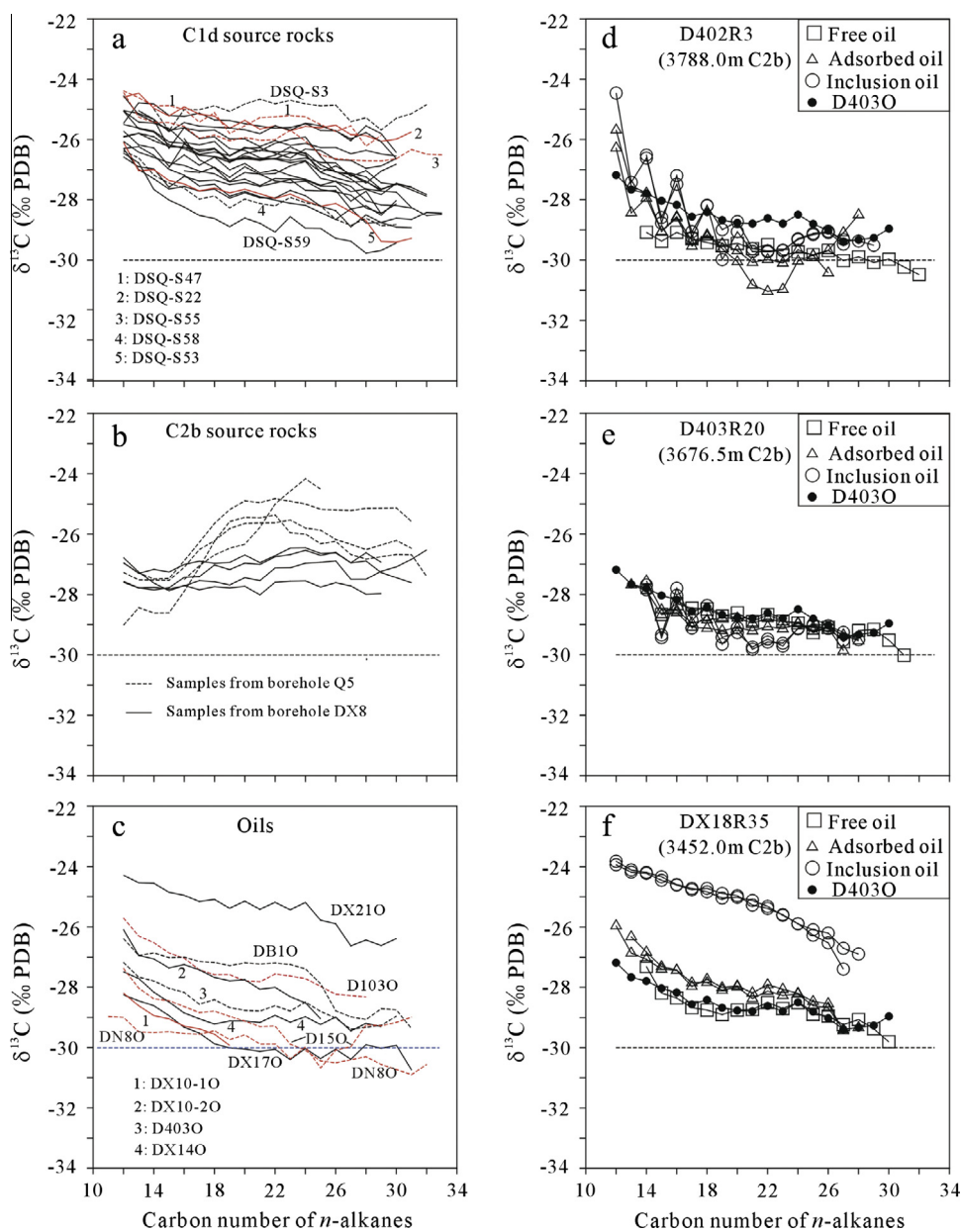
For reservoir rock D402R3, the ratios of C<sub>23</sub> tricyclic terpene/(C<sub>23</sub> tricyclic terpene + C<sub>30</sub> hopane), C<sub>21</sub>/(C<sub>21</sub> +  $\Sigma$ C<sub>29</sub>) steranes, C<sub>20</sub>/(C<sub>20</sub> + C<sub>23</sub>) and C<sub>21</sub>/(C<sub>21</sub> + C<sub>23</sub>) tricyclic terpenes decrease from the free oil, through the adsorbed oil to the inclusion oil (Table 4, Fig. 10). In contrast, these ratios vary in an opposite sequence for reservoir rock D403R20 (Table 4, Fig. 11). These results can be mainly ascribed to maturity variation of oil components during filling process of the reservoir, demonstrating that the maturity of the initial charging oil was lower than the later charging oil for reservoir rock D402R3 while opposite for reservoir rock D403R20 (e.g., Aquino Neto et al., 1983; Shi et al., 1988; Van Graas, 1990; Farrimond et al., 1999; Pan and Yang, 2000; Yu et al., 2011; Jin

et al., 2014). For both reservoir rocks D402R3 and D403R20, ratios of Ts/(Ts + Tm) and C<sub>30</sub> diahopane/(C<sub>30</sub> diahopane + C<sub>30</sub> hopane) decrease from the free oil, through the adsorbed oil to the inclusion oil, demonstrating the facies variation of source rocks for the

**Table 5**

Terpane and sterane assignments for Figs. 6 and 8–12.

Peak	Terpenes ( $m/z$ 191)
1	C <sub>19</sub> tricyclic terpene
2	C <sub>20</sub> tricyclic terpene
3	C <sub>21</sub> tricyclic terpene
4	C <sub>22</sub> tricyclic terpene
5	C <sub>23</sub> tricyclic terpene
6	C <sub>24</sub> tricyclic terpene
7	C <sub>25</sub> tricyclic terpene
8	C <sub>24</sub> tetracyclic terpene
9	C <sub>26</sub> tricyclic terpene
10	C <sub>27</sub> 18 $\alpha$ (H)-22,29,30-trisnorneohopane (Ts)
11	C <sub>27</sub> 17 $\alpha$ (H)-22,29,30-trisnorhopane (Tm)
12	C <sub>29</sub> 17 $\alpha$ (H),21 $\beta$ (H)-30-norhopane
13	C <sub>29</sub> 18 $\alpha$ (H)-30-norneohopane (C <sub>29</sub> Ts)
14	C <sub>30</sub> 17 $\alpha$ (H)-diahopane
15	C <sub>29</sub> 17 $\beta$ (H),21 $\alpha$ (H)-30-norhopane
16	C <sub>30</sub> 17 $\alpha$ (H),21 $\beta$ (H)-hopane
17	C <sub>30</sub> 17 $\beta$ (H),21 $\alpha$ (H)-hopane
18	C <sub>31</sub> 17 $\alpha$ (H),21 $\beta$ (H)-homohopane 22S
19	C <sub>31</sub> 17 $\alpha$ (H),21 $\beta$ (H)-homohopane 22R
20	Gammacerane
21	C <sub>32</sub> 17 $\alpha$ (H),21 $\beta$ (H)-homohopane 22S
22	C <sub>32</sub> 17 $\alpha$ (H),21 $\beta$ (H)-homohopane 22R
	Steranes ( $m/z$ 217)
23	C <sub>21</sub> sterane
24	C <sub>27</sub> 13 $\beta$ (H),17 $\alpha$ (H)-diacholestane 20S
25	C <sub>27</sub> 13 $\beta$ (H),17 $\alpha$ (H)-diacholestane 20R
26	C <sub>29</sub> 24-ethyl-13 $\beta$ (H),17 $\alpha$ (H)-diacholestane 20S
27	C <sub>27</sub> 5 $\alpha$ (H),14 $\alpha$ (H),17 $\alpha$ (H)-cholestane 20R
28	C <sub>29</sub> 24-ethyl-13 $\beta$ (H),17 $\alpha$ (H)-diacholestane 20R
29	C <sub>28</sub> 24-methyl-5 $\alpha$ (H),14 $\alpha$ (H),17 $\alpha$ (H)-cholestane 20R
30	C <sub>29</sub> 24-ethyl-5 $\alpha$ (H),14 $\alpha$ (H),17 $\alpha$ (H)-cholestane 20S
31	C <sub>29</sub> 24-ethyl-5 $\alpha$ (H),14 $\beta$ (H),17 $\beta$ (H)-cholestane 20R
32	C <sub>29</sub> 24-ethyl-5 $\alpha$ (H),14 $\beta$ (H),17 $\beta$ (H)-cholestane 20S
33	C <sub>29</sub> 24-ethyl-5 $\alpha$ (H),14 $\alpha$ (H),17 $\alpha$ (H)-cholestane 20R



**Fig. 13.**  $\delta^{13}\text{C}$  values of individual  $n$ -alkanes for the extracts of source rocks, crude oils and oil components from reservoir rocks. (a)  $\text{C}_{1d}$  source rocks; (b)  $\text{C}_{2b}$  source rocks; (c) crude oils; (d) reservoir rock D402R3; (e) reservoir rock D403R20; (f) reservoir rock DX18R35.

charging oil during reservoir filling process (Table 4, Figs. 10 and 11).

For reservoir rock D402R3, most  $\delta^{13}\text{C}$  values of individual  $n$ -alkanes for the free, adsorbed and inclusion oils are higher than  $-30\text{‰}$ , with minor  $\delta^{13}\text{C}$  values for the free and adsorbed oils are close to or lower than  $-30\text{‰}$  (Fig. 13d). For reservoir rock D403R20, all  $\delta^{13}\text{C}$  values of individual  $n$ -alkanes for the free, adsorbed and inclusion oils are higher than  $-30\text{‰}$  (Fig. 13e). For both reservoir rocks,  $\delta^{13}\text{C}$  values of individual  $n$ -alkanes generally decrease with carbon number, and are relatively lower than those of most  $\text{C}_{1d}$  source rocks from Dishuiquan outcrop.

In summary, the free, adsorbed and inclusion oils from reservoir rocks D402R3 and D403R20 were most likely generated from  $\text{C}_{1d}$  source rocks. However, substantial facies variation occurs between the source rocks for these oil components and the  $\text{C}_{1d}$  samples from Dishuiquan outcrop.

### 5.3.2. Oil components from reservoir rock DX18R35

The free, adsorbed and inclusion oils of reservoir rock DX18R35 are characterized by (1) lower  $\text{Pr}/n\text{-C}_{17}$  and  $\text{Ph}/n\text{-C}_{18}$  ratios, (2) higher ratios of  $\text{Ts}/(\text{Ts} + \text{Tm})$  and  $\text{C}_{30}$  diahopane/ $(\text{C}_{30}$  diahopane +  $\text{C}_{30}$  hopane) and (3) lower concentration of  $\text{C}_{24}$  tetracyclic terpane relative to  $\text{C}_{23}$  and  $\text{C}_{26}$  tricyclic terpanes (Table 4, Figs. 4 and 12). They correlate better with  $\text{C}_{1d}$  source rocks than do oil components from reservoir rocks D402R3 and D403R20 based on these molecular parameters. The ratios of  $\text{C}_{23}$  tricyclic terpane/ $(\text{C}_{23}$  tricyclic terpane +  $\text{C}_{30}$  hopane),  $\text{C}_{21}/(\text{C}_{21} + \Sigma\text{C}_{29})$  steranes,  $\text{C}_{20}/(\text{C}_{20} + \text{C}_{23})$  and  $\text{C}_{21}/(\text{C}_{21} + \text{C}_{23})$  tricyclic terpanes decrease from the free oil, through the adsorbed oil to the inclusion oil, demonstrating that the initial charging oil had lower maturity than the later charging oil for this reservoir rock, just as reservoir rock D402R3.

$\delta^{13}\text{C}$  values of individual  $n$ -alkanes for the free, adsorbed and inclusion oils are higher than  $-30\text{‰}$  and decrease with carbon

number (Fig. 13f). These values are substantially higher for inclusion oil than the free and adsorbed oils while slightly higher for the adsorbed oil than free oil. In addition, Pr/n-C<sub>17</sub> and Ph/n-C<sub>18</sub> ratios for inclusion oil are substantially lower than the free and adsorbed oils (Fig. 12a, d and g). Isotopic composition and molecular parameters demonstrate that the initial charging oil and later charging oil for this reservoir rock were generated from C<sub>1d</sub> source rocks but varied significantly in source facies.

## 6. Conclusions

Clear differences between the Lower Carboniferous C<sub>1d</sub> and Upper Carboniferous C<sub>2b</sub> source rocks can be observed in molecular and carbon isotopic compositions of individual *n*-alkanes. C<sub>1d</sub> source rocks are characterized by lower Pr/n-C<sub>17</sub> ratio, higher ratios of Ts/(Ts + Tm), C<sub>30</sub> diahopane/(C<sub>30</sub> diahopane + C<sub>30</sub> hopane) and gammacerane/C<sub>31</sub> hopanes, lower concentration of C<sub>24</sub> tetracyclic terpanes relative to C<sub>23</sub> and C<sub>26</sub> tricyclic terpanes compared with C<sub>2b</sub> source rocks. δ<sup>13</sup>C values of individual *n*-alkanes decrease with carbon number for C<sub>1d</sub> source rocks while opposite for C<sub>2b</sub> source rocks.

Ten oils from the Upper Carboniferous volcanic reservoirs and the Upper Permian, Upper Triassic, Lower Jurassic and Lower Cretaceous sandstone reservoirs were generated exclusively from C<sub>1d</sub> source rocks, or mainly from C<sub>1d</sub> source rocks but contaminated by minor oil components derived from source rocks within C<sub>2b</sub>, Middle Permian Pingdiqian Formation (P<sub>2p</sub>), and Lower Jurassic strata.

Molecular and carbon isotopic compositions of individual *n*-alkanes of the free, adsorbed and inclusion oils from three selected volcanic reservoir rocks demonstrate that the charging oils for these reservoirs were generated from C<sub>1d</sub> source rocks but varied significantly in facies and maturity during the filling process of the reservoirs.

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## Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.orggeochem.2014.09.011>.

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