Compound-specific stable carbon isotope analysis as a tool for correlating coal-sourced oils and interbedded shale-sourced oils in coal measures: an example from Turpan basin, north-western China

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Abstract

Molecular geochemical methods have shown that it is often difficult to differentiate between coal- and interbedded shale-sourced oils, even though coals and interbedded shales may exhibit considerable organic influx variation (e.g. land plant vs algal organic matter) due to the changes of depositional setting. However, compound-specific stable carbon isotopic compositions are sensitive to the source input variations. Typically, specific molecules are more depleted in ^{13}C with increasing content of aqueous biota. This hypothesis is examined and exemplified by comparing the stable carbon isotopic ratios of *n*-alkanes from source rock extracts and related oils of the Turpan basin, north-western China. Stable carbon isotopic values of *n*-alkanes extracted from coals and interbedded shales show that δ^{13}C values of *n*-alkanes with less than 20 carbon atoms vary only slightly. However, there are dramatic changes in the isotopic compositions of higher molecular weight *n*-alkanes. Furthermore, *n*-alkanes from coal extracts are enriched in ^{13}C relative to that of interbedded shales with excursions up to 2–3‰. This comparison enables the differentiation of coal- and interbedded shale-sourced oils, and provides information useful in assessing the hydrocarbon system of a basin. © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: Compound specific stable carbon isotope analysis; Coal-sourced oils; Interbedded shale-sourced oils; Productivities of biota; Turpan basin; China

1. Introduction

Recent work by Isaksen et al. (1998), Inan et al. (1998) and Killops et al. (1998) have unraveled the key controls on the oil and gas potential of humic coals. Isaksen et al. (1998) showed that the concentration of long-chained aliphatic hydrocarbons in the coal matrix is the main control on the oil potential of humic coals, and that a "bulk" measure of liptinite content may prove misleading when assessing the oil potential. However, questions remain as to how to characterize oils derived from coals. Powell and Boreham (1994) argued that the composition of coal-sourced oils represented the average of the hydrocarbons expelled from

the drainage area of a structure, which may not be represented by a single sample or a group of samples due to the strong heterogeneity in organic matter content and composition in coal-bearing sequence. Accordingly, It is difficult to pin the source of this type of oil down to a specific stratigraphic intervals or particular lithofacies. A key problem is that common molecular geochemistry correlation techniques may not differentiate between coals and associated shales containing dispersed organic matter from terrigenous higher plants. The traditional "correlation" of this type of oil consists of (i) the similarity between the heavy *n*-paraffin distribution of source extracts and produced crude oils (Gordon, 1985), (ii) specific biomarker compound classes (e.g. sesqui-, di-, tetra-, and pentacyclic terpanes), which can be linked back to land plant communities (e.g. Philp, 1994; Isaksen, 1995). Although similarities may exist between crude oils and potential source rock

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extracts in the biomarker assemblages, it does not support a genetic relationship because both coals and interbedded shales are usually the source beds (Hunt,1991), and they often have the same or similar biomarker distributions. One exception to this was presented by Curiale and Lin (1991). In their study, three distinctly different organic facies were identified among coals and shales in a deltaic setting. By comparing the pyrolysis-gas chromatography results of a collection of coals, Katz et al. (1991) concluded that the shales associated with the coals are possibly the primary source rocks for the most petroleum systems in coal-bearing basin in the southeastern Asian and southwestern Pacific region, particularly in the Gippsland basin (Brooks,1970; Curry et al., 1994) and in central Sumatra (Williams et al., 1985). Since the petroleum reserves of a basin strongly rely upon the source rock attributes, including organic richness, type of organic matter, source rock volume (areal extent and stratigraphic thickness), level of thermal maturity and expulsion efficiency, it is necessary and useful to develop new methods to distinguish between coal-derived oils and interbedded shale-sourced oils. Such methods will allow more reliable assessment of the source rock potential of coal-bearing basins as well as reserve estimations.

To date, much of the literature data concerning the origins of these types of oils is based on biomarker correlation techniques. However, the use of stable carbon isotopic analysis of bulk and individual compounds has shown to complement the molecular approach, and in certain instances to lead greater insights in oil-oil and oil-source correlation in terrigenous sequences (Boreham and Summons, 1999). Earlier work on the isotopic characterization of crude oils focused on the analysis of the total carbon or compound-type fractions (saturate, aromatic, polar and asphaltene) of crude oils, particularly on the use of stable carbon isotope type-curves (Stahl, 1978; Schoell, 1983; Weston et al., 1988; Clayton,1991). Given that both coals and interbedded shales can be the source for oil in coal-bearing sequence, the stable carbon isotopes of bulk kerogen or compound classes do not provide the required resolution due to the slight variation of organic matter types between coals and interbedded shales. As a support of this contention, Zhang (1997) compared the stable carbon isotopes of kerogen, total solvent extracts and bulk compound classes for coals and interbedded shales from Jurassic coal measures in the Turpan basin, north-western China. The study showed that the carbon isotopic compositions were similar between coal and interbedded shale (e.g. an average of −23.5‰ vs −23.8‰ for kerogens, and −26.6‰ vs −26.0‰, for extracts respectively). On the other hand, coals and interbedded shales in a fluvial/lacustrine bog succession may show differences in organic influx, which is reflected by the productivities of different biota (e.g. land plants vs algae). It is possible

that such variations in the gross composition of the organic matter result in little, or no difference in the molecular compositions and bulk or fraction carbon isotopes (Boreham et al., 1994). However, such differences in organic matter input may be reflected in the isotopic composition of specific compounds derived from some specific organisms thought to be dominant in the depositional environment (Boreham et al., 1994). A greater resolution of this variation in organic matter type will enhance oil-source correlations in coal-bearing basin.

Compound-specific isotope analysis can provide additional information for this purpose at the molecular level (Freeman et al., 1990). Hence, a greater understanding into the source of crude oils may be possible. In this paper, we have utilized the compound specific stable carbon isotope technique to further enhance the oil-oil and oil-source correlations (e.g. Karlsen et al., 1995; Stoddart et al., 1995). The aim of present study is to determine whether different environments of source rock deposition in coal-bearing sequence can be identified from the compound-specific stable carbon isotopic composition of source extracts and corresponding oils. The approach taken to answer this question is based on the assumption that the organic influx variations in a fluvial/lacustrine bog setting will affect the stable carbon isotopic compositions of specific compounds. Here, we concentrate our study on the stable carbon isotopic analysis of individual *n*-alkanes. The reasons for this selection are: (1) oils derived from coal measures are commonly dominated by paraffinic hydrocarbons, except for the naphthenic-aromatic oils and condensates found in the Beaufort-Mackenzie basin (Snowdon,1991); and (2) the distributions of *n*-alkanes from ancient depositional environments have been widely used to infer biological inputs, maturity, and proportions of terrestrial *vs* marine input (Tissot and Welte, 1984), and the precursor-product relationship has been well established (Peters and Moldowan, 1993). Several studies have utilized the stable carbon isotopic compositions of individual *n*-alkanes to resolve source of *n*-alkanes or *n*-alkane precursors in ancient sediments (Kennicutt and Brooks, 1990; Collister et al., 1994; Boreham et al., 1994). These results indicated that the stable carbon isotopic compositions of individual *n*-alkanes can be used as a criteria for distinguishing coal- and interbedded shale-sourced oils.

Oils and related source rocks from the Turpan basin, north-western China have been used to illustrate the application of this methodology. During the past decade, moderate-size accumulations of petroleum have been discovered in the Turpan basin. Most crude oils from the Turpan basin are volatile, light oils with API gravities in the 30–60° range and typically range in color from amber to yellow. *n*-Alkanes are the major class of compounds of the oils, and no evidence can be observed

for the oil biodegradation and phase fractionation (Chen and Zhang, 1994). Previous studies showed that the Lower and Middle Jurassic coal-bearing sequences likely act as the primary source for petroleum. Two types of potential source rocks have been identified within this section: (1) coal measures from Lower Jurassic Badaowan Formation and Middle Jurassic Xishanyao Formation, mainly deposited under limnic condition; and (2) Middle Jurassic mudstones (Qiketai Formation) deposited in a brackish lacustrine environment. Except the Qiketai lacustrine sediments containing type II organic matter, the organic matter in Badaowan and Xishanyao Formations is attributed to humic material (type III) (Huang et al., 1991; Chen and Zhang, 1994, Hendrix et al., 1995; Sun, 1999).

Although there is little doubt that oils discovered in the Turpan basin are derived mainly from the Lower and Middle Jurassic sequence, attempts to characterize a coal- or associated shale-sourced oil have proven unsuccessful. In this paper, we report the preliminary results from the identification of coal- and interbedded shale-sourced oils using stable carbon isotopic compositions of n-alkanes from n-C_{10} to n-C_{29}.

2. Analytical procedures

Oil and source rock sample information is listed in Tables 1 and 2, respectively, and sample locations are shown in Fig. 1. Sediments were soxhlet extracted with chloroform for approx. 72 h. Asphaltenes were removed from the total chloroform extracts and oils by precipitation with n-hexane followed by filtration. The de-asphalted extracts and oils were then separated into saturate, aromatic, and polar (NSO) fractions by column chromatography, using hexane, benzene, and alcohol as the solvents, respectively.

2.1. Gas chromatography

Aliphatic hydrocarbon fractions were analyzed by GC using a Hewlett-Packard 5890II Gas Chromatograph equipped with a 25 m×0.25 mm i.d. fused silica capillary column coated with a 0.17 μm film of the HP-1 phase. The temperature program was from 40°C (for oil) or 80°C (for source extracts), held isothermally for 2 min and then increased to 290°C at a rate of 4°C/min, followed by a 30 min hold at 290°C. The carrier gas was nitrogen with a flow rate of 1.0 ml/min.

2.2. Gas chromatography–mass spectrometry

GC–MS analyses were conducted on a Voyger 1000 mass spectrometer coupled to a Carlo Erba GC 8000 TOP gas chromatograph with an on-column injection system. Chromatographic separations were performed with a 30 m×0.32 mm i.d. fused silica capillary column coated with a 0.25 μm film of the CB-5 phase (Chrompack). The oven temperature program was from 55 to 205°C at 15°C/min and maintained for 5 min, and then from 205 to 300°C at 2°C/min, followed by a 30 min hold. Helium was used as carrier gas with a flow rate of 1.0 ml/min. The transfer line temperature was 250°C, and the ion source temperature was 200°C. The ion source was operated in the electron impact (EI) mode at an electron energy of 70 eV. Relative amounts of biomarker compounds were determined by multiple ion detection.

Table 1
Jurassic oil sample information and bulk composition[a]

Sample no.	Oil field	Depth (m)	Formation	API	Wax (%)	Sats (%)	Arom (%)	Res (%)	Asph (%)	Oil group (%)
TH01	Hongtai	1394–1400	Sanjianfang	43	12.9	81.8	11.4	3.5	3.3	A
TH02	Sakeshang	3087.5–3098.1	Xishanyao	43	9.9	81.0	10.4	6.5	2.1	A
TH03	Qiketai	Oil mine	Sanjianfang	n.d.	n.d.	66.2	17.4	4.0	12.4	A
TH04	Hongfu	2140.6–2188	Sanjianfang	42	3.91	78.8	14.8	3.3	3.1	A
TH05	Wenjisang	2828–2856.7	Sanjianfang	n.d.	n.d.	75.4	13.9	5.1	5.6	A
TH06	Qiuling-shanshan	3236–3247.4	Xishanyao	37	11.92	82.1	13.6	3.7	0.6	A
TH07	Bake	2379–2391	Xishanyao	43	6.69	78.5	11.8	2.6	7.1	A
TH08	Shanle	2692–2737	Xishanyao	44	19.3	82.6	7.1	8.6	1.7	A
TH09	Hongnan	2679.2–2859.6	Sanjianfang	43	21.0	82.6	10.6	5.7	1.1	A
TH10	Lianmoxin	3372–3379	Xishanyao	39	27.7	82.2	13.0	4.2	0.6	A
TH11	Shengnan	2320–2330.4	Sanjianfang	40	13	72.8	17.4	5.3	4.5	A
TH12	Shengbei	2953–2992.5	Kalaza	50	9.3	86.5	10.7	2.3	0.5	B
TH13	Shengjinkou	?	Sanjianfang	n.d.	n.d.	78.5	13.7	5.5	2.3	B
TH14	Pubei	3345.6–3351.2	Qiketai	43	9.5	78.2	15.3	4.4	2.1	B
TH15	Shenqie	2489–2493	Sanjianfang	52	5.12	79.6	13.1	3.6	3.7	B

[a] API = API gravity, Sats = saturates, Arom = aromatics, Res = resins, Asph = asphaltenes, n.d. = not determined. Saturates, aromatics, resins, and asphaltenes are in normalized percent. Oil group = interpreted genetic oil families (see text for discussion).

Table 2
Jurassic source rock description, TOC, rock-eval pyrolysis and vitrinite reflectance data[a]

| Sample no.	Oil field	Lithology	Depth (m)	Formation	TOC (wt.%)	HI (mg HC/org C)	S1 (mg/g)	S2 (mg/g)	Tmax (C)	Ro range (%)	Ro average (%)	Number of vitrinite particles
TH16	Hongfu	Coal	2742-2751	Xishanyao	79.48	197	10.96	151.11	436	0.66–0.77	0.73	38
TH17	Wenjishang	Coal	3877	Xishanyao	76.78	244	11.00	187.60	444	0.70–0.78	0.72	29
TH18	Sakeshang	Coal	3175	Xishanyao	42.87	117	5.92	50.37	439	0.45–0.57	0.51	32
TH19	Bake	Coal	2966.13	Xishanyao	62.11	187	8.20	116.20	429	0.47–0.62	0.54	38
TH20	Lianmoxin	Shale	3408.5	Xishanyao	3.62	386	0.51	12.99	429	n.d.	n.d.	n.d.
TH21	Hongnan	Shale	2789.49	Xishanyao	1.54	210	0.11	3.66	442	0.59–0.65	0.60	40
TH22	Lianmoxin	Shale	3077-3079	Qiketai	0.45	116	0.05	0.52	443	0.60–0.67	0.61	35
TH23	Qiuling-shanshan	Shale	2780-2785	Qiketai	3.92	454	0.44	17.80	431	n.d.	n.d.	n.d.
TH24	Shengbei	Shale	3996-4014	Qiketai	1.07	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.

[a] n.d. = not determined.
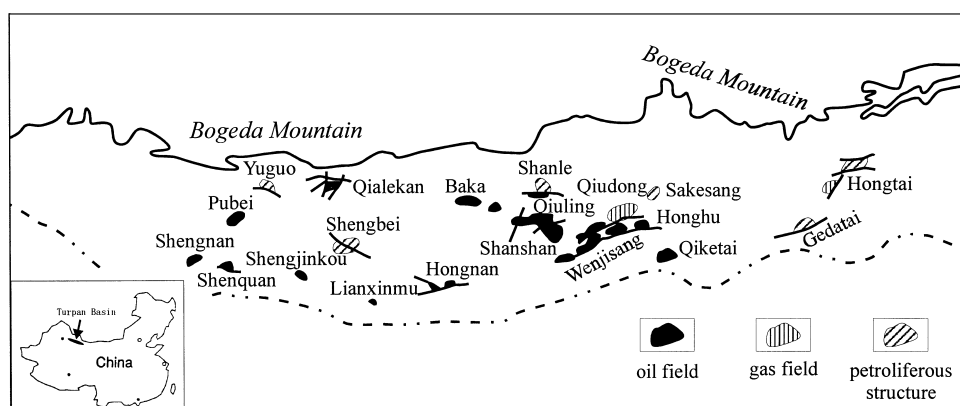
Fig. 1. Map showing location of oil fields from which samples were obtained for geochemical analysis.

2.3. Gas chromatography–isotope ratio mass spectrometry

The GC–IRMS analyses were performed on a VG Isochrom II system interfaced to a Hewlett-Packard 5890 gas chromatograph. The GC was fitted with a fused silica column (OV1, 50 m×0.32 mm i.d.) leading directly into the combustion furnace. Initial temperature of the GC oven was 70°C and held isothermally for 5 min, and then programmed to 290°C at 3°C/min, followed by a 30 min hold. Helium (12 psi) was used as a carrier gas. The injection of samples was conducted in split mode at a ratio of 35:1. The isotope values were calibrated against the reference gas and are reported in the usual "del" notation relative to PDB.

2.3.1. Controls on the instrument accuracy and data quality

The accuracy of the instrument was tested daily before sample analysis, by analyzing a mixture of *n*-alkanes and isoprenoid alkanes with known δ^{13}C values.

The reproducibility, run at different times, was typically less than ±0.3‰. For calibration, a CO_2 reference gas, which is calibrated against the Charcoal Black (a national calibrated standard, with a value of-22.43‰ based on the PDB standard), was automatically introduced into the IRMS in a series of pulses before and after the array of peaks of interest (Fig. 2).

There are two problems in GC–IRMS analysis. One is integrating the isotopic distribution across single-component GC peaks (Hayes et al., 1990); while the other is associated with the identification and the correction required for coeluting, unresoved baseline components (Collister et al., 1992). The solution of these problems is achieved using background subtraction software supplied with the Isochrom II instrument. This can be performed automatically or manually.

Saturated fractions analyzed in this study are dominated by *n*-alkanes with a carbon distribution ranging from n-C_{10} to n-C_{30}, and have a low level background matrix or UCM. Fig. 2 shows the typical m/z 44 mass chromatograms of oils and source extracts. As seen in
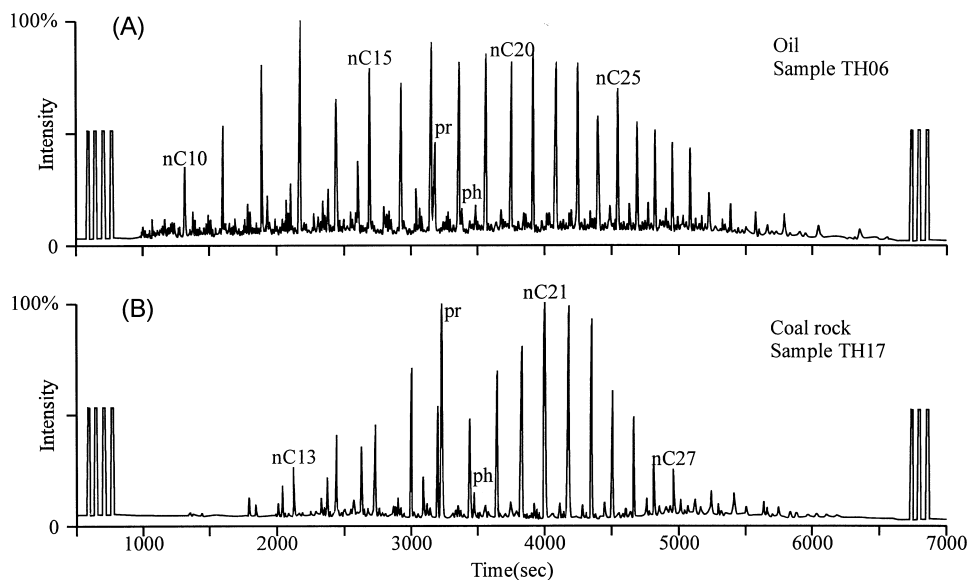
Fig. 2. Typical *m/z* 44 mass-chromatograms of crude oil (a) and source extracts (b) by GC–IRMS analysis.

Fig. 2, *n*-alkanes are well resolved from neighboring peaks and no major co-elution is observed. For these characteristics, comparison of the measured isotope values for the *n*-alkane components in the saturated fractions and isolated from the molecular sieve and urea adducted sample fractions revealed a very close match, most components however, agreed to within 0.3–0.5‰ on average, a value considered well within the instrument precision error (Wilhelms et al., 1994; Ellis and Fincannon, 1998), thus allowing accurate isotopic analyses. Alkanes with more than 29 carbons commonly coelute with compounds in the "biomarker range" (e.g. steranes and hopanes) and are therefore, not analyzed isotopically in this paper.

3. Results and discussion

3.1. Turpan basin oil characterization

3.1.1. Bulk properties

Bulk geochemical data for the oils are given in Table 1. All have a predominance of saturated hydrocarbons relative to aromatics and non-hydrocarbons. API gravities range from 37 to 52° at reservoir depths of 1394–3379 m, which suggests that the reservoired petroleum is dominated by light oil. There is very little correlation between API gravity and depth. An extremely wide variation in the wax content is typical among these crude oils, which ranges from 3.91 to 27.7%. This variation is most likely attributed to variations in coaly source rock organic matter (Isaksen et al., 1998) since phase fractionation (Thompson, 1987) and biodegrada-

tion are of no consequence in the Turpan basin (Chen and Zhang, 1994). We did not determine sulfur content of the oils, but most of Turpan basin oils have a low sulfur content (often < 0.1 wt.%) according to a previous study (Huang et al., 1991). The low sulfur contents in the oils are consistent with the proposed nonmarine effective source rocks in the Turpan basin (Huang et al., 1991; Chen and Zhang, 1994; Hendrix et al., 1995; Sun, 1999).

3.1.2. Molecular properties

Gas chromatography data from the analyses of saturated hydrocarbons of the oils are listed in Table 3. Most Turpan basin oils contain a typical *n*-alkane distribution with a predominance of a lower carbon number range (n-C_{10}–n-C_{21}), and the ratio of $C_{21}+/C_{21}$- ranging from 0.26 to 0.70. This is in agreement with the API gravity of the oils. Oils produced from various oil fields have variable pristane/phytane (Pr/Ph) ratios (2.67–6.60) indicating terrestrial organic matter input under oxic-suboxic conditions (Peters and Moldowan, 1993; Powell and Boreham, 1994). In all of the oils, the Pr/n-C_{17} and Ph/n-C_{18} ratios are less than 1.0 except for the oil produced from Sakeshang oil field, which has a Pr/n-C_{17} ratio of 1.26. The CPI values in the range of n-C_{22}–n-C_{30} varies from 1.06 to 1.14, suggesting relatively low oil maturities. There is no correlation between Pr/ Ph ratios and CPI values.

Representative mass chromatograms showing the molecular distribution of steranes (*m/z* 217) and terpanes (*m/z* 191) for oils are shown in Fig. 3. The proportions of individual classes of sterane compounds (C_{27}–C_{29}) among the Turpan basin oils exhibit

Table 3
Geochemical results for crude oils from Turpan basin[a]

| Sample no. | Pr/Ph | Pr/nC17 | Ph/nC18 | Hopanoids | | | | | | Steroids | | | | |
				CPI	C21+/C21-	22S/22R	$Ts/Ts+Tm$	GI	C30*/C29Ts	20S/20R	$\beta\beta$	C27%	C28%	C29%
TH01	6.60	0.74	0.12	1.11	0.56	0.59	0.15	3.2	1.43	0.40	0.49	12.89	16.33	70.78
TH02	6.45	1.26	0.18	1.11	0.70	0.60	0.09	3.2	1.13	0.44	0.58	11.40	13.40	75.20
TH03	4.67	0.42	0.09	1.08	0.36	0.58	0.30	4.0	1.38	0.40	0.52	13.76	21.28	64.96
TH04	4.78	0.48	0.11	1.06	0.34	0.59	0.29	3.2	1.44	0.40	0.53	12.50	22.12	65.38
TH05	4.70	0.48	0.11	1.09	0.39	0.59	0.28	3.2	1.73	0.41	0.54	12.58	22.52	64.90
TH06	3.91	0.36	0.09	1.12	0.53	0.58	0.23	3.2	1.36	0.41	0.53	12.04	22.15	65.81
TH07	5.92	0.60	0.10	1.13	0.44	0.60	0.19	2.4	1.43	0.43	0.53	14.72	17.26	68.02
TH08	5.08	0.50	0.10	1.14	0.45	0.59	0.18	2.8	1.77	0.38	0.52	13.40	18.90	67.70
TH09	4.58	0.45	0.10	1.12	0.38	0.58	0.28	4.0	0.93	0.36	0.51	20.19	19.87	59.94
TH10	5.13	0.53	0.11	1.12	0.63	0.59	0.19	4.0	1.11	0.39	0.44	16.81	21.59	61.59
TH11	3.92	0.43	0.12	1.07	0.26	0.58	0.35	6.4	0.70	0.38	0.52	18.48	22.12	59.39
TH12	2.67	0.48	0.20	1.13	0.36	0.59	0.58	8.0	0.33	0.35	0.40	35.86	20.12	44.02
TH13	2.59	0.40	0.17	1.10	0.38	0.59	0.48	7.2	0.31	0.31	0.38	31.36	20.56	48.08
TH14	3.13	0.44	0.16	1.10	0.47	0.58	0.46	6.8	0.42	0.34	0.45	29.55	19.60	50.86
TH15	3.70	0.42	0.13	1.11	0.30	0.59	0.48	7.2	0.39	0.33	0.42	29.00	20.76	50.25

[a] Pr/Ph = pristane/phytane; 22S/22R = C31 homohopane 22S/(22S + 22R); $Ts/(Ts + Tm)$ = 18α(H)-trisnorneohopane/[18α(H)-trisnorneohopane + 17α(H)-trisnorhopane]; GI = gammacerane index (gammacerane/hopane × 100); C30*/C29Ts = 17α(H)-diahopane/18α(H)-30-norneohopane; 20S/20R = 20S/(20S + 20R) C29 14α(H), 17α(H) steranes; ββ = C29 20R 14β(H),17β(H)/[14β(H),17β(H) + 14α(H),17α(H)]; C29, C28, C27 = normalized percentages in sum of C27-C29 steranes; CPI = 2 (nC23 + nC25 + nC27 + nC29)/[nC22 + 2 (nC24 + nC26 + nC28) + nC30]; C21+/C21- = (sum of the *n*-alkanes carbon number less than 21)/(sum of the *n*-alkanes carbon number more than 21).

considerable variability, but all with a predominance of C29 ethylcholestane. Additionally, almost all of the oils have a relative high amount of diasteranes, and the ratio of diasteranes/steranes ranges from 0.45 to 0.64. The pentacyclic triterpanes in the Turpan basin oils are mainly from the common hopane series. A significant feature of the m/z 191 mass chromatograms is the abundance of 17α(H)-diahopane and 18α(H)-30-nor-neohopane. Other terrigenous triterpanes such as 18α(H)-oleanane and ursane have not been detected, which is consistent with the Jurassic age of the sediments (Grantham et al., 1983; ten Haven and Rullkotter, 1988). Gammacerane is also present, but varies considerably in concentration among samples. There are minor amounts of tricyclic terpanes (C19–C26) in the oils although their intensities in the m/z 191 mass chromatograms are weak and usually clear mass spectra are not available.

3.1.3. Thermal maturity

Several molecular maturation parameters from GC and GC/MS analyses were calculated to estimate relative thermal maturities of the oils (Table 3). We have not attempted to relate these relative maturity levels to specific levels of maturation that could be measured in their source rocks. Our objective is to assign relative maturites and provide a general framework for interpreting the genetic oil families.

Ratios of pristane/n-C17 and phytane/n-C18 can be used to evaluate maturity as well as depositional environment. Maturity can be assessed where the oils come from one source rock facies (Connan, 1980). Different

maturity trends may indicate differences in depositional environment (Le Tran and Philippe, 1993). Fig. 4 reveals that all of the analyzed oils follow two maturity trends, which indicates that there are probably two main source rock types. The trend A→B is interpreted as lacustrine-sourced oils with relative low pristane/phytane ratios (2.67–3.70), and the trend A→C probably reflects oils sourced from coal measures. The latter have a larger content of land plant organic matter and higher pristane/phytane ratios than the trend A→B (see later discussion). The difference cannot be due to maturity alone and it will be argued that these oils are from different source facies of the Lower to Middle Jurassic sequence.

A better understanding of the oil maturities can be seen using hopane and sterane maturity parameters. The 22S/(22S + 22R) ratios for the C31 17α(H)-homohopanes in the Turpan basin oils are at equilibrium having a range from 0.58 to 0.60 with an average of 0.59 (Table 3), and indicate maturities at or past the onset of oil generation (Seifert and Moldowan, 1986). Typical sterane maturity parameters such as the 20S/(20S + 20R) ethylcholestane ratio and the ββ/(ββ + αα) ethylcholestane ratio are cross-plotted in Fig. 5. C29 steroid isomerization ratios 20S/(20S + 20R) are in the range of 0.31–0.44, less than the pseudo-equilibrium value of 0.55 (Seifert and Moldowan, 1986). The low isomerization ratios for these oils indicate relatively low levels of thermal stress, and below an effective maturity of 0.9% vitrinite reflection. The trend due to maturity shown in Fig. 5 suggests two oil families: Group A and B. Group
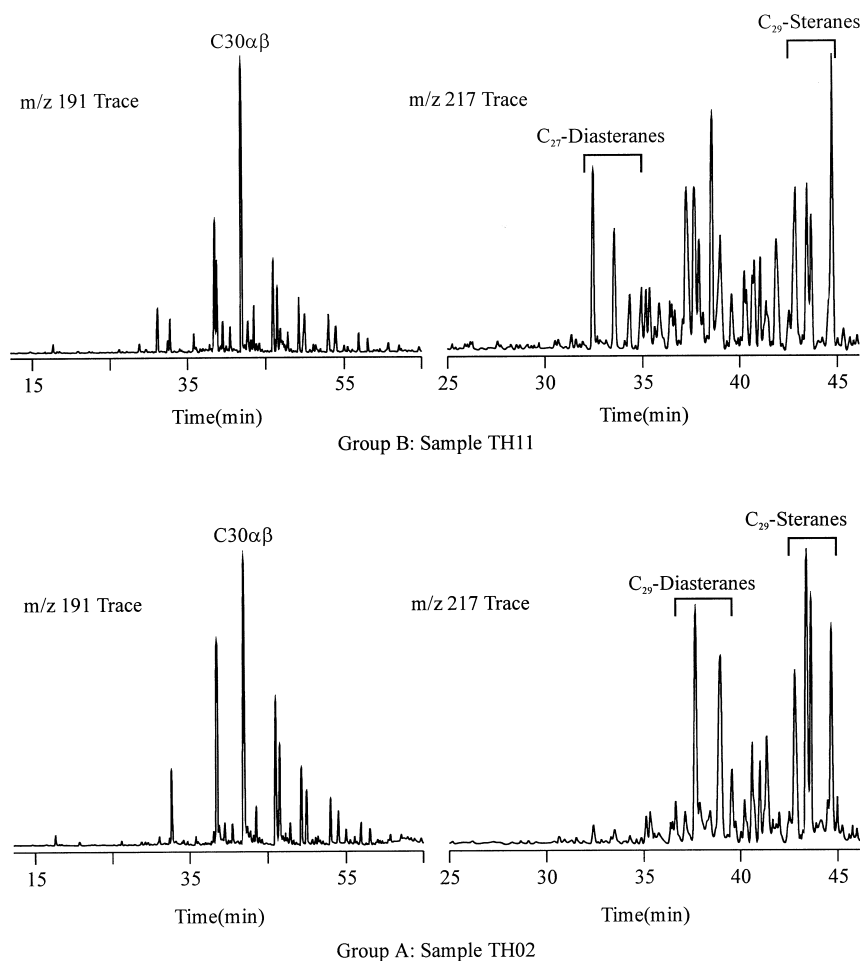
Fig. 3. Biomarker fragmentograms for *m/z* 191 hopane, *m/z* 217 sterane scans for typical Group A and B oils.
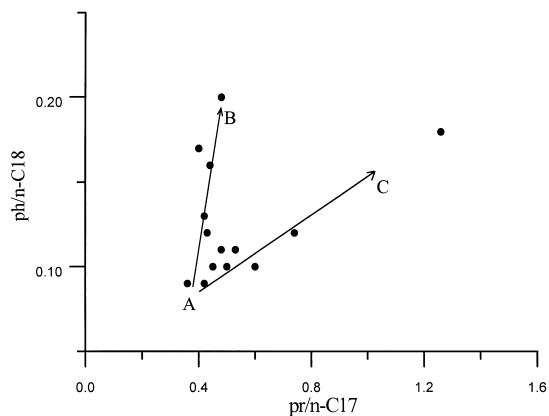

Fig. 4. Pristane/n-C_{17} ratio vs phytane/n-C_{18} ratio calculated from FID gas chromatograms of saturated hydrocarbons of oils.

A oils plot fairly close together, while Group B oils have lower maturity levels than Group A. These two oil families are considered to correlated with the Xishanyao Formation and Qiketai Formation respectively (see later discussion).

Although Ts/(Ts + Tm) ratio has been used successfully as an indicator of thermal maturity for oils and source rocks (Seifert and Moldowan, 1978), it has also been shown to have a strong source influence (Moldowan et al., 1986). Fig. 6 shows the correlation of the 20S/(20S + 20R) ethylcholestane ratios and $Ts/(Ts + Tm)$ ratios. With increasing maturity, an inverse trend is observed. Therefore, the $Ts/(Ts + Tm)$ ratio of Turpan basin oils is significantly influenced by organic facies rather than by thermal history.

3.1.4. Source rock and oil families

The thermal maturity difference between the oils suggests that there is more than one source kitchen. Molecular parameters of source rocks are summarized in
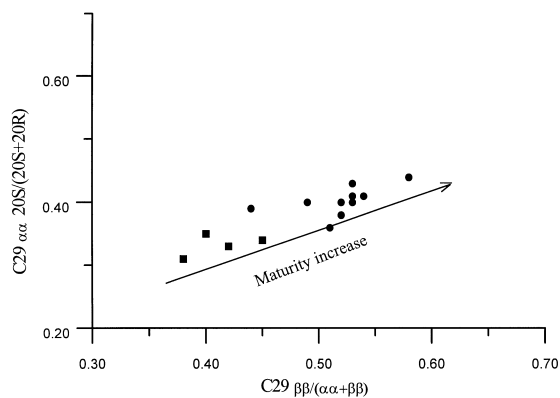
Fig. 5. C_{29} $\beta\beta/(\beta\beta + \alpha\alpha)$ vs C_{29} $\alpha\alpha\alpha$ 20S/(20S + 20R) of crude oils, calculated from MID, GC–MS analysis. ●, Group A oils; ■, group B oils.
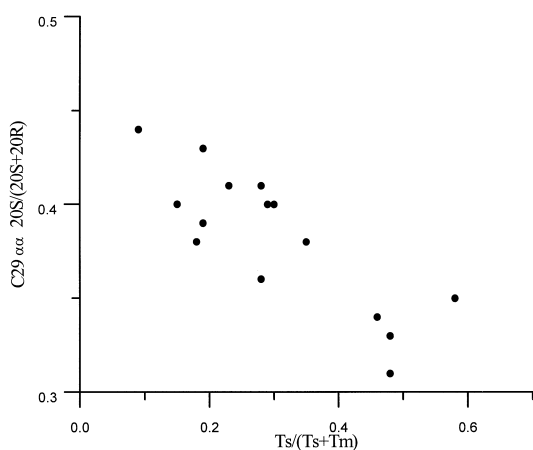

Fig. 6. C_{29} $\alpha\alpha$ 20S/(20S + 20R) vs $Ts/(Ts + Tm)$ ratio from Turpan basin oils, calculated from m/z 191 and 217 fragmentograms in the MID, GC–MS analysis.

Table 4. Fig. 7 plots the relative abundance of the C_{27}–C_{28}–C_{29} steranes of oils and source extracts, and clearly demonstrates that the oils/source rocks separate into two populations, and support the distinctions made above. Oils from Group A are well correlated with the Xishanyao coal and interbedded shale samples, whereas the Group B oils are well correlated with the Qiketai source rocks. All oils are characterized by a high concentrations of C_{29} steranes, indicating a land-plant source (Huang and Meinshein, 1979; Mackenzie et al., 1983). Of significance, however, are the variations of C_{29}- vs C_{27}-steranes of oils and related source rocks (Tables 3 and 4). It is noted that the Group B oils contain lower C_{29} sterane and higher C_{27} sterane contents than the Group A oils, indicating that depositional environments associated with the source inputs show considerable variablility in this two source kitchens. Previous studies show that the Qiketai Formation is

deposited in a semi-deep lacustrine facies (Huang et al., 1991; Wang et al., 1994), and the Xishanyao Formation represents a series of fluvial/lacustrine and swamp deposits (Wang et al., 1994; Hendrix et al., 1995). The relative contribution of land plants to these two facies is clearly evident in Fig. 7, with relative C_{29} ethylcholestane concentration increasing from the lacustrine facies (Qiketai Formation) to the swamp deposits (Xishanyao Formation). At the same time, the influence of water-column biota (measured from C_{27} cholestane) becomes evident in lacustrine facies (Qiketai Formation).

Further evidence for the differentiation of Turpan basin oils comes from the crossplot of the ratio of 18α(H)-trisnorneohopane/(18α(H)-trisnorneohopane + 17α(H)-trisnorhopane) $[Ts/(Ts + Tm)]$ against the 17α(H)-diahopane/18α(H)-30-norneohopane $[C_{30}^*/C_{29}Ts]$ (Fig. 8). Although there is possible influence of thermal maturity on the $Ts/(Ts + Tm)$ and $C_{30}^*/C_{29}Ts$ ratios, the depositional environment appears to exert the principal control on the relative Ts and Tm concentrations in this study as discussed above, and it can be used as a source-dependant parameter. On the other hand, the relative amounts of 17α(H)-diahopane and 18α(H)-30-norneohopane also depend most strongly on depositional environments, and the $C_{30}^*/C_{29}Ts$ ratios can be considered to be a function of Eh/pH in the depositional environments. High $C_{30}^*/C_{29}Ts$ ratios indicate that source rocks were deposited under oxic-suboxic conditions (Moldowan et al., 1991; Peters and Moldowan, 1993). In contrast with the Group A oils and related Xishanyao source rocks, Group B oils and related Qiketai source extracts have higher $Ts/(Ts + Tm)$ ratios and lower $C_{30}^*/C_{29}Ts$ ratios. It suggests that the Xishanyao source rocks accumulated under more oxic conditions compared to the Qiketai source rocks. This is also supported by variations in pristane/phytane and gammacerane index (Fig. 9). The depositional environments for Xishanyao and Qiketai Formations, inferred from biomarker assemblages of oils, are in accordance with the result of sedimentary facies analysis (Wang et al., 1994).

Of significance in Fig. 8, however, is the more scattered distribution of Group A oils compared to the Group B oils due to the wider range of $C_{30}^*/C_{29}Ts$ ratios (0.70–1.77) (Table 3). Although application of the $C_{30}^*/C_{29}Ts$ parameter has not been well established, the similar maturities of Group A oils (Fig. 5) suggests that the effects of thermal maturity on $C_{30}^*/C_{29}Ts$ ratios is small. An alternative explanation can be attributed to the heterogeneous nature of depositional environment in coal measures which results in the changeable redox potential of the source sediments, and thereby having a significant effect on the $C_{30}^*/C_{29}Ts$ values. The tighter cluster of Group B oils indicates the relative homogeneous nature of source rock depositional environment. Again, these findings are in agreement with the

Table 4
Biomarker data for source rocks from Turpan basin[a]

| Sample no. | Pr/Ph | Pr/nC17 | Ph/nC18 | Hopanoids | | | | | | Steroids | | | | |
				22S/22R	$Ts/Ts+Tm$	GI	C30*/C29Ts	C29Ts/C29αβ	20S/20R	ββ	C27%	C28%	C29%
TH16	11.10	2.70	0.20	0.59	0.02	2.0	1.33	0.03	0.43	0.23	5.41	14.11	80.48
TH17	10.85	1.86	0.16	0.59	0.09	2.4	0.83	0.07	0.44	0.53	9.89	15.83	74.28
TH18	3.90	0.50	0.20	0.56	0.16	9.6	0.67	0.19	0.21	0.39	20.49	19.27	60.24
TH19	3.50	0.40	0.10	0.53	0.15	4.8	0.95	0.21	0.19	0.34	17.54	18.13	64.33
TH20	1.38	0.35	0.32	0.56	0.08	14.4	0.55	0.13	0.31	0.38	21.42	29.73	48.85
TH21	2.65	0.44	0.20	0.46	0.31	5.6	0.77	0.36	0.21	0.36	13.20	26.90	59.90
TH22	2.11	1.00	0.39	0.52	0.03	7.2	0.80	0.06	0.16	0.25	32.14	20.41	47.45
TH23	1.40	0.70	0.60	0.33	0.39	11.2	0.16	0.86	0.05	0.25	38.09	15.28	46.63
TH24	1.40	0.40	0.50	0.60	0.01	4.7	0.50	0.02	0.37	0.36	23.18	26.61	50.21

[a] Pr/Ph = pristane/phytane; 22S/22R = C31 homohopane 22S/(22S + 22R); $Ts/(Ts + Tm)$ = 18α(H)-trisnorneohopane/[18α(H)-trisnorneohopane + 17α(H)-trisnorhopane]; GI = gammacerane index (gammacerane/hopane×100); C30*/C29Ts = 17α(H)-diahopane/18α(H)-30-norneohopane; C29Ts/C29αβ = 18α(H)-30-norneohopane/17α(H)-30-norhopane; 20S/20R = 20S/(20S + 20R) C29 14α(H), 17α(H) steranes; ββ = C29 20R 14β(H),17β(H)/[14β(H),17β(H) + 14α(H),17α(H)]; C29, C28, C27 = normalized percentages in sum of C27–C29 steranes.
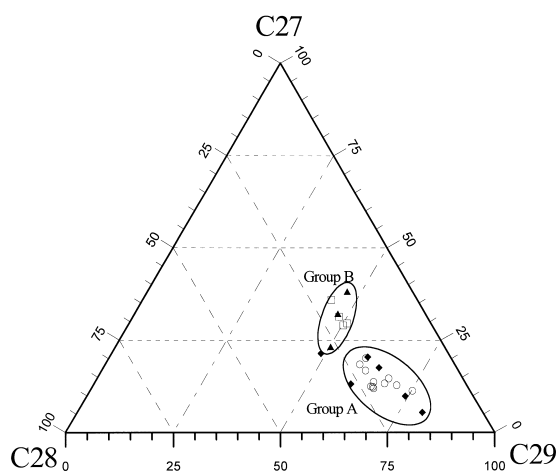

Fig. 7. Ternary diagram showing the relative abundances of C27, C28, and C29 regular steranes in the saturated hydrocarbons of oils and source extracts determinded by MID, GC–MS analysis. ○, Group A oils; □, group B oils; ◆, source rocks from Xishanyao Fm.; ▲, source rocks from Qiketai Fm.

Fig. 8. $C_{30}*/C_{29}Ts$ vs $Ts/(Ts + Tm)$ ratio calculated from m/z 191 fragmentograms in MID, GC–MS analysis. ●, Oils from group A; ■, oils from group B; ○, source rocks from Xishanyao Fm; □, source rocks fom Qiketai Fm.

Fig. 9. Variations in Pr/Ph and gammacerane index for the Turpan basin oils. GI = gammacerane index (gammacerane/hopane×100). ●, Group A oils; ■, group B oils.

descriptions of source rock depositional environments (Wang et al., 1994).

3.2. Compound specific isotope analysis

It has been shown that the traditional molecular organic geochemical parameters have separated the Turpan basin oils into two families. However, similar maturities and a broad range for some source-dependant parameters suggests heterogenous source facies for Group A oils. Based on the oil-oil and oil-source correlations, Group A oils are considered to be derived from coal measures. Therefore, both coals and associated shales are possible sources for the oils. In order to
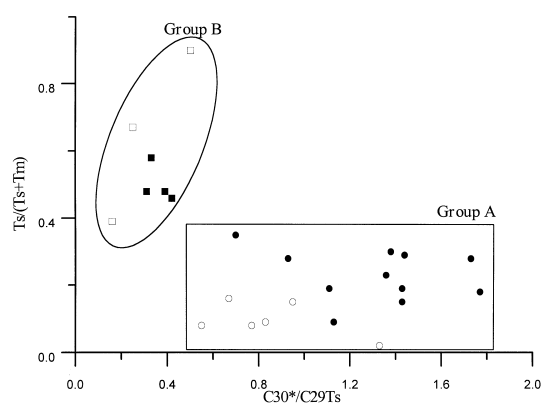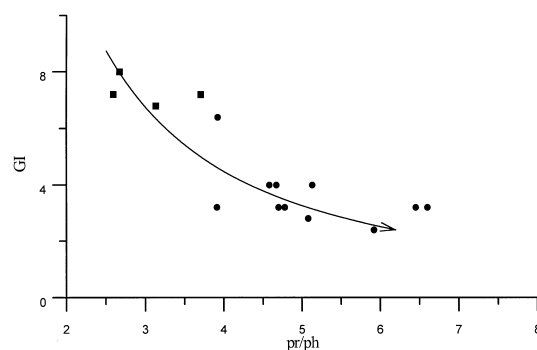
Table 5
$\delta^{13}C$ values (‰) for n-alkanes of the oils and source extracts from Turpan basin[a]

ID	TH01	TH02	TH03	TH04	TH05	TH06	TH07	TH08	TH09	TH10	TH11	TH16	TH17	TH18	TH19	TH20	TH21
n-C10	−25.06		−26.75	−25.61	−24.88	−25.47	−26.12	−26.45									
n-C11	−24.63		−26.47	−25.10	−24.96	−25.63	−25.17	−27.13	−24.52	−25.89	−24.06						
n-C12	−25.39	−24.95	−26.89	−25.64	−25.98	−26.33	−25.85	−26.62	−25.05	−25.71	−24.44						
n-C13	−25.19	−25.15	−26.65	−25.29	−25.31	−26.13	−25.40	−27.06	−24.96	−25.51	−24.61						
n-C14	−24.46	−24.85	−26.53	−25.45	−25.43	−26.54	−25.34	−26.85	−25.03	−25.93	−24.51	−26.20	−24.81	−24.26	−25.22		−25.02
n-C15	−25.25	−25.18	−26.75	−26.48	−25.35	−26.27	−25.58	−27.12	−25.38	−25.90	−24.72	−26.24	−25.25	−24.97	−25.36		−25.16
n-C16	−25.14	−25.38	−26.01	−25.61	−25.62	−26.71	−25.77	−26.91	−25.22	−25.67	−24.99	−25.81	−24.86	−25.29	−25.50	−25.87	−24.54
n-C17	−24.74	−25.37	−26.18	−25.48	−25.77	−26.70	−25.29	−25.20	−25.84	−24.76	−25.52	−25.51	−25.24	−25.82	−26.10	−25.40	
n-C18	−24.98	−25.58	−25.98	−25.55	−25.30	−26.84	−26.20	−26.82	−25.33	−25.95	−25.26	−25.60	−25.06	−25.73	−24.67	−26.44	−25.67
n-C19	−24.55	−25.39	−26.09	−25.61	−25.54	−26.93	−26.02	−26.63	−25.59	−26.29	−25.15	−25.47	−25.52	−25.54	−25.63	−26.40	−25.49
n-C20	−24.98	−25.18	−25.97	−25.61	−25.42	−26.26	−25.36	−26.09	−25.38	−26.18	−25.39	−26.03	−25.31	−24.82	−24.47	−26.18	−25.38
n-C21	−24.93	−25.15	−26.79	−25.53	−25.61	−26.03	−25.22	−26.34	−25.48	−26.13	−25.62	−25.14	−25.42	−24.92	−24.90	−26.57	−25.98
n-C22	−24.92	−24.74	−26.23	−25.47	−25.64	−26.28	−26.08	−26.62	−25.53	−26.32	−25.28	−25.12	−25.06	−24.66	−25.03	−26.59	−26.28
n-C23	−24.79	−24.51	−25.79	−25.72	−25.48	−26.35	−25.43	−26.68	−25.72	−26.40	−25.60	−25.42	−25.20	−24.63	−25.14	−27.07	−26.55
n-C24	−25.38	−25.15	−26.31	−25.81	−25.91	−26.84	−25.82	−26.98	−25.99	−26.93	−26.15	−25.76	−25.52	−24.96	−25.71	−28.08	−27.16
n-C25	−24.90	−24.75	−26.04	−25.77	−26.54	−26.37	−25.35	−26.94	−26.21	−26.76	−26.00	−25.32	−25.26	−24.46	−24.63	−27.82	−26.80
n-C26	−25.47	−24.63	−26.47	−26.70	−25.42	−26.90	−25.76	−26.65	−26.01	−27.01	−25.94	−25.16	−25.59	−25.13	−25.83	−28.17	−27.06
n-C27	−25.37	−25.00	−27.01	−26.05	−25.94	−27.32	−25.46	−27.22	−26.47	−27.50	−26.34	−25.40	−25.17	−25.37	−25.31	−27.97	−26.84
n-C28	−25.33	−25.07	−27.12	−26.41	−26.52	−27.36	−26.26	−27.80	−26.54	−27.60	−27.14	−25.93	−25.62	−25.57	−25.47	−27.95	−26.90
n-C29	−25.49	−24.74	−26.48	−26.89	−26.69	−27.43	−25.85	−26.99	−26.78	−27.42	−26.92	−26.36	−25.33	−26.10	−26.18	−27.75	−26.90

[a] n-CX = n-alkane of carbon number X.

attempt a differentiation of coal-sourced oils and interbedded shale-sourced oils, we utilized the GC/IRMS technique to gain a better understanding of the source variations of the Group A oils.

Stable carbon isotopic compositions for n-alkanes from oils and source extracts are summarized in Table 5. n-Alkane isotope profiles for the source extracts are plotted in Fig. 10, and two distinct isotope curves can be identified. Shales show a trend toward isotopically lighter values with increasing n-alkane chain length (negative slope) with an isotopic range being typically 2–3‰. This phenomenon has been observed previously and is a characteristic of terriestrial organic matter and related oils (Bjorøy et al., 1991; Dzou and Hughes, 1993; Wilhelms et al., 1994; Murray et al., 1994). Coals show that n-alkane isotope curves are relatively flat across the range. This trend is a characteristic of a homogenous precursor for the derivation of n-alkanes. Under such circumstances there is typically not much of an isotopic differences between long and short chain alkanes (Eglinton, 1994; Murray et al., 1994). Alternatively, n-alkanes, particularly those with carbon number less than 21, could be derived from algae that happen to produce isotopically similar alkanes as the land plants (Boreham et al., 1994). In addition to chain length dependant isotope effects, there are dramatic changes in the n-alkane isotope ratios in the range n-C_{20}–n-C_{29} for coal and interbedded shale extracts. n-Alkanes from coal extracts are enriched in ^{13}C relative to those of interbedded shale extracts. Carbon isotope excursions are in the range of 2–3‰. This finding is in

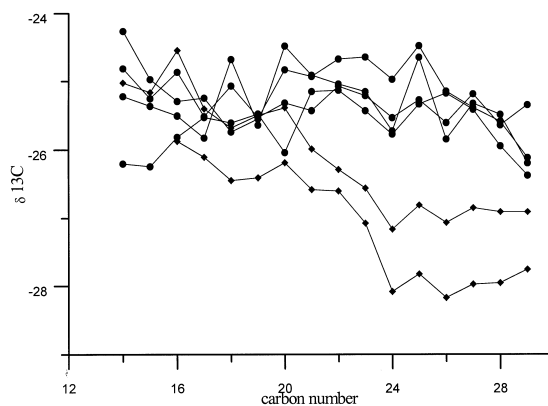Fig. 10. Range of isotope values of n-alkanes in coal and interbedded shale samples. ●, Coals; ◆, inerbedded shales.

contrast to that of a study of the Permian Toolachee coal measures from Cooper and Eromanga basin, Australia, where the n-alkanes isotopic profiles of shale extracts is heavier than that of coal extracts (Boreham and Summons, 1999), and highlights the value of site-specific geochemical analysis.

In view of the similar maturities of source rocks (Table 2), cracking and other secondary processes cannot have significantly influenced the n-alkane isotope ratios (Bjorøy et al., 1992; Clayton and Bjorøy, 1994). Since vitrinite reflectance values for most studied samples suggest that these source rocks are close to or at the onset of the conventional oil window (Ro% 0.5–0.7), there may be a difference between the proportion of free

n-alkanes (bitumen) and those released from the kerogen, which will be generated by the different chemical kinetics between the coal and shale (Collister et al., 1992; Boreham et al., 1994; Murray et al., 1994). However, the difference of isotope profiles for coal and interbedded shale extracts has to be mostly due to the source input variations. The source of organic matter in the sedimentary system include predominantly algae and the remains of terrestrial vegetation. Terrestrial organic matter exhibits varying $\delta^{13}C$ values, depending on the photosynthetic pathways used by the plants. The observed range of $\delta^{13}C$ values for C3 plants is about-23‰ to-34‰ with an average value of about-27‰ (Smith and Epstein, 1971; Benedict, 1978). Our result is in generally agreement with C3 photosynthetic pathways. Planktonic algae can have $\delta^{13}C$ values more negative than those of terrestrial plants due to the high concentrations of dissovled CO_2 in fluvial/lacustrine bog systems (Herczeg and Fairbanks, 1987; Hollander and Mckenzie, 1991; Mayer and Schwark, 1999). Thus, more negative ratios are predicted if algal productivity prevails

in the sedimentary record, whereas more positive $\delta^{13}C$ values can be indicative of input from predominantly terrestrial biomass. A study of the Green River oil shale revealed that the isotopic compositions of the *n*-alkanes are different between algae-sourced organic matter and terrestrial plant-sourced organic matter. Comparatively, *n*-alkanes derived from algae are more depleted in ^{13}C (Collister, 1992). Considerable organic influx variations, particularly algae *vs* land plants, are likely to take place in a fluvial/lacustrine bog setting. We attributed a high proportion of land plant organic matter for the coals, and greater contents of algal material for the interbedded shales. Hence, source-dependant isotope effects are expected to cause an enrichment of ^{13}C for *n*-alkanes extracted from coals with respect to that of interbedded shales.

The data in Fig. 10 suggests that the shapes of the *n*-alkane isotope profiles from coals and interbedded shales have the potential to differentiate the coal-sourced oils from interbedded shale-sourced oils. Stable carbon isotopic compositions of the *n*-alkanes from n-C_{10} to n-C_{29} of Group A oils are determined (Table 5). Because there is no significant difference between
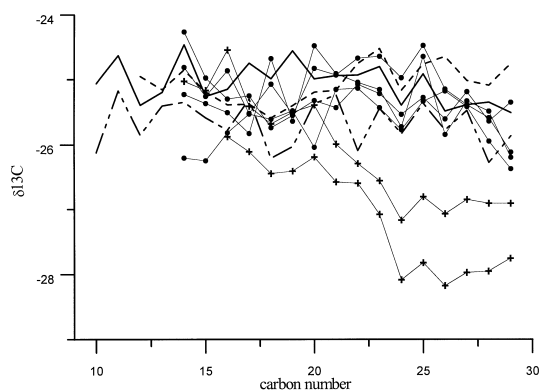

Fig. 11. *n*-Alkane isotope profiles of coal-sourced oils in Turpan basin. ●, Coals; +, interbedded shales; ----, oil TH01; —, oil TH02; – - –, oil TH07.
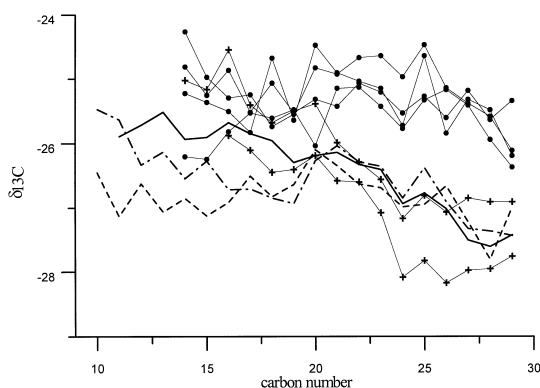

Fig. 12. *n*-Alkane isotope profiles of interbedded shale-sourced oils in Turpan basin. ●, Coals; +, interbedded shales; —, oil TH10; – - –, oil TH06; ----, oil TH08.
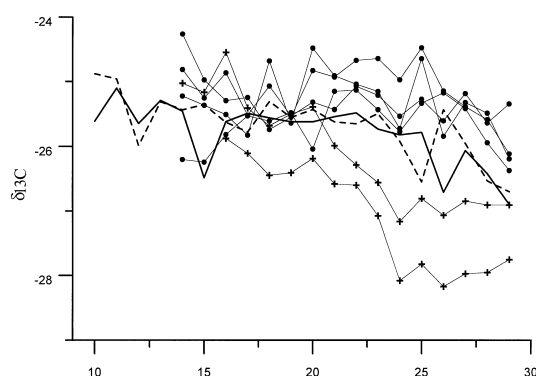

Fig. 13. *n*-Alkane isotope profiles of mix-sourced oils from Wenjishang and Honghu oil fields. ●, Coals; +, interbedded shales; —, oil TH04, ----, oil TH05.
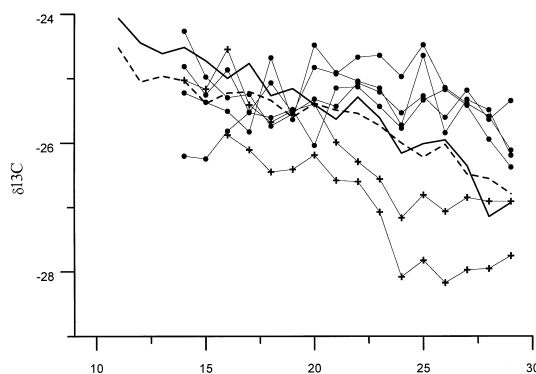

Fig. 14. *n*-Alkane isotope profiles of mix-sourced oils from Hongnan and Shengnan oil fields. ●, Coals; +, interbedded shales; ----, oil TH09; —, oil TH11.
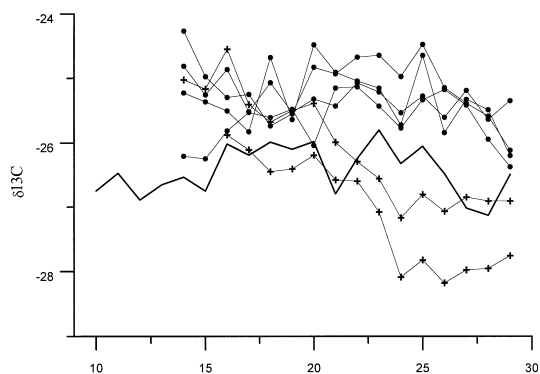
Fig. 15. *n*-Alkane isotope profiles of mix-sourced oils from Qiketai oil mine. ●, Coals; +, interbedded shales; —, oil TH03.

maturity of oils and source rocks based on the maturity parameters derived from biomarkers (Tables 3 and 4), oil-source correlations can be undertaken using *n*-alkane isotopic curves of oils and sediments, and shown in Figs. 11–15. Most oils are characterized by a n-C_{10}–n-C_{29} *n*-alkane isotopic compositions with a range from-28 to-24‰, yet show wide variations in *n*-alkane isotope profiles. In view of the fairly similar maturities of Group A oils (based on the various maturity parameters discussed above, see Table 3 and Fig. 5), this change is considered to be due to the variation of source input/depositional setting. The oils from Sakeshang oil field, Bake oil field and Hongtai oil field, have similar isotopic profiles to the coal extracts (Fig. 11), indicating these oils are mainly generated from coal seams. In contrast, the oils from Qiuling-shanshan oil field, Lianmoxin oil field and Shanle oil field, show isotopic patterns were closely related to the shale extracts (Fig. 12), indicating these oils are mainly derived from shales. The remaining Group A oils, however, are believed to be of mixed source from both coals and interbedded shales (Figs. 13–15). The n-C_{20}–n-C_{29} *n*-alkanes of oils from Wenjishang oil field and Hongfu oil field, are slightly more depleted in ^{13}C than that of coal extracts (Fig. 13), which could be interpreted as a considerable contribution from interbedded shales. Other mix-sourced oils include the Hongnan oil field, Shengnan oil field (Fig. 14) and Qiketai oil mine (Fig. 15).

4. Summary

Selected oil samples from Turpan basin have been characterized by organic geochemistry techniques and correlated to their source rocks. Two major oil families are differentiated:

- Group A oils, derived from coal measures deposited in a strong oxic environment with organic matter from higher land plants (Xishanyao Formation).

- Group B oils, derived from a lacustrine source rock of a suboxic, brackish environment with organic matter bearing a relatively high amount of plankton and low amounts of land-plant input (Qiketai Formation).

Compound-specific isotope analysis has been used to further differentiate coal-sourced and interbedded shale-sourced Group A oils. Based on the carbon isotopic pattern of *n*-alkanes from source rock extracts and oils in the Turpan basin, some important conclusions are drawn: (i) coal-sourced oils are mainly present in Bake, Sakeshang and Hongtai oil fields, (ii) shale-sourced oils occur in the Qiuling-shanshan, Lianmoxin and Shanle oil fields and, (iii) mixed oils were found in Wenjisang, Honghu, Shengnan, Hongnan oil fields and Qiketai oil mine.

Acknowledgements

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