Chinese Science Bulletin 2004 Vol. 49 Supp. I 19—26

Constraints on the diversity of crude oil types in the Lunnan Oilfield, Tarim Basin, NW China

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Abstract **Hydrocarbons, include heavy oils, normal oils, waxy oils and light oils, condensate oils and gases were all found in Lunnan Oilfield, the occurrences of hydrocarbons with complicated physical properties indicates a complicated distribution of reservoirs. By the drilling data, the distribution pattern had been found that, in plan view, the density of crude oils in the western part is heavier than that of oils in the eastern part in Lunnan region, namely the crude oils were mainly found in the western part while the natural gases were mainly found in the eastern, which shows that an obvious differentiation in the east and west part was presented in the Lunnan region. Furthermore, in vertical view, the light hydrocarbons were mainly found in the Carboniferous reservoir while the heavier hydrocarbons were mainly found in the Ordovician and Triassic reservoirs. By analysis of the semi-logarithm distribution of *n*-alkanes, biomarker ratios and stable carbon isotopic compositions of crude oils, we have found that two oil families and discontinuous distribution were presented in hydrocarbons in Lunnan Oilfield. This kind of discontinuous distribution of geochemical parameters was not from continuous fractionation in migration process. The biodegraded heavy oil reservoirs in western part belong to the lower matured oil family, while the light or condensate oils reservoirs belong to higher matured oil family. Thus, a mixed filling of hydrocarbons was occurred in the middle part in Lunnan Oilfield and resulted in the occurrence of waxy oil. The mixed filling model may help to explain the distribution pattern of hydrocarbon reservoirs in Lunnan Oilfield, especially can lead to a relatively consistent explanation for all kinds of geochemical data, such as physical properties, pyrrolic nitrogen compounds, biomarker parameters and stable carbon isotopic compositions.**

Keywords: mixing, differentiation in eastern and western part, oil family, light oil, high waxy oil, semi-logarithm distribution of *n*-alkanes.

DOI: 10.1360/04wd0344

Hydrocarbons, include heavy oils, normal oils, waxy oils and light oils and condensate oils together with natural gases were all found in Lunnan Oilfield, the occurrences of hydrocarbons with complicated physical properties indicates a complicated distribution of reservoirs. In vertical view, hydrocarbons were produced in the Ordovician carbonate reservoir, Carboniferous, Triassic and Jurassic clastic reservoirs. And the light crude oils were mainly found in the Carboniferous reservoirs while the heavy oils were mainly found in the Ordovician and Triassic reservoirs, which showed a distribution characteristic of light oils in middle production formation. Especially, the oils with medium-high content of wax were mainly distributed in the Ordovician and Carboniferous carbonate reservoirs.

In plan view, the Lunnan tectonic structure can be divided into the eastern and western part by the fault Lunnan 40. By the drilling data, the light oils, condensate oils, high waxy oils and normal oils were mainly found in the eastern part, while the normal oils and heavy oils were mainly presented in the western part. Especially, the high waxy oils were mainly found in the middle part. As a whole, a distribution pattern of hydrocarbons has been found that, the density of crude oils in the western part is heavier than that of oils in the eastern part in Lunnan region, namely the crude oils were mainly found in the western part while the natural gases were mainly found in the eastern, which shows that an obvious differentiation in the eastern and western part was existed in the Lunnan region.

For the above mentioned distribution patterns of hydrocarbons in Lunnan Oilfield[1,2], evaporation fractionation[3—7], migration fractionation[8,9] and gas washing[1,2,10] were successively brought forward to explain the distribution pattern of hydrocarbons in the Lunnan region, and they had been widely accepted. However, some unexplainable and paradoxical phenomenon or data were existed in these models. In this paper, on the basis of systemic sampling and multiple-technologic measurement, especially by the organic geochemical analyses for crude oils, a mixed filling model was put forward to explain the distribution pattern and diversity of crude oil types in Lunnan Oilfield.

1 Samples and analyses

All the analyzed 48 crude oil samples were collected from the Lunnan Oilfield, Tarim Basin, NW China. They mainly came from the Ordovician, Carboniferous and Triassic production formations, several samples in Jurassic production formations. Asphaltenes for all oils were removed by precipitation with *n*-hexane followed by filtration. The deasphalted oils were then separated into saturated, aromatic and polar (NSO) fractions by silica column chromatography, using *n*-hexane, benzene and ethanol as the solvents, respectively. The saturated and aromatic fractions were further injected into gas chromatography-mass spectrometry (GC-MS) for analyses.

The whole hydrocarbon chromatographic quantitative analysis method: the weighted oils were added into all-deuteriumed $nC_{20}d_{42}$ and known-volumed dichloromethane, and then preserved in sample vials with teflon cushion. After GC analysis, the mole fraction of *n*-alkanes was obtained with inner standard ($nC_{20}d_{42}$) and its logarithm values were plotted with the carbon number to gain the distribution curve of *n*-alkanes.

Whole hydrocarbon analysis were finished by GC using a Hewlett-Packard 6890 gas chromatograph equipped with a 30 m×0.32 mm i.d. fused silica capillary column coated with a 0.25 μm film of DB-5. The temperature program started at 65℃, held isothermally for 1 min and then increased to 290℃ at the rate of 3℃/min, followed by a 20 min hold at 290℃. The carrier gas was helium at a flow rate of 1.0 mL/min.

GC-MS analysis was conducted on a Micromass Platform II mass spectrometer coupled to a Hewlett-Packard 6890 gas chromatograph. Chromatographic separation was achieved with a 30 m×0.32 mm i.d. fused silica capillary column coated with a 0.25 um film of DB-5 (J&W). The oven temperature program started at 65 to 290℃ at 3℃/min and held for 30 min. Helium was used as the carrier gas at a flow rate of 1.0 mL/min. The transfer line temperature was 250℃, and the ion source temperature was 200℃. The ion source was operated in the electron impact (EI) mode at 70 eV.

The molecular weight analysis of asphaltene was finished by the Gel Permeation Chromatography (GPC) method which conducted on a Hewlett-Packard 1100 High Performance Liquid Chromatograph equipped with DAD detector. Chromatographic separation was achieved with a 30 m×7.5 mm i.d. fused capillary column coated with a 5 μm film of Plgel MIXCD-D type (purchased from Polymer Co.). HPLC graded Tetrahydrofuran (THF) was used as the mobile phase at a flow rate of 1.0 mL/min. The detected wavelength was set at 254 nm. Polystyrene Standards (0.5, 1, 2, 3, 5, 10, 20, 30, 70, 100 K, purchased from Fluka Co., USA) were used to correct the molecular weight value of asphaltene.

2 Results

(i) Distribution of asphaltene content. The characteristics of differentiation in eastern and western part in Lunnan Oilfield, to some extents, can be embodied in the asphaltene content of crude oils. As shown in Fig. 1, asphaltene fraction cannot be separated in the crude oils in wells LN4, LN30, LN9 and LN22 in the eastern part because of its presence with minimum content. The low asphaltene contents in crude oils were 1.53% in well LN10, 2.34% in well LN14 and 1.86% in well LN48, respectively. On the contrary, the asphaltene content in crude oils in western part was relatively higher, for example, the value was 3.83% in well LN19, 4.61% in well LN44, respectively. Especially, the asphaltene content can be reached 10.13% and 19.08% for crude oils in well LN11 and well LN1.

(ii) Semi-logarithm distribution of *n*-alkanes in crude oils. As shown in Fig. 2, the logarithm values of mole fraction of *n*-alkanes in crude oils in wells LN4, LN9 and LN22 in eastern part decreased sharply with increased carbon number. For example, the logarithm value of mole fraction can be decreased from −1.0 at C_8 to −4.0 at C_{30}, which shows an evident characteristic with high slope. On the contrary, the values in crude oils in wells LN1, LN8 and LN19 in western part have little change, which shows a mild distribution curve with a low slope. The variation in semi-logarithm distribution of *n*-alkanes in crude oils in eastern and western part is coincident with the above-mentioned distribution pattern of hydrocarbons in Lunnan Oilfield.

(iii) Distribution of biomarker ratios. In distribution of biomarker ratios, an evident differentiation was presented in crude oils in eastern and western part in

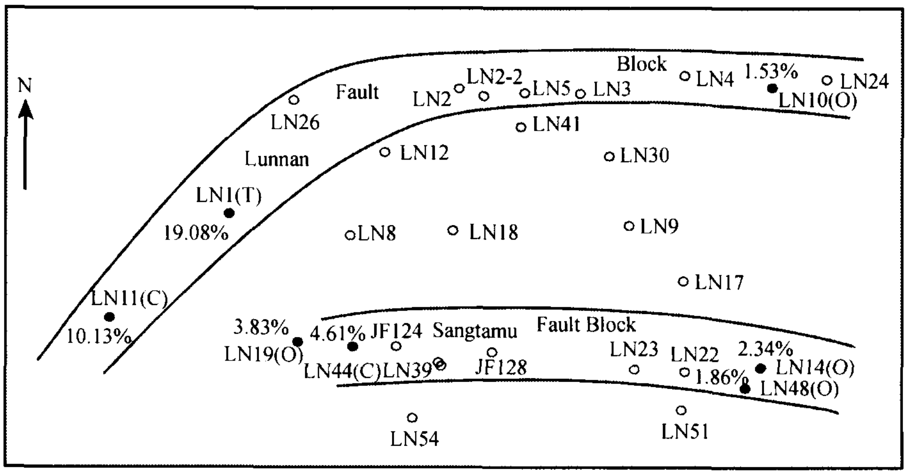

Fig. 1. Distribution of asphaltene content in crude oils in eastern and western part in Lunnan Oilfield, Tarim Basin.

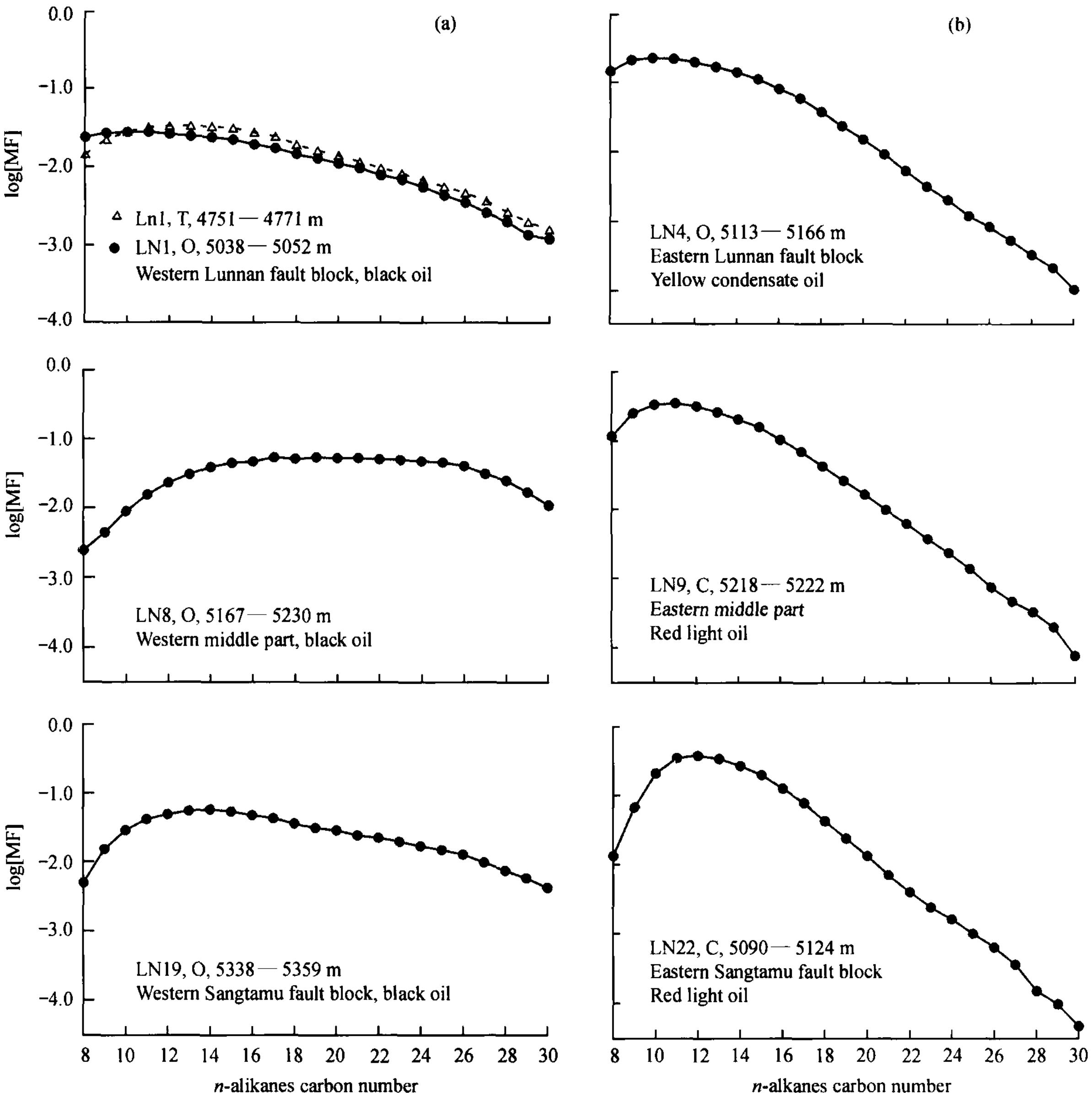

Fig. 2. Semi-logarithm distribution of mole fraction for *n*-alkanes of crude oils in western and eastern part of Lunnan Oilfield, Tarim Basin.

Lunnan Oilfield. For example, in saturated fraction, many biomarker parameters, such as 4,4,8,8,9-rearranged drimane/C_{15}-β(H) drimane, C_{15}-α(H) drimane/C_{15}-β(H) drimane, C_{20}/C_{21}-terpane, hopane/seisquiterpane, hopane/terpane, Ts/(Ts+Tm) and C_{29}-ααα sterane 20S/(20S+20R) etc., large variation were existed in crude oils in eastern and western part in Lunnan Oilfield (Fig. 3(a)). In the same time, in aromatic fraction, 1/4-methyldibenzothiophene, iC_{15}/C_{13}-aryl isoprenoids, C_{13}/C_{14}-aryl isoprenoids, iC_{15}/C_{15}-aryl isoprenoids, MPI_3 and F_1 etc., great variations were existed in crude oils in eastern and western part in Lunnan Oilfield (Fig. 3(b)). In crude oils in the middle part, the distribution of biomarkers exhibits a mixed characteristic of hydrocarbons in eastern and eastern and western part.

3 Discussion

(i) Reasons for differentiation in eastern and western part. Firstly, in the semi-logarithm distribution curve of mole fraction for *n*-alkanes of crude oils as shown in Fig. 2, under the condition of the same source, a larger slope curve means a relatively high maturity while a small slope curve means a relatively low maturity. In the same time, high content asphaltene were found in lower matured oils and few asphaltene were found in high matured oils. Especially, many ratios, such as 4,4,8,8,9-rearranged drimane/C_{15}-β(H) drimane, C_{15}-α(H) drimane/C_{15}-β(H) drimane, Ts/(Ts+Tm), C_{29}-ααα sterane 20S/(20S+20R)

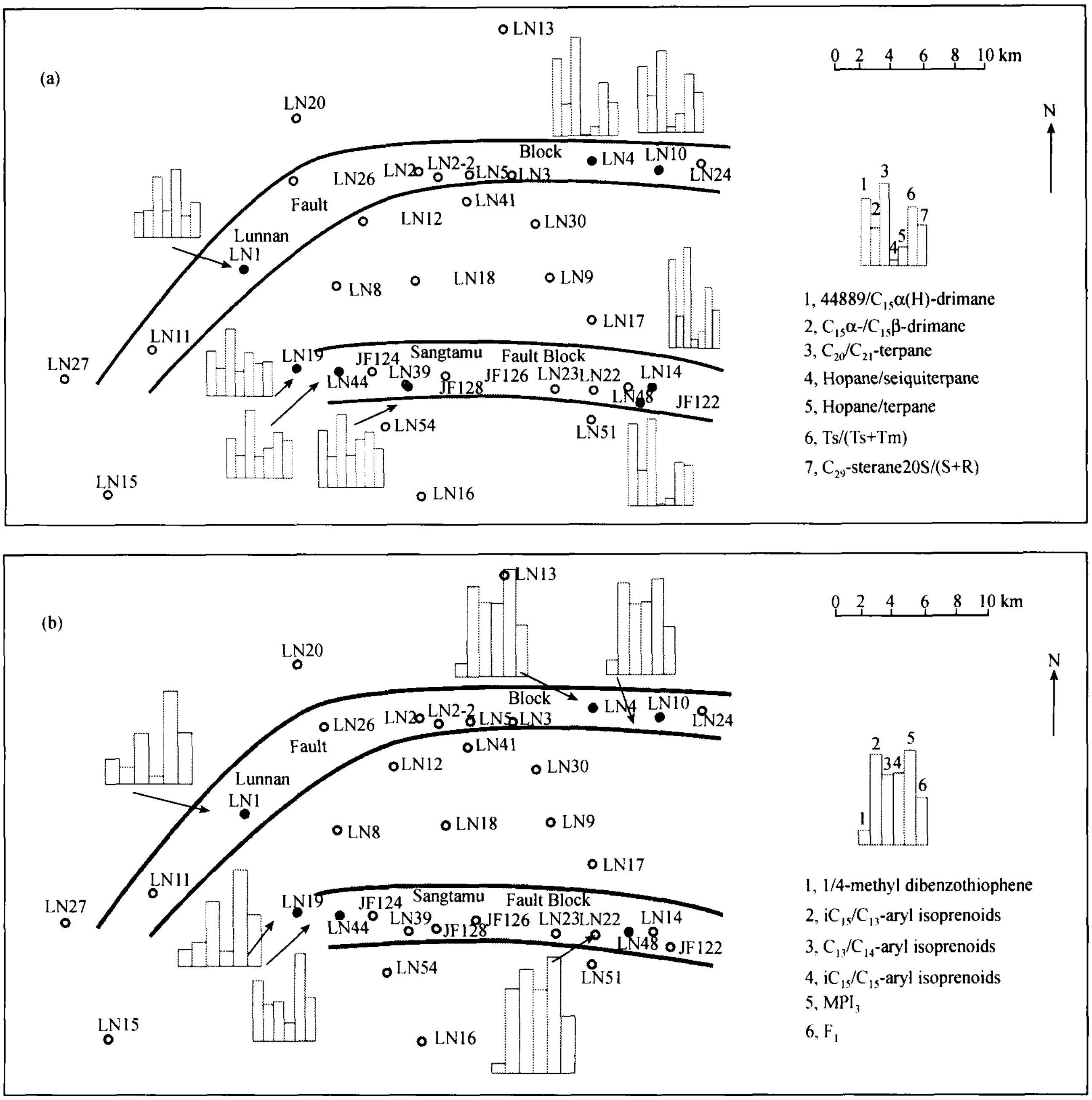

Fig. 3. Distribution of some geochemical parameters of compounds in saturated (a) and aromatic (b) fractions of crude oils in Lunnan Oilfield, Tarim Basin. C_{13}, C_{14}, C_{15}-aryl isoprenoids are mainly with 2,3,6-thimethyl structure, iC_{15}-aryl isoprenoids are with speculated 2,3,4-thimethyl structure. MPI_3= (3-MP+2-MP)/(9-MP+1-MP); F_1= (3-MP+2-MP)/(3-MP+2-MP+9-MP+1-MP).

and 1/4-methyldibenzothiophene, MPI_3 and F_1 etc., are all widely accepted parameters with mature meanings. To some extent, the ratios such as iC_{15}/C_{13}-aryl isoprenoids, C_{13}/C_{14}-aryl isoprenoids and iC_{15}/C_{15}-aryl isoprenoids are relative to the mature stage of hydrocarbons. Thus, the variations in eastern and western part in the above-mentioned parameters with mature meaning, obviously indicate the differentiation of hydrocarbon in eastern and western part.

Secondly, the other selected parameters, such as C_{20}/C_{21}-terpane, hopane/seisquiterpane, hopane/terpane etc., are all relative to the source rock and depositional environment. Thus, the variation of such parameters in eastern and western part, possibly indicates a differences in eastern and western part in source rock and depositional environment.

Moreover, obviously, by biomarkers and stable carbon isotopic analysis as shown in Fig. 4, two oil family

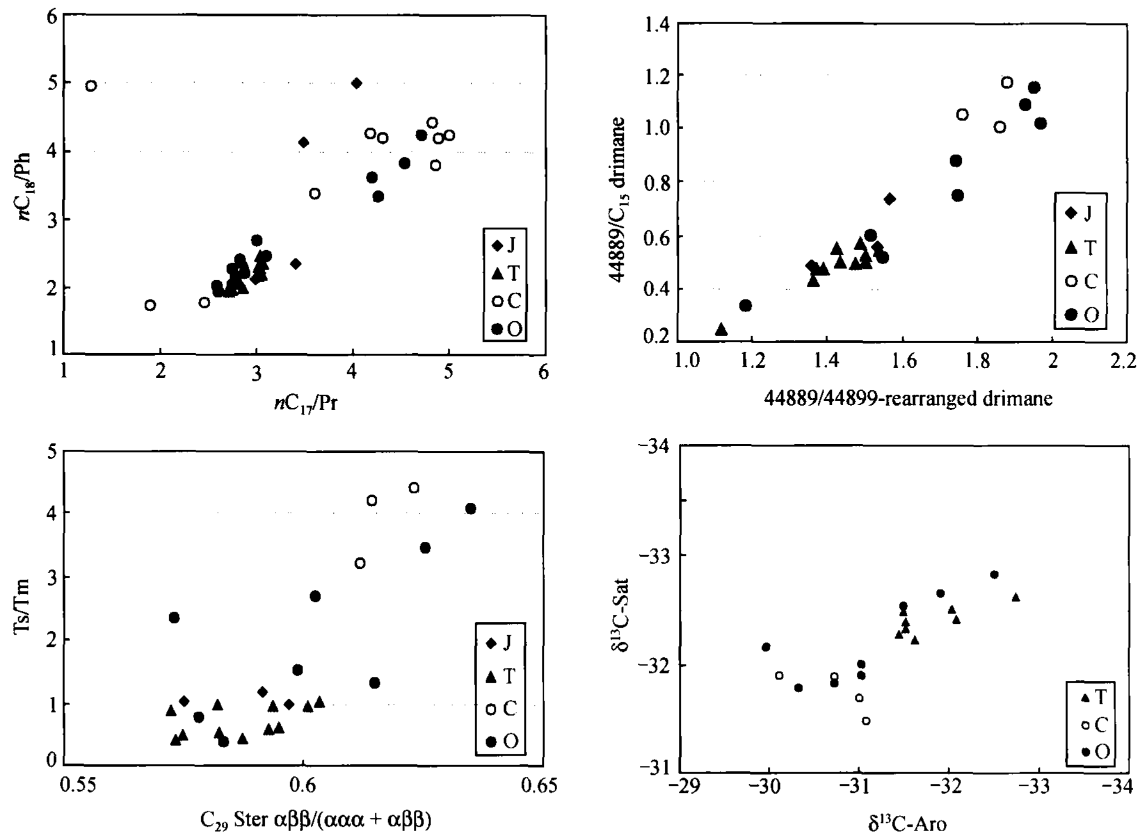

Fig. 4. Distribution of molecular geochemical parameters in crude oils in Lunnan Oilfield. It can be seen that two oil families and discontinuous distribution was presented in the Ordovician and Carboniferous crude oils. (44889/44899=4,4,8,8,9-rearranged drimane/4,4,8,9,9-rearranged drimane).

and discontinuous distribution were presented in crude oils in Ordovician and Carboniferous reservoirs in Lunnan oilfield[12].

Obviously, the above-mentioned discontinuous distribution phenomenon in maturity parameters cannot come from continuous process of migration fractionation. For the Sangtamu fault block, although the western part is locally higher than that of the eastern part in early Nogene period, and the eastern part is locally higher than that of the western part in late Nogene period, hydrocarbons may seems to suffer a possible process of migration and regulation from west to east. However, the above-mentioned variation in maturity index in eastern and western part, evidently, is not produced by a continuous migration fractionation process, because a continuous migration process cannot produce large differences in physical properties of crude oils, moreover, the maturity ratios should be sequentially decreased from west to east.

It has been approved by the fluid-inclusion data that, an uniform distribution was presented in the homogenization temperature data in western part which shows a single period of hydrocarbon filling, while a characteristic with two or more periods of hydrocarbon filling was existed in the homogenization temperature data in light or condensate oil reservoirs in eastern part[13,14].

In conclusion, the differentiation in eastern and western part, two oil families and discontinuous distribution reflected by the physical properties and geochemical parameters proved that, two oil families were presented in the Lunnan oilfield, the heavy oil reservoirs in western part belong to the lower matured oil family, while the light or condensate oils reservoirs belong to relatively high matured oil family. The variations between these two oil families in source rocks, depositional environment or period of hydrocarbon filling, especially the difference in maturity, were possibly the main reasons of hydrocarbon distribution.

(ii) Explanation for diversity in crude oil types. In physical properties, light oils, condensate oils, waxy oils, normal oils and natural gases were mainly found in eastern part while normal oils and heavy oils were mainly presented in western part. Thus, a mixed filling process between light or condensate oils and normal or heavy oils can be occurred in the middle part.

The mixing between light oils and heavy oils may have a wide effect on crude oil properties. Typically, when the light or condensate oils and natural gases were filled into the heavy oil or normal oil reservoirs, the increased pressure of reservoir will largely lead to increasing the solubility of higher carbon numbered *n*-alkanes, as a result, waxy oils can be possibly produced. In the same time, a deasphaltened phenomenon will be occurred in subsurface reservoir as observed in precipitation experiment in laboratory.

For example, in Ordovician and Carboniferous carbonate reservoirs in well LN14 in Sangtamu fault block, the semi-logarithm distribution curve of mole fraction for *n*-alkanes was changed by the occurrence of invasion of light oils and natural gases, the increased logarithm values of mole fraction for C_{21}—C_{29} *n*-alkanes reflect that an obvious wax deposit phenomenon occurred in reservoirs, as a result, waxy oils were produced.

In fact, the following phenomenon can also be occurred in the wax deposit process:

(1) Asphaltene deposit. According to principle of dissolution, asphaltene should not dissolve in nonpolar solvent such as pentane, hexane and heptane etc., it should dissolve in polar solvent such as benzene, toluene and chloroform etc., because it belongs to organic macromolecular with a polar group. As a result, asphaltene will be separated when hexane or heptane was added into crude oils under the laboratory condition. For the same reason, asphaltene will be precipitated in the bottom of reservoir or relatively higher porous and permeable zone when light or condensate oils and natural gases with large quantitative *n*-alkanes were filling into reservoirs. Moreover, wax with high molecular weight (>C_{30}) will also be deposited (as shown in Fig. 5, about 23% calculated by peak areas) in the precipitation of asphaltene. As a result of coprecipitation, asphaltene content is generally low in high waxy crude oils. According to the Gel Permeation Chromatography (GPC) analysis, molecular weights of asphaltene in crude oils in Ordovician and Carboniferous reservoirs in well LN14 are 1149.4, 1085.1, 1072.0, 732.9 Dalton, respectively. These values decreased sequentially in vertical view and are evidently smaller than that (1305.4 Dalton) of crude oils in Triassic reservoir in the same well, which is another evidence of asphaltene precipitation occurred in reservoirs.

(2) Loss of light hydrocarbons. As shown in Fig. 5, the logarithm values of mole fraction for C_8—C_{15} *n*-alkanes decreased sharply not as a mild curve described in literatures[15—18], which reflects an obvious loss of light hydrocarbons occurred in reservoirs. According to calculation by peak area, about 6% light *n*-alkanes were lost. Though loss of light hydrocarbons can be occurred in the transportation, preservation and experiment process, little loss of light *n*-alkanes were indeed existed in crude oils in the Triassic reservoir in the same well, indicating that the loss of light hydrocarbons in the transportation, preservation and experiment process can be neglected in some extent.

Because light hydrocarbons and natural gases cannot perforate the unconformity surface between Paleozoic and Mesozoic systems, coprecipitation of asphaltene together with loss of light hydrocarbons did not occurred in Triassic reservoirs. As a result, high asphaltene content and abundant pyrrolic nitrogen compounds were presented in crude oils in Triassic reservoirs. Thus, the semi-logarithm values of mole fraction for C_8—C_{30} *n*-alkanes in crude oil in Triassic reservoir in well LN14 decreased with increased carbon number, and a single bias line in the distribution curve is evidently different from that of crude oils in Paleozoic reservoirs in well LN14.

Theoretically, light oils and gas washing both can produce the waxy oils in the above mixed filling model. However, the wax deposition and asphaltene precipitation observed in well LN14 possibly are not the result of gas washing. After systematic experiment for asphaltene precipitation under the condition of 60℃ and 2000 psi, Rassamdana et al. (1999) found that no asphaltene precipitation occurred when content of added dry methane is more than 85%, while several drop of asphaltene will be deposited when ethane was added and the content of methane ranged in 75%—85%. This experiment confirmed that the coprecipitation of high molecular weight wax and asphaltene in reservoirs cannot come from the invasion of dry natural gases, more probably come from the invasion of wet gases and light or condensate oils. Especially, the defined light or condensate oils under the surface probably existed in the gas phase under the subsurface reservoir. By the way, this experimental result also explained the phenomenon observed by Thompson (1987)[20,21] in the evaporation fractionation experiment, which only observed the shift of max peak carbon number from C_{13} to C_{17} and no asphaltene precipitation occurred.

(iii) Rationality discussion of explanation for diversity in crude oil types. By analysis of asphaltene content, semi-logarithm distribution curve of *n*-alkanes, biomarker parameters and stable carbon isotopic compositions, we have found that differentiation in eastern and western part, two oil families and discontinuous distribution were presented in the crude oils in Lunnan Oilfield. On the basis of this, we think that two oil families were existed in the Lunnan Oilfield, the heavy oil reservoirs in western part belong to the lower matured oil family, while the light or condensate oils reservoirs belong to relatively higher matured oil family. The waxy oils in the middle part were the result of mixing by the heavy oils. This kind of mixed filling model may help to explain the characteristics of hydrocarbon reservoir distribution and almost all geochemical data. The followings were included:

(1) Distribution of physical property of crude oils. Two oil families with variations in maturity index may help to explain the distribution pattern of light oils in eastern part and heavy oils in western part. The mixed filling model may help to explain the occurrence of marine waxy crude oils in the middle part.

(2) Distribution of pyrrolic nitrogen compounds. Pyrrolic nitrogen compounds were mainly presented in the

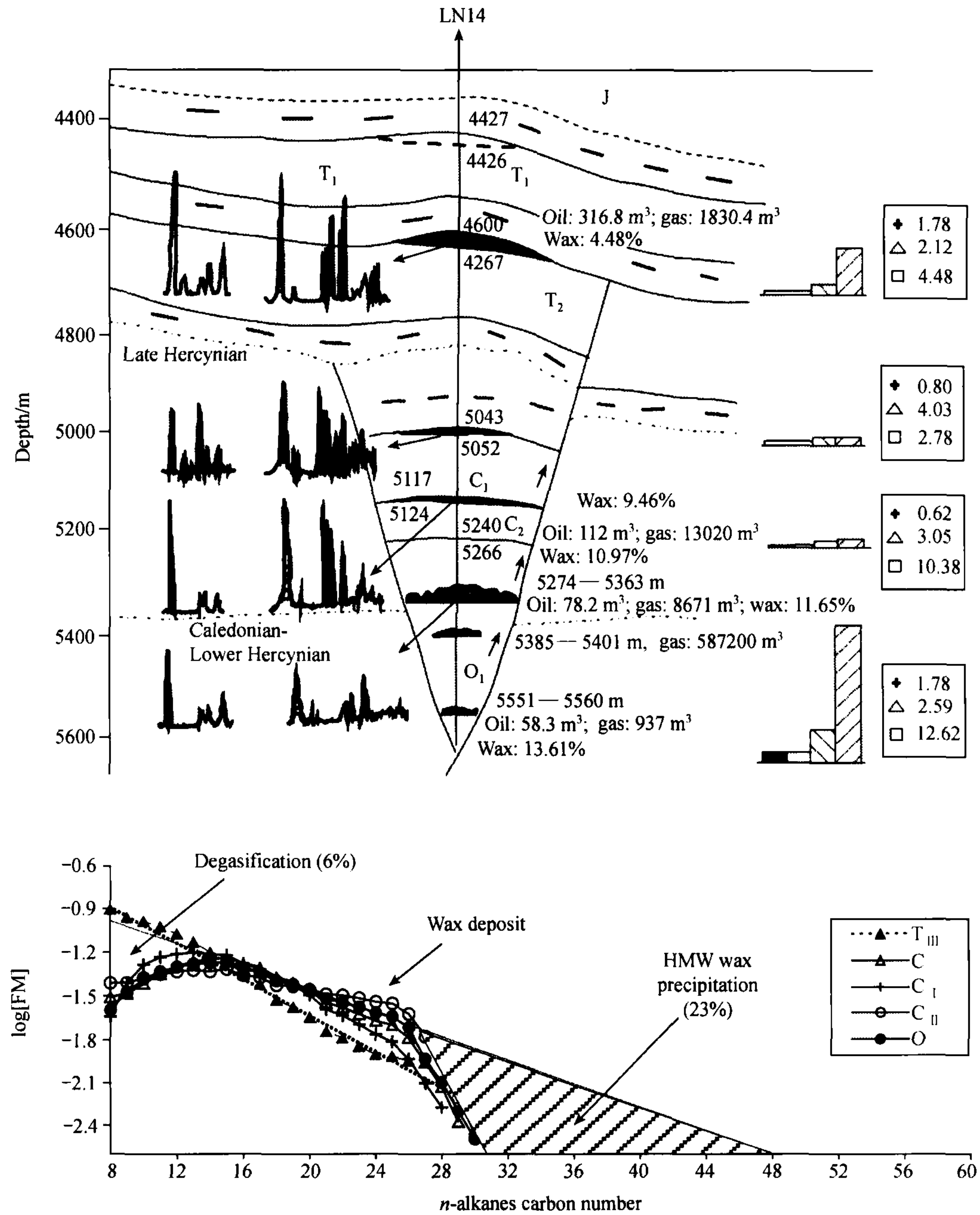

Fig. 5. Reservoir section and the semi-logarithm distribution of mole fraction for *n*-alkanes of crude oils in well LN14 (revised according to Liang et al., 1998). The chromatograms in the left were distribution of carbazole compound, the bar chart in the right were distribution of ratios. +, benzocarbazole [a]/benzocarbazole [c]; □, 1,8-dimethyl carbazole/1,7-dimethyl carbazole; Δ, 1,8-diemthyl carbazole/2,7-dimethyl carbazole.

resin and asphaltene fraction of crude oils and few in light or condensate oils. In the western part, concentration of pyrrolic nitrogen compounds in normal oils and heavy oils must be higher than that in light oils and condensate oils in eastern part. Thus two oil families may help to explain the distribution pattern of pyrrolic nitrogen compounds[22].

(3) Distribution of stable carbon isotopic compositions. Stable carbon isotopic compositions of hydrocarbons should be enriched with increase in maturity. This can help to theoretically explain the fact of enrichment about 1‰ in stable carbon isotopic compositions of condensate oils and high waxy oils in Lunnan Oilfield[8]. Evaporation fractionation in vertical view in some local regions cannot be neglected in Lunnan Oilfield. However, the stable carbon isotopic compositions of hydrocarbons resulted from evaporation fractionation should be depleted,

because evaporation fractionation belonging to physical fractionation just produced large quantitative of light hydrocarbon in condensate oils and high waxy crude oils.

4 Conclusions

(i) By analysis of asphaltene content, semi-logarithm distribution of mole fraction of *n*-alkanes, biomarkers parameters etc., hydrocarbons in eastern part were evidently different from that of hydrocarbons in western part. Two oil families and discontinuous distribution were obviously presented in hydrocarbons in Lunnan Oilfield. As a whole, the maturity in hydrocarbons in eastern part is higher than that of hydrocarbons in western part, which is consistent with the distribution pattern of hydrocarbon reservoirs in Lunnan oilfield.

(ii) Various geochemical analyses indicate that the heavy oils in western part have a relatively low maturity. The light oils or condensate oils in eastern part have a relatively high maturity. They belong to different oil families. The mixed filling of hydrocarbons in the middle part in Lunnan Oilfield may lead to coprecipitation of high molecular weight waxes and asphaltene, as a result, waxy oils were produced in the middle part.

(iii) The mixed filling model may help to explain the distribution pattern of hydrocarbon reservoirs in Lunnan Oilfield, especially can lead to a relatively consistent explanation for all kinds of geochemical data, such as physical properties, pyrrolic nitrogen compounds, biomarker parameters and stable carbon isotopic compositions.

Acknowledgements Thanks are given to Dr. Liu Jinzhong, Dr. Li Chao and Xiang Tongshou for their technological helps in the analysis of all samples. This work was supported jointly by the Knowledge Innovation Project of the Chinese Academy of Sciences (Grant No. KZCX1-SW-18) and the State 973 Project (Grant No. G1999043308).

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(Received August 5, 2004; accepted October 25, 2004)