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ORIGINAL ARTICLE

# Carbon isotopic composition and genetic types of natural gas in the Sichuan Basin, China

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**Abstract** The origin and genetic types of natural gas in the Sichuan Basin are still disputed. To classify the origin and genetic types in different areas, the paper analyzes the carbon isotopic composition of gases and geologic features in the Sichuan Basin. The results showed that the gas sourced from terrestrial layers is typically characterized by terrestrial origin and was mainly accumulated nearby to form reservoir. The carbon isotopic composition of gas showed a normal combination sequence distribution, suggesting that natural gas in continental strata is not affected by secondary alteration or that this deformation is very weak. The gas source is singular, and only gas from the southern and northern Sichuan Basin shows the characteristic of mixed sources. However, marine gas presents the characteristics of an oil-formed gas. The carbon isotopic composition of natural gas in the western and central part of the basin mostly distributes in a normal combination sequence, and few of them showed an inversion, indicating that the gas perhaps had not experienced secondary alteration. The carbon isotopic composition of marine-origin gas in the southern, northern and eastern Sichuan Basin displays a completely different distribution pattern, which is probably caused by different mixing ratio of gas with multi-source and multi-period.

**Keywords** The Sichuan Basin · Alkane · Carbon isotope · Genetic type

## 1 Introduction

The Sichuan Basin is one of major basins in China where natural gas is produced (Ma et al. 2010; Dai et al. 2001; Zhu et al. 2006), where many gas fields are distributed among the structural units (Fig. 1). Amounts of gases economically have been produced in multiple strata, including Late Sinian ($Z_2$), Cambrian ($\in$), Silurian (S), Middle Carboniferous ($C_2$), Permian (P), Early and Middle Triassic ($T_{1-2}$), Early and Middle Jurassic ($J_1$–$J_2$). The main producing horizons of marine natural gas are $C_2$, P and $T_{1-2}$, while continental gas has been formed in the Xujiahe Formation of Late Triassic ($T_3x$) and Penglaizhen Formation of Late Jurassic ($J_3p$) (Yao et al. 2003; Ma et al. 2005a, b; Ma and Cai 2006; Jiang et al. 2006; Hu et al. 2005; Xu et al. 2004; Yang et al. 2004; Xu 1999) (Fig. 1). Previous researchers have discussed the origin and genetic types of natural gas in the Sichuan Basin (Liu et al. 2002; Xie et al. 2007; Wang et al. 2006; Li et al. 2006, 2007; Dai et al. 2009; Xiao et al. 2008a, b; Fan et al. 2005; Wang 2001, 2002; Huang et al. 1991; Li et al. 2005; Dai et al. 1993). This paper, based on 89 gas samples and 145 test data collected from the predecessors, combines geological structure conditions and geochemical data, the origin and genetic types of natural gas in marine and continental strata of Sichuan Basin and discusses these comprehensively, thus providing some guidance and reference for future exploration in the study area.

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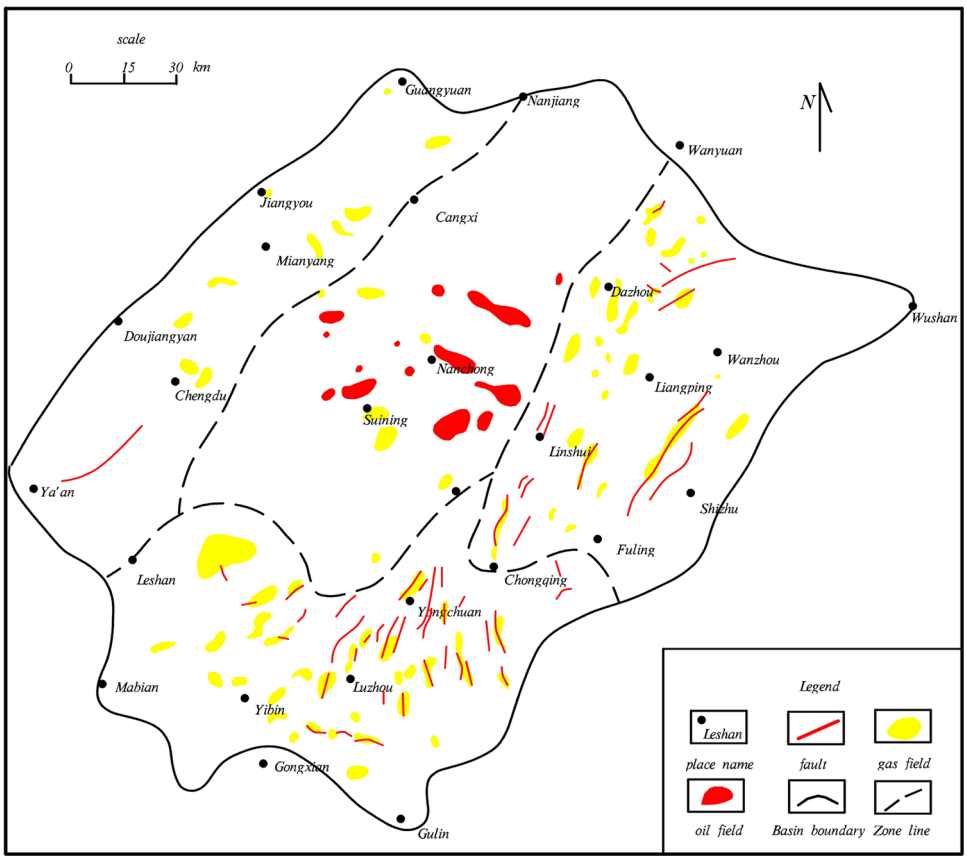


**Fig. 1** The distribution of oil and gas fields in the Sichuan Basin

## 2 Gas compositions

The dry coefficient of terrestrial-origin gases in the Sichuan Basin varies greatly, ranging from 0.8627 to 0.9882, while the dry coefficient of marine gas is generally higher than 0.98 (Fig. 2). Compared with marine methane gas, the relative abundance of heavy hydrocarbon in continental methane gas is high, and the value of log $C_2$/log $C_3$ is nearly equal to 0 (Fig. 3). Non-hydrocarbon contents of continental natural gas are presented by low contents of nitrogen ($N_2$) and carbon dioxide ($CO_2$) and super-low hydrogen sulphide ($H_2S$). By contrast, the contents of non-hydrocarbon (such as $N_2$, $CO_2$ and $H_2S$) of marine natural gas vary greatly. The contents of $CO_2$ and $H_2S$ show a positive correlation, especially the gas from eastern Sichuan Basin (Fig. 4).

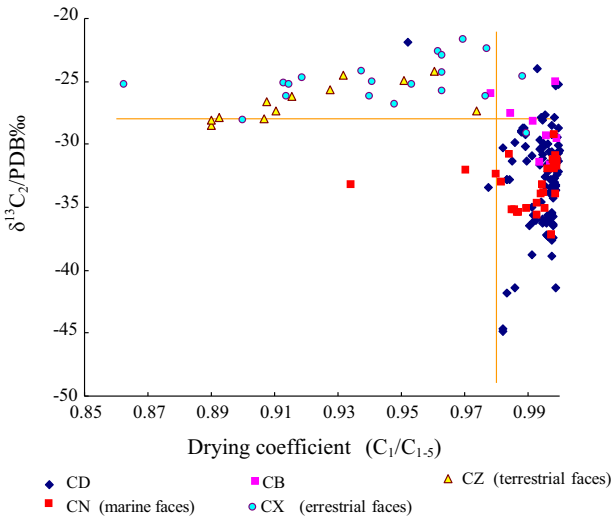


**Fig. 2** Correlation diagram between dry coefficient and ethane carbon isotope of natural gas in Sichuan Basin. *Note*: "CD" represents eastern Sichuan Basin, "CB" represent northern Sichuan Basin, "CN" represent southern Sichuan Basin, "CX" represent western Sichuan Basin, and "CZ" represent middle of Sichuan Basin

## 3 The features of carbon isotopic compositions of alkanes

### 3.1 Gas area in the West of Sichuan Basin

The carbon isotopic composition of methane gas in the west of Sichuan Basin ranges from −38.9‰ to −30.8‰, with an average of −34.9‰. Carbon isotopic composition of ethane is relative heavier with a range of −29.1‰ to −21.5‰. Most of them higher than −28.0‰ and the mean

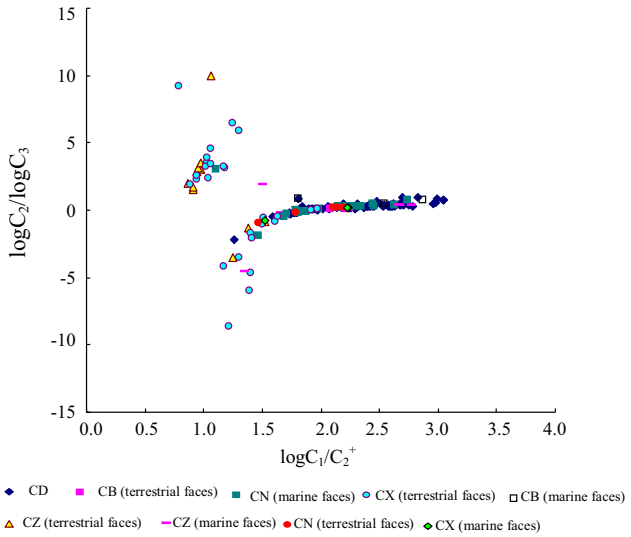


**Fig. 3** Correlation diagram between log $C_1/C_2^+$ and log $C_2/C_3$ of the natural gas in Sichuan Basin. *Note*: "CD" represents eastern Sichuan Basin, "CB" represent northern Sichuan Basin, "CN" represent southern Sichuan Basin, "CX" represent western Sichuan Basin, and "CZ" represent middle of Sichuan Basin

is −24.0‰, showing a typical characteristics of terrestrial sourced gases (Figs. 5, 6). The type of organic matter is dominated by III and $II_2$, which is corresponding to the coal-related and lacustrine source-rocks from the Xujiahe Formation. The terrestrial gas show typical features of humic kerogen (type III) in Qiongxi, Pingluoba, Baimasonghua, Daxingchang, Tuobachang, and Wenxingchang formations. However, most of natural gases from Zhongba, Xinchang and Luodai are located in the high-over mature oil type gas and coal-related gas mixed source areas. It demonstrates that the lacustrine mudstone and shale is the main source supply for natural gas, and it may be related to the geological conditions and facies of the foreland basin in later Triassic. The main facies of the former is shore shallow lacustrine–swamp, while the latter is dominated by shore shallow and semi-deep lake sediments, which reveal that the source rocks were mainly accumulated nearby to form reservoir.

The carbon isotope series for methane gas in the western Sichuan Basin mostly appeared to be a normal combination sequence ($\delta^{13}C_1 < \delta^{13}C_2 < \delta^{13}C_3 < \delta^{13}C_4$), while partial "reversal" between ethane and propane, and propane and

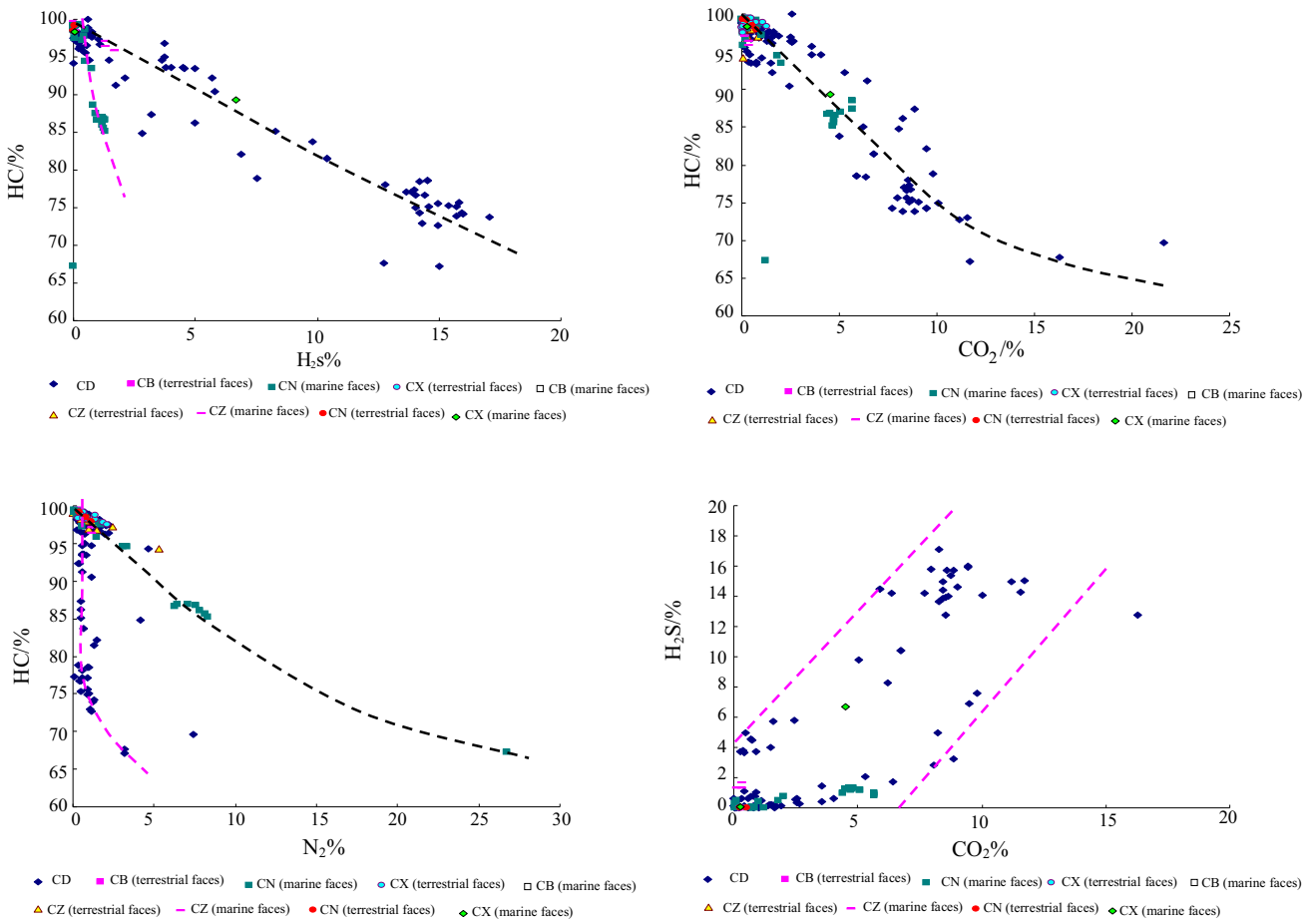


**Fig. 4** Correlation diagram between non-hydrocarbon and alkane contents of the natural gas in Sichuan Basin

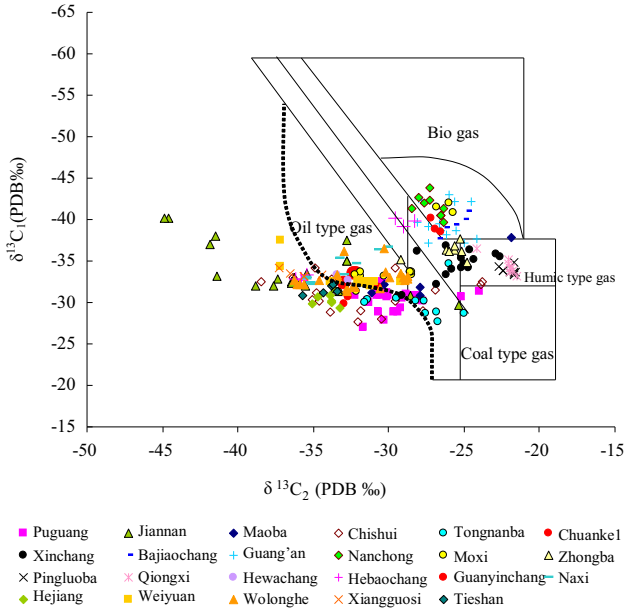


**Fig. 5** The genetic type of natural gas in Sichuan Basin

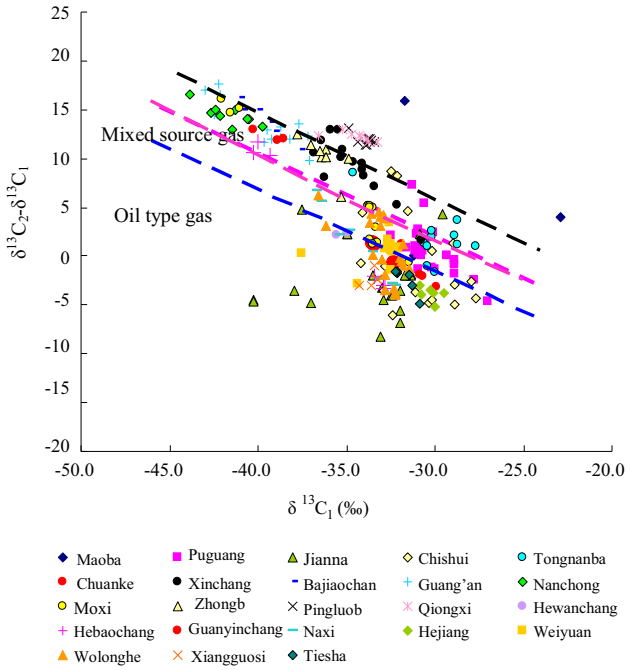


**Fig. 6** Correlation diagram between $\delta^{13}C_1$ and $\delta^{13}C_2$–$\delta^{13}C_1$ of the natural gas in Sichuan Basin

butane, occurred in a few samples, and the magnitude was generally less than 1‰ (Fig. 7). It showed that less or no secondary alteration function occurred for natural gas in the western Sichuan Basin, and the gas source was single.

The carbon isotopic compositions of methane for marine gas in the west Sichuan Basin (including the Ma'antang Formation of Chuanke1 well) are between −35.5‰ and

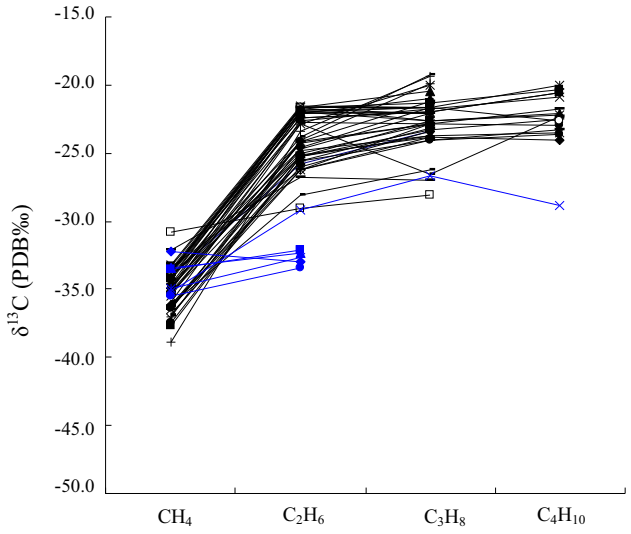


**Fig. 7** Alkane carbon isotope series of natural gas in western Sichuan Basin

−32.3‰ with an average of −34.9‰. Compared with the continental gas, these values are relatively heavier, signifying that they have a slightly higher degree of thermal evolution. The carbon isotopic compositions of ethane in marine gas range from −33.4‰ to −29.2‰ with a mean of −32.1‰. These values are lighter than the terrestrial gas by 7.2‰, and they belong to oil type gas. The normal distribution carbon isotope composition of alkane no longer hold, but partial "reversal" had occurred within few samples.

### 3.2 Gas area in the central part of the Sichuan Basin

In the middle of the Sichuan Basin, the carbon isotopic compositions of methane for terrestrial gas range from −43.8‰ to −35.8‰, and the average is −40.2‰. It is obviously lighter than that of the western part and shows a low value of thermal evolution degree. The values of ethane are between −28.5‰ and −24.2‰. Most of them are generally heavier than −28.0‰, with an average of −26.3‰. It presents a characteristic of continental gas. The dominant samples exist among source-mixed gas area of low and mature oil type and coal related gases (Figs. 5, 6), revealing that it may be accumulated nearby. Moreover, the degree of thermal evolution for hydrocarbon source rocks in the Xujiahe Formation is clearly lower than of west Sichuan Basin.

Carbon isotope compositions of natural gas mostly display a normal combination sequence, while partial "reversal" occurs between carbon isotopes for propane and butane in few samples, perhaps showing that non-secondary reconstruction had been happened basically (Fig. 8).

The methane carbon isotopic ratio of the gas derived from marine layer (Leikoupo Formation in Moxi gas field)

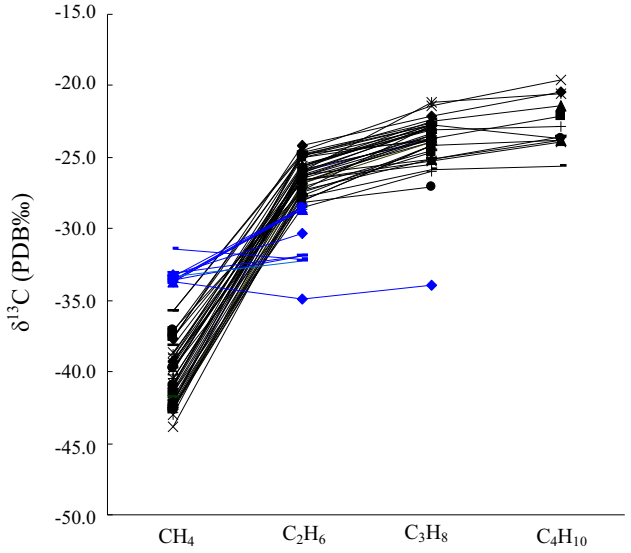


**Fig. 8** Alkane carbon isotope series of natural gas in middle of Sichuan

is mainly distributed from −33.7‰ to −31.4‰, with an average of −33.3‰. These values are heavier than that of terrestrial sourced gases by 6.9‰, reflecting a relatively higher degree of thermal evolution. The range of ethane Carbon isotopic composition is larger −32.2‰ to −28.5‰) and the average is −30.1‰, which is a typical characteristic of oil-type gas. Compared with terrestrial-origin gas, its dry coefficient is higher in marine gas, and the contents of heavy hydrocarbon are lower. Propane carbon isotope can be only detected in individual samples. Methane and ethane carbon isotope composition series generally distribute in appositive sequence, and slight reversal is found in a few samples.

### 3.3 Gas area in the southern Sichuan Basin

Carbon isotope composition of natural gas from the Southern Sichuan Basin is very complex. Among the 22 samples in the continental production layer, 6 samples have ethane carbon isotope as more than −28.0‰ (the average of −26.5‰), accounting for 27.3%, which is similar to the composition of methane and ethane carbon isotope from terrestrial sourced gases in the middle and west of Sichuan Basin, presenting continental origin gas (Figs. 5, 6). The samples with ethane carbon isotope (<−28‰) are divided into two groups. One methane carbon isotope is between −43.1‰ and −34.1‰, with an average of −38.4‰; ethane carbon isotope ranges between −30.7‰ and −28.3‰, with an average of −29.5‰. Compared with terrestrial gas, methane carbon isotope is similar, and the values of ethane carbon isotope are less than 3.0‰, indicating that the source rocks were derived from lacustrine organic matter of Xujiahe Formation, where the type of organic matter is II. The values of methane carbon isotope from another type are between −34.2‰ and −30.0‰, significantly higher than the first type, with ethane carbon isotope ranging between −36.4‰ and −32.8‰ with the average of −34.3‰. It is obviously different from the terrestrial natural gas and similar to the methane and ethane carbon isotope composition in the Permain production layer from the south of Sichuan Basin, thus belonging to an oil type gas. All these data showed that the source rocks were not derived from continental strata, but from an underlying layer. Additionally, hydrocarbon preservation system had once opened in the marine strata from south of Sichuan Basin, with the natural gas shifting upward to the continental strata and accumulating there.

The terrestrial-origin gas can be divided into three groups according to their carbon isotopic composition of hydrocarbon gases: (a) $\delta^{13}C_1 > \delta^{13}C_2 < \delta^{13}C_3$ (seen in the Chishui and Hejiang gas fields), (b) $\delta^{13}C_1 < \delta^{13}C_2 < \delta^{13}C_3$ (seen in the Danfengchang, Guanyinchang, Weidong and Naxi gas fields) and (c) $\delta^{13}C_1 < \delta^{13}C_2 > \delta^{13}C_3$ (only seen in the Guan9 well and the Chishui and Shaximiao gas reservoirs) (Fig. 9). The first type of gas is similar to the main part of the Permian production layer from the Southern Sichuan, showing that they have the same gas sources. In gas fields from the south, the marine gas-producing layer is mainly comprised of the Dengying Formation, Maokou Formation, Changxing Formation, Jialingjiang Formation, etc. The range of distribution for methane and ethane carbon isotope is limited, from the Dengying Formation to lower Cambrian production in the Weiyuan gas field with methane carbon isotope ranging between −32.7‰ and −32.0‰ (an average of −32.4‰),

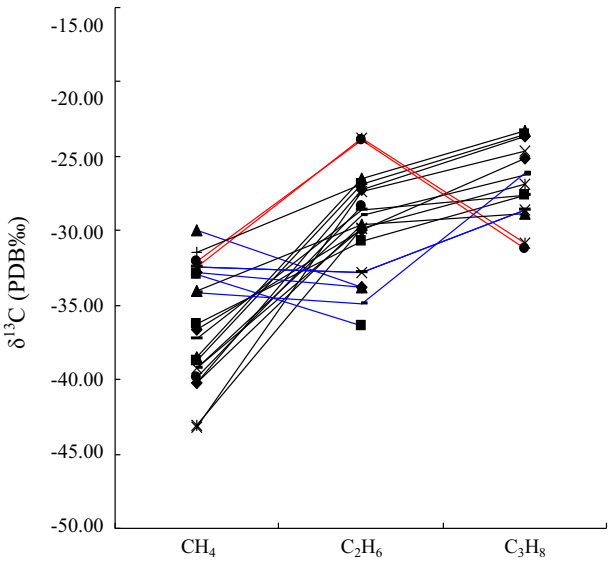


**Fig. 9** Alkane carbon isotopic composition of natural gas derived-from the continental production layer in Southern Sichuan

and the value of ethane falling between −33.9‰ and −29.2‰ (an average of −31.5‰). It shows the oil-prone gas is singular and has a similar evolution degree. Carbon isotope composition series have mostly normal distribution, and only a few individual samples demonstrated a slight reversal (1.5‰, Wei 39 well), which showed no secondary alteration or permeation and also revealed that superimposition by multisource had happened (Fig. 10). Methane carbon isotope of Permian production is between −37.5‰ and −29.8‰ (with an average of −32.4‰), ethane carbon isotope distributes within −37.4‰ to −30.7 ‰ (with an average of −35.1‰), and carbon isotope with "∨"type is presented by $\delta^{13}C_1 > \delta^{13}C_2 < \delta^{13}C_3$, indicating that maybe they did not experience secondary modification and superimposition by multiple source. The methane carbon isotope of the middle-lower Triassic production presents between −35.3‰ and −27.7‰ (with an average of −31.5‰), and ethane carbon isotope is distributed within −35.1‰ to 27.7 ‰ (with an average of −31.1‰). The carbon isotope can be divided into two groups ($\delta^{13}C_1 < \delta^{13}C_2 < \delta^{13}C_3$ and $\delta^{13}C_1 > \delta^{13}C_2 < \delta^{13}C_3$), showing that they may be regarded as two kinds of gases, followed as single source gas and mixed gases, which were formed as a result of penetration and were superimposed by multiple stage tectonics.

### 3.4 Gas area in the northern Sichuan Basin

The methane Carbon isotopic composition of natural gas from the Fen 1 well ($T_3x$, 2533-2553m) at Yuanba and Tongnanba areas is −34.6‰ and the ethane carbon isotope is −26.0‰, which is similar to that of terrestrial-origin gases from the Xinchang, Luodai, and Zhongba areas. Most of the samples were collected from the mixed areas with high-over-matured oil-formed gas and coal-related gas, which showed that the source rocks were probably derived from lacustrine mud shale of the foreland basin of Late Triassic to Jurassic (Fig. 6).

The natural gas from $T_3x$ and $J_1z$ of Chuanfu 82 well and the Chuanfu 190 well is distributed in the mixed areas of oil-formed gas and coal-related gas, which is similar to methane and ethane carbon isotope composition in the Changxing Formation of Puguang2 well and the Feisan Formation of the Jian51well. However, the natural gas in $T_3x$ of the Ma1 well is typical oil-formed gas with Permian source rocks. The data indicated that the preservation conditions in this area were similar to the southern Sichuan Basin. Hydrocarbon preservation system once had been opened during marine periods, and the natural gas shifted up to the continental strata and accumulated there.

The carbon isotope of marine natural gas varies greatly. For example, methane and ethane carbon isotope range from −30.5‰ to −27.7‰ (the average is −29.3 ‰) and −29.6‰ to −25.0‰ respectively (Fig. 11). The genetic type of natural gas also varies. Carbon isotope composition of natural gas of the Yuanba1 well from the Feisan Formation is similar to that of the Puguang2 well and the Qingxi1 well from the Changxing Formation. They all belong to coal-related gas. Natural gas in $T_1j$ of the Chuanfu 82 well and in $T_1f_{1-2}$ of the Yuanba1 well and the Ce1 well is oil-type gas, while natural gas in $T_1j_2$ of the Heba 1 well and the Changxing Formation of the Yuanba1 well belongs to high-over mature oil-type and coal-related mixed gas.

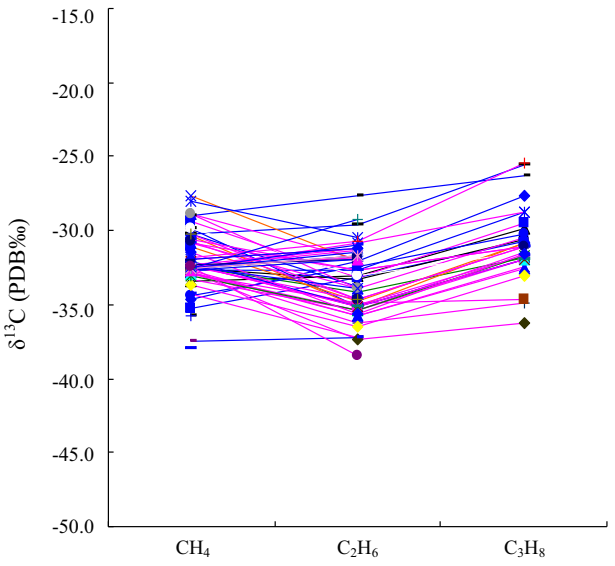


**Fig. 10** Alkane carbon isotopic composition of natural gas derived-from marine production layer in Southern Sichuan

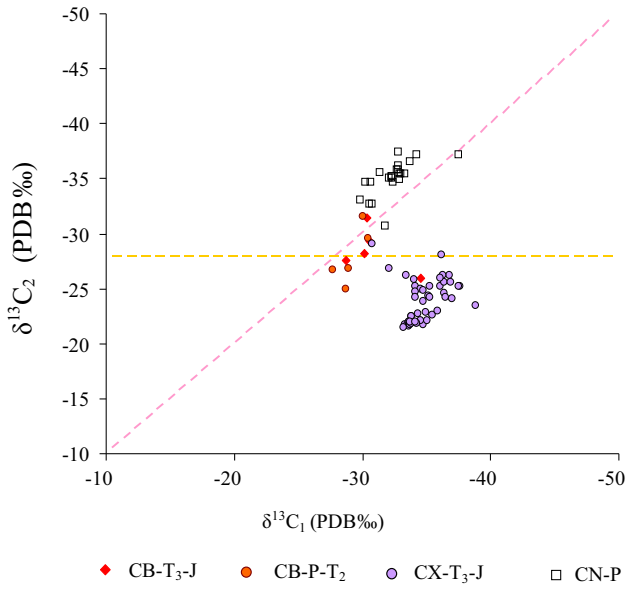


**Fig. 11** Correlation diagram of $\delta^{13}C_1$–$\delta^{13}C_2$ of natural gas between Yuanba and Tongnanba areas

As shown in Fig. 12, carbon isotope composition series display a positive distribution sequence ($T_3x$ of the Ma1 well, $T_1f_{1-2}$ of the Yuanba1 well and the Ce1 well appeared as slight "reversal"). However, carbon isotope "reversal" between ethane and propane is obvious. Compared to the series of marine natural gas in southern Sichuan, the distribution pattern of carbon isotope is generally type "∧", showing that there are some differences in gas sources.

### 3.5 Gas area in the eastern Sichuan Basin

Natural gas in the eastern Sichuan Basin mainly sources from the Huanglong Formation, Changxing Formation, Feixianguan Formation and the Jialingjiang Formation etc. A few gas fields are characterized by gas reservoir of $T_3x$ and $J_1z$ (Chuanyue83well, Chuanyue84 well, Maoba7 well, Woqian1 well, etc.). Apart from continental natural gas of the Maoba7 well, which belongs to humic-type natural gas, all gases from the Chuanyue 83 well, the Chuanyue 84 well and the Woqian1 well are oil-type gas. It shows that marine natural gas upwardly migrated to the continental strata and accumulated nearby (Fig. 5). Carbon isotope compositions of the Woqian1 well and the Maoba7 well display a positive sequence, while "upside down" occurred in gas from the Chuanyue83 well and the Chuanyue84 well. Natural gas in the Shaximiao Formation from the Wubaochang structural belt in NE Sichuan is mainly derived from the Xujiahe Formation and lower Jurassic, and gas of the Zhenzhuchong area from the Dukouhe structural belt is mainly from the Xujiahe Formation (Xiao and Ma 2007; Ma et al. 2005b). It indicates that the marine hydrocarbon preservation conditions are generally good in NE Sichuan, only some regions may had been transformed by fault activities which had occurred for gas migration to terrestrial horizons upwardly and accumulated there.

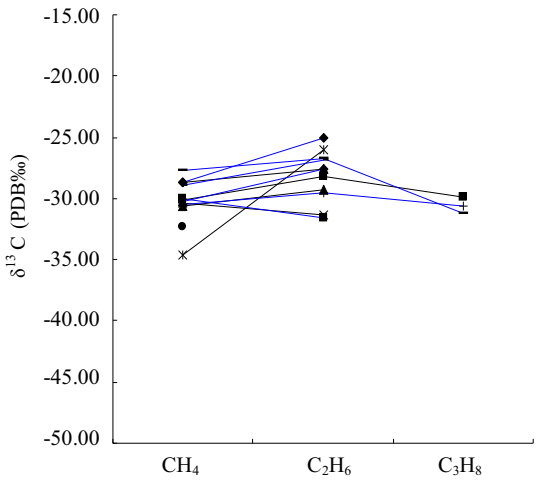


**Fig. 12** Alkane Carbon isotopic composition of natural gas in northern Sichuan

The natural gas in the marine production layer is mostly singular in origin, belonging to oil-prone gas, but there are also exceptions: the gas from the Puguang2 well ($P_3ch$), the Dawan1 well ($P_3ch$), and the Jian61 well ($T_1f_3$) belongs to coal type gas. Moreover, the gas from the Maoba1 well ($P_3ch$), the Maoba6 well ($T_1f_{1-2}$), the Puguang9 well ($T_1f_3$, $P_3ch$) and the Puguang2 well ($T_1f$) belong to source-mixed and coal-bearing, which were produced at the high-and over-mature stage. Among 100 samples, 37 samples have such characteristics that the carbon isotope of methane and ethane shows a positive sequence distribution, and the remaining samples have the reverse result (Figs. 13, 14). Also among the 100 samples, the carbon isotope of propane was detected from 18 samples. Judged from the carbon isotopic composition of methane, ethane and propane, out of 18 samples, 3 samples are normal sequence distribution, 2 samples show a normal sequence distribution between methane and ethane, but the reversal had been happened between ethane and propane, looking like a "∧" tendency, which is similar to the character of alkane carbon isotopic composition from the Ma1 well($T_3x$), the Yuanba1 and well Ce1 ($T_1f_{1-2}$) in northern Sichuan. 13 out of 18 samples turned out to be a reverse sequence distribution between methane and ethane whereas the distribution between ethane and propane was normal, with the tendency shape appearing like "∨", which is similar to the main distribution character of marine gas-production layer in the southern Sichuan. The polyline of alkane carbon isotope was not consistent, which may have resulted from multi-source origin, multi-stages gas mixture or mixed varies with mixture proportion of different gas origin.

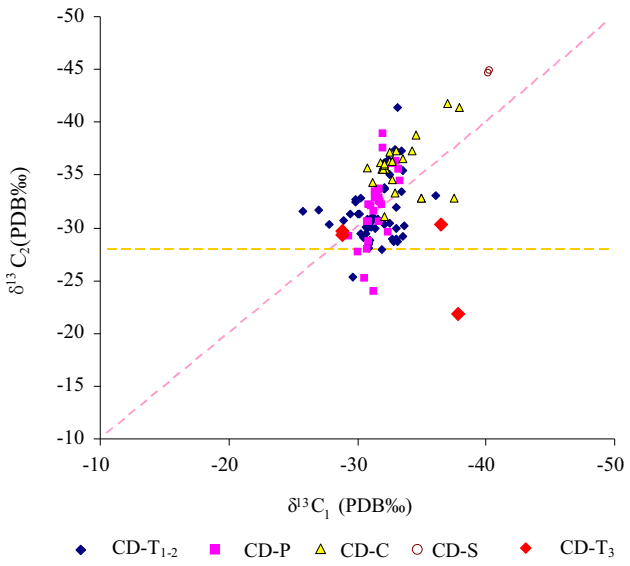


**Fig. 13** Correlogram of $\delta^{13}C_1$–$\delta^{13}C_2$ of natural gas in eastern Sichuan

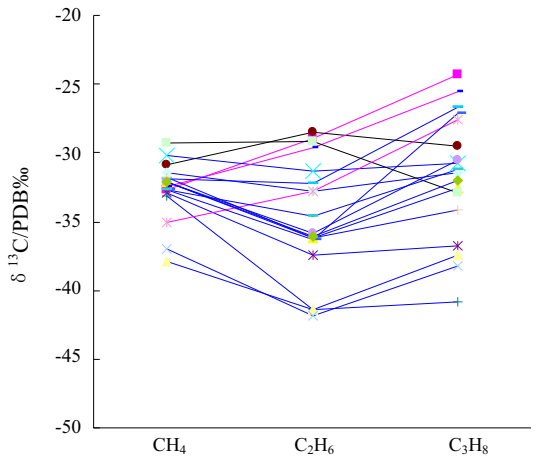


**Fig. 14** Alkane Carbon isotopic composition of natural gas in marine production layer in eastern Sichuan

## 4 The correlation between carbon isotopic compositions of ethane and propane

Although the dry coefficient of natural gas in the Sichuan Basin is high, carbon isotope of propane has also been detected in some natural gas. The cross-plot shown in Fig. 15 indicates that: ethane and propane are linearly related with a high correlation coefficient; the carbon isotope of ethane and propane changes simultaneously, revealing there was not obvious gas-mixture effect or secondary alteration; and carbon isotopes data of ethane and propane form continental production layer is mostly heavy, plotting on the upper-right corner of the diagram. However some data plotting on the marine zone with the carbon isotopes is lighter, such as the gas produced in the Woqian1 well, the Naqian1 well, the Na14 well, the Yin10 well, and the Yin17 well from the Xujiahe Formation, which is determined as marine origin, with the result being consistent with the result of carbon isotope of methane and ethane. The range of distributions for carbon isotope of ethane and propane is wide (ethane: −27.7‰ to −37.4‰, propane: −23.5‰ to −36.8‰) which was produced by marine production layer (C, P, $T_{1-2}$). Of all the samples, gas from the Carboniferous layer (the Jian28 well and the Jian32-1 well) gas form the 3rd member of the Feixianguan Formation (Jian61 well), gas from the Silurian layer (Jianshen1 well) is abnormally light in carbon isotope of ethane and propane, plotting on the bottom-left corner of the diagram, which is perhaps caused by the mixture of different gas genesis.

## 5 Conclusions

(1) The carbon isotopic composition for marine-genetic gas and continental gas in the Sichuan Basin are quite different according to their composition characters. The former presents a typical character of terrestrial source, which reveals that gas would have been reservoired near the source rocks. The carbon isotopic series presents a normal sequence for methane gas. Although a few exceptions exist where the phenomenon "reversal" occurred among ethane and propane and propane and butane, all of these indicate that terrestrial-origin gas did not experience secondary alteration or less changed, and they also

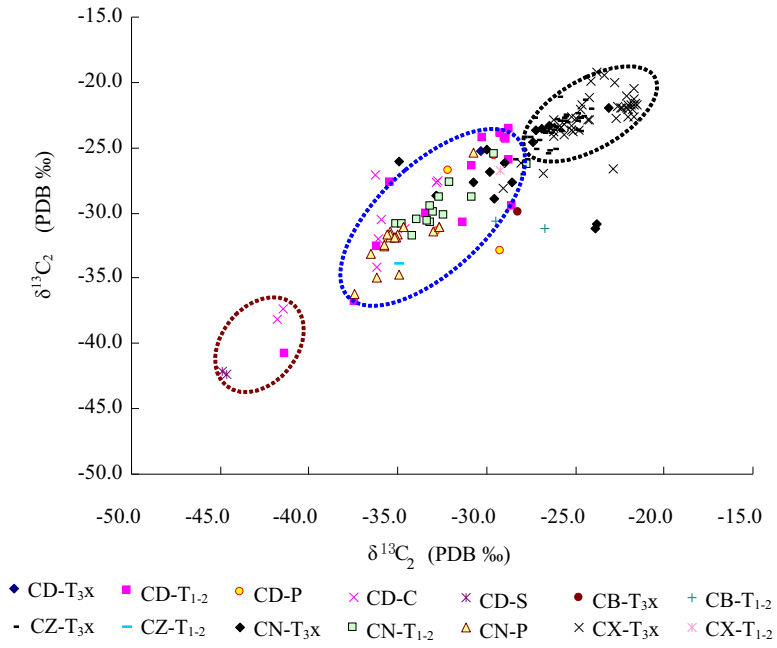


**Fig. 15** Correlation diagram between ethane and propane carbon isotopes of natural gas in Sichuan Basin

demonstrated that they have single source. However, only the southern and northern parts have mixed source gases; marine origin gas has the character of oil type gas, and in the western and middle Sichuan, alkane carbon isotopic composition series mostly show a normal sequence with only a few samples turning to reverse sequence, demonstrating that the natural gas nearly has not been altered. In the southern, northern and eastern parts, the alkane carbon isotopic composition of marine origin gas displays a quite different distribution, which is perhaps caused by the gas origin or the inconsistency in source-mixture, hydrocarbon generation stage and the proportion of mixture.

(2) There is an obvious difference between marine-origin gas and terrestrial-origin gas in the Sichuan Basin. The terrestrial producing layer is typically characterized by terrestrial genesis gas, which was formed at near horizon, and has a single source. However, marine-origin gas presents oil-type gas, for which there are differences for source property in various areas.

(3) Gas composition also may reflect good or bad conditions for gas preservation in marine layers. On the whole, marine origin gas was mainly accumulated in the layers below the source rocks of the Xujiahe Formation, for which only a portion of gas had locally migrated to the Xujiahe Formation of Jurassic upward and formed reservoirs but had limited accumulation. As a result, we can conclude that the preservation condition for marine-origin gas is generally good in the Sichuan Basin, but only in local areas that were disturbed by certain geological events, which resulted in small-scale accumulation of marine origin gas in continental strata.

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