

# Genetic Type and Source Analysis of Natural Gas in the Leikoupo Formation of the Sichuan Basin in China

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The Middle Triassic Leikoupo Formation is largely extended through the Sichuan Basin, SW China. In this formation, several commercially exploited gas reservoirs have been discovered in western and central parts of the basin. Due to the complicated geochemical signatures of the natural gases in these reservoirs, there are contrasted interpretations about their sources, which hamper the evaluation and exploration for new gas resources in the area. To obtain complete understanding of the natural gas sources, the Leikoupo Formation gas reservoirs discovered so far in the Zhongba, Yuanba, Longgang, and Moxi gas fields were selected as the research object of this study. The genetic types and sources of the natural gases in the Leikoupo Formation are discussed based on gas geochemistry combined with their geological background. The natural gas in the top members of the Leikoupo Formation  $(T_2|^4 \text{ or } T_2|^3)$  is partially originated from a humic kerogen contained in the source rocks from the overlying Upper Triassic Xujiahe Formation and from sapropel kerogen from the source rocks of the Leikoupo Formation itself. The natural gas of  $T_2 l^1$  member in the lower part of the Leikoupo Formation is mainly sapropeltype probably from the source rock of the Permian Wujiaping Formation, where the Permian Longtan Formation undergoes a phase change into the Wujiaping Formation The reversed  $\delta^{13}C_1$  and  $\delta^{13}C_2$  trend in the Leikoupo Formation of the Yuanba gas field is due to the more sapropelic source rocks and higher degree of maturity.

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# **1 INTRODUCTION**

The Middle Triassic Leikoupo Formation is the latest set of marine strata developed in the Sichuan Basin, China, which is not the main strata for oil and gas exploration, and the exploration and research conducted for the Leikoupo Formation is relatively low. Back in the 1970s, small-scale gas pools were discovered in this set of strata, for example, the third member  $(T_2l^3)$  of the Leikoupo Formation in the Zhongba gas field in NW Sichuan (Qin et al., 2007), and the first member  $(T_2l^1)$  of the Leikoupo Formation in the Moxi gas field in central Sichuan. With the increasing natural gas exploration, commercial gas reservoirs have been found in the fourth section  $(T_2l^4)$  of the Leikoupo Formation in the Longgang and Yuanba gas fields successively in this century, showing that the Leikoupo Formation has good potential for gas exploration. Since the Leikoupo Formation is

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dominated by evaporative platform facies as a whole, and evaporites mainly composed of gypsum and dolomite are deposited, it is doubtful whether the Leikoupo Formation formed in such an environment can develop effective source rocks. In addition, the geochemical characteristics of natural gas in the Leikoupo Formation are complex, and the types of natural gas origin are diverse. It is difficult to identify the gas sources, and there are different opinions on the source of natural gas. For example, in the Leikoupo Formation in the Zhongba gas field in western Sichuan, it was believed that all natural gas came from the underlying Permian hydrocarbon source rocks (Dai, 1980). It was also believed that the gas came from biogenic limestone and mudstone of the Leikoupo Formation (Li, 1993; Qin et al., 2007). Some studies suggested that the gas came from Permian sapropel source rock, mixed with a small amount of gas from coal measures (Liao et al., 2013), and some even thought that the gas mainly came from overlying coal measures (Wang, et al., 1989; Zheng, et al., 1990). In the central Sichuan Basin, recoverable gas reservoirs have been found in both the T<sub>2</sub>l<sup>4</sup> and T<sub>2</sub>l<sup>1</sup> members of the Leikoupo Formation, but the gas geochemical characteristics differ significantly between the reservoirs, and the gas source is highly controversial (Huang, 2014; Liu, et al., 2014; Zhou, et al., 2015; Liu, et al., 2019). Even within the  $T_2l^4$  member, the gas' geochemical signatures are highly variable; the gas indicated to be not only humic-type gas but also sapropel-type, and it is difficult to explain the source of gas.

In this study, the gas reservoirs of the Leikoupo Formation in the Zhongba, Moxi, Longgang, and Yuanba gas fields that have been discovered so far are systematically analyzed using natural gas geochemical methods. The source of natural gas has been elucidated based on the comparative study of Triassic and Permian natural gas types and their source rocks, which in turn provides a basis for natural gas exploration in the Leikoupo Formation.

#### 2 GEOLOGICAL BACKGROUND

The Indo-Chinese early episodic movement at the end of the Middle Triassic made the central Sichuan area rise to land, the seawater withdrew, and the large inland lake basin began to appear, which was an important turning period from marine sedimentation to lacustrine sedimentation in the Sichuan Basin as it ended the deposition of the carbonate platform in Sichuan. The uplift of the Sichuan Basin suffered from denudation, forming the erosion surface at the top of the Middle Triassic Leikoupo Formation. Since the Late Triassic, it has received continental deposits and developed multiple sets of coal-measure source rocks and multiple sets of interbedded sedimentary assemblages of sandstones.

#### 2.1 Strata

There are many oil- and gas-bearing layers in the Sichuan Basin. This study focuses on the gas reservoir in the Leikoupo Formation. The related layers span the Triassic and the upper Permian. The strata from top to bottom are the Upper Triassic Xujiahe Formation  $(T_3x)$ , Middle Triassic Leikoupo Formation  $(T_21)$ , Lower Triassic Jialingjiang Formation  $(T_1J)$  and Feixianguan Formation  $(T_1f)$ , and Upper Permian Changxing Formation  $(P_3ch)$  and Longtan Formation  $(P_31)$  (**Figure 1**).

The Xujiahe Formation is a fluvial–swamp–lacustrine deposit developed in a humid environment (Luo, 1983, 2011; Li, 2011). Several sets of coal-measure source rocks and sandstone reservoirs are developed. The coal-measure source rocks and sandstones were stacked on top of each other. The Xujiahe Formation is subdivided into  $T_3x^1$  to  $T_3x^6$  members from bottom to top, in which  $T_3x^1$ ,  $T_3x^3$ , and  $T_3x^5$  are mainly coal-measure sources, and the organic matter is mainly kerogen-III type, which is a gas dominated source rock. The  $T_3x^2$ ,  $T_3x^4$ , and  $T_3x^6$  members are dominated by sandstone (Wang, et al., 1997; Yang, et al., 2005; Li et al., 2010) and are good reservoirs for gas storage.

The top of the Leikoupo Formation  $(T_2l)$  suffered from dissolution and contact with the overlying Xujiahe Formation in parallel unconformity. The  $T_2l$  is mainly composed of grayish-white medium-thick microcrystalline dolomite, argillaceous dolomite, and gray dolomite, with light gray gypsum and thin gray-black shale, which is a high-quality caprock in the whole area, in which the gray-black shale can be used as a hydrocarbon source rock. It is subdivided into  $T_2l^1$  to  $T_2l^4$  members from bottom to top, among which the natural gas reservoirs have been discovered in  $T_2l^1$ ,  $T_2l^3$ , and  $T_2l^4$ members.

The Jialingjiang Formation  $(T_1j)$  is composed of gray micrite limestone and dolomite interbedded with a gypsum layer, dolomitic gypsum, and argillaceous dolomite, which is not only a high-quality caprock in the whole area but also a gas reservoir in the Jialingjiang Formation. The top of the Feixianguan Formation ( $T_1f$ ) consists of mudstone, dolomitic mudstone, gypsum, dolomicrite, micrite, and marlstone. The middle and lower part of the Feixianguan Formation is karst oolitic dolomite and limestone, which has good porosity and permeability and a regional high-quality reservoir rock (Wen, et al., 2012; Zhang, et al., 2013; Zhu, et al., 2013).

The Upper Permian ( $P_3$ ) includes the Changxing Formation ( $P_3$ ch) and the Longtan Formation ( $P_3$ 1) from top to bottom. The Changxing Formation is mainly composed of bioclastic micrite limestone, reef limestone, and dolomite, which is also an important reservoir in central Sichuan (Peng, et al., 2011). The Longtan Formation is mainly composed of marine–continental transitional coal measures and marine biological limestone, which is an important regional hydrocarbon source rock series and the main source rock of Changxing and Feixianguan gas reservoirs such as Longgang and Yuanba gas fields. It should be pointed out that the coal-measure source rocks of the Longtan Formation have undergone phase transformation in some areas and become marine source rocks of the Wujiaping Formation ( $P_3$ w), and the hydrocarbon-generating material changed from humic to sapropel-type.

#### 2.2 Gas Reservoir Types

The Leikoupo Formation gas reservoirs include  $T_2l^1,\,T_2l^3,$  and  $T_2l^4$  members. A weathering crust and a karst reservoir are



developed at the top of the Leikoupo Formation, and their controlled distribution are by lithology and paleogeomorphology of exposed strata at the top (Bian, et al., 2019). The  $T_2l^3$  gas reservoir of the Leikoupo Formation in the Zhongba gas field is distributed in the dolomite layer, which is an anticline reservoir type. The top is in an unconformable contact with the overlying Xujiahe Formation, missing the  $T_2l^4$  member (Zeng, et al., 2007). The top surface of the Leikoupo Formation in the Yuanba gas field was affected by denudation and karstification, and a dolomite karst reservoir was formed in the  $T_2l^4$ member. The reservoir is controlled bv paleogeomorphology and has strong lateral heterogeneity (Fan, 2014), there is no unified gas-water interface, and natural gas tends to be distributed in structural highs (Liu, et al., 2019); therefore, the reservoir is a karst-controlled structural and lithologic gas reservoir. The reservoir of the T<sub>2</sub>l<sup>4</sup> in the Longgang gas field is related to Indosinian denudation and karstification (Yang, et al., 2014), and the natural gas between different wells in the lateral direction is quite different, with obvious heterogeneity, which is a structural and lithologic gas reservoir. The T<sub>2</sub>l<sup>1</sup> gas reservoir is currently mainly discovered in the Moxi gas field. This reservoir has a large trap area and is an anticline porous carbonate gas reservoir with a high degree of fullness. It has a unified gas-water interface and is a monolithic gas reservoir.

# 2.3 Possible Source Rocks

The reason why it is difficult to judge the source of natural gas in the Leikoupo Formation is that there are several sets of different types of source rocks developed in the Leikoupo Formation and it's overly and underlying strata, which may provide a gas source for the Leikoupo gas reservoirs under appropriate geological conditions. This increases the difficulty for identification of Leikoupo gas sources.

The natural gas generated by coal-measure source rocks of the Xujiahe Formation has formed many large- and mediumsized gas fields in the Sichuan Basin. The source rocks directly cover the unconformities of the Leikoupo Formation and might partly provide gas for the karst reservoirs of the Leikoupo Formation.

The Jialingjiang, Feixianguan, and Changxing formations under the Leikoupo Formation did not develop source rocks. Under the Changxing Formation, the marine-terrestrial transitional coal-measure source rocks developed in the Formation had acted as a source rock for many large gas reservoirs in Changxing and Feixianguan formations of Longgang, Yuanba, and Puguang gas fields (Hao, et al., 2008; Wu et al., 2015; Qin, et al., 2016a; Deng, et al., 2018). If conditions permitted, it might also be possible to indirectly supply gas sources to the Leikoupo gas reservoir through the Changxing, Feixianguan, and Jialingjiang formations.

The Leikoupo Formation was formed in a salinized evaporation environment. Many researchers questioned that such an environment could form high-quality source rocks. Theoretically, such a saline environment has weak hydrodynamics, limited seawater circulation, high salinity, and alternating deposition of gypsum salt and carbonate rocks, which is conducive to the preservation of organic matter, and some of the strata can develop high-quality source rocks. Early studies believed that the Leikoupo Formation in the western Sichuan Basin was an algae-rich carbonate rock deposited in deep-water lagoon facies on a confined-evaporative platform and had favorable conditions for forming the main source rock of large- and medium-sized gas fields (Xu et al., 2013). The gypsum-bearing carbonate samples of the Leikoupo Formation in the western Sichuan Basin have relatively high TOC, and the gypsum-dolomite flat and gypsum-bearing lagoon facies with more evaporative platform facies are deposited, which are favorable sedimentary facies belts and lithologic assemblages for the development of high-quality source rocks (Yang, 2016; Wang, et al., 2018b). Other studies have concluded that the TOC of organic-rich shale in the Leikoupo Formation in the Sichuan Basin is 0.49%-1.08%, with an average of 0.77%, and the Ro values are 2.36%-2.40% (Sun, et al., 2021). According to the evaluation standard of a highly mature source rock (Dai, et al., 2008), it is evaluated as a high-quality source rock. The source rock was formed in a dry heat, salt water, and anoxic environment. Drilling revealed that the actual drilling thickness of the Leikoupo Formation in the central and northern Sichuan area is 960 m, of which the  $T_2l^3$  member is the most likely the source rock with a thickness of 530 m, and the lithology is argillaceous limestone, micrite limestone, organicrich shale rock, salt rock, and gypsum. The  $T_2l^1$ ,  $T_2l^2$ , and T<sub>2</sub>l<sup>4</sup> members are dominated by dolomite and gypsum rock, and no source rocks have been found (Sun, et al., 2021).

In fact, it is not uncommon to form source rocks in evaporative environments. In both the Bohai Bay Basin and the tertiary of the Qaidam Basin, the symbiotic phenomenon of evaporite and the source rock had been found (Jin, et al., 2006). In the Junggar Basin (Yu, et al., 2018), Polish Basin (Krzywiec, et al., 2017), Mexico Basin (Xie, et al., 2019), and West Texas-New Mexico (Hussain, et al., 1991), source rocks had been found in an evaporative environment.

# **3 SAMPLING AND ANALYTICAL METHODS**

#### 3.1 Sample Collection

Natural gas samples were taken from the Xujiahe, Leikoupo, Jialingjiang, and Changxing formations in the central Sichuan Basin. To eliminate the interferences of external factors and ensure representativeness of the natural gas in these reservoirs, all samples were collected from wells with long-term normal production without application of de-foaming or any other chemical agents recently.

Gas samples from the reservoirs were taken at the wellheads by using double valve steel cylinders. To take such samples, the pressure gauge was dismantled before connecting the steel cylinder with the sampling tubing. Prior to taking samples, wellhead natural gas was used to flush the steel cylinder thoroughly for about 3 min. The sampling steel cylinder was then filled with natural gas equilibrated to the wellhead pressure.

# **3.2 Analytical Methods**

Natural gas compositions were determined using an Agilent 6890N gas chromatograph (GC) with He and N<sub>2</sub> as the carrier gases. Double thermal conductivity detectors (TCD) and a 30 m  $\times$  0.25 mm  $\times$  0.25 µm quartz capillary column were used. The inlet temperature was 150 °C, and the TCD temperature was 200 °C. The initial oven temperature was maintained at 40 °C for 7.5 min isothermally, then rose from 40 °C to 90 °C at 15 °C/min, and finally rose from 90 °C to 180 °C at 6 °C/min.

An on-line analysis was conducted for the measurement of carbon isotopic compositions with a MAT 253 gas isotopic mass spectrometer. Natural gas samples were separated to methane, ethane, propane, butane, and CO<sub>2</sub> using the chromatography column of an SRI 8610C gas chromatograph, which were then transferred into a combustion furnace by the carrier gas (He) and oxidized into CO<sub>2</sub> by CuO at 850°C. All of the converted species were transferred by the carrier gas (He) into MS to measure the isotopic compositions. A dual inlet analysis was performed with the international measurement standard of NBS-19 CO<sub>2</sub>  $(\delta^{13}\text{CVPDB}=1.95 \pm 0.04\%)$ , International Atomic Energy Agency, 1995), and the stable carbon isotopic values were reported in the  $\delta$  notation in per mil (%) relative to the Peedee belemnite standard (VPDB). Reproducibility and accuracy were estimated to be  $\pm$  0.2‰ with respect to the VPDB standard.

# **4 ANALYTICAL RESULTS**

# 4.1 Natural Gas Components

The hydrocarbon gas contents of the 38 samples in this study range from 60.19% to 99.71%, with an average of 92.77%. Samples also contain a small amount of  $N_2$ ,  $CO_2$ , and  $H_2S$  (Table 1). A small number of samples have N2 content exceeding 10%; other samples have  $N_2$  content of 0–3.8%, with an average of 1.01%. Most of the samples have low CO<sub>2</sub> content, less than 5%. A few samples have higher CO<sub>2</sub> content. The marine carbonate rocks in the reservoirs are accompanied by gypsum salt. This results in the gas reservoirs generally containing H<sub>2</sub>S. Most of the natural gas containing H<sub>2</sub>S in the world is distributed in such type of strata, and it is considered to be formed by TSR action (Krouse, et al., 1988; Worden, et al., 1995; Machel, 2001; Cross et al., 2004). The content of heavy hydrocarbon gas such as ethane in the hydrocarbon gas is very low, and the dry coefficient  $(C_1/C_{1+})$ of natural gas is very high, ranging from 0.940 to 0.998, with an average of 0.983. Taking more than 0.95 as the criterion for dry gas, all the natural gas in the Leikoupo Formation is dry gas. Among them, the  $T_2 l^1$  gas reservoir in the Moxi gas field has the highest dry coefficient, with an average of 0.998, and the  $T_2l^3$  gas reservoir in the Zhongba gas field is the lowest, with an average of 0.97. The  $T_2 l^4$  gas reservoirs in the Longgang and Yuanba gas

ABLE 1   Composition and isotopic data of Permian and Triassic natural gas in the central Sichuan Basin.

Gas Field	Well	/ell Strata	Main Molecular Composition (%)									δ <sup>13</sup> C (VPDB)‰			References
			N <sub>2</sub>	CO <sub>2</sub>	H₂S	CH₄	$C_2H_6$	C <sub>3</sub> H <sub>8</sub>	iC <sub>4</sub> H <sub>10</sub>	$nC_4H_{10}$	$\delta^{13}C_1$	$\delta^{13}C_2$	$\delta^{13}C_3$	$\delta^{13}C_4$	
Zhongba	Zhong 29	T <sub>3</sub> x <sup>2</sup>	0.39	0.28	0.00	87.86	6.53	2.10	0.60	0.83	-36.7	-25.5	-23.3	-23.5	Qin et al. (2007)
	Zhong 34	$T_3 x^2$	0.70	0.44	0.00	90.71	5.53	1.65	0.31	0.36	-36.1	-25.6	-23.2		Qin et al. (2007)
	Zhong 31	T <sub>3</sub> x <sup>2</sup>	0.22	0.47	0.00	90.49	6.00	1.62	0.32	0.35	-37.8	-23.0	-29.4	-22.5	Qin et al. (2007)
	Zhong 39	$T_3 x^2$	0.03	0.32	0.00	87.82	6.36	2.70	0.93	1.38	-35.8	-26.0	-23.4	-23.7	Qin et al. (2007)
	Zhong 37	$T_3x^2$	0.21	0.48	0.00	90.44	5.83	1.62	0.33	0.37	-38.0	-24.4	-25.9	-22.3	Qin et al. (2007)
	Zhong 9	Τ <sub>3</sub> x <sup>2</sup> τ ι <sup>3</sup>	0.20	0.44	0.00	90.65	5.91	1.61	0.31	0.35	-38.0	-23.9	-25.8	-22.4	Qin et al. (2007)
	Zhong 18	T <sub>2</sub> I <sup>3</sup> T <sub>2</sub> I <sup>3</sup>	1.69	4.86	3.30 1.78	86.88	1.66	0.53 0.54	0.39	0.00	-36.9	-27.7	-22.1 -30.3	-29.6 -29.8	Qin et al. (2007)
	Zhong 21 Zhong 24	$T_2 I^3$	1.78 0.22	3.65 4.69	4.11	87.92 87.78	1.82 1.88	0.54 0.56	0.39 0.40	0.00 0.00	-35.4 -35.7	-31.1 -30.3	-30.3 -27.9	-29.8	Qin et al. (2007) Qin et al. (2007)
Longgang	LG 3	$T_{3}x^{6}$	1.18	0.39	0	92.62	4.42	0.90	0.40	0.14	-37.1	-25.4	-23.8	-22.1	This study
201.990.19	LG 12	T <sub>3</sub> x <sup>6</sup>	1.03	0.88	0	95.54	2.07	0.13	0.01	0.01	-37.8	-23.4	-22.2		This study
	LG 17	T <sub>3</sub> x <sup>6</sup>	0.45	0.43	0	92.16	5.38	0.84	0.19	0.14	-38.7	-25.1	-23.6	-21	This study
	LG 18	$T_3 x^6$	0.5	0.38	0	93.83	4.4	0.55	0.09	0.06	-38.1	-23.6	-21.7	-23.6	This study
	LG 20	T <sub>3</sub> x <sup>6</sup>	0.87	0.39	0	87.65	7.42	2.15	0.49	0.38	-42.2	-25.6	-22	-21.8	This study
	LG 29	T₃x <sup>6</sup>	0.23	0.08	0	97.24	2.13	0.17	0.03	0.02	-35.4	-20.8	-21.6	-18.3	This study
	LG172	T <sub>3</sub> x <sup>6</sup>	6.99	0.38	0	81.04	6.87	2.32	0.64	0.41	-38.6	-24.9	-23.3	-20.9	This study
	LG176	T <sub>3</sub> x <sup>6</sup>	0.29	0.54	0	91.32	5.88	1.19	0.21	0.18	-39.9	-24.7	-22.7	-21.5	This study
	LG 177	T <sub>3</sub> x <sup>6</sup>	0.34	0.54	0	92.64	4.99	0.9	0.17	0.14	-38.9	-25	-23.5	-22.5	This study
	LG 171	T <sub>3</sub> x <sup>4</sup> T <sub>3</sub> x <sup>2</sup>	0.19	0.43	0	91.07	6.1	1.37	0.28	0.23	-38.9	-24.2	-21.8	-20.3	This study
	LG 30 LG176	$T_3 x$ $T_3 x^2$	0.26 0.85	0.55 0.27	0 0	95.28 90.69	3.07 5.99	0.43 1.32	0.09 0.23	0.07 0.22	-34.6 -37.9	-23.5	-22.7 -22.2	-20.6 -21.7	This study
	LG170 LG160	$T_3 x^2$	12.2	0.27	0	90.09 77.09	6.81	1.78	0.23	0.22	-37.9 -38.8	-24.3 -24.2	-22.2 -20.9	-21.7	This study This study
	LG100	$T_{2}I^{4}$	1.39	0.19	Nd*	92.81	4.28	0.83	0.00	0.30	-36.3	-24.2 -25.1	-20.9 -23.8	-21.3	This study
	LG7	$T_2 I^4$	0.47	21.65	Nd	76.59	0.9	0.1	0.04	0.03	-37.2	-32.2	-24.3	-22.0	This study
	LG12	$T_2 l^4$	0.31	8.65	Nd	88.1	2.2	0.37	0.06	0.04	-35.5	-26.2	-23.8	-21.7	This study
	LG17	$T_2 I^4$	0.51	35.61	Nd	63.06	0.51	0.02	0	0.01	-35.8	-35.3			This study
	LG18	$T_2l^4$	0.12	4.59	0.01	94.34	0.79	0.07	0.01	0.01	-36.5	-35.5	-30.5	-27.1	This study
	LG20	$T_2 l^4$	0.01	13.36	0.01	83.46	2.23	0.38	0.09	0.09	-38.4	-29.0	-25.5	-22.8	This study
	LG22	$T_2l^4$	0.39	1.76	Nd	95.78	1.72	0.19	0.04	0.03	-37.7	-30.8	-27.2		This study
	LG160	$T_2l^4$	1.27	1.84	Nd	94.18	2.14	0.32	0.06	0.06	-35.3	-26.6	-24.3	-23.8	This study
	LG172	$T_2l^4$	0.8	1.37	0.01	94.92	2.31	0.35	0.06	0.05	-36.3	-25.3	-24.4	-20.4	This study
	LG176	T <sub>2</sub>   <sup>4</sup>	0.34	2.42	Nd	95.16	1.71	0.23	0.02	0.02	-37.8	-32.5	-30.6		This study
	LG173	T <sub>2</sub>   <sup>4</sup> T 1 <sup>4</sup>	2.87	0.15	Nd	92.01	2.96	0.65	0.14	0.15	-37.9	-28.5	-24.8	01.4	This study
	LG 022-H6 LG 022-H2	T <sub>2</sub> 1 <sup>4</sup> T <sub>2</sub> 1 <sup>4</sup>	0.84	0.71 0.79	Nd	92.25 95.29	4.41	0.83 0.21	0.17 0.04	0.14	-34.9 -37.2	-26.5	-23.5 -27.9	-21.4 -21.1	This study
	LG 022-H2 LG 022-H8	$T_2^{1}$	0.91 1.70	0.79	Nd Nd	95.29 91.50	1.51 4.33	0.21	0.04 0.16	0.04 0.13	-37.2 -38.4	-32.2 -26.7	-27.9 -24.3	-21.1 -21.4	This study This study
	LG 022-H3	$T_2^{14}$	0.46	0.24	Nd	96.55	1.69	0.21	0.04	0.04	-36.7	-31.6	-24.3	-26.7	This study
	LG 1	T <sub>1</sub> f	1.44	2.82	Nd	94.02	0.07	0.00	0.00	0.00	-29.5	-25.0	-20.6	20.1	This study
	LG 001–7	T <sub>1</sub> f	1.1	2.37	Nd	94.49	0.08	0.01	0.00	0.00	-29.4	-25.2	-23.3	-24.1	This study
	LG 2	T₁f	0.2	4.77	3.06	91.90	0.05				-28.5	-24.3			Qin, et al. (2016a
	LG 3	T <sub>1</sub> f	1.76	15.84	0.04	81.96	0.11				-31.0	-22.8			Qin, et al. (2016a
	LG 12	T <sub>1</sub> f	2.84	1.12	Nd	95.70	0.09				-30.5	-27.3			Qin, et al. (2016a
	LG 26	T <sub>1</sub> f	0.59	7.09	2.75	89.48	0.06	0.01			-29.1	-25.8			Qin, et al. (2016a
	LG 1	P <sub>3</sub> ch	0.7	4.41	2.48	92.33	0.07				-29.4	-24.3			Qin, et al. (2016a
	LG 2	P <sub>3</sub> ch	0.31	6.07	4.52	89.03	0.06				-28.5	-21.7			Qin, et al. (2016a
	LG 8	P₃ch	0.25	8.63	7.24	83.8	0.05				-29.0	-22.1			Qin, et al. (2016a
	LG 11 LG 26	P <sub>3</sub> ch	0.17	6.08	9.09	84.56	0.07	0.01			-27.8	-27.0			Qin, et al. (2016a
	LG 28 LG 28	P₃ch P₃ch	0.64 0.58	4.71 2.48	1.67 0.7	92.88 96.15	0.08 0.07				-29.4 -29.3	-23.0 -24.7			Qin, et al. (2016a Qin, et al. (2016a
	LG 29	P <sub>3</sub> ch	1.46	4.98	4.78	88.52	0.07	0.01			-29.3	-24.7			Qin, et al. (2016a
	LG 23 LG 001–2	P <sub>3</sub> ch	0.25	4.36	Nd	92.04	0.07	0.00	0.00	0.00	-29.4	-25.3			This study
	LG 001-23	P <sub>3</sub> ch	1.42	4.79	Nd	90.11	0.37	0.06	0.00	0.00	-28.8	-27.8	-26.6	-26.1	This study
Moxi	M 38-H	$T_2 ^1$	2.28	0.00	Nd	96.90	0.39	0.08	0.02	0.02	-32.8	-29.7	-25.1	-24.1	This study
	Mo 30-24H	$T_2 ^1$	3.80	0.25	Nd	94.70	0.23	0.01	0.00	0.00	-33.2	-33.0	-31.9	-31.0	This study
	M 030-H21	$T_2 ^1$	1.36	0.11	Nd	97.23	0.20	0.01	0.00	0.00	-35.8	-33.8	-29.3	-29.4	This study
	Mo 140	$T_2 l^1$	0.23	0.05	Nd	99.54	0.17				-35.0	-32.4			This study
	M 144	T <sub>2</sub> I <sup>1</sup>	0.75	0.16	Nd	98.90	0.18				-34.9	-32.1			This study
	M 004-H9	$T_2 l^1$	0.59	0.13	Nd	99.12	0.16				-35.0	-32.8			This study
	MS 1	T₁j	2.31	0.46	Nd	96.71	0.28	0.01	0.00	0.00	-33.1	-34.0	-33.9	-28.7	This study
	MS 005-1	T₁j T∵	1.47	0.21	Nd	97.30	0.21	0.01	0.00	0.00	-32.4	-32.9	-32.4	-29.0	This study
	M 150	T₁j T∶	0.21	0.08	Nd	99.50	0.21				-34.7	-33.7			This study
	M 5 M 005 H10	T₁j Ti	0.80	0.18	Nd	98.85	0.17				-34.6	-33.2			This study
	M 005-H10	T₁j	0.44	0.10	Nd	99.24	0.21				-34.6	-34.6	(		This study on following page)

(Continued on following page)

Gas Field	Well	Strata	Main Molecular Composition (%)										C (VPDB)	References	
			N <sub>2</sub>	CO <sub>2</sub>	H₂S	CH₄	$C_2H_6$	C <sub>3</sub> H <sub>8</sub>	$iC_4H_{10}$	$nC_4H_{10}$	$\delta^{13}C_1$	$\delta^{13}C_2$	$\delta^{13}C_3$	$\delta^{13}C_4$	
	M 005-H9	T₁j	0.81	0.18	Nd	99.12	0.16				-34.8	-33.6			This study
Yuanba	YB 222	$T_3 x^4$	0.31	0.47	0	97.43	1.47	0.18	0.03	0.03	-3.98	-25.24			Hu, et al. (2014
	YB 2-CP1	T <sub>3</sub> x <sup>3</sup>	0.80	2.43	0	95.38	1.13	0.03	0	0.01	-30.9	-25.2	-24.4	-20.6	Hu, et al. (2014
	YB 3	$T_3x^4$	0.17	0.58	0	97.80	1.32	0.13			-31.4	-21.5	-23.9		Hu, et al. (2014
	YB2-C1	$T_3x^1$	0.48	0.29	0	98.07	1.01	0.09	0.01	0.01	-31.7	-30.2	-26.5		Hu, et al. (2014
	YL 1	$T_3 x^2$	0.86	0.85	0	96.82	1.23	0.14	0.01	0.01	-32.1	-28.0			Hu, et al. (2014
	YL 3	$T_3 x^4$	0	0.25	0.29	98.39	0.93	0.09	0	0.01	-30.6	-24.8			Hu, et al. (2014
	YL 10	T <sub>3</sub> x <sup>2</sup>	1.40	0.14	0	97.14	1.05	0.09			-31.8	-32.6	-32.7		Liu, et al. (2014
	YB 05	$T_3 x^3$	0	2.43	0	95.38	1.13	0.057	0.004	0.0071	-30.9	-25.2	-24.4		Liu, et al. (2011
	YB 06	$T_3 x^2$	0	0	0	96.6	2.39	0.35	0.04	0.04	-32.0	-27.0	-23.4		Liu, et al. (2011
	YB 1	$T_3 x^2$	0.58	0	0	96.6	2.39	0.35	0.04	0.04	-31.9	-28.5			Wu, et al. (2015
	YL 10	$T_3 x^4$	0.20	0.67	0	98.05	0.93	0.09	0.01	0.01	-32.0	-25.7	-27.3		Wu, et al. (201
	YL 9	$T_3 x^2$	1.29	8.13	0	89.71	0.72	0.06	0.01	0.01	-30	-33	-33.6		Wu, et al. (201
	YB 27	$T_3 x^2$	16.5	1.43	0	80.71	1.11	0.11			-31.8	-30.8			Yin, et al. (2013
	YB 3	$T_3 x^1$	1.12	0.59	0	95.56	2.36	0.28	0.03	0.03	-33.9	-24.4	-23.9		Yin, et al. (2013
	YB 4	$T_3 x^4$	0.68	0.35	0	97.46	1.25	0.14			-31.7	-28	-26.9		Yin, et al. (2013
	YB 4	$T_3 x^2$	0.53	0.53	0	97.86	0.91	0.08			-33.5	-29.7			Yin, et al. (2013
	YB 11	$T_3 x^2$	0.27	0	0	98.35	1.07	0.12	0.01	0.01	-30.3	-25.4			Yin, et al. (2013
	YB 2	$T_3 x^3$	0.8	2.43	0	95.38	1.13	0.06			-30.9	-25.2	-24.4		Yin, et al. (2013
	YB 2	T <sub>3</sub> x <sup>1</sup>	0.48	0.29	0	98.07	1.01	0.09			-31.7	-30.2	-26.5		Yin, et al. (2013
	YB 22	$T_3 x^2$	0.36	0.67	0	98.21	0.67	0.04	0	0	-34.5	-35.4			Yin, et al. (2013
	YL 6	$T_3 x^2$	0.31	0.64	0	97.71	1.16	0.11	0.01	0.01	-31.3	-31.4	-31.7		Yin, et al. (2013
	YB 221	$T_2l^4$	1.14		Nd	97.36	1.00	0.10			-33.2	-28.5	-27.5		Liu, et al. (2014
	YB 223	$T_2l^4$	0.86	2.42	Nd	95.97	0.65	0.05			-35.6	-36.7			Liu, et al. (2014
	YB 07	$T_2l^4$	0	2.07	Nd	96.41	0.63	0.05	0	0.01	-35.3	-36.0			Liu, et al. (2011
	YB 10	$T_2 I^4$	1.52	4.67	Nd	92.50	1.15	0.13	0.03	0.01	-33.6	-29.6	-29.4	-25.9	Qin, et al. (201
	YB 13	$T_2 I^4$	0.33	3.30	Nd	95.24	1.05	0.08	0		-31.7	-27.7	2011	2010	Qin, et al. (201
	YB 17	$T_2 I^4$	1.55	9.30	Nd	88.34	0.72	0.07	0.02		-32.11	-32.8	-33.41		Qin, et al. (2010
	YB 222	$T_2l^4$	0.27	1.04	Nd	97.65	0.93	0.09	0.02		-32.91	-28.6			Qin, et al. (2010
	YB 224	$T_2 I^4$	13.7	26.14	Nd	59.79	0.37	0.03	0		-35.55	-36.23			Qin, et al. (2010
	YB 23	$T_2 I^4$	0.37	0.77	Nd	98.08	0.73	0.05	0		-34.3	-35.1			Qin, et al. (201
	YB 3	$T_2 I^4$	0.74	3.28	Nd	95.38	0.58	0.02	0		-34.2	-36.5			Qin, et al. (201
	YB 5	$T_2 I^4$	0.86	2.42	Nd	96.02	0.65	0.05	0		-35.3	-36.0			Qin, et al. (201
	YB 6	$T_2 I^4$	2.69	5.89	Nd	90.88	0.50	0.04	0		-34.0	-34.5			Qin, et al. (201
	YL 17	$T_2 I^4$	1.07	0.00	Nd	98.02	0.81	0.08	0.02		-35.1	-32.7			Qin, et al. (2010
	YL 2	$T_2 I^4$	11.3	21.77	Nd	66.48	0.42	0.04	0.02		-35.39	-36.61			Qin, et al. (201
	YB 1	P3ch	30.2	3.04	13.33	53.25	0.09	0.09	0.02		-30.2	-27.6			Hu, et al. (2014
	YB 11	P3ch	11.8	0.23	7.37	80.55	0.05	0.03			-27.9	-27.0			Hu, et al. (2014
	YB 27	P3ch	3.12	0.23	5.14	90.71	0.03	0			-28.9	-26.6			Guo, et al. (2012
	YB 221	P <sub>3</sub> ch	15.1	22.09	Nd	61.98	0.04	0			-28.9 -29.2	-20.0 -28.6	-26.9		Wu, et al. (201
	YB 222	P <sub>3</sub> ch	0.28	0.07	Nd	99.15	0.04	0.02			-29.2 -30.9	-28.0 -29.7	-20.9 -29		Wu, et al. (201 Wu, et al. (201
	YB 222 YB 224	P <sub>3</sub> ch	0.20	4.68	6.85	99.15 88.46	0.47	0.02			-30.9 -28.3	-29.7 -25.9	-23		Wu, et al. (201) Wu, et al. (201)
	YB 224 YB 273	-	0.84		0.85										
	10213	P <sub>3</sub> ch	0.04	6.04	0.40	92.57	0.05				-28.6	-25.4			Wu, et al. (201

TABLE 1 | (Continued) Composition and isotopic data of Permian and Triassic natural gas in the central Sichuan Basin.

\*Nd = not determined.

fields are 0.971 and 0.991, respectively. Although both belong to  $T_2l^4$  gas reservoir, the dry coefficient of the Yuanba gas field is slightly larger than that of the Longgang gas field, reflecting the difference in the maturity of the source rocks.

The natural gas in the upper and lower strata of the Leikoupo Formation is mainly hydrocarbon gas, the content of nonhydrocarbon gas is relatively low, and there are obvious differences in natural gas components between different strata.

The most important feature of natural gas in the Xujiahe Formation is that it does not contain  $H_2S$ ; the content of  $N_2$  is 0–16.5%, with an average of 1.36%, and the content of  $CO_2$  is 0–8.13%, with an average of 0.78%. The content of ethane and other heavy hydrocarbon gas in alkane gas varies greatly for different gas reservoirs. The content of heavy hydrocarbon in the

Zhongba gas field and the Longgang gas field is similar. The content in the Zhongba gas field and the Longgang gas field is between 2.22% and 11.37%, respectively, with an average of 7.32%, and the dry coefficient is between 0.89% and 0.98%, with an average of 0.92. There are both dry gas and wet gas in natural gas, indicating that the characteristics of gas source rocks are more complex. The content of heavy hydrocarbon gas in the Yuanba gas field is not only relatively low but also relatively concentrated, 0.71%–2.82%, with an average of 1.40%, and the dry coefficient is 0.97–0.99, with an average of 0.99. This shows that the source rock characteristics of natural gas in the Yuanba gas field are located under and adjacent to the  $T_2l^1$  gas reservoir. The composition characteristics of natural gas are highly consistent with that of the  $T_2l^1$  gas reservoir, and the dry





coefficient of natural gas is close to 1.0%. The average content of  $N_2$  is 1.1% and that of  $CO_2$  is 0.2% (**Table 1**).

The gas reservoirs of Feixuan and Changxing formations under the Jialingjiang Formation have the same source of natural gas (Qin, et al., 2016a), and the geochemical characteristics of natural gas are completely the same. The natural gas dry coefficient is close to 1; contains higher H<sub>2</sub>S, nitrogen, and carbon dioxide content of most samples less than 5%; and also there are some samples have high content (**Table 1**).

#### 4.2 Carbon Isotope of Natural Gas

The carbon isotopes of natural gas in the Leikoupo Formation are generally less negative, and the overall characteristics are that the  $\delta^{13}C_1$  value is relative concentrated, ranging from -38.4% to -31.7%, with an average of -35.4%, but the  $\delta^{13}C_2$  values vary widely, ranging from -36.7% to -25.1%, with an average of -31.4%. As the propane content is very low, only part of the



samples were detected, with the  $\delta^{13}C_3$  values ranging from -33.4‰ to -22.1‰, with an average of -26.9‰.

The carbon isotopes of different gas reservoirs are obviously different. The carbon isotope values of the  $T_2l^1$  gas reservoir in the Moxi gas field are the most concentrated, with  $\delta^{13}C_1$  ranging from -35.8% to -32.8%, with an average of -34.4%, and  $\delta^{13}C_2$  ranging from -33.8% to -29.7%, with an average of -32.3%. Although the  $\delta^{13}C_1$  values are concentrated in the  $T_2l^3$  gas reservoir in the Zhongba gas field and the  $T_2l^4$  gas reservoir in Longgang and Yuanba gas fields, the range of the  $\delta^{13}C_2$  value is widely distributed, ranging from -36.7% to -25.1%. The carbon isotope characteristics of natural gas in the Jialingjiang Formation are consistent with those of the  $T_2l^1$  reservoir, with  $\delta^{13}C_1$  ranging from -34.8% to -32.4%, with an average of -34.1%, and  $\delta^{13}C_2$  ranging from -34.6% to -32.9%, with an average of -33.7%.

The  $\delta^{13}C_1$  of Changxing and Feixianguan formations in Yuanba and Longgang fields are the least negative in this area, and the variation range is relatively narrow, indicating the relatively consistent gas source rocks. The  $\delta^{13}C_1$  ranges from -31%to -27.8%, with an average of -29.2%, and  $\delta^{13}C_2$  ranges from -29.7% to -21.7%, with an average of -25.8% (**Table 1**).

The  $\delta^{13}C_1$  of the Xujiahe Formation gas reservoir ranges from -42.2‰ to -30.0‰, with an average of -34.7‰, and the  $\delta^{13}C_2$  ranges from -35.4‰ to -20.8‰, with an average of -26.3‰. The carbon isotopes of natural gas vary widely, especially the ethane carbon isotopes that reflect the genetic types of natural gas. The natural gas of the Xujiahe Formation mainly comes from its own coal-measure source rocks, and its carbon isotopes vary greatly, which reflects that the source rock type of natural gas may not be single.

In addition, the natural gas in Leikoupo and Xujiahe gas reservoirs in the Yuanba gas field show the inversion of  $\delta^{13}C_1$  and  $\delta^{13}C_2$ , that is,  $\delta^{13}C_1 > \delta^{13}C_2$  (**Table 1**; Figure 2).

#### **5 GENETIC TYPES OF NATURAL GAS**

According to the carbon isotope distribution chart, it can be seen that among the natural gas in the Leikoupo Formation, the



Longgang gas field is the most negative, followed by the Zhongba gas field. Yuanba and Moxi gas fields are relatively less negative (**Figure 2**). The source maturity of natural gas in the Leikoupo Formation of Longgang, Zhongba, Yuanba, and Moxi gas fields increases sequentially. The gas samples of  $T_{1j}$  and  $T_2l^1$  gas reservoirs in the Moxi gas field completely overlap in Whiticar's chart. The natural gas of Changxing and Feixiangguan formations in Yuanba and Longgang gas fields also fall in the same area, and the maturity of their source rocks are much higher than that of the Leikoupo gas reservoir (**Figure 3**). In order to judge the genetic type of natural gas more precisely, this study adopts the identification standard of genetic type of natural gas proposed by Dai, that is, the value of  $\delta^{13}C_2$  more negative than -28.8% indicate the sapropel-type gas (Dai, 1993).

It can be seen from **Table 1** that the  $\delta^{13}C_2$  values of natural gas in the  $T_2l^1$  reservoir of the Moxi gas field are more negative than -28.8‰, the genetic type is relatively simple, and all samples are sapropel gas. Among the three natural gas samples of the  $T_2l^3$ member in the Zhongba gas field, two samples are sapropel gas and one is humic. The genesis of natural gas in the  $T_2l^4$  reservoirs of Longgang and Yuanba gas fields is complicated; some samples are humic gas and some are sapropel-type, and the heterogeneity of the gas reservoir is very obvious. Half of the natural gas samples of the T<sub>2</sub>l<sup>4</sup> gas reservoir in the Longgang gas field are humic-type, while only a few samples of the  $T_2 l^4$  gas reservoir in the Yuanba gas field are humic-type, and the natural gas is mainly sapropeltype (Figure 4). The carbon isotope of alkane gas series can better reflect the overall appearance of natural gas. Half of the samples of the  $T_2l^4$  gas reservoir in the Longgang gas field are humic-type and the other half are sapropel-type, while the gas samples of the  $T_2l^4$  gas reservoir in the Yuanba gas field are mainly sapropeltype, and only a few are humic gas (Figure 5).

# 6 DISCUSSION ON NATURAL GAS SOURCES

### 6.1 The T<sub>2</sub>l<sup>4</sup> Gas Reservoir

The paleokarst on the top of the Leikoupo Formation developed and formed a good karst reservoir, which showed an unconformable contact with the coal-measure source rocks of the overlying Xujiahe Formation (Song, et al., 2012; Wang, et al., 2018a). Some corrosion ditches are formed in the Leikoupo Formation, and the corrosion ditches will be filled by the Xujiahe Formation. Theoretically, the natural gas generated by the source rocks of the Xujiahe Formation has the opportunity to migrate to the top reservoir of the Leikoupo Formation; therefore, the samples showing humic-type in  $T_2$ l<sup>4</sup> reservoir should come from the source rocks of the Xujiahe Formation.

It is researched that there is a close relationship between the  $\delta^{13}C_2$  values and the  $C_2H_6$  content in the  $T_2l^4$  reservoir of Longgang and Yuanba gas fields. The higher the ethane content, the less negative of the carbon isotope, showing the characteristics of humic gas. Taking the Longgang gas field as an example, the samples with ethane content more than 2% in the  $T_2l^4$  reservoir are humic gas, and on the contrary, those with the ethane content less than 2% are sapropel gas. A similar situation has occurred in the Yuanba gas field. Because the maturity of the source rocks is higher than that of the Longgang gas field, the ethane content is generally lower than that of the Longgang, but the  $\delta^{13}C_2$  also tends to become less negative with the increase of the ethane content. The ethane content of several humic-type gas samples in  $T_2l^4$  is more than 1%. The ethane content of sapropel gas samples is less than 1% (**Figure 4**). The  $\delta^{13}C_2$  value can best reflect the type of source rock, and the content of ethane can often reflect the content of heavy hydrocarbon gas, which indirectly reflects the dry coefficient (or humidity coefficient) of natural gas; the higher value of the ethane content, the lower the dry coefficient (high humidity coefficient). The humidity coefficient is related to the maturity of source rocks. For the same type of source rocks, the smaller the humidity coefficient, the higher the maturity of source rocks (Dai, et al., 2016b). Under the same degree of evolution, different types of source rocks have different dry coefficients, the sapropel-type is lower than humictype. The ethane content of sapropel gas is less than that of sapropel gas in  $T_2l^4$ , indicating that the maturity of sapropel gas of hydrocarbon source rocks is higher than that of humic gas.

According to the carbon isotope of the alkane gas series of the  $T_2l^4$ , there are obvious differences in carbon isotopes of heavy

hydrocarbons such as ethane between sapropel and humic gas; however, there is no obvious difference in the  $\delta^{13}C_1$  values (Figure 5). Based on this, it is concluded that the sapropel gas in  $T_2l^4$  can only come from the source rocks of the Leikoupo Formation or other underlying strata, mainly sapropel organic matter. Due to the fact that the  $\delta^{13}C_1$  value generated by sapropelic source rocks is more negative than that generated by humic source rocks under the same maturity, the  $\delta^{13}C_2$  value is highly related to the type of organic matter and does not change obviously with the increase of the maturity of source rocks, and the genetic type of natural gas is therefore often identified according to the ethane carbon isotope.

As the Xujiahe Formation is the first terrestrial strata deposited after the end of marine sedimentation in the Sichuan Basin, in addition to humic source rocks, the T<sub>3</sub>x<sup>1</sup> member may also develop marine sapropel source rocks at some local area. It is unlikely that the sapropel gas in the T<sub>2</sub>l<sup>4</sup> reservoir come from the sapropel source rocks that may undergo phase transformation corresponding to the T<sub>3</sub>x<sup>1</sup> member in contact with it. If the sapropel gas in the  $T_2l^4$  comes from the sapropel source rock in the  $T_3x^1$  member, the maturity of this source rock should be similar to other humic hydrocarbon source rocks in the  $T_3x^1$ . As mentioned before, the  $\delta^{13}C_1$  from rocks with the same maturity should be lighter than those from humic source rocks. In fact, the  $\delta^{13}C_1$  of sapropel and humic gas in  $T_2l^4$  are similar, but the  $\delta^{13}C_2$  is quite different (Figure 5). The ethane content of sapropel gas in the  $T_2l^4$  reservoir is less than that of humic gas (Figure 4). Therefore, it is judged that the sapropel gas in  $T_2l^4$  comes from source rocks with higher maturity than the source rocks in the  $T_3x^1$  member.

It is not possible that the sapropel gas in  $T_2l^4$  came from the source rocks of the Longtan Formation below. This is because the hydrocarbon source rocks of the Longtan Formation and the T<sub>2</sub>l<sup>4</sup> reservoir are separated by multiple sets of gypsum strata from the Jialingjiang Formation and  $T_2l^1$  to  $T_2l^3$  members, and there are many sealing layers, making it difficult for natural gas to migrate to the  $T_2l^4$  reservoir. In addition, the natural gas from the Changxing and Feixianguan formations in Longgang and Yuanba gas fields all come from coal-measure source rock of the Longtan Formation; the carbon isotopes of methane and ethane are much less negative, and their characteristics are very different from those in T<sub>2</sub>l<sup>4</sup> reservoirs, uncorrelated with the gas in  $T_2l^4$  reservoir (Figures 3, 5). Collectively, it is judged that the sapropel gas in  $T_2l^4$  should come from the source rocks developed in the T<sub>2</sub>l<sup>3</sup>, and the humic gas comes from the Xujiahe coal-measure source rock.

**6.2 The T\_2 l^3 Gas Reservoir** The forming condition of  $T_2 l^3$  gas reservoir is similar to that of the  $T_2l^4$ , in which the humic gas comes from the overlying Xujiahe Formation source rocks and the sapropel gas comes from the  $T_2l^3$  source rocks. In this study, the  $T_2l^3$  gas reservoir occurs in the Zhongba gas field, where the  $T_2 l^4$  member is depleted and the T<sub>2</sub>l<sup>3</sup> section is in unconformity contact with the overlying Xujiahe Formation. The T<sub>3</sub>x<sup>2</sup> gas reservoir develops above the  $T_3x^2$  member in the gas field. The  $\delta^{13}C_1$  of the gases in  $T_2l^3$  and  $T_3x^2$  reservoirs are similar, with the former



averaging -37% and the latter -36%, but  $\delta^{13}C_2$  are more different, with the Leikoupo Formation being more negative, averaging -29.7%, and the T<sub>3</sub>x<sup>2</sup> averaging -24.7%, both of which are from different hydrocarbon source rocks. The  $T_2l^3$ itself has source rock, and the sapropel-type gas should come from the  $T_2x^3$  member source rock.

# 6.3 The T<sub>2</sub>I<sup>1</sup> Gas Reservoir

In this study, all gas data of  $T_2 l^1$  come from the Moxi gas field, where the  $T_2l^1$  gas reservoir is adjacent to the underlying Jialingjiang gas reservoir, and its geochemical characteristics are highly consistent with the natural gas in the Jialingjiang Formation based on the analysis of the samples in this study (Figures 2, 3, 6). The gas in both reservoirs should be from the same source, but it is difficult to determine which set of hydrocarbon source rocks the gas comes from. First, it is impossible for the gas to come from the coal-measure source rocks of the Xujiahe Formation because the gas characteristics of the  $T_2l^1$  do not match the humic gas and there are effective interlayers between the Xujiahe Formation and  $T_2l^1$ ,  $T_2l^2$ ,  $T_2l^3$ , and  $T_2l^4$  members, and it is difficult for the gas of the Xujiahe Formation to traverse downward through the multilayered gypsum rocks to reach the  $T_2l^1$  reservoir.

Comparing the isotopic data of typical sapropel gas samples from the  $T_2l^4$  section of the Longgang and Yuanba gas fields with the  $T_2l^1$  gas reservoir, it is found that the  $T_2l^1$  gas is closer to the sapropel gas of the  $T_2l^4$  reservoir in Yuanba. The difference is that most of the sapropel gas samples from the  $T_2l^4$  reservoir in Yuanba show carbon isotope inversion, while the carbon isotopes of gas from the  $T_2l^1$  reservoir in Moxi do not. The  $\delta^{13}C_2$ (average -32.3‰) is significantly less negative than that of sapropel gas (average 35.3‰) in the  $T_2l^4$  gas reservoir in Yuanba (Figure 7), and the source rock may be relatively humic. Although there are some similarities between the carbon isotopes of  $T_2l^1$  gas and  $T_2l^4$ , we cannot suggest that T<sub>2</sub>l<sup>1</sup> gas may also come from T<sub>2</sub>l<sup>3</sup> hydrocarbon source rocks because the direct caprock of the T<sub>2</sub>l<sup>1</sup> gas reservoir is the gypsum layer and gypsum dolomite above the gas reservoir. There are



three layers of anhydrite layers with a total thickness of about 5 m on the gas reservoir, and 20 layers of anhydrite and gypsum dolomite are sandwiched up together with the  $T_2l^1$  member, with a thickness of 160 m, which together constitute a good direct cover for the  $T_2l^1$  gas reservoir (Dai, et al., 1996). Therefore, it is difficult for natural gas in the  $T_2l^3$  member to migrate down to the  $T_2l^1$  reservoir across such a thick high-quality caprock.

Earlier studies suggested that the natural gas in the  $T_2l^1$  gas reservoir of the Moxi gas field was mainly from the coal-measure source rock of the Longtan Formation based on the less negative of  $\delta^{13}C_2$  value in some drilled wells (Wang, et al., 1998). However, the average  $\delta^{13}C_2$  value of  $T_2l^1$  gas is -32.3‰, which is typical of sapropel gas (Figure 6). Its methane and ethane carbon isotopes are very different from the natural gas from the Longtan Formation coal-measure source rocks in the Changxing and Feixianguan formation gas reservoirs of the Longgang and Yuanba gas fields, and the carbon isotopes of the Changxing and Feixianguan natural gas are much less negative (Figure 7). Therefore, the natural gas in T<sub>2</sub>l<sup>1</sup> gas reservoir does not come from the coal-measure rocks of the Longtan Formation. However, the coal-measure hydrocarbon source rocks of the Longtan Formation would have undergone a phase change in some areas of the Sichuan Basin and become marine hydrocarbon source rocks of the Wujiaping Formation (the Upper Permian), with a shift in organic matter type from humic to sapropelic. Compared with the coal measures of the Longtan Formation, the carbon isotope of the natural gas generated by the source rocks of the Wujiaping Formation would be relatively more negative, which can completely match the natural gas of the  $T_2 l^1$ member. Therefore, it is believed that the natural gas from the  $T_2l^1$  member of the Moxi gas field comes from source rocks of the Wujiaping Formation.

# 6.4 Causes of Carbon Isotope Inversion in Natural Gas in $T_2 I^4$ in the Yuanba Gas Field

The phenomenon of carbon isotope inversion in alkane gas has been reported for a long time (Stahl et al., 1975; Fuex, 1977; Burruss et al.,

2010) and later studied by Tilley et al. (2011), Zumberge et al. (2012), and many others. This phenomenon is often found in major oil and gas basins in China, especially shale gas, where carbon isotopes are mostly in reverse order (Dai et al., 2016a). There are various explanations for the isotope inversion, but none is convincing. Most of the carbon isotope inversions in this study occurred in the  $T_2l^4$  member of the Yuanba gas field, nine of the 14 samples have carbon isotope inversions of methane and ethane, that is,  $\delta^{13}C_2 < \delta^{13}C_1$ . The inverted samples were nearly 65% (**Table 1**; **Figure 7**). Some samples from the Xujiahe Formation in the Yuanba gas field have carbon isotope inversions of methane and ethane, and the samples are all distributed in the  $T_3x^2$  member, such as wells YL 9 and YL 10 (**Table 1**). No carbon isotope inversions were found in either  $T_2l^4$  or Xujiahe Formation gas reservoirs in the Longgang gas field, and no carbon isotope inversions occurred in any of the other gas reservoirs.

First, we think that the carbon isotope inversion presented in this study can rule out the reason caused by mixing different natural gases. The natural gas generated from the Xujiahe Formation can migrate downward to the T<sub>2</sub>l<sup>4</sup> gas reservoir, which is common in the Longgang gas field. Half of the samples from the  $T_2l^4$  gas reservoir in the Longgang gas field are humic gas, while only a few samples from T<sub>2</sub>l<sup>4</sup> gas reservoir in the Yuanba gas field are humic gas (Figure 8). This indicates that the scale of humic gas of the Xujiahe Formation mixed into the T<sub>2</sub>l<sup>4</sup> gas reservoir in Longgang is larger than that in Yuanba. However, the natural gas of the Longgang T<sub>2</sub>l<sup>4</sup> member does not appear reversed, while most samples of the Yuanba T<sub>2</sub>l<sup>4</sup> member with little humic gas appear carbon isotope inversion. In addition, no sapropel gas samples were found in the Xujiahe Formation gas reservoir in the Longgang gas field, indicating that the natural gas in  $T_2l^4$  member did not migrate upward obviously. However, more than half of the samples from the Xujiahe Formation gas reservoir in the Yuanba gas field are sapropel gas, indicating that the natural gas from the underlying  $T_2l^4$  gas reservoir has massively upwardly migrated into the Xujiahe Formation gas reservoir (Figure 9). Only a few individual samples showed carbon isotope inversions in the case of large amounts of sapropel gas mixed into the Xujiahe Formation in the Yuanba gas field, and the inverted samples were all found in the  $T_3x^2$  member,





which is close to the  $T_2l^4$  member, and showed typical sapropel gas, which had already inverted before leaving the  $T_2l^4$  member and entering the Xujiahe Formation.

From the carbon isotope of sapropel gas in the  $T_2l^4$  member of Longgang and Yuanba gas fields, the  $\delta^{13}C_2$  in  $T_2l^4$  member of Yuanba is 2‰ more negative than that of Longgang, while the methane carbon isotope is 2‰ less negative than that of Longgang. This indicates that the hydrocarbon source rock type in the  $T_2l^3$ member of the Yuanba gas field is of a higher quality than that of Longgang gas field. The hydrocarbon source rock maturity is higher than Longgang. Therefore, it can be concluded that the carbon isotope inversion of natural gas in the Yuanba  $T_2l^4$  member occurred because of its good hydrocarbon source rock type is good and very favorable for oil generation, the ethane carbon isotope will be relatively more negative and will change less with increasing evolution, while methane carbon isotopes become less negative rapidly and become less negative than ethane as source rock maturity reaches a certain level. It is difficult for humic gas to have the carbon isotope inversion of methane and ethane. Because the  $\delta^{13}C_2$  in humic gas is inherently much less negative, the carbon isotope of methane generated during the evolution of source rocks is difficult to be less negative than ethane. This also explains why the methane and ethane carbon isotopes in the Changxing and Feixianguan formation gas reservoirs did not reverse.

### **7 CONCLUSION**

The natural gas of the Leikoupo Formation in the Sichuan Basin has complex genetic types and various gas sources. The natural gas in the  $T_2l^1$  gas reservoir of the Moxi gas field is all sapropel-type, which comes from the sapropel source rock of the Upper Permian Wujiaping Formation. The natural gas in the  $T_2l^3$  gas reservoir of the Zhongba gas field is mainly sapropel gas, which comes from hydrocarbon source rocks of  $T_2l^3$  itself. Half of the natural gas in  $T_2l^4$ gas reservoir of the Longgang gas field is humic gas from the humic source rocks of the Xujiahe Formation, and the other half is sapropel gas from the  $T_2l^3$  source rocks. The gas in  $T_2l^4$  gas reservoir of the Yuanba gas field is mainly sapropel gas from the  $T_2l^3$  hydrocarbon source rocks, and a very small portion is humic gas from the Xujiahe Formation source rocks. The natural gas with inversed carbon isotopes of methane and ethane was formed from favorable quality source rocks at a higher evolution stage.

# DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/Supplementary Material; further inquiries can be directed to the corresponding author.

## AUTHOR CONTRIBUTIONS

SQ put forward the opinion of article and wrote the manuscript. BZ compiled the diagrams. CH collected data. JL collected sample, performed analysis, and helped with manuscript translation. JW helped with sample analysis and manuscript translation. GT helped with sample analysis and manuscript translation. ZZ helped some data interpretation and manuscript revision.

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**Conflict of Interest:** Authors SQ, BZ, and CH were employed by the company Southwest Oil and Gas Company, PetroChina.

The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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