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Geochemical Comparison of the Deep Gases From the Sichuan and Tarim Basins, China

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In order to have a better understanding of the geochemical characteristics of gases from deep depths, gases from the clastic sandstone reservoirs in the Dabei and Keshen gas fields in the Kuqa depression, Tarim Basin, and gases from the marine carbonate reservoirs (Ordovician and Cambrian) in the craton area of Tarim Basin and Sichuan Basin (Yuanba, Longgang, Puguang gas fields) are investigated based on the molecular composition, stable carbon and hydrogen isotopes. Deep gas, either from the clastic sandstone reservoirs or from the marine carbonate reservoirs, is dominated by alkane gas. Gases from Kuqa depression and Sichuan Basin are dry gas, with high gas dryness coefficient, 0.976 and 0.999, respectively. Deep gas from the craton area in Tarim Basin includes both dry and wet gases. N_2 and CO_2 are the common non-hydrocarbon components in the deep gas. Gases from the continental sandstone reservoirs have no H_2S, while gases from the marine carbonate reservoirs often have H_2S. The relatively high $\delta^{13}C_2$ value in the Kuqa depression indicates the gas was generated from humic type III kerogen, while the relatively low $\delta^{13}C_2$ value in the craton area of Tarim Basin indicates most of the gas was generated from the marine sapropelic organic matter. Deep gas in Sichuan Basin, which has medium $\delta^{13}C_2$ value, was generated from both humic type III and sapropelic type II organic matter. Carbon isotopic anomaly such as partial carbon isotopic reversal or relatively heavy carbon isotope is common in the deep gas, which is caused by secondary alteration. Gases from the Dabei gas field have a mean δ^2H_1 value of −156‰, while gases from the craton area of Tarim Basin, and Yuanba and Puguang gas fields in Sichuan Basin have relatively heavier δ^2H_1 value, i.e., average at −130 and −122‰, respectively. The abnormally heavier δ^2H_1 value in Dabei gas field is due to the high thermal maturity and possible saline depositional environment of the source rocks. This study performed a comprehensive comparison of the geochemical characteristics of the deep gases with different origins, which may provide a hint for future exploration of deep gas in the world.

Keywords: deep gas, ultra-deep gas, tight gas, hydrogen isotope, carbon isotope, Sichuan Basin, Tarim Basin

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INTRODUCTION

After massive exploitation of conventional oil and gas, exploration has been increasingly difficult and more attention has been paid to the deep and ultra-deep resources, especially in the mature exploration area, deep and ultra-deep area has been a significant target. Debates exist in the definition of exact burial depth of the deep and ultra-deep resources. From a geological perspective, "deep gas" is generally referred to the gas with reservoir depth greater than 4,500 m and "ultra-deep gas" is generally referred to the gas with reservoir depth greater than 6,000 m (He et al., 2016; Li, 2016; Sun et al., 2017; Dai et al., 2018). It is also proposed that definition of the exact reservoir depth of the deep and ultra-deep oil and gas resources should consider the influence of geothermal gradient of the basin (Dai et al., 2004a; Chinese Petroleum Society, 2016). For example, the geothermal gradient is 2.26°C/100 m (Feng et al., 2009) and 2.28°C/100 m (Xu et al., 2011) in the Tarim and Sichuan basins, respectively, so in these two basins, deep gas is referred to the gas with reservoir depth greater than 4,500 m and ultra-deep gas is referred to the gas with reservoir depth greater than 6,000 m (Dai et al., 2018). While in eastern China, due to the relatively high geothermal gradient (i.e., 3.58°C/100 m in the North China Basin) (Chen, 1988), "deep" is referred to reservoir depth greater than 3,500 m and "ultra-deep" is referred to reservoir depth greater than 4,500 m (Chinese Petroleum Society, 2016).

To date, a number of countries have carried out deep gas exploration and great breakthrough has been achieved (Tuo, 2002). During 2008–2018, the global proven oil and gas reserves in the deep and ultra-deep strata with burial depth >4,000 m were around 14×10^9 t and 9.1×10^9 t (oil equivalent), respectively, accounting for 66 and 61% of the total increased oil and gas proven reserves, respectively (Zhao, 2019). In China, most exploration has focused on the shallow-middle depths and few exploration has been carried out on the deep oil and gas resources, so the exploration degree of deep resources is low. However, the deep oil and gas resources in China have great potential. The geological oil and gas reserves of China's deep and ultra-deep depths are 26.6×10^9 t and 49.7×10^{12} m^3, respectively, which account for 21 and 55% of the total oil and gas geological reserves in China, respectively (Strategic Research Center of Oil and Gas Resources, 2017). The exploration degree is only 13 and 10% for the deep and ultra-deep oil and gas, respectively (Strategic Research Center of Oil and Gas Resources, 2017). Recently, significant discoveries have been made in the exploration of deep gas in the Tarim and Sichuan basins in China. For example, a number of marine carbonate oil/gas zones have been discovered, such as Lunnan-Tahe, Tazhong, and Halahatang in the Tarim Basin and Puguang, Longgang and Yuanba in the Sichuan Basin, and terrestrial clastic gas fields such as Dabei and Keshen in the Kuqa depression in Tarim Basin (Sun et al., 2013). In China, the deepest gas field is the Keshen gas field in Tarim Basin with reservoir depth over 8,000 m (Well Keshen 902, 8,038 m).

With increasing improvements in the technological development and theoretical understanding, exploration of deep gas will become more and more practical. Moreover, natural gas may form in the deep parts of the sedimentary basin and migrate upward and accumulate in the shallow layers. A full understanding of the origin of deep gas will shed light for the research of natural gas in both shallow and deep basins. However, knowledge about the deep gas in many sedimentary basins is still poor and geochemical information about the deep gas is limited. Previous study has indicated that a number of factors contribute to the origin of deep gas such as the type of organic matter, thermal maturity, oil stability, the influences of minerals, water, non-hydrocarbon gas, etc. (Dyman et al., 2003). This makes the study of the origin and source of deep gas very difficult. Dai et al. (2018) carried out a geochemical study of the ultra-deep gas in the Sichuan Basin and proposed that most of these deep gases were sourced from humic organic matter. In order to have a better understanding of the origin and distribution of deep gas, this study performed a comprehensive comparative investigation of the geochemical characteristics of the deep gases from different types of reservoirs in China. First of all, gases from the clastic sandstone reservoirs in the Dabei gas field in Kuqa depression, Tarim Basin were emphatically investigated (**Figure 1A**). Dabei gas field is one of the 15 large tight gas fields in China, which is also the only large tight gas field in Tarim Basin (Dai et al., 2014). It is also the one with largest burial depth and most complex structures among the continental large gas fields that have ever been found. The sandstone reservoir under the salts in the Dabei gas field generally has depth more than 5,300 m, porosity of 1∼8%, and permeability of $0.01 \times 10^{-15} \sim 1.0 \times 10^{-15}$ m^2, which belongs to typical deep tight gas field (Dai et al., 2014). For comparison, literature data of deep gases from the sandstone reservoir in Keshen gas field in the same depression, and deep gases from the marine carbonate reservoirs in the craton area of Tarim Basin (**Figure 1B**) and Sichuan Basin (Yuanba, Longgang, Puguang gas fields) (**Figure 2**) are also investigated. A detailed comparative geochemical research about the deep gases with different origins from different basins may deepen our understanding of the origin and accumulation of deep gas and shed a light on the future exploration of deep gas in the world.

GEOCHEMISTRY OF SOURCE ROCKS

Tarim Basin

Tarim Basin is located in northwest China and has an area of 560,000 km^2. A series of giant gas fields have been discovered in this basin, which made it one of the most important gas basins in China. Tarim Basin is a large superimposed and composite basin, and it consists of a Paleozoic cratonic basin in the south (**Figure 1B**) and a Mesozoic–Cenozoic foreland basin in the north (**Figure 1A**). The basin was deposited with sediments from Sinian to Quaternary with maximum thickness up to 15,000 m. The main gas-bearing reservoirs include the strata of Ordovician, Carboniferous, Triassic, Jurassic, Cretaceous, and Tertiary. Potential source rocks include the Cambrian–Ordovician highly mature–over mature marine carbonates, Carboniferous–Permian marine–terrestrial transitional highly mature–middle mature carbonates and mudstones, and the Triassic–Jurassic low mature–middle mature lacustrine mudstones and lacustrine-swamp coal measures.

In the Kuqa depression, the widely distributed Middle-Lower Jurassic coal measures deposited in the lacustrine-swamp environment have been regarded as the main source rocks for the coal-derived gases (Liang et al., 2003b; Qin et al., 2007). The thickness of coal bed usually ranges from 6 to 29 m. The Middle-Lower Jurassic source rocks are dominated by type III kerogen and have $\delta^{13}C$ values −22∼−26‰ (Liang et al., 2003a;

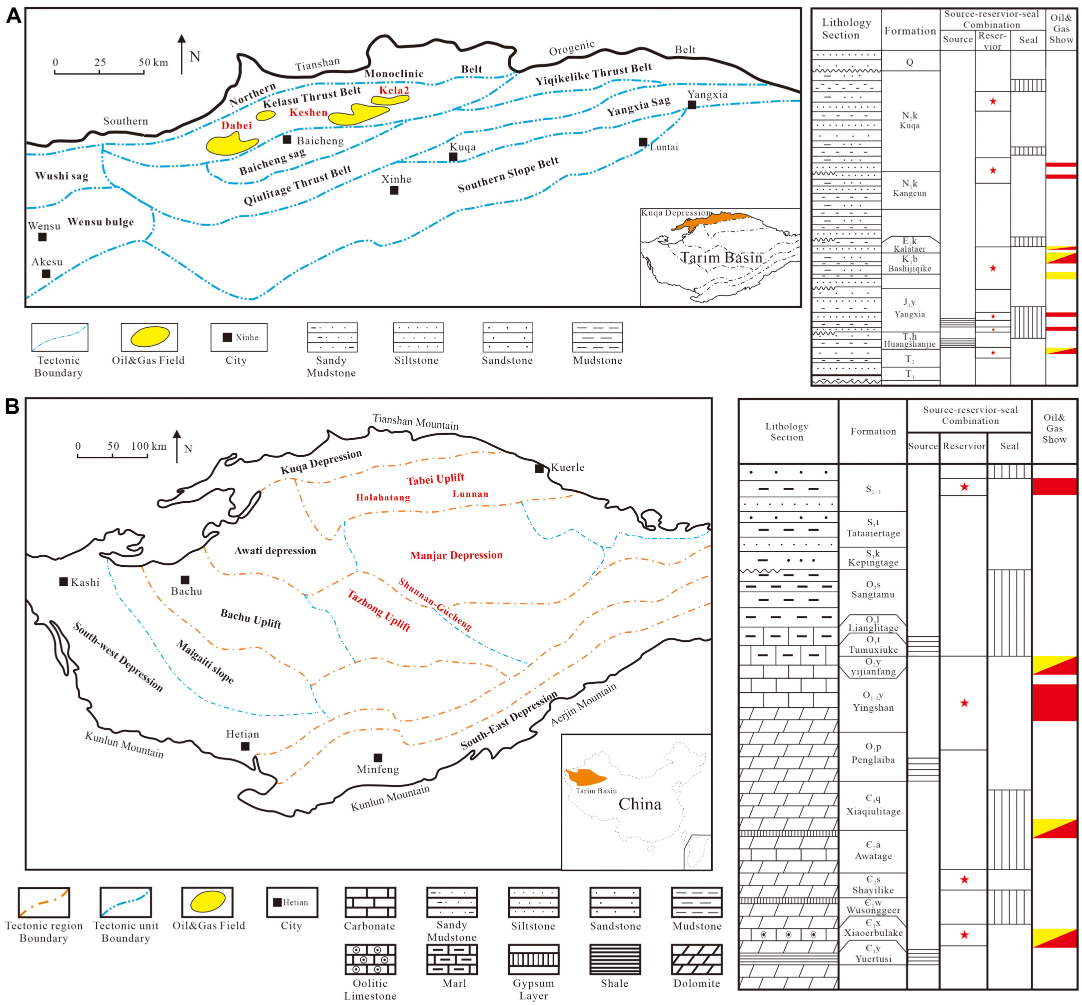

FIGURE 1 | (A) Location map of the Dabei, Keshen, and Kela2 gas fields in the Kuqa depression, Tarim Basin and the stratigraphic column of the Kuqa depression (Wang J. et al., 2018; Zhao et al., 2018). **(B)** Location map of the cratonic Tarim basin and the corresponding stratigraphic column (Ni et al., 2013; Zhu et al., 2019b).

Wang et al., 2005). Thermal maturity of the source rocks in the depression is generally greater than 0.6%, and in the Baicheng sag, thermal maturity of the source rocks is very high, i.e., 1.5~3.5% (Dai, 2016).

The Cambrian-Lower Ordovician carbonates have been considered as the main source rocks of the oil-derived gases in the Paleozoic cratonic Tarim Basin (Liang et al., 2002). The Cambrian-Lower Ordovician source rocks are deposited in a sub-compensation and evaporating lagoon environment and the water medium is saline water. It is dominated by type I kerogen and it has total organic carbon of 0.27~5.52%. The carbon isotope value of kerogen is −32.5~−27.0‰ (Chen et al., 2000, 2001). The source rocks are at high to over mature stage and the vitrinite reflectance value (Ro%) is 1.8~3.0% (Wang et al., 2002; Zhang Z. et al., 2006).

Sichuan Basin

The Sichuan Basin is located in eastern Sichuan and has an area of 18 × 10^4 km^2 (**Figure 2**). A number of giant gas fields have been discovered in this basin and recent exploration success of the deep gases with reservoir depth more than 4,500 m has made Sichuan Basin one of the most important gas producing basins in China. The Sichuan Basin is a tectonic superimposed basin, where the late Mesozoic–Cenozoic foreland basin overlying on a Sinian–Middle Mesozoic passive margin (Ma et al., 2007). It has deposited super-thick Sinian-Middle Triassic marine carbonates (4~7 km), the early stage of Late Triassic marine-continental transitional deposition (300~400 m) and the middle stage of Late Triassic-Eocene terrestrial clastic rocks (2~5 km) (Ma et al., 2010).

In the Sichuan Basin, Permian is an important strata developing two sets of source rocks including Middle Permian carbonates and Upper Permian coal measures with Upper Permian Dalong Formation marine mudstone source rocks in local areas (Chen et al., 2018b). Among them, the Upper Permian Longtan Formation has been considered to make main contribution to the gas source of Puguang, Yuanba and Longgang gas fields (Ma, 2008; Ma et al., 2008; Wu et al., 2015a;

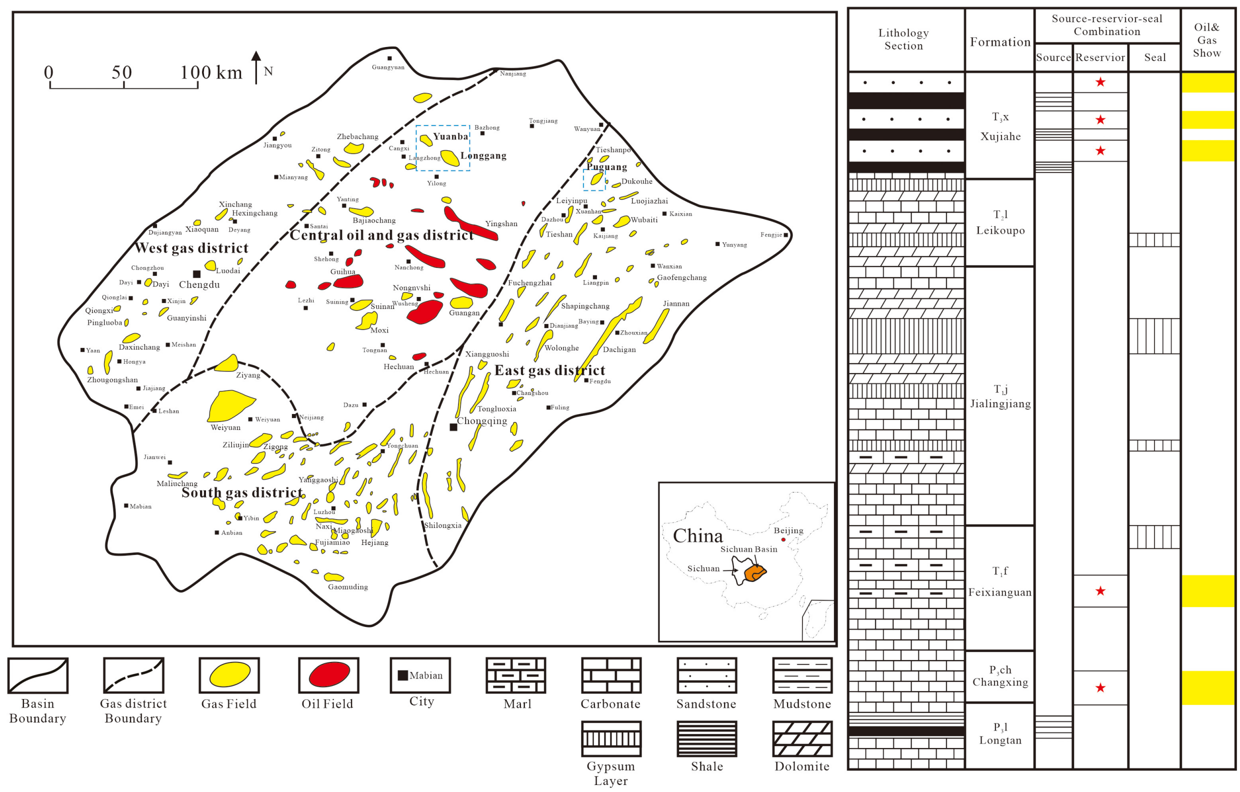

FIGURE 2 | Location map of the Puguang, Yuanba and Longgang gas fields in the Sichuan Basin, the stratigraphic column and source-reservoir-seal combination (Dai et al., 2012a; Qin et al., 2016). Reservoir layer is marked as "*".

Qin et al., 2016; Guo, 2011; Zhao et al., 2011). The marine-continental transitional coal-bearing deposits of the Upper Permian Longtan Formation include mudstone, carbonaceous mudstone, coal and carbonates. The Longtan mudstone source rocks are widely distributed in the whole basin and the coal bed has thickness of 2∼5 m in the northeastern Sichuan Basin, 5∼15 m in the central and southeastern Sichuan Basin, and less than 1 m in the north and southwestern Sichuan Basin (Chen et al., 2018b). According to the analysis of 372 samples, the TOC of Longtan source rocks has an average value of 2.83% (Chen et al., 2018a). In the southwestern part of eastern Sichuan Basin, the Longtan coal measure source rocks have TOC up to 7.47% with an average of 2.91%, $\delta^{13}C$ of kerogen of −24.0∼−23.5‰, thermal maturity (Ro%) of 1.89∼2.63% (Yang et al., 2002).

SAMPLES AND METHODS

276 gas samples with reservoir depth more than 4,500 m from Sichuan and Tarim basins were analyzed in this study, including 11 gas samples collected from the Dabei gas field in the Kuqa depression, Tarim Basin and 265 samples from published literature (see **Table 1** for the natural gas from Dabei gas field). All the gases were called as deep gas, which includes both deep gas and ultra-deep gas. Among the 276 samples, 53 samples were from the Kelasu thrust belt in the Kuqa depression, i.e., 47 samples from the Dabei gas field (Li et al., 2005; Zhang et al., 2011; Dai, 2016; Wei et al., 2019; Zhu et al., 2019a), and 6 samples from the Keshen gas field (Wei et al., 2019), marked as Kuqa-Dabei and Kuqa-Keshen, respectively in all figures; 153 gas samples were from the Tabei and Tazhong uplifts in the Tarim Basin (Wang, 2005; Ni et al., 2013; Wang et al., 2014; Wu et al., 2014; Zhu et al., 2014, 2015; Wang Y. et al., 2018; Zhou et al., 2019), marked as Tarim-craton in all figures; and 70 samples were from Longgang, Yuanba, and Puguang gas fields in Sichuan Basin (Li et al., 2015, 2016; Wang et al., 2015; Wu et al., 2015b; Qin et al., 2016; Dai et al., 2018), marked as Sichuan in all figures.

The 11 gas samples were analysed in the PetroChina Research Institute of Petroleum Exploration and Development (RIPED). Gas molecular composition was conducted on a two-channel Wasson-Agilent 7890 gas chromatograph, equipped with one auxiliary oven, two capillary and six packed analytical columns, one flame ionization detector (FID) and two thermal conductivity detectors (TCD). The carrier gas was high-pure N_2 for FID and He for TCD. GC oven temperature was initially set at 68°C for 7 min, and then increased to 90°C at 10°C/min, held at 90°C for 1.5 min, ramped to 175°C at 15°C/min and held at this temperature for 5 min.

Compound specific stable carbon isotopic composition was determined on a Thermo Delta V mass spectrometer interfaced with a Thermo Trace GC ultra-gas chromatograph (GC). Individual hydrocarbon gas components (C_1-C_5) and CO_2 were separated by using a fused silica capillary column (PLOT Q 27.5 m × 0.32 mm × 10 μm) on a gas chromatograph with He as the carrier gas, then converted into CO_2 in a combustion interface and introduced into the mass spectrometer. The GC oven temperature was increased from 33 to 80°C at 8°C/min, then to 250°C at 5°C/min, and maintained at this temperature for 10 min. All analyses were analysed in triplicate, and the stable carbon isotopic values were reported in the δ-notation in per mil (‰) relative to VPDB. The analytical precision was estimated to be ± 0.3‰.

Compound specific stable hydrogen isotopic composition was measured on a MAT253 isotope ratio mass spectrometer interfaced with a Trace GC ultra-gas chromatograph (GC) and a micropyrolysis furnace (1,450°C). Gas components were separated on a HP-PLOT Q column (30 m × 0.32 mm × 20 μm). The carrier gas was helium with flow rate of 1.2 ml/min. A split injection sample introduction mode with split ratio of 1:10 was used for methane and a non-split injection mode was used for ethane and propane. The GC oven temperature program was the same as that for the carbon isotope. Natural gas was separated into individual gas components via the gas chromatograph and then transformed into C and H_2 in the micropyrolysis furnace at 1,450°C. H_2 went into the mass spectrometer and was determined for δ^2H. The stable hydrogen isotope value (δ^2H) was reported in the δ-notation in per mil (‰) relative to VSMOW and the analytical precision was expected to be ± 3‰.

The laboratory working standard was a coal-derived hydrocarbon gas which was obtained from the Ordos basin in China. An international laboratory standard comparison has been conducted for the working standard, for which two-point calibrations were performed with international measurement standards for both carbon isotope ratios (NBS19 and L-SVEC CO_2) and hydrogen isotope ratios (VSMOW and SLAP) (Dai et al., 2012b).

GEOCHEMICAL CHARACTERISTICS OF DEEP GASES

Molecular Composition

Methane is the dominant gas component of the gases, 1.39∼99.34% with an average of 83.80% (n = 269) (**Figure 3**). Among them, 236 gas samples have methane content more than 70%, and 110 gas samples have methane more than 90%, accounting for 87.73 and 40.89% of the total, respectively. C_{2+} heavy hydrocarbons range between 0.01 and 36.26% with an average of 4.06% (n = 266). 209 out of 266 samples have C_{2+} heavy hydrocarbons less than 5%, accounting for 78.57%. In general, samples from the Kelasu thrust belt in the Kuqa depression have relatively high content of methane (average 95.76%, n = 49), with average gas dryness coefficient of 0.976 (n = 49), indicative of dry gas. While samples from the craton area of Tarim Basin have average methane content of 81.98% (n = 153) and C_{2+} heavy hydrocarbons of 6.24% (n = 153). The gas dryness coefficient varies greatly, except for sample from Well LN44 in the Tabei uplift which has gas dryness coefficient of 0.238, the gas dryness coefficient mainly ranges between 0.594 and 0.999, indicating the existence of both dry gas and wet gas. Among the gases from the craton area of Tarim Basin, gas in the Shunnan-Gucheng area has relatively very low C_{2+} heavy hydrocarbon (average 0.69%, n = 19) and high gas dryness coefficient (average 0.992, n = 19), belonging to dry gas; while gas from Halahatang area has relatively very high content of C_{2+} heavy hydrocarbons (average 25.50%, n = 9) and low gas dryness coefficient (average 0.713, n = 9), belonging to wet gas. Gases from Puguang, Yuanba and Longgang gas fields in the Sichuan Basin have very low C_{2+} heavy hydrocarbons (0.01∼0.78% with average value of 0.11%, n = 66) and high gas dryness coefficient (average 0.999, n = 66).

Non-hydrocarbon gas has an average content of 11.56% (n = 260). 162 out of 260 samples have non-hydrocarbon gas less than 10%, 82 out of 260 samples have non-hydrocarbon gas of 10∼30%, and only 16 samples have non-hydrocarbon gas more than 30%. N_2 and CO_2 are common non-hydrocarbon gas, and nearly exist in all samples. 215 out of 258 samples have N_2 less than 5%, 31 out of 258 samples have N_2 of 5∼10%; 171 out of 252 samples have CO_2 less than 5%, 61 out of 252 samples have CO_2 of 5∼15% and 20 out of 252 samples have CO_2 more than 15% (**Figure 3**). H_2S only presents in the gases with marine carbonates as reservoirs, such as gases from Sichuan Basin and the craton area of Tarim Basin. In those gases from Kelasu thrust belt which has sandstone as reservoirs, there is no sign of the presence of H_2S. 76 out of 115 samples have H_2S less than 5%, 28 out of 115 samples have H_2S of 5∼15%, 9 out of 115 samples have H_2S of 15∼30% and two samples from Puguang gas field in the Sichuan Basin have H_2S even more than 45%. In general, gases from the Dabei gas field have the lowest content of non-hydrocarbon gas, i.e., 0.01∼2.94% with an average value of 1.37% (n = 34). Followed by the gas from Keshen gas field, which has non-hydrocarbon gas of 3.50% (average, n = 6). Then followed by the gas from the craton area of Tarim Basin (average 11.17%, n = 153) and Sichuan Basin (average 18.39%, n = 67).

Carbon Isotope

The stable carbon isotopes of methane, ethane and propane vary greatly for gases from different areas. In total, $\delta^{13}C_1$ varies from −26.5 to −54.9‰ with an average of −35.4‰ (n = 253), and $\delta^{13}C_2$ varies from −16.1 to −41.1‰ with an average of −29.7‰ (n = 226). Among them, gas from Dabei and Keshen gas fields in the Kuqa depression has $\delta^{13}C_1$ of −26.5∼−33.1‰ (average −29.7‰, n = 50), $\delta^{13}C_2$ of −16.1∼−24.2‰ (average −21.1‰, n = 49) and $\delta^{13}C_3$ of −18.9∼−25.7‰ (average −21.0‰, n = 38). A small number of samples from the Dabei gas field have slight carbon isotopic reversal between ethane and propane. Except one sample has carbon isotopic reversal magnitude up to 5.9‰, most samples have reversal magnitude less than 2.2‰. While most gases from the Keshen gas field have carbon isotopic reversal between ethane and propane and the reversal magnitude can be up to 3.6‰. Gas from the craton area of Tarim Basin has much lower carbon isotopes, i.e., $\delta^{13}C_1$ of −30.2∼−54.9‰ (average −40.5‰, n = 133), and $\delta^{13}C_2$ of −26.2∼−41.1‰ (average −34.2‰, n = 125). Most samples have normal carbon isotopic distribution among C_1∼C_3 alkane gases, i.e., methane and its homologues become more and more enriched in ^{13}C with increasing carbon

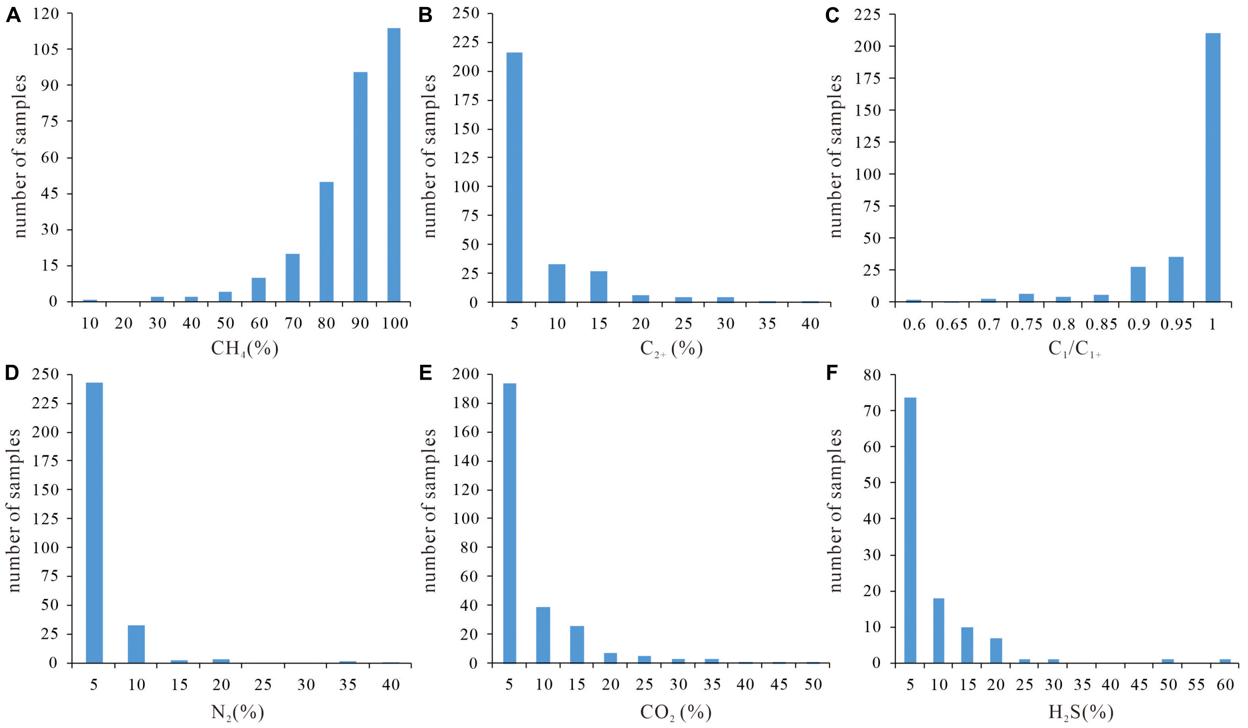

FIGURE 3 | Histogram of the content of CH_4 **(A)**, C_{2+} **(B)**, C_1/C_{1+} **(C)**, N_2 **(D)**, CO_2 **(E),** and H_2S **(F)** of the deep gases from Tarim and Sichuan basins. Data source see section "Samples and Methods" in the text.

number ($\delta^{13}C_1$ < $\delta^{13}C_2$ < $\delta^{13}C_3$). Samples from Sichuan Basin have $\delta^{13}C_1$ of −27.4∼−33.7‰ (average −29.9‰, n = 70), and $\delta^{13}C_2$ of −21.0∼−32.2‰ (average −27.0‰, n = 52). The average C_{2+} heavy hydrocarbon content is only 0.11% (n = 66), so only 4 samples have $\delta^{13}C_3$ values with average of −28.0‰.

The $\delta^{13}C$ of CO_2 varies greatly, i.e., −2.8∼−31.0‰ with an average of −6.0‰ (n = 110). Gas from the Dabei and Keshen gas fields has $\delta^{13}C_{CO2}$ values of −3.6∼−31.0‰ (average −15.1‰, n = 19). Among them, one sample has $\delta^{13}C_{CO2}$ value of −3.6‰, 3 samples have $\delta^{13}C_{CO2}$ values of −8.5∼−9.7‰, and all other 15 samples have $\delta^{13}C_{CO2}$ values lower than −10‰. Gas from the craton area of Tarim Basin has $\delta^{13}C_{CO2}$ values of −25.0∼0.6‰ (average −7.1‰, n = 41). Among them, 27 samples have $\delta^{13}C_{CO2}$ values greater than −8‰, 5 samples have $\delta^{13}C_{CO2}$ values of −8.8∼−9.8‰ and 9 samples have $\delta^{13}C_{CO2}$ values lower than −10‰.

Hydrogen Isotope

The stable hydrogen isotopic values vary between −111∼−158‰ with an average of −134‰ (n = 44) for methane, between −94∼−145‰ with an average of −124‰ (n = 29) for ethane and between −91∼−130‰ with an average of −111‰ (n = 18) for propane. Gas from the Dabei gas field has relatively very high δ^2H value, δ^2H value ranges −153 to −158‰ with an average of −156‰ (n = 11) for methane and from −115 to −122‰ with an average of −119‰ (n = 11) for ethane. Gas from the craton area of Tarim Basin has δ^2H_1 value of −121∼−140‰ (average −130‰, n = 18) and δ^2H_2 value of −94∼−145‰ (average −126, n = 18). Due to the relatively small amount of C_{2+} heavy hydrocarbons in the samples from Sichuan Basin but relatively large required sample amount for the hydrogen isotopic analysis, gas from Sichuan Basin only has hydrogen isotopic values for methane, which ranges from −111 to −156‰ with an average of −122% (n = 15). All the samples have normal hydrogen isotopic distribution pattern among C_1∼C_3 alkane gases, i.e., methane and its homologues become more enriched in D with increasing carbon numbers (δ^2H_1 < δ^2H_2 < δ^2H_3).

ORIGIN OF DEEP GASES

Type and Origin of Deep Gas

Natural gas in nature can be divided into biotic and abiotic origin, and the biotic gas can be divided into microbial gas and thermogenic gas. According to the type of organic matter, thermogenic gas can be further divided into oil-related gas in sapropelic origin (including oil-associated gas and oil-cracked gas) and coal-derived gas in humic origin. The oil-related gas is produced by the thermal decomposition of sapropelic kerogen or the thermal cracking of oil which was generated from sapropelic kerogen. The coal-derived gas is generated from the thermal degradation of humic kerogen or the thermal cracking of heavy hydrocarbons generated from humic organic matter (Dai et al., 1992). Since natural gas is composed of a few simple small molecules, gas origin studies are mainly dependent on its molecular composition, stable carbon and hydrogen isotopes.

A number of studies using molecular composition, carbon and hydrogen isotopes have been performed to investigate the gas origin (Bernard et al., 1976; James, 1983; Clayton, 1991; Dai, 1993; Prinzhofer and Huc, 1995; Lorant et al., 1998; Whiticar, 1999; Tang et al., 2000; Dai et al., 2005a, 2012a, 2016a; Milkov and Etiope, 2018).

The $C_1/(C_2 + C_3)$ vs. $\delta^{13}C_1$ diagram has been widely used to investigate the gas origin (Bernard et al., 1976; Whiticar, 1999; Milkov and Etiope, 2018). In this diagram (**Figure 4**), all the deep gas samples fall in the field of thermogenic gas and far away from the field of primary microbial gas. This indicates there is no mixing with primary microbial gas. Some gas samples from the craton area of Tarim Basin fall in the oil-associated gas field, but most of the samples are more toward the field of late mature thermogenic gas, especially the gas from Sichuan Basin, which shows particularly low content of C_{2+} heavy hydrocarbons and the $C_1/(C_2 + C_3)$ ratio can be up to 4689 (Well Y123 in Yuanba gas field). Though there is significant overlap of thermogenic, primary microbial and abiotic gases in the $\delta^{13}C_1$ vs. $\delta^{13}C_{CO2}$ diagram (**Figure 5**), this diagram is very useful for distinguishing secondary microbial gas. As seen in **Figure 5**, all gas samples are away from the field of secondary microbial gas. This further indicates that all the deep gas has no mixing with neither primary microbial gas or secondary microbial gas. This agrees well with the geological conditions. The burial depth of the produced layer of all gas samples was more than 4,500 m, microbial activities are unlikely to be active at such depth.

With the increase of thermal evolution degree of the organic matter, natural gas become more and more enriched in ^{13}C, plot of $\delta^{13}C_1$ vs. $\delta^{13}C_2$ has been widely used as a gas maturity trend and classification diagram for natural gas origin (Jenden et al., 1988; Dai and Qi, 1989; Rooney et al., 1995; Berner and Faber, 1996; Huang et al., 1996; Tang et al., 2000). A theoretical slope value of 2.4 was calculated for the correlation line between $\delta^{13}C_1$ and $\delta^{13}C_2$ (Xia and Gao, 2017). Different source rocks, secondary alteration or different gas generation mechanism may cause deviation from the theoretical linear trend. As shown in **Figure 6**, gases from the continental sandstone reservoirs (Dabei and Keshen gas fields) in the Kuqa depression have $\delta^{13}C_2$ values far greater than −28‰, and all fall in the up-left area in the $\delta^{13}C_1$ vs. $\delta^{13}C_2$ diagram for gases generated from type III kerogen (Jenden et al., 1988; Dai and Qi, 1989; Rooney et al., 1995; Berner and Faber, 1996). Compared to the gases generated from type III kerogen, gases from Dabei gas field were the most similar to the gases generated from type III kerogen in Niger Delta (Rooney et al., 1995), but still much heavier carbon isotope of ethane, which result in the fact that only two gas samples from the Dabei gas field obey the evolution trend of gas generated from type III kerogen in Niger Delta and all other gas samples fall above the gas maturity trend (**Figure 6**). Gases from the Keshen gas field have even heavier carbon isotope of ethane (up to −16.1‰ of $\delta^{13}C_2$) and fall far above the evolution trend of gas generated from type III kerogen in Niger Delta of type III kerogen (Rooney et al., 1995). Gases with reservoir depth shallower than 4,500 m from Kela2 gas field in Kuqa depression were also shown for comparison. In general, gases from Kela2 gas field have the heaviest carbon isotope of methane (−25.1∼−30.8‰,

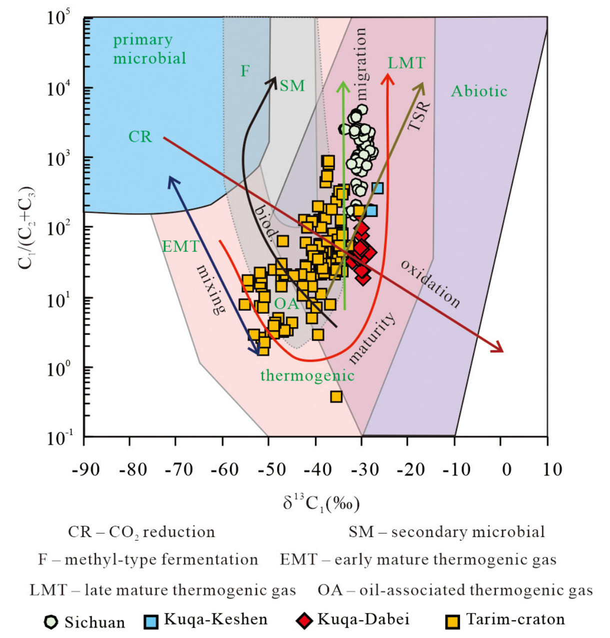

FIGURE 4 | Genetic diagram of $C_1/(C_2 + C_3)$ vs. $\delta^{13}C_1$ of gases from Dabei and Keshen gas fields in the Kuqa depression, craton area of Tarim Basin and Sichuan Basin, China (Milkov and Etiope, 2018). Data source see section "Samples and Methods" in the text.

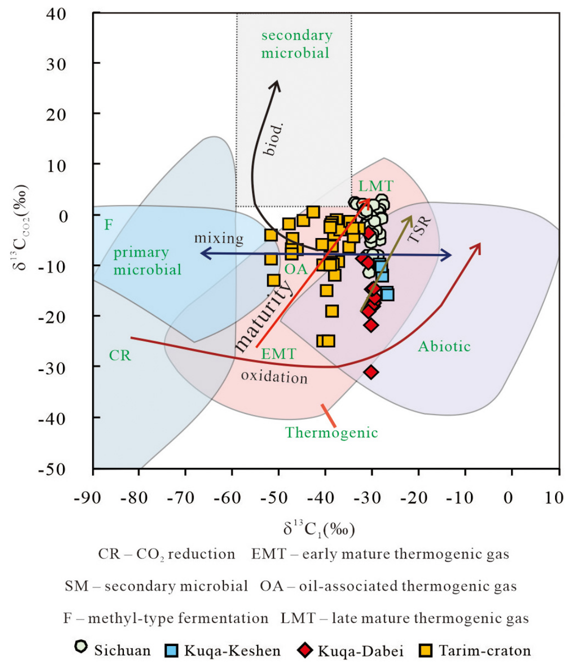

FIGURE 5 | Genetic diagram of $\delta^{13}C_{CO2}$ vs. $\delta^{13}C_1$ of gases from Dabei and Keshen gas fields in the Kuqa depression, craton area of Tarim Basin and Sichuan Basin, China (Milkov and Etiope, 2018). Data source see section "Samples and Methods" in the text.

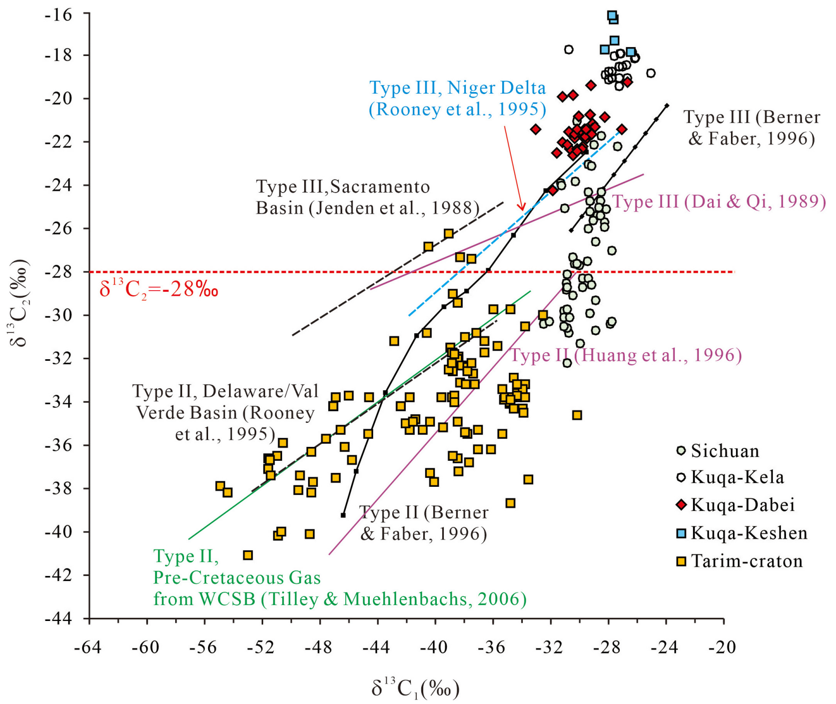

FIGURE 6 | $\delta^{13}C_1$ vs. $\delta^{13}C_2$ cross-plot of gases from Dabei, Keshen and Kela2 gas fields in the Kuqa depression, craton area of Tarim Basin, and Sichuan Basin. Also shown are published maturity trends of type II kerogen (Rooney et al., 1995; Huang et al., 1996; Tilley and Muehlenbachs, 2006) and type III kerogen (Jenden et al., 1988; Dai and Qi, 1989; Rooney et al., 1995; Berner and Faber, 1996). Data source see section "Samples and Methods" in the text.

average −27.4‰, n = 24), gases from the Keshen gas field have the heaviest carbon isotope of ethane (−16.1∼−17.8‰, average −17.0‰, n = 5) and gases from the Dabei gas field have the lightest carbon isotope of methane and ethane. All these characteristics indicate that gases from the continental sandstone reservoirs (Dabei and Keshen gas fields) in the Kuqa depression were generated from higher plant-derived type III kerogen. Gases from Sichuan Basin can be divided into two groups, the first group falls in the region of gases generated from type III kerogen (Dai and Qi, 1989; Rooney et al., 1995; Berner and Faber, 1996), and the second group falls in the region of gases generated from type II kerogen (Huang et al., 1996). Gases in the first group were generated from the humic type III kerogen and gases in the second group were produced from the sapropelic organic matter. Most gas samples from the craton area of Tarim Basin have $\delta^{13}C_2$ values lower than −28‰ and match better with the evolution trend of gas generated from type II kerogen (Rooney et al., 1995; Berner and Faber, 1996; Huang et al., 1996; Tilley and Muehlenbachs, 2006), and only several samples have $\delta^{13}C_2$ values greater than −28‰ and follow the evolution trend of gases generated from the type III kerogen (Jenden et al., 1988; Rooney et al., 1995; **Figure 6**). This indicates that most gases from the craton area of Tarim Basin were generated from the sapropelic organic matter. Compared to the deep gases from the continental sandstone reservoirs in the Kuqa depression and marine carbonate reservoirs in the craton area of Tarim Basin, gases from Sichuan Basin have relatively medium values of carbon isotope of ethane. For the comparison of gases generated from type III kerogen, several samples from the Longgang gas field in Sichuan Basin have $\delta^{13}C_2$ values greater than −25‰, which is close to the $\delta^{13}C_2$ values of gases from Dabei gas field (−19.2∼−24.2‰, average −21.6‰, n = 47). Except these gases, all other coal-derived gases from the marine carbonate reservoirs in Sichuan Basin have lower $\delta^{13}C_2$ values than that of the gases from the continental sandstone reservoirs. However, for the comparison of gases generated from type II kerogen, most gases from Sichuan Basin have greater $\delta^{13}C_2$ values than that of the gases from the craton area of Tarim Basin.

Humic organic matter is dominated by aromatic structures which is enriched in ^{13}C, while sapropelic organic matter is dominated by aliphatic structures which is enriched in ^{12}C. During the decomposition process of kerogen, carbon isotope of methane is more inclined to the influences of thermal maturity while carbon isotope of ethane can better reflect the features of its precursor. Therefore, gas sourced from humic organic matter generally has heavier carbon isotope of ethane compared to that from sapropelic organic matter at similar thermal maturity. A number of cut-off values of $\delta^{13}C_2$ have been proposed for the distinguishing of gas origin in sapropelic or humic according to numerous empirical studies in the Chinese sedimentary basins (Wang, 1994; Gang et al., 1997; Liang et al., 2002; Dai et al., 2005b; Xiao et al., 2008). According to the detailed gas study in the Kuqa depression by Liang et al. (2002), a cut-off value of −28‰ of $\delta^{13}C$ for ethane was proposed to distinguish the gas sources (sapropelic vs. humic). Based on the study of the Sinian-Jurassic gases in the Sichuan Basin, Wang (1994) suggested a cut-off value of −29‰ of $\delta^{13}C$ for ethane to distinguish the gas origin. Dai et al. (2005b) proposed a $\delta^{13}C_2$ >−27.5‰ for gases sourced from humic organic matter and a $\delta^{13}C_2$ <−29‰ for gases sourced from sapropelic organic matter according to a comprehensive study on the natural gas in China, and Dai et al. (2012a) suggested a cut-off value of $\delta^{13}C_2$ >−28‰ for gases sourced from humic organic matter in the Sichuan Basin. Taking −28‰ of $\delta^{13}C_2$ as a cut-off value, combined with the $\delta^{13}C_1$ vs. $\delta^{13}C_2$ diagram (**Figure 6**), gas from the continental sandstone reservoirs in the Dabei and Keshen gas fields in the Kelasu thrust belt in the Kuqa depression were generated from the type III kerogen and belong to coal-derived gas (Zhang et al., 2011; Dai et al., 2014; Wang, 2014; Wei et al., 2019), gases from the marine carbonate reservoirs (Ordovician and Cambrian) in the craton area in the Tarim Basin are mostly sourced from the sapropelic organic matter and belong to oil-related gas (Wang et al., 2014; Zhu et al., 2014; Chen F. et al., 2018). Gases from the marine carbonate reservoirs (Permian Changxing Formation and Triassic Feixianguan Formation) from the Longgang, Yuanba and Puguang gas fields in Sichuan Basin were complicated (Yang et al., 2002; Zhu et al., 2005; Hao et al., 2008; Ma, 2008; Zhao et al., 2011; Hu et al., 2014; Qin et al., 2016; Dai et al., 2018). Gases from the Longgang gas field were mainly generated from the coal measure of Permian Longtan Formation (Zhao et al., 2011; Qin et al., 2016; Dai et al., 2018), or with minor contribution of oil cracked gas derived from the Upper Permian carbonaceous source rocks (Deng et al., 2018). Different opinions exist for the origin of gases from Yuanba gas field. Some scholars proposed that gases from Yuanba gas field were mainly coal-derived gas or with minor contribution from oil-related gas, which were sourced from the coal measure of Permian Longtan Formation and/or organic matter in Permian Dalong Formation (Hu et al., 2014; Dai, 2016; Dai et al., 2018). However, some scholars proposed that gases from Yuanba gas field were mainly oil cracked gas from oil cracking which was generated from the Permian source rocks (Guo, 2011; Guo et al., 2014; Li et al., 2015; Wu et al., 2015b). Gases from Yuanba gas field were also interpreted as oil cracked gas sourced from the sapropelic organic matter in Sinian/Lower Cambrian Qiongzhusi Formation (Wu et al., 2017). Gases from the Puguang gas field were mainly sourced from the marine sapropelic organic matter with minor contribution from the humic organic matter in Permian Longtan Formation (Hao et al., 2008; Ma, 2008). There is also study indicating that gases in the Puguang gas field were generated from multiple sources such as Permian, Silurian and Cambrian (Zhao et al., 2011). The gas origin distinguishing can be further evidenced in the $\delta^{13}C_1 \sim \delta^{13}C_2 \sim \delta^{13}C_3$ genetic diagram (**Figure 7**). In **Figure 7**, gas from the Keshen gas field has very heavy carbon isotope of ethane, which is also clearly reflected in **Figure 6**. Gas from the craton area of Tarim Basin has relatively heavy carbon isotope of propane and some samples even fall in the area of coal-derived origin.

Carbon Isotope Anomaly of the Deep Gas

Partial carbon isotopic reversal is common in the deep gas. The coal-derived tight gas from the Kuqa depression shows partial carbon isotopic reversal between ethane and propane, especially gas from the Keshen gas field, where 4 out of 5 samples have partial carbon isotopic reversal between ethane and propane ($\delta^{13}C_1 < \delta^{13}C_2 > \delta^{13}C_3$). A few samples from the marine

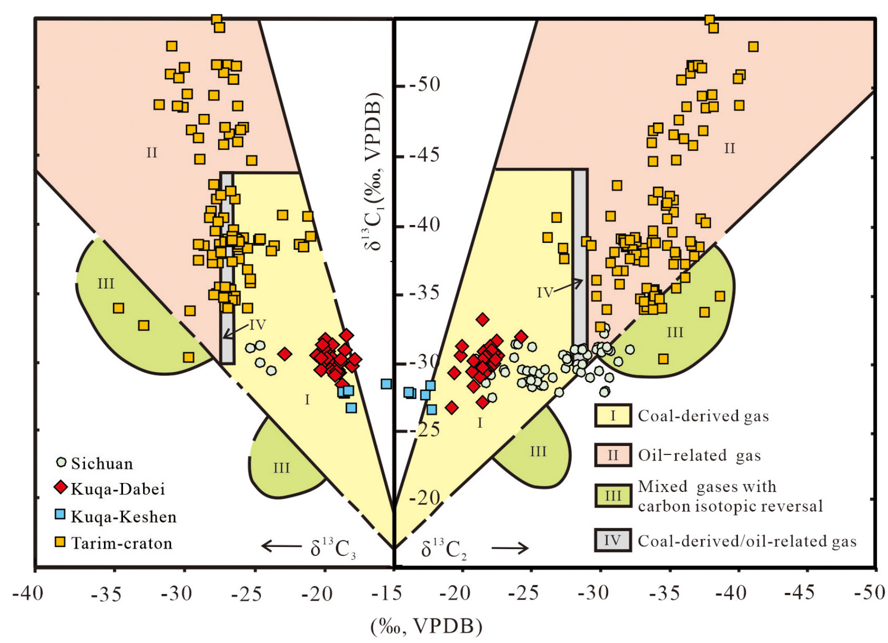

FIGURE 7 | Plot of $\delta^{13}C_1$ vs. $\delta^{13}C_2$ vs. $\delta^{13}C_3$ of gases from Dabei and Keshen gas fields in the Kuqa depression, craton area in the Tarim Basin, and Sichuan Basin (Dai et al., 1992). Data source see section "Samples and Methods" in the text.

carbonate reservoirs either in the craton area of Tarim Basin ($\delta^{13}C_1 < \delta^{13}C_2 > \delta^{13}C_3$) or Sichuan Basin ($\delta^{13}C_1 > \delta^{13}C_2$) also have partial carbon isotopic reversal.

Partial carbon isotopic reversal among $C_1 \sim C_3$ alkane gases are usually interpreted as a result from the mixing of gases generated from different types of organic matter or at different thermal maturity (biotic vs. abiotic, thermogenic vs. microbial, humic vs. sapropelic, gases with different maturities), bacterial oxidation, high thermal maturity, thermochemical sulfate reduction (Dai et al., 2004b; Hao et al., 2008). Some gas samples from the Keshen and Dabei gas fields have partial carbon isotopic reversal between ethane and propane, i.e., $\delta^{13}C_1 < \delta^{13}C_2 > \delta^{13}C_3$. Due to the limited amount for C_{3+} heavy hydrocarbon gas, hydrogen isotopic distribution pattern between ethane and propane is not available. Such partial carbon isotopic reversal has also been reported for gases from other wells with burial depth either more than 4,500 m or less than 4,500 m in the Kelasu-Yiqikelike thrust belt and South Slope Belt (Liu et al., 2007). The partial carbon isotopic reversal between ethane and propane of gases from the Kuqa depression has been explained as abiotic gas (Zhou, 1998; Zhang, 2002). The $\delta^{13}C_1$ vs. δ^2H_1 diagram (**Figure 8**) can better interpret the abiotic methane. As shown in **Figure 8**, gas from the craton area of Tarim Basin and Sichuan Basin falls in the area of thermogenic gas, while gas from the Dabei gas field falls in the overlapped area of abiotic gas and thermogenic gas. However, counterview existed according to the helium isotope. Researches found that natural gas in the Kuqa depression has R/Ra values of 0.03~0.108 with an average of 0.05 (Qin, 1999) or $^3He/^4He$ values of $n \times 10^{-8}$ (Liu et al., 2007), indicating the absence of mantle-derived helium, hence proposed that the partial carbon isotopic reversal was not caused by the mixing between biotic and abiotic gases. Mixing of gases generated from different sources or generated at different thermal maturity levels have been advocated to interpret the partial carbon isotopic reversal. Zhang et al. (2011) proposed that the partial carbon isotopic reversal of gases from Dabei gas field were caused by the mixing between coal-derived gas sourced from the Jurassic coal measures and oil-related gas that was sourced from the Triassic lacustrine source rocks. Dai et al. (2014) indicated that the partial carbon isotopic reversal of gases from Dabei gas field was due to the mixing of gases with the same origin but different maturity levels. Isotopic fractionation caused by gas leakage has also been proposed to interpret the partial carbon isotopic reversal of gases from the Kuqa depression (Qin, 1999). Except these single factor, multi-factor analysis has also been carried out. For example, Liu et al. (2007) proposed that the partial carbon isotopic reversal of gases from the Kuqa depression was caused by the mixing with gas of the same origin but different maturity or resulting from the high temperature, high pressure, multistage hydrocarbon accumulation and diffusion. A recent study proposed that the partial carbon isotopic reversal was due to the mixing between gases of the same origin but different sources or mixing between oil-related and coal-derived gases or high temperature and high pressure (Wei et al., 2019).

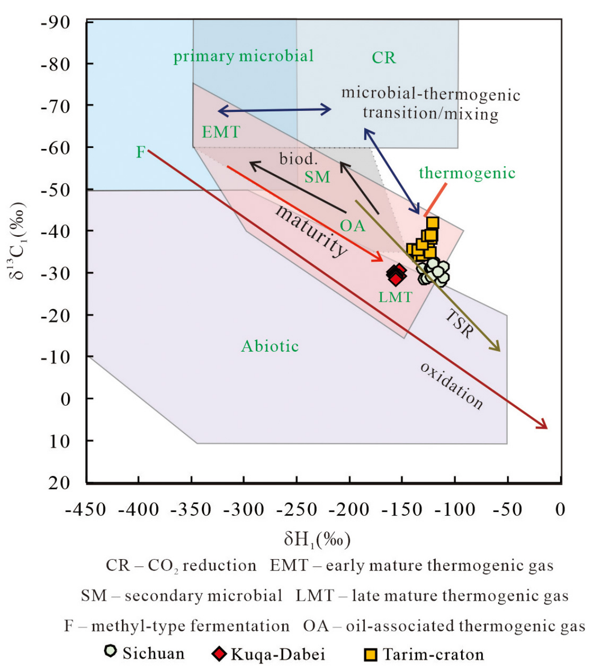

FIGURE 8 | Plot of δ^2H_1 vs. $\delta^{13}C_1$ of gases from Dabei gas field in the Kuqa depression, craton area of Tarim basin and Sichuan basin (Milkov and Etiope, 2018). Data source see section "Samples and Methods" in the text.

Partial carbon isotopic reversal between methane and ethane also exists in the deep gases from marine carbonate reservoirs in the Sichuan Basin (Wang, 1994; Zhu et al., 2006; Hao et al., 2008). Mixing of gases from different sources/maturity levels has been advocated to interpret the partial carbon isotopic reversal found in the gases from marine carbonate reservoirs (Wang, 1994; Hao et al., 2008; Ma, 2008; Deng et al., 2018). For example, partial carbon isotopic reversal between methane and ethane was found in the gases from Puguang gas field, which has been interpreted as mixing of oil cracked gases with different sources, mixing between secondary-cracking and primary cracking gases, in-reservoir mixing of oil cracked gases with different thermal maturities (Hao et al., 2008), or mixing of gases generated from different sources (Wang, 1994; Ma, 2008). In the Longgang gas field, partial carbon isotopic reversal between methane and ethane has been interpreted as a result from the isotopic fractionation during the migration of coal-derived gas (Dai et al., 2018), or mixing of gases generated from humic organic matter and gases generated from the cracking of oil (Deng et al., 2018).

One biggest difference between the gases from continental sandstone reservoirs and marine carbonate reservoirs is the content of H_2S. Gases from the continental sandstone reservoirs have no H_2S, while gases from marine carbonate reservoirs have varying content of H_2S. In the Puguang gas field in Sichuan Basin, the content of H_2S can be up to 58.34%. In most cases, the high content of H_2S in deeply buried gas reservoirs are due to the thermochemical sulfate reduction (TSR). A number of studies have demonstrated the existence of TSR in gases from the craton area of Tarim Basin and northeastern Sichuan Basin (Cai et al., 2001, 2007, 2009, 2013; Zhu et al., 2005, 2006, 2019b; Hao et al., 2008; Liu et al., 2013). Previous studies found that TSR may affect the carbon isotopic composition of hydrocarbon gases

(Krouse et al., 1988; Worden and Smalley, 1996; Cai et al., 2004; Hao et al., 2008; Pan et al., 2006; Zhu et al., 2006). After the liquid hydrocarbon are exhausted, $C_2 \sim C_5$ heavy hydrocarbon gases become the dominant reactants for TSR, then there will be a significant increase of CO_2 and H_2S content (Hao et al., 2008). At this stage, due to the kinetic isotopic fractionation, ^{12}C-^{12}C bond will break faster than ^{12}C-^{13}C bond, resulting in the preferentially loss of C_{2+} heavy hydrocarbon gases enriched in ^{12}C (Worden and Smalley, 1996; Krouse et al., 1988). Methane is the least reactive compound and its involvement in TSR as a reductive reactant has long been in dispute. Therefore, during the stage with $C_2 \sim C_5$ heavy hydrocarbon gases as the dominant reactants for TSR, the increasing extent of TSR will cause the increase of $\delta^{13}C_2$ values but not affect the $\delta^{13}C_1$ values, which will change the partial carbon isotopic reversal between methane and ethane into normal trend (Hao et al., 2008). A gas souring index [$GSI = H_2S/(H_2S + \Sigma Cn)$] has been used to show the extent of TSR (Worden et al., 1995). During this stage, CO_2 should positively correlate with H_2S, GSI should positively correlate with gas dryness coefficient (C_1/C_{1+}), $\delta^{13}C_2$ values correlate negatively with $\delta^{13}C_1$-$\delta^{13}C_2$ values. As seen in **Figure 9**, the relatively high content of CO_2 and H_2S in the gases from Sichuan Basin (H_2S: 9.78%, $n = 51$; CO_2: 8.93%, $n = 67$) and the craton area of Tarim Basin (H_2S: 2.2%, $n = 64$; CO_2: 5.38%, $n = 152$) indicates the existence of TSR. In general, high H_2S content accompanied with high CO_2 content, however, such correlation between H_2S and CO_2 is not very strong (**Figure 9**). The correlation between GSI and C_1/C_{1+}, GSI and $\delta^{13}C_2$, GSI and $\delta^{13}C_1 - \delta^{13}C_2$ values is also not very apparent. This has been explained as that TSR has not affected the hydrocarbon gas (Puguang gas field) (Hu et al., 2010), or differences in the depositional environment and thermal maturity of source rocks in different gas fields (Wu et al., 2015a). Previous study in the Puguang gas field proposed that hydrocarbon gas was generated from the oil cracking and altered by TSR, and the transformation from a C_{2+} heavy hydrocarbon dominated TSR stage to a methane dominated stage caused different effects on the $\delta^{13}C$ of different hydrocarbon gas (Hao et al., 2008).

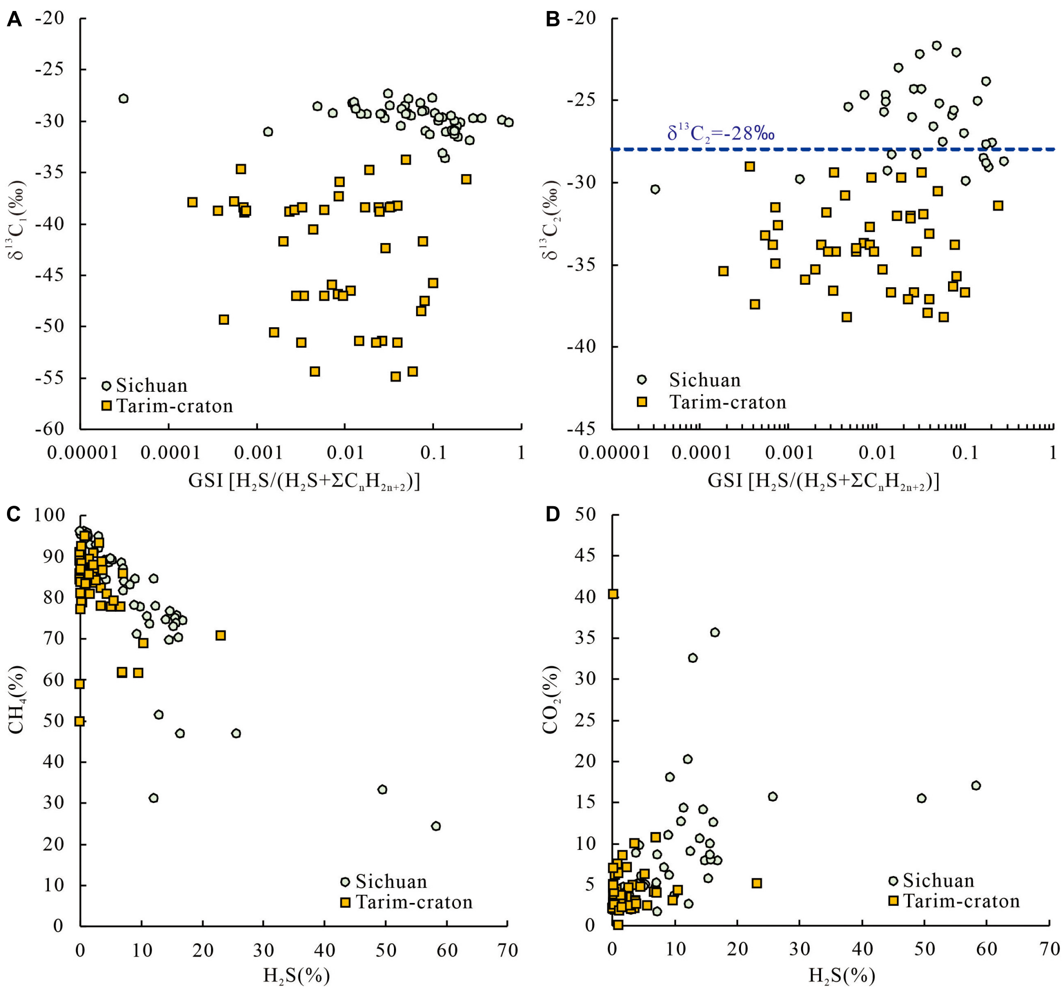

FIGURE 9 | Plot of $\delta^{13}C_1$ vs. GSI [$H_2S/(H_2S + \Sigma C_nH_{2n+2})$] **(A)**, $\delta^{13}C_2$ vs. GSI [$H_2S/(H_2S + \Sigma C_nH_{2n+2})$] **(B)**, CH_4 vs. H_2S **(C)**, and CO_2 vs. H_2S **(D)** of gases from the craton area of Tarim Basin and Sichuan Basin. Data source see section "Samples and Methods" in the text.

Considering the geological background, extremely high thermal maturity of source rocks might play a key role. The present burial depth of these marine carbonates is of 4,775~7,150 m and the present thermal maturity is between 2.0 and 3.0% (Hao et al., 2008; Qin et al., 2016). However, the compression from the movement of the Pacific plate resulted in the entire uplift of Sichuan Basin during the Himalayan orogeny and the uplift amplitude reached 2,400 m (Liu et al., 2008; Deng et al., 2009). Before the uplift, the marine carbonate reservoir (Changxing and Feixianguan formations) in the Puguang gas field reached a maximum burial depth of 7,000 m with temperature up to 220°C (Hao et al., 2008). The maximum burial depth in the Longgang gas field is also estimated to be over 7,000 m and the present thermal maturity is 2.81% (Ro%) for the reservoir bitumen in the Changxing Formation with burial depth of 6118.81~6119.89 m (Qin et al., 2016). The deep gases from Sichuan Basin all show extremely high gas dryness coefficient (average 0.999, $n = 67$) and the average content of C_{2+} heavy hydrocarbon is only 0.11%. Both molecular and isotopic compositions of hydrocarbon gases are affected by the thermal evolution degree of source rocks, especially with such high gas dryness coefficient, it is hard to determine the effects from thermal maturity or TSR.

TSR will cause the increase of carbon isotope of hydrocarbon gases, which may contribute to the relatively heavy carbon isotopic values of ethane in the gases from Sichuan Basin (**Figure 6**). As methane can be the dominant reducing reactant for TSR after most C_{2+} heavy hydrocarbons are exhausted at elevated temperature (Hao et al., 2008), in this case, TSR will result in the increase of carbon isotope of methane. However, great care has to be paid to the explanation of the carbon isotope of methane since other processes also exert influences on the $\delta^{13}C_1$ values. For example, the relatively heavy carbon isotope of methane in the Longgang gas field in Sichuan Basin was interpreted as a result of high thermal maturity of the source rocks and the mixing of gas degassed from water (Qin et al., 2016).

Origin of Non-hydrocarbon Gas CO_2

Milkov and Etiope (2018) found that there is significant overlap of $\delta^{13}C_{CO2}$ values for the primary microbial gas, thermogenic gas and abiotic gas in the $\delta^{13}C_{CO2}-CO_2$ plot (**Figure 5**). According to the carbon isotopic studies of 390 CO_2 gas samples from different sedimentary basins in China, Dai et al. (1996) suggested a distinguishing criterion for CO_2, i.e., CO_2 of biotic origin (including thermogenic and microbial): CO_2 content <15% and $\delta^{13}C_{CO2} < -10‰$; CO_2 of abiotic origin: CO_2 content >60% or $\delta^{13}C_{CO2} > -8‰$ (**Figure 10**). CO_2 in the deep gas has wide variation of carbon isotope (**Figures 5**, **10**). It is clear that contributions from microbial effects can be ignored (**Figure 5**). In general, samples from the continental sandstone reservoirs have relatively light carbon isotope of CO_2 (average −15.1‰, $n = 19$), indicative of the thermogenic origin (**Figure 10**). Gases from the Sichuan Basin have $\delta^{13}C$ value of −1.77‰ ($n = 50$) for CO_2, indicating that most CO_2 gases are of abiotic origin. As shown in **Figure 10**, most deep gases from Sichuan Basin fall in the region of abiotic CO_2. The content of CO_2 in the deep gas from Sichuan Basin has a peak range of 5~10%. The $\delta^{13}C$ values of CO_2 have an average of −1.8‰ ($n = 50$) and a peak range of −5‰ ~ + 3‰, which is close to the $\delta^{13}C$ value of marine carbonate reservoir (0.9~3.7‰) (Hu et al., 2010). A number of studies have demonstrated the abiotic CO_2 was generated from the dissolution of marine carbonates through acid fluid (Hao et al., 2008, 2015; Huang et al., 2010; Liu et al., 2014). Dai et al. (2018) proposed that the CO_2 was generated from the decomposition of marine carbonates during the over mature thermal evolution stage.

THERMAL MATURITY OF DEEP GASES

Gas Dryness and Gas Wetness

Due to the decomposition of heavy hydrocarbons at elevated temperature, with increasing thermal maturity of source rocks, the relative content of methane of hydrocarbon gases will increase and the content of C_{2+} heavy hydrocarbon gases decrease. Gas dryness coefficient $[D = C_1/(C_1\text{-}C_5)]$ or gas wetness coefficient $[W = (C_2\text{-}C_5)/(C_1\text{-}C_5)]$ has been used to evaluate the gas maturity. According to compositional study of the natural gases from Ordos, Sichuan, Bohaibay, Qiongdongnan, Junggar, and Turpan-Hami basins in China, a negative correlation between gas wetness and gas maturity was established for the coal-derived gases sourced from the humic organic matter ($Ro\% = -0.419\ln W + 1.908$) (Dai et al., 2016b). In general, gases with gas dryness coefficient (D) greater than 0.95 is defined to be dry gas and gases with gas dryness coefficient (D) lower than 0.95 is wet gas. Gas generation during the thermal stage with vitrinite reflectance greater than 2.0% is usually dominated by methane and the produced gas is commonly dry gas. According to the delineation of dry gas via gas dryness coefficient, deep gases from Dabei and Keshen in the Kelasu thrust belt in Kuqa depression, Longgang, Yuanba and Puguang gas fields in Sichuan Basin are dry gas, while gases from the craton area of Tarim Basin are composed of both dry gas and wet gas. For example, gases from Shunnan-Gucheng area are mostly dry gas, while gases from Halahatang are mostly wet gas.

Theoretically, with increasing burial depth of source rocks, the geothermal temperature will increase and the thermal maturity of source rocks will increase as well. Therefore, gas dryness coefficient commonly increases with increasing burial depth. However, processes such as migration, leakage and TSR may affect the molecular composition of natural gases, thus result in the changes of gas dryness coefficient. Therefore, great care has to be paid to the researches of natural gases with gas dryness coefficient. **Figure 11A** shows the changes of gas dryness coefficient with burial depth of the coal-derived deep gases from Dabei and Keshen gas fields in the Kuqa depression. Gases with different burial depth of reservoirs from Dawanqi oil field (Zhou and Bao, 2011; Wang et al., 2012) and Kela2 gas field (Qin et al., 2007) in the Kelasu thrust belt were shown for comparison. According to previous studies, due to the barrier of the gypsum-salt layer with massive thickness between the Jurassic-Triassic source rocks and the Neogene Kangcun Formation reservoirs, gases generated from the Jurassic-Triassic source rocks could migrate into the Kangcun Formation reservoirs through the Tuzimaza large fault and form the Dawanqi oil field, while it

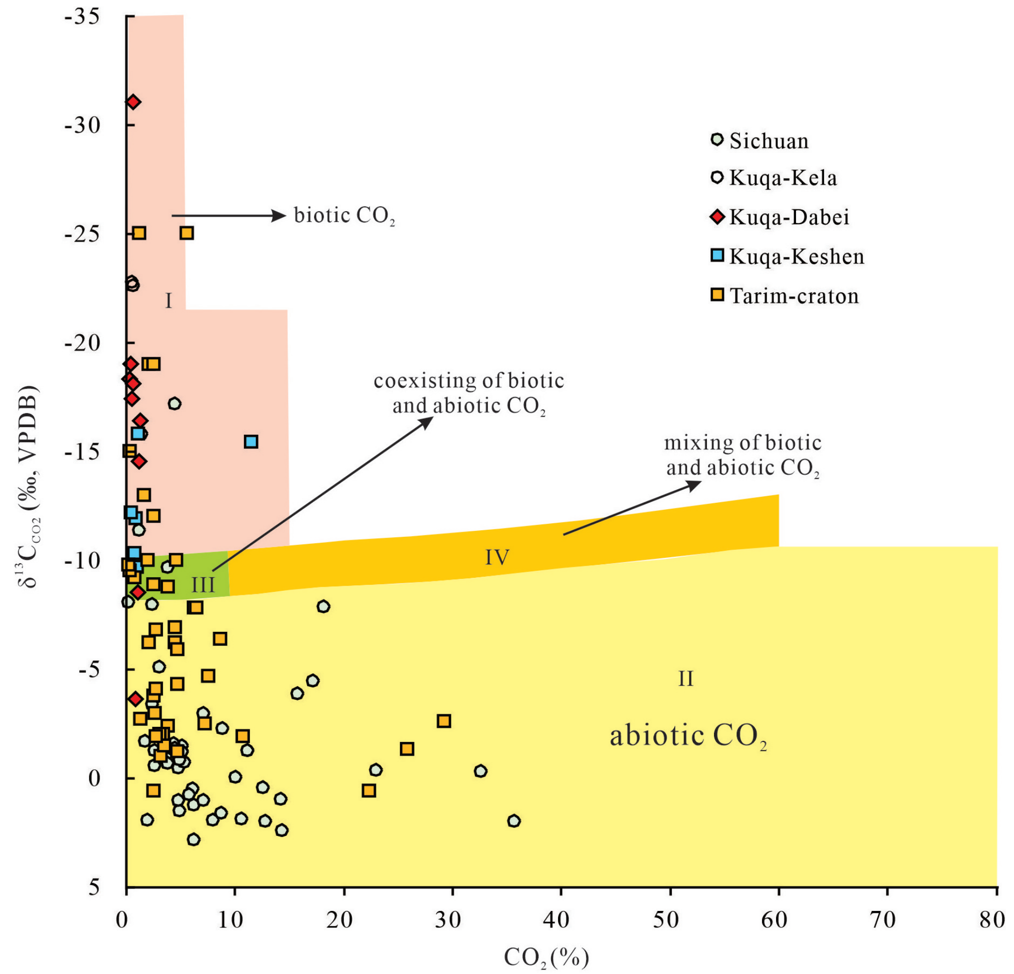

FIGURE 10 | Plot of $\delta^{13}C_{CO2}$ vs. CO_2 of gases from Dabei, Keshen and Kela2 gas fields in the Kuqa depression, craton area of Tarim Basin and Sichuan Basin, distinguishing the origin of CO_2 (Dai et al., 1992). Data source see section "Samples and Methods" in the text.

formed the highly mature-over mature Dabei gas field under the gypsum-salt layer (Liu et al., 2005). Because of the influences of fault and erosion of cap layer, leakage of hydrocarbon gases occurred for the shallow gas, which result in the gas distribution with wet gas in the top and dry gas in the bottom vertically (Tian et al., 2001; Zhou and Bao, 2011). As shown in **Figure 11**, gases from the Dawanqi gas field have relatively low $C_1/(C_1\text{-}C_5)$ ratios and shallower burial depth, while with increasing burial depth, gases from Kela2, Dabei and Keshen gas fields have much higher $C_1/(C_1\text{-}C_5)$ ratios. Actually, gases from Dawanqi oil field have relatively heavy carbon isotope of methane (−28∼−33‰) and the $\delta^{13}C_1$ gets lighter with increasing burial depth (Zhao, 2003), so the apparently low dryness coefficient of gases from Dawanqi oil field is mainly caused by the leakage of light composition in the shallow layers. The deep gases from Dabei and Keshen gas fields demonstrate a slight increasing trend of gas dryness coefficient with increasing burial depth, indicating the influences from thermal maturity.

TSR commonly occurred in the northeastern Sichuan Basin (Cai et al., 2003, 2013; Zhu et al., 2005, 2006; Hao et al., 2008). Since TSR consumed liquid hydrocarbon in the first, followed by C_{2+} heavy hydrocarbon and then methane (Hao et al., 2008), the extremely high gas dryness coefficient in the Longgang, Yuanba and Puguang gas fields not only reflects the high thermal maturity of source rocks to some extent. **Figure 11B** shows the natural gases from Xujiahe, Leikoupo, and Changxing-Feixianguan formations in the Yuanba gas field in Sichuan Basin. In general, all gases in the Yuanba gas field belong to dry gas and have very high dryness coefficient, indicating the high thermal maturity. Gases from Changxing-Feixianguan formations have relatively higher dryness coefficient values (nearly 1) than those from Xujiahe and Leikoupo formations, however, the difference is usually not more than 0.02. Therefore, it is hard to tell the exact contribution of thermal maturity or TSR to the high dryness coefficient of gases from Changxing-Feixianguan formations.

Logarithmic Linear Correlation Between $\delta^{13}C_1$ and Ro%

Due to the kinetic isotopic fractionation effect during the hydrocarbon generation process, a logarithmic linear relation

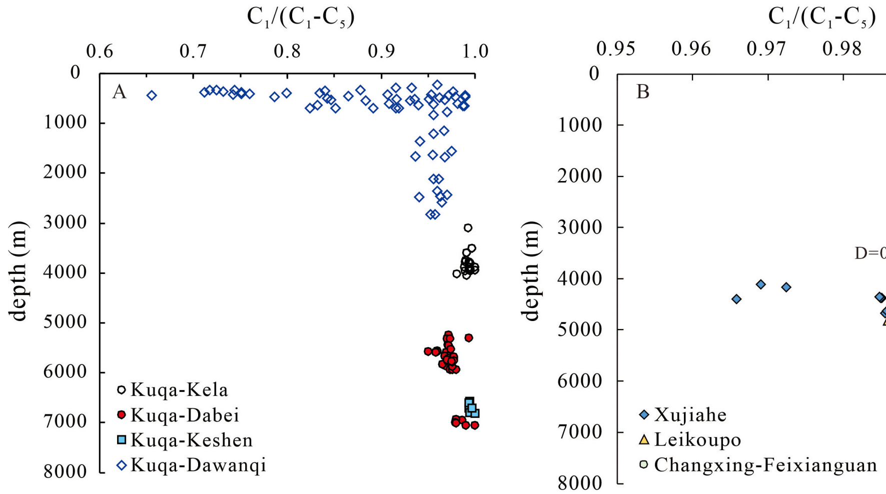

FIGURE 11 | (A) Plot of $C_1/(C_1$-$C_5)$ vs. burial depth of natural gases from the Dabei, Keshen, Kela2 and Dawanqi gas fields in the Kuqa Depression, Tarim Basin. **(B)** Plot of $C_1/(C_1$-$C_5)$ vs. burial depth of natural gases from the Xujiahe, Leikoupo and Changxing-Feixianguan formations of Yuanba gas field in Sichuan Basin. Data of Kela2 gas field were from Qin et al. (2007), data of Dawanqi oil field were from Zhou and Bao (2011) and Wang et al. (2012), data of Yuanba gas field were from Dai et al. (2013), Yin et al. (2013), Huang (2014), Wu et al. (2015b) and Li et al. (2016).

exists between $\delta^{13}C_1$ and Ro% (Stahl and Carey, 1975; Schoell, 1980). A ^{12}C-^{12}C bond is easier to break than a ^{12}C-^{13}C bond, which makes the residual precursors and late gaseous products more enriched in ^{13}C (Berner and Faber, 1996; Cramer et al., 1998; Tang et al., 2000). Different $\delta^{13}C_1$–Ro% models have been developed for gases derived from different types of organic matters (Stahl and Carey, 1975; Schoell, 1980; Dai and Qi, 1989). So based on the relation between carbon isotope of methane and the thermal maturity of gas source rocks, one can obtain the thermal maturity level of gas source rocks by using the measured $\delta^{13}C$ values of methane. Recently new equations of $\delta^{13}C_1$–Ro% [$\delta^{13}C_1 = 25\lg(Ro) - 37.5$ for gases sourced from the humic organic matter; $\delta^{13}C_1 = 25\lg(Ro) - 42.5$ for gases sourced from the sapropelic organic matter] have been established for primary gases which have not undergone secondary alteration according to abundant measured carbon isotopic values of methane in China (Chen et al., 2020). According to the $\delta^{13}C_1$–Ro% equation for gases from humic organic matter, deep gas from the Kuqa depression was calculated based on its carbon isotopic values of methane. The gas maturity is estimated to be 1.50~2.75% with an average of 2.06%. This is consistent with the local geological background. In the Kuqa depression, thermal maturity is higher in the east than that in the west, and in the deposition centre such as Baicheng sag, thermal maturity of source rocks (Ro%) is 1.5~3.5% (Dai, 2016). The gas dryness coefficient of 0.974 also implies the relatively high thermal maturation.

According to the calculation with the above $\delta^{13}C_1$–Ro% equations, deep gases from the Yuanba, Longgang and Puguang gas fields in the Sichuan Basin also have high gas maturity around 2.0% Ro%. However, methane in these gas fields might be altered by secondary processes such as TSR, water dissolution, resulting in the changes of $\delta^{13}C$ values of methane. Therefore, errors may exist for the gas maturity calculation based on the $\delta^{13}C$ values of methane for these gases.

HYDROGEN ISOTOPES

Carbon isotope of natural gas is mainly affected by the type of organic matter and the level of thermal maturity of source rocks. However, unlike the carbon isotope, the gas hydrogen isotope is highly dependent on the thermal maturity of source rocks and salinity of the water medium of the formation environment of the source organic matters (Schoell, 1980; Dai, 1993; Ni et al., 2015; Wang et al., 2015). Influences from the type of organic matter on the gas hydrogen isotope is weak, which will be easily masked by the influences from the water salinity of the depositional environment of the source organic matter (Ni et al., 2019).

In the Tarim Basin, a boundary δ^2H_1 value of −150‰ has been found between the gases sourced from the Middle-Lower Jurassic humic organic matter in the Kuqa depression (<−150‰) and the oil-cracked gases sourced from Cambrian-Lower Ordovician marine sapropelic organic matter in the Lunnan low uplift and the Ordovician oil-cracked gases in the Tazhong low uplift (>−150‰) (Ni et al., 2013). As shown in **Figure 12**, such boundary value also exists between the deep gases from the Dabei gas field in the Kuqa depression and those from the craton area of Tarim Basin. The deep gases from the Dabei gas field have δ^2H value of methane of −153~−158‰ with an average value of −156‰, while the deep gases from the craton area of Tarim Basin have relatively heavier hydrogen isotope of methane, with an average value of −130‰. Deep gases from the Yuanba and Puguang gas fields have even heavier δ^2H_1 value, with an average of −122‰. According to previous studies, bacterial methane sourced from the marine and saline-water lacustrine organic matter ($\delta^2H > -190$ or −200‰) has heavier hydrogen isotope compared to those from continental freshwater source rocks ($\delta^2H < -190$ or −200‰) (Schoell, 1980; Shen, 1995). Apparently deep gases from the Dabei gas field have δ^2H_1 value much heavier than

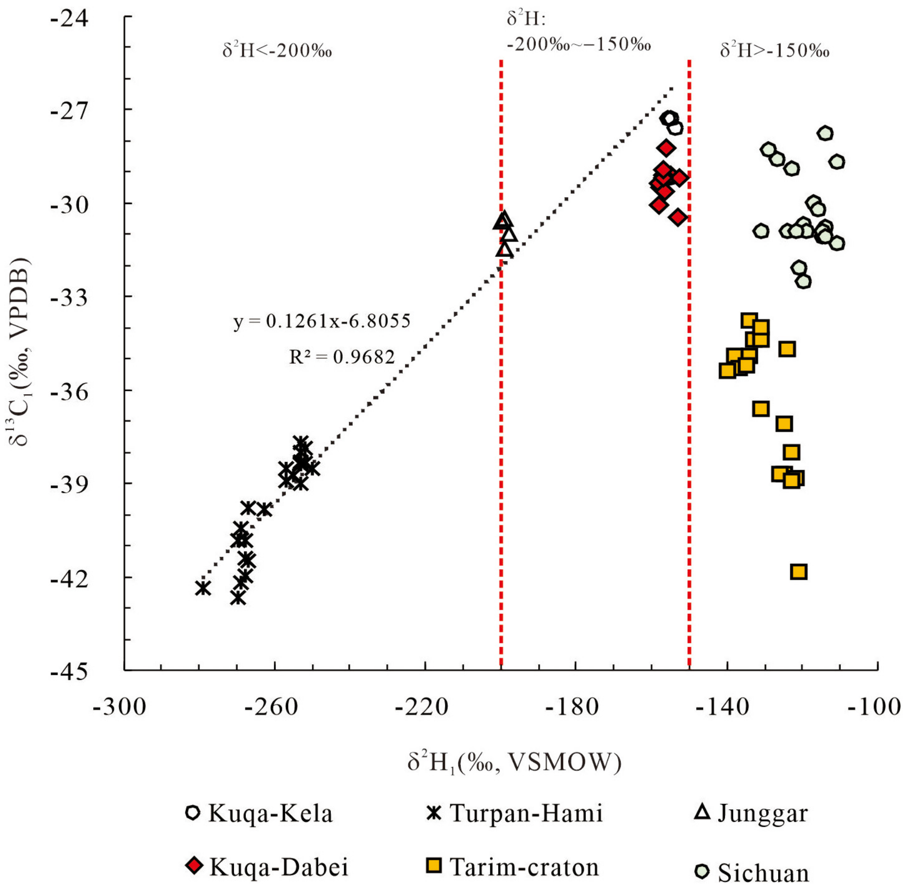

FIGURE 12 | Plot of $\delta^{13}C_1$ vs. δ^2H_1 of gases from Dabei and Kela2 gas fields in the Kuqa depression, craton area of Tarim Basin, Turpan-Harmi and Junggar basins in China. Gas data of the Dabei and craton area of Tarim Basin see section "Samples and Methods" in text, gas data of Kela2 gas field were from Qin et al. (2007), gas data of the Turpan-Hami basin were from Ni et al. (2015) and gas data of Junggar basin were from Wang et al. (2013).

this determining value for gases sourced from the continental freshwater organic matter.

The deuterium concentration in methane was found to increase with increasing thermal maturity (Schoell, 1980; Ni et al., 2011), methane in the Dabei gas field was expected to have relatively higher δ^2H values due to the high thermal maturity of their source rocks (Ro%: 1.5~3.5%). Because gas hydrogen isotope was mainly dependent on the thermal maturity and the water salinity of depositional environment of source organic matters, if one wants to investigate any influences from the thermal maturity or the water medium of depositional environment, a comparison has to be made with one fixed influencing factor. So gas samples with different thermal maturity but similar depositional environment from various locations in China were compared here (**Figure 12**).

Deep gases in the Dabei gas field are considered mainly sourced from the Middle-Lower Jurassic coal measures deposited in the lacustrine-swamp environment with freshwater medium (Zhang et al., 2011; Guo et al., 2016). Coal resources are widely distributed in the northwestern China and it is mainly accumulated in the Middle-Lower Jurassic formations. Actually in China the Early Middle Jurassic was one of the four major coal forming periods among the 14 coal forming periods in the geological history. The Middle-Lower Jurassic coal-bearing strata which was formed during the Early Middle coal forming period were widely distributed in the Tarim, Junggar, Turpan-Hami, Qaidam basins in northwestern China with similar depositional environment. Gases from the Turpan-Hami Basin (Ni et al., 2015), Junggar Basin (Wang et al., 2013) and Kela 2 gas field in the Kuqa depression in Tarim basin (Dai, 2016) are coal-derived gas sourced from the Jurassic coal measures. Under similar depositional environment, the differences in deuterium concentration of gases from Kela 2 gas field, Junggar and Turpan-Hami basins are mainly due to the differences in the level of thermal maturity of their source organic matters. There is a very good correlation between hydrogen and carbon isotopic compositions of methane ($y = 0.1261 \times -6.8055$, $R^2 = 0.9682$). This may indicate a possible influence of thermal maturity on gas isotopes since organic matter $^2H/^1H$ and $^{13}C/^{12}C$ ratios increase systematically throughout the thermal maturation (Schoell, 1980; Dai, 1993; Li et al., 2001).

Compared to the gases from Kela 2 gas field, Turpan-Hami and Junggar basins, considering the influences from thermal maturity of source rocks, the Dabei gases are relatively much more enriched in D at similar thermal maturity level and show remarkable offset toward the maturity trend (**Figure 12**). However, gases from these four areas demonstrate consistently quite good correlation between $\delta^{13}C_1$ and $\delta^{13}C_2$ (R^2 = 0.9007), δ^2H_1 and δ^2H_2 (R^2 = 0.9975) (**Figure 13**). This implies that the carbon isotope is affected by one type of factor while the hydrogen isotope is affected by another type of factor. Therefore, under one type of influencing factor, the correlation can be linear between methane and ethane for both carbon and hydrogen isotopes. If the carbon isotope was plotted vs. hydrogen isotope, then offset will occur (**Figure 12**).

Considering the heavy carbon isotopes of methane (average: –30.0‰) and ethane (average: –21.7‰), it is unlikely that the heavy hydrogen isotope is due to the mixing with oil-cracked gases. Gas derived from marine source rocks normally has much heavier hydrogen isotope due to the saline water depositional environment, for example, gases from the Lunnan and Tazhong areas in the Tarim basin are gases sourced from the Cambrian-Lower Ordovician marine sapropelic organic matter with saline water depositional environment, so they are more enriched in D but depleted in ^{13}C (**Figure 12**; Ni et al., 2013). Oil-cracked gas normally has much heavier carbon isotope of ethane compared to the oil-associated gas but normally still less than –28‰, and it also contains trace amount of ethane, i.e., the oil-cracked gas from the Sinian strata in the Weiyuan gas field (Dai et al., 2003). The carbon isotopic distribution pattern among methane and its homologues is normal in general and only a few samples have carbon isotopic reversal between ethane and propane of the deep gases from Dabei gas field (**Table 1**). To some extent, mixing of gases with different origins often causes a partial reversed carbon isotopic distribution pattern among methane and its homologues (Dai et al., 2004b). However, mixing with gases from sapropelic organic matter from deeper strata is unlikely. Though potential mixing with gases from the marine sapropelic organic matter will cause the enrichment of D, but it will also cause the depletion of ^{13}C, especially for ethane, which is not consistent with the heavy carbon isotope of the deep gases in the Dabei gas field.

Except the thermal maturity, one other potential cause for the relatively heavy hydrogen isotope is the saline depositional environment. The Kela 2 gases are on the maturity trend, while the gases from Dabei gas field have a big offset (**Figure 12**), which indicates that differences exist in the depositional environment of their source rocks. Based on the studies of paleontological fossil, mineralogy and petrology, and geochemistry (biomarkers, trace elements and carbon/hydrogen isotopes), a number of studies have demonstrated that marine transgressive happened several times during the Triassic-Jurassic period in the Kuqa depression (Chen, 1995; Zhou et al., 1999; Zhang B. et al., 2006). For example, based on the research of paleontological fossils and trace elements, Zhang B. et al. (2006) proposed that there were three big transgressive during the Triassic-Jurassic period in the Kuqa depression. According to the research based on stromatolite and trace elements, Chen (1995) thought that marine transgressive existed during the Middle Jurassic in the Tarim Basin. The author calculated the paleosalinity to be 33.4‰ according to the boron content, which is very similar to the modern seawater salinity. Extensive transgressive-regressive cycles during the Jurassic have been reported in the worldwide since Jurassic sea-level curves were first proposed several decades ago (Hallam, 1978, 2001). Major episodes of eustatic rise occurred in the Early Hettangian, Early Sinemurian, Early Pliensbachian, Early Toarcian, Early and Late Bajocian, Middle Callovian and Late Oxfordian to Kimmeridgian, among them, the Early Bajocian transgessive/deepening event can be recognised widely across the world (Hallam, 2001). Therefore, the relatively heavy hydrogen isotope in the Dabei gases may result from a possible saline depositional environment. Compared to the gases from Kela 2 gas field, the deep gases from Dabei gas field are more inclined to the influences from the transgressive, which caused the apparent enrichment in D in the deep gases in the Dabei gas field.

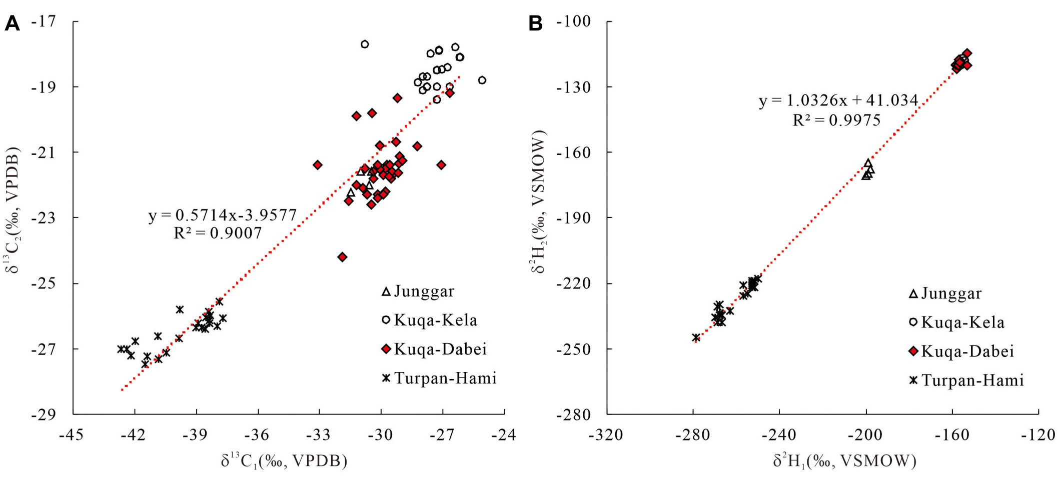

FIGURE 13 | Plots of $\delta^{13}C_1$ vs. $\delta^{13}C_2$ **(A)** and δ^2H_1 vs. δ^2H_2 **(B)** of gases from Dabei and Kela2 gas fields in the Kuqa depression in Tarim Basin, also from the Turpan-Harmi and Junggar basins in China. Gas data of Kela2 gas field were from Qin et al. (2007), gas data of the Turpan-Hami Basin were from Ni et al. (2015) and gas data of Junggar Basin were from Wang et al. (2013).

TABLE 1 | Molecular composition and stable carbon and hydrogen isotopic values of deep gas from Dabei gas field, Kuqa depression, Tarim basin.

Well	Strata	Depth (m)	Main composition (%)								$\delta^{13}C$ (‰,VPDB)					δ^2H (‰, VSMOW)		C_1/C_{1+}	Ro%[a]	References
			CH_4	C_2H_6	C_3H_8	C_4H_{10}	C_5H_{12}	N_2	CO_2	H_2S	CH_4	C_2H_6	C_3H_8	C_4H_{10}	CO_2	CH_4	C_2H_6			
DB1	K_1bs	5,552–5,586	94.50	3.45	0.22	0.28	0.05	1.08	0.42									0.959		Li et al., 2005
Dabei-1	K	5568.0–5620	91.87			4.84					−29.7	−21.4	−20.8					0.950	2.05	Zhang et al., 2011
DB1	E	5568.08–5,620									−29.7	−21.4	−20.8	−21.9	−18.0					Li et al., 2005
Dabei-1	K	5,576–5,586	95.04			3.96					−29.8	−21.5	−21.8					0.960	2.03	Zhang et al., 2011
Dabei-1	K	5,576–5,586	93.57			3.90					−29.9	−21.7	−21.8					0.960	2.01	Zhang et al., 2011
DB1	K_1bs	5,584–5,586	95.28	3.56	0.31	0.20	0.06	0.22	0.37									0.958		Li et al., 2005
DB1	K_1bs	5,586	94.80	2.30	0.36	0.35		1.12	0.44		−33.1	−21.4						0.969	1.50	Zhu et al., 2019a
DB1	K_1bs	5,865–5,872	94.13	3.00	0.03			2.85										0.969		Li et al., 2005
DB101-1	K_1bs	5,248–5,384 5,379–5,384	96.43	2.19	0.39	0.19	0.08	0.38	0.35	0.00	−30.1	−21.5	−21.3			−158	−121	0.971	1.98	This study
DB101-1	K_1bs	5,730	96.10	2.24	0.37	0.31		0.33	0.54		−31.6	−22.5	−21.7	−22.6				0.971	1.72	Zhu et al., 2019a
DB101-2	K_1bs	5,314–5,435,	96.14	2.25	0.41	0.19	0.07	0.46	0.47	0.00	−29.5	−21.8	−20.8			−158	−122	0.970	2.09	This study
DB101-3	K_1bs	5,302–5,380	99.34	0.64	0.02	0.00	0.00			0.00	−30.5	−19.8	−25.7			−153	−115	0.993	1.91	This study
DB101-5	K_1bs	5,438–5,540	96.54	2.19	0.41	0.20	0.09	0.57		0.00	−29.4	−21.7	−20.5			−158	−120	0.971	2.11	This study
DB101-5	K_1bs	5,744	95.10	2.11	0.45	0.36		0.92	0.62		−29.3	−20.7	−22.1	−20.6				0.970	2.13	Zhu et al., 2019a
DB101	K_1bs	5,725	95.70	2.22	0.40	0.29		1.20	0.22		−30.5	−22.6	−21.8	−22.8	−18.3			0.970	1.91	Dai, 2016
DB102	K_1bs	5,315	96.01	2.08	0.38	0.18		0.91	0.44		−29.5	−21.6	−21.0	−22.6	−17.4			0.973	2.09	Wei et al., 2019
DB102	K_1bs	5,451–5,479	96.24	2.17	0.39	0.19	0.07	0.52	0.42	0.00	−29.6	−21.7				−157	−120	0.972	2.06	This study
DB102	K_1bs	5,451	95.30	2.22	0.28	0.25		1.29	0.71		−30.7	−22.3	−21.5	−25.8	−3.6			0.972	1.87	Dai, 2016
Dabei-103	K	5,677–5,687	94.30			2.92					−31.9	−24.2	−19.7					0.970	1.67	Zhang et al., 2011
DB103	K_1bs	5,677	95.67	2.21	0.43	0.21		0.95	0.53		−30.2	−22.3	−21.1	−22.5	−18.1			0.971	1.96	Wei et al., 2019
DB103	K_1bs	5,832	94.30	2.45	0.51	0.45		1.27	0.99		−31.9	−24.2	−19.7		−8.5			0.965	1.67	Dai, 2016
DB104	K_1bs	5,922–5949									−27.1	−21.4							2.61	Dai, 2016
DB104	K_1bs	5,981–5,985									−26.7	−19.2							2.70	Dai, 2016
DB2	K_1bs	5,658–5,669	95.26	2.25	0.38	0.53		1.19	0.39		−30.8	−21.5	−19.8					0.968	1.85	Dai, 2016
DB2	K	5,658–5669.5	96.11	2.18	0.38	0.19	0.14	0.62	0.38		−30.8	−21.5	−19.8		−9.5			0.971	1.85	Li et al., 2005
DB201	K_1bs	5,658	95.26	2.25	0.38	0.53		1.27	0.31		−30.8	−21.5	−19.8		−19.0			0.968	1.85	Wei et al., 2019
DB201	K_1bs	5,932–6,010	96.10	1.98	0.37	0.31		0.64	0.54		−31.2	−22.0	−21.0					0.973	1.79	Dai, 2016
DB201	K_1bs	5,932–6,112	95.80	1.90	0.27	0.18		1.42	0.44		−30.9	−22.1	−21.9					0.976	1.84	Dai, 2016
Dabei-201	K	5932.45–6,145	96.10			1.96					−31.2	−19.9	−22.0					0.980	1.79	Zhang et al., 2011
DB201-1	K_1bs	5,876–5,976	97.13	1.89	0.33	0.16	0.06	0.43		0.00	−29.1	−21.1	−20.3			−155	−120	0.976	2.16	This study
DB202	K_1bs	5,711–5,845	97.59	1.84	0.33	0.17	0.07	0.01		0.00	−29.2	−21.3	−20.5			−157	−119	0.976	2.16	This study
DB202	K_1bs	5,711–5,845	94.80	1.57	0.36	0.30		1.27	1.67		−30.4	−21.8	−22.5					0.977	1.92	Dai, 2016
DB202	K_1bs	5,711	95.10	1.93	0.45	0.36		0.92	1.19		−29.3	-20.7	−22.1	−19.6	−16.4			0.972	2.13	Wei et al., 2019
DB202	K_1bs	5,763	94.80	2.16	0.36	0.30		1.27	0.45		−30.4	−21.6	−22.5	−21.4				0.971	1.92	Zhu et al., 2019a
DB205	K_1bs	5787.5–5,941	97.10	1.89	0.33	0.16	0.06	0.46		0.00	−29.2	−21.6	−21.0			−153	−120	0.975	2.15	This study

(Continued)

TABLE 1 | Continued

Well	Strata	Depth (m)	Main composition (%)								$\delta^{13}C$ (‰,VPDB)					$\delta^{2}H$ (‰, VSMOW)		C_1/C_{1+}	Ro%[a]	References
			CH_4	C_2H_6	C_3H_8	C_4H_{10}	C_5H_{12}	N_2	CO_2	H_2S	CH_4	C_2H_6	C_3H_8	C_4H_{10}	CO_2	CH_4	C_2H_6			
DB207	K_1bs	5,527	95.80	2.20	0.17	0.17		0.64	0.51		−29.8	−22.2						0.974	2.03	Zhu et al., 2019a
DB207	K_1bs	5,746–5,888	96.62	2.08	0.47	0.27	0.14	0.43		0.00	−29.2	−19.4	−20.6			−157	−118	0.970	2.14	This study
DB208	K_1bs	5,681	96.50	2.22	0.01	0.01		0.51	0.59		−30.2	−21.4	−21.6					0.977	1.96	Zhu et al., 2019a
DB208	K_1bs	5,755–5,830	96.80	1.73	0.30	0.15	0.07	0.44	0.51	0.00	−29.0	−21.3	−20.8			−157	−120	0.977	2.19	This study
DB209	K_1bs	5,776–5,878	97.09	1.91	0.33	0.16	0.07	0.44		0.00	−28.3	−20.8	−20.1			−156	−119	0.975	2.34	This study
DB3	$E_{1-2}km$	6,950	97.20	1.23	0.09	0.03					−29.9	−22.3	−20.3		−14.7			0.986	2.01	Wei et al., 2019
DB3	$E_{1-2}km$	7,058	96.70					0.71	1.08		−29.6	−21.4	−19.2	−20.1	−14.5			1.000	2.07	Wei et al., 2019
DB3	K_1bs	7,058–7,090									−29.6	−21.4	−19.2						2.07	Dai, 2016
Dabei-3	E	7,058–7090.88	96.90		0.98						−30.1	−20.8	−18.9					0.990	1.98	Zhang et al., 2011
DB301	K_1bs	6,930–7,012	96.20	1.66	0.17	0.17		1.26	0.51		−29.9	−22.3						0.980	2.01	Dai, 2016
DB301	$E_{1-2}km$	7,000	92.50	1.66	0.24	0.06					−30.1	−20.8	−20.3	−20.3	−21.7			0.979	1.98	Wei et al., 2019
DB301	$E_{1-2}km$	7,012	96.20	1.69	0.17	0.17		1.26	0.51		−30.2	−22.4	−22.1	−21.2	−31.0			0.979	1.96	Wei et al., 2019

[a]*Ro% values were calculated according to the formula by Chen et al. (2020), $\delta^{13}C_1 = 25lg(Ro)-37.5$.*

CONCLUSION

With increasing improvements in technological development and theoretical understanding, deep and ultra-deep areas have become more and more important, especially in the mature exploration area. A comprehensive knowledge about the geochemical characteristics of deep gas from continental sandstone reservoirs and marine carbonate reservoirs may improve our understanding of the origin and accumulation of deep gas. This study carried out a detailed comparison of the molecular composition, stable carbon and hydrogen isotopes of deep gases from the clastic sandstone reservoirs in the Dabei and Keshen gas fields in the Kuqa depression, Tarim basin, and deep gases from the marine carbonate reservoirs in the craton area of Tarim Basin and Sichuan Basin (Yuanba, Longgang, Puguang gas fields). The following conclusions were obtained.

(1) Deep gas, either from the continental sandstone reservoirs of Dabei and Keshen gas fields in the Kuqa depression, Tarim Basin or from the marine carbonate reservoirs of the craton area of Tarim Basin and Puguang, Yuanba and Longgang gas fields in the Sichuan Basin, is dominated by alkane gas. The average content of methane is about 83.80%. Deep gas from the Kuqa depression in Tarim Basin has gas dryness coefficient of 0.976, while gas from the Sichuan Basin has even higher gas dryness coefficient of 0.999, indicative of dry gas. Deep gas from the craton area of Tarim Basin has various gas dryness coefficient, mainly 0.594∼0.999, indicating the existence of both dry gas and wet gas. N_2 and CO_2 are the common non-hydrocarbon gases in the deep gas. One important difference between the deep gas from sandstone reservoirs and that from carbonate reservoirs is the content of H_2S. Deep gas from the continental sandstone reservoirs have no H_2S, while deep gases from the marine carbonate reservoirs often have H_2S, which can be even higher than 45%.

(2) Deep gas from the continental sandstone reservoirs in Dabei and Keshen gas fields in the Kuqa depression has $\delta^{13}C_2$ values much higher than −28‰, and has $\delta^{13}C_1 \sim \delta^{13}C_2$ maturity trend similar to the gases generated from type III kerogen, indicative of coal-derived origin. Most gas samples from the craton area of Tarim Basin have $\delta^{13}C_2$ values lower than −28‰ and match better with the evolution trend of gas generated from type II kerogen. Deep gases from the Sichuan Basin include two types: gases generated from humic type III kerogen, and gases generated from sapropelic type II kerogen. Carbon isotopic anomaly such as relatively heavy carbon isotope of methane or/and ethane and carbon isotopic reversal is common in the deep gas. Carbon isotopic reversal between ethane and propane exists in the deep gas from Kuqa depression, and carbon isotopic reversal between methane and ethane is often found in the deep gas from Sichuan Basin. Secondary alteration such as mixing, TSR, diffusion, high temperature may have important contribution to such carbon isotopic anomaly.

(3) A boundary δ^2H_1 value of $-150‰$ is found between the deep gases from the continental sandstone reservoirs in the Kuqa depression ($<-150‰$) and the deep gases from the marine carbonate reservoirs in the craton area of Tarim Basin and Sichuan Basin ($>-150‰$). The deep gas from Kuqa depression has average δ^2H value of methane of –156‰, while the deep gas from the craton area of Tarim Basin has average δ^2H value of methane of $-130‰$ and the deep gas from Yuanba and Puguang gas fields has even heavier δ^2H_1 value of $-122‰$. The extremely high hydrogen isotope of methane in Dabei gas field is due to the high thermal maturity and possible saline depositional environment of the source rocks.

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/s.

AUTHOR CONTRIBUTIONS

YN: manuscript writing. LY, FL, and JC: manuscript discussion. CY: experimental analyses. GZ: assistance in sample collection. All authors contributed to the article and approved the submitted version.

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Conflict of Interest: All authors are affiliated with the company institution Research Institute of Petroleum Exploration and Development, PetroChina.