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Geochemical Characteristics and Gas Source Contributions of Noble Gases of the Sulige Large Tight Gas Field of Upper Paleozoic in Ordos Basin, China

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Tight gas is the fastest developing unconventional natural gas resource, becoming the principal part for gas reserves and production growth in China. The Sulige gas field is the largest gas field and also the typical low porosity and low permeability tight sandstone gas field discovered in China, with an annual natural gas output exceeding 300 billion and cumulative output exceeding 290 billion, playing an important role in ensuring national energy provision, helping China's energy transformation, and promoting green, low-carbon, environmental protection and high-quality development. Based on sample collection and laboratory analysis, natural gas compositions including hydrocarbons, non-hydrocarbons, light hydrocarbons, and noble gases of the Sulige gas field are systematically analyzed, their genetic identifications are identified, and finally gas source originations and contribution proportions are comprehensively discussed from the perspectives of noble gases and hydrocarbon gases. The main achievements are as follows: 1) natural gases in the Sulige gas field are mainly alkane gases, with high methane content, high drying coefficient, low heavy hydrocarbon contents, low non-hydrocarbon gas contents of CO_2 and N_2, and relatively low noble gas contents. The helium content is relative 2 order of magnitude higher than the atmospheric value, while neon, argon, krypton, and xenon are relatively about 1–2 orders of magnitudes lower than the atmospheric values. 2) The carbon and hydrogen isotopes of alkanes are generally positive sequence distributions with some part inversion. The $^3He/^4He$ values are mainly distributed in magnitude of 10^{-8}, the $^{40}Ar/^{36}Ar$ is ranged from 506 to 1940, the ^{129}Xe is relative loss, and the ^{132}Xe is relative surplus. 3) Natural gases in the Sulige gas field are typical coal-formed gases generated from a humic organic mother material with maturity from high mature to over mature according to C_7 and C_8 light hydrocarbons and alkane carbon isotopes. Noble gases are typical crustal genesis, mainly originating from the radioactive decay of crustal source materials. 4) The gas source correlations of noble gases and alkane gases and their quantitative evaluations on source contributions show that natural gases in the Sulige gas field are originated from Carboniferous-Permian coal measure source rocks in Ordos Basin, mainly contributed by coals and supplemented by mudstones, accounting for 55–60% and 40–45%, respectively.

Keywords: Ordos basin, Sulige gas field, tight gas, noble gases, C_7 and C_8 light hydrocarbon, genetic identification, gas source correlation

1 INTRODUCTION

Tight gas is a natural gas stored in overburden matrix permeability less than or equal to 0.1 md (air permeability less than 1 md) in tight sandstone formations. As an important unconventional gas resource, tight gas has played an important role in China's natural gas development and been in a leading position in the process of unconventional gas development in China, becoming a major growth point for increasing natural gas reserves and production. Accelerating the exploration and development of tight gas is strategically important for the development of natural gas and the adjustment of the energy structure in China. At present, tight gas in China is widely distributed in Sichuan, Ordos, Tuha, Songliao, and Junggar basins, and mainly accumulated in Ordos and Sichuan basins. As early as 1971, China discovered the Zhongba tight gas field in the western part of Sichuan Basin. Since the mid-1990s, great breakthroughs were made in the exploration of Upper Paleozoic gas in Ordos Basin, and a large number of tight gas fields such as Yulin, Sulige, Wushengqi, Mizhi, Shenmu, and Zizhou were discovered successively. The Sulige gas field is the largest onshore mono-block natural gas field and also the largest unconventional tight gas field in China. The proven (including basic proven) gas reserves is $4.77\times10^{12}\,m^3$ (Fu et al., 2019; Jia et al., 2021), the gas production is more than $300\times10^8\,m^3$ in 2021 and the cumulative gas production is more than $2{,}900\times10^8\,m^3$. It plays an important role in ensuring national energy provision security, helping China's energy transformation and promoting green, low-carbon and high-quality development. Noble gases are considered as valuable tracers for the studies of geological processes, having great effects on gas origins and migration, crustal–mantle interaction, geotectonic etc. (Xu et al., 1979, Xu et al., 1998; Allegre et al., 1983, 1993; Ballentine et al., 1992; Battani et al., 2000; Burnard et al., 2013; Clarke et al., 1976; Du, 1989; Honda et al., 1991, 1993; Hiyagon et al., 1992; Kennedy et al., 1990; Lee et al., 2006; Liu and Xu, 1987, 1993; Lupton, 1983; Lupton and Evans, 2004; Mamyrin et al., 1970, 1984; Mark et al., 2011; Ozima and Podesek, 1983; Poreda and Farley, 1992; Poreda et al., 1986; Prinzhofer, 2013; Sano et al., 2008; Sarda et al., 1988; Shen et al., 1995; Sun et al., 1991; Wang, 1989; Wang et al., 2013, Wang et al., 2015, Wang et al., 2016, Wang et al., 2018; Wei et al., 2014; Welhan and Craig, 1983; Welhan et al., 1983; Xu et al., 1996; Xu et al., 1996, 1997). A lot of researches have been done on the geochemical characteristics, genesis, and sources of natural gas in the Sulige gas field (Dai et al., 1997; Fu et al., 2000; He et al., 2003; Dai et al., 2005; Feng et al., 2007; Hu et al., 2007; Liu et al., 2007; Hu et al., 2008; Lin et al., 2009; Dai et al., 2012; Hu et al., 2012; Li et al., 2012; Fu et al., 2013; Liu et al., 2013; Wang et al., 2013; Yu et al., 2013; Dai et al., 2013; Dai, 2014; Dai, 2014; Yang et al., 2014; Dai et al., 2016; Jia et al., 2018; Fu et al., 2019; Guo et al., 2020; Dai et al., 2021; Jia et al., 2021), but relatively little work has been done on the genesis and sources of natural gases by using the full components and isotopes of noble gases and C_8 light hydrocarbons. Hence, the authors intend to systematically analyze the hydrocarbon, non-hydrocarbon, noble gas components, and isotopic characteristics of natural gases in the Sulige gas field based on a large amount of alkane gases, non-hydrocarbon gases, noble gases, and light hydrocarbons, to comprehensively identify the natural gas genesis in the largest tight gas field from the perspectives of alkane gases, noble gases, and C_8 light hydrocarbons, and finally to explore the natural gas sources and their contribution proportions in the Sulige tight sandstone gas field from various perspectives, such as comparison of noble gases and conventional hydrocarbon gases. This study aims to deepen the understandings of gas genesis and sources in the Sulige tight sandstone gas field, providing references for tight sandstone gas field exploration in China.

2 REGIONAL GEOLOGICAL BACKGROUND

The Ordos Basin is a multi-rotation evolutionary craton basin with stable subsidence and depressional migration, covering an area of about $37\times10^4\,km^2$, which is ranked the second largest oil and gas-bearing basin in China after Tarim Basin (the distribution area of Paleozoic sedimentary rocks is about $25\times10^4\,km^2$). In general, it has the characteristics of active surrounding structures, developed faults, stable internal structures, high in the West and low in the East, and gently inclined to the West. Tectonically, the Ordos Basin can be divided into six primary tectonic units (**Figure 1**), namely, Yimeng Uplift, Weibei Uplift, Western Shanxi Flexural Fold Belt, Yishan Slope, Tianhuan Depression, and the Western Margin Thrust Belt, and several secondary tectonic units. On the basis of Archean and Lower Proterozoic metamorphic rock basement, the basin has undergone five stages of tectonic evolution: Middle and Late Proterozoic Aula Valley, Early Paleozoic Shallow sea platform, Late Paleozoic Coastal Plain, Mesozoic Inland Basin, and Cenozoic Peripheral Fault Depression (Compilation group of petroleum geology of Changqing Oilfield, 1992; Yang and Pei, 1996; Kang et al., 2000; Liu et al., 2000; Yang et al., 2000; He et al., 2003; Yang et al., 2006; Hu, 2009).

The Ordos Basin is mostly developed from bottom to top with the Middle and Upper Paleozoic Changcheng and Jixian; Lower Paleozoic Cambrian and Ordovician; Upper Paleozoic Carboniferous Benxi Formation; Permian Taiyuan Formation; Shanxi Formation; Shihezi Formation and Shiqianfeng Formation; Mesozoic Triassic, Jurassic, and Cretaceous; Cenozoic Tertiary and Quaternary; missing Upper Ordovician; and Silurian and Devonian, and the average thickness of sedimentary rocks reaches 6000 m (Hu, 2009). Marine carbonate deposits are well developed in Lower Paleozoic, and fluvial and lacustrine clastic deposits are well developed in Upper Paleozoic and Mesozoic. The Shanxi Formation of Lower Permian is divided into Shan 2 and

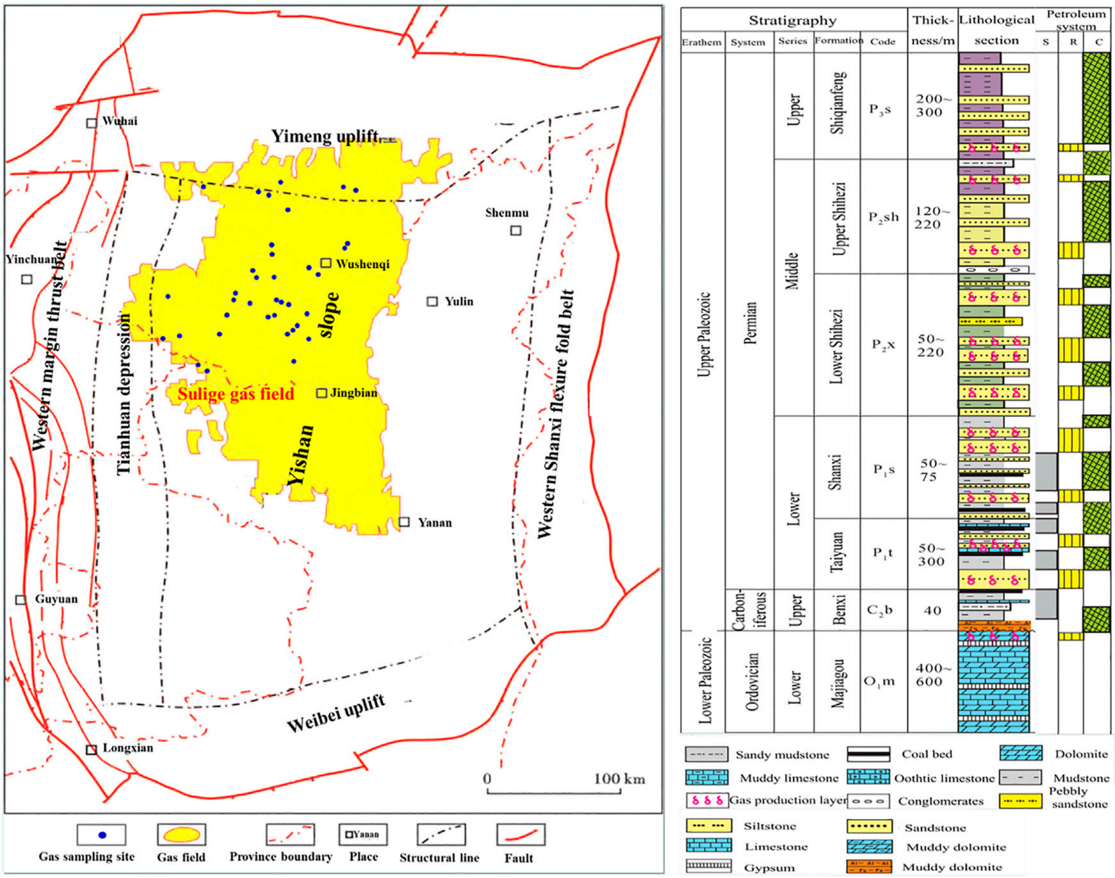

FIGURE 1 | Geological unit classification and the Sulige gas field distribution in Ordos Basin (Dai, 2014; Fu et al., 2019, Modifed).

Shan 1 members from bottom to top. The Shan 2 member which is a set of deltaic coal-formed clastic sediments, with lithology mostly of clastic sandstones or quartz sandstones, interspersed with thin layer of siltstones, mudstones, and coal seam, is one of the main gas layers in Upper Paleozoic. The Lower Shihezi Formation of Middle Permian is a set of stable distribution of fluvial–deltaic sediments and can be divided into He 8, He 7, He 6, and He 5 members. The He 8 member which is mostly composed of quartz sandstones and mudstones with unequal thickness alternate layer is one of the main gas layers in Upper Paleozoic. The Carboniferous-Permian coal measures hydrocarbon source rocks of Upper Paleozoic, with a thickness of 60–100 m, high organic matter content, high thermal evolution from high to over mature stage, and strong generation capacity are widely covered in the whole basin. The early Cretaceous is the main gas generation period for the Carboniferous-Permian coal measures hydrocarbon source rocks. The source rocks are in the high over mature stage and enter the peak of gas generation, which provides sufficient gas source for the formation of Paleozoic gas reservoir. The vertical superposition relationship between Permian-Carboniferous coal measures hydrocarbon source rocks and main reservoir such as He 8 and Shan 1 members of Permian are favorable for vertical migration and accumulation of natural gases generated in the peak of hydrocarbon generation of the source rocks. On the one hand, driven by the residual pressure of the source reservoir, the natural gases generated from the underlying Carboniferous-Permian coal measure source rocks accumulate and dissipate to the overlying Permian reservoir with the main mechanism of volumetric flow Darcy or non-Darcy seepage migration. On the other hand, due to the differences of hydrocarbon concentration caused by hydrocarbon generation, the natural gases generated in the source rock continuously diffuses to the overlying low porosity and permeability tight sandstone reservoir driven by concentration gradient (Wang et al., 2014; Wang et al., 2015). In addition, Upper Shihezi Formation has a stable distribution of fluvial and lacustrine mudstones with a thickness of about 60–120 m and constitutes the regional cover of Upper Paleozoic gas reservoir. The overlying mudstones and updip directional dense sandstones provide good direct covering and lateral sealing abilities (Yang and Pei, 1996; Kang et al., 2000; Yang et al., 2006; Hu, 2009).

The Sulige gas field is located in the north central part of Ordos Basin, tectonically in the northwestern part of the Yishan Slope, straddling three regional tectonic units: Yishan Slope, Yimeng Uplift, and Tianhuan Depression. Its administrative area is located in the Inner Mongolia Autonomous Region and the Shaanxi Province, consisting of four blocks: Central, West, East, and South. The Paleozoic strata of the Sulige gas field are lack of middle and Upper Ordovician, Silurian, Devonian, and Lower Carboniferous. The Majiagou Formation of Lower Ordovician, Benxi Formation of Upper Carboniferous, Taiyuan Formation and Shanxi Formation of Lower Permian, Lower Shihezi Formation of Middle Permian, and Upper Shihezi Formation and Shiqianfeng Formation of Upper Permian are developed from bottom to top. The Sulige gas field is mostly developed in Upper Paleozoic, and its main reservoir is the He 8 member of Middle Permian Lower Shihezi Formation of Middle Permian and the Shan 1 member of Shanxi formation of Lower Permian, with a depth of 3,200–3,500 m and a thickness of 80–100 m, followed by Shan 2 member of Shanxi Formation of Lower Permian and Ma 5 member of Majiagou Formation of Lower Ordovician. The Sulige gas field is generally characterized by a large gas-bearing area, many formations, thin single layer thickness, poor reservoir properties, strong non-homogeneity, generally low porosity and permeability, low pressure coefficient, low reserve abundance, and low production. It is a typically large distributed dense sandstone gas reservoir with low permeability, low pressure, low abundance, and river sand body as the main reservoir. In 2000, the Su6 well obtained a high production rate of 120×10^4 m^3 per day in Shihezi Formation, which kicked off the large-scale exploration of the Sulige gas field and marked a major discovery (Dai, 2014). Since 2007, the Sulige gas field has insisted on the integration of exploration and development, and the scale of new gas reserves exceeded $5{,}000\times10^8$ m^3 per year for seven consecutive years (Yang and Liu, 2014). At present, the exploration area of the Sulige gas field is about 5.5×10^4 km^2, with total gas resources of 6×10^{12} m^3, proven gas-bearing area is 3.88×10^4 km^2, accumulated tertiary reserves reaches 4.77×10^{12} m^3, and the natural gas production reaches 2.7×10^{10} m^3 in 2020, which becomes a key area for increasing natural gas storage and production in the Changqing Oilfield and an important natural gas gathering area in China (Fu et al., 2019; Jia et al., 2018; Jia et al., 2021). It makes a significant contributions on guaranteeing a safe and stable gas supply in the Capital, North China, and areas around the field.

3 SAMPLES AND ANALYTICAL METHODS

The noble gas content in natural gas is low but relatively high in air (especially argon content is as high as 0.934%), so it can easily be contaminated, which can lead to experimental analysis data to severely deviate from the real value. In order to minimize the contamination of noble gases in air, special measures are required for sampling natural gases for noble gases analysis (Wang et al., 2013; Wang et al., 2016; Wang et al., 2018):① choose a double valve high pressure–resistant steel cylinder; ② evacuate a sampling steel cylinder to below 10^{-1} Pa using a mechanical pump; ③ repeatedly flush the steel cylinder with natural gas for 4–6 times during wellhead sampling and intercept the middle section of gas flow. For the aforementioned noble gas analysis and testing requirements, more than 50 gas samples were collected from the Sulige gas field in Ordos Basin using high-pressure cylinder sampling, involving Permian Taiyuan Formation, Shanxi Formation, Shihezi Formation, and Shiqianfeng Formation.

The sample collection tests were completed at the Key Laboratory of Gas Formation and Development of China National Petroleum Corporation (CNPC).

(1) The natural gas component analysis was performed using an Agilent 7890A gas chromatograph, separating components by using a PLOT Al_2O_3 capillary column with a specification of 50 m × 0.20 mm × 0.5 μm. The sample inlet temperature was 300°C, carrier gas was helium, flow rate was 1 ml/min, splitting ratio was 50:1, and the temperature program was set to keep 10 min from 30°C, then the temperature was increased to 260°C at the heating rate of 3°C/min.
(2) Noble gas components and isotopes were analyzed using a noble gas sampling system and a Noblesse isotope mass spectrometer. The high vacuum of the system was achieved by mechanical, molecular, and ion pumps, and the sample volume of natural gas samples was controlled by an injection control system; a zirconium-based furnace and a suction pump were used to purify hydrocarbon gas, nitrogen (N_2), oxygen (O_2), carbon dioxide (CO_2), hydrogen sulfide (H_2S), trace hydrogen (H_2), and other active gases in natural gas samples; enrich noble gases; measure the content of noble gas components; and further separate components according to different boiling points of noble gases. The noble gas isotope composition was determined based on the ion flow signal intensity peak height ratio method by sending it to the Noblesse noble gas isotope mass spectrometer for isotope analysis. The relative deviations of the noble gases helium, neon, argon, krypton, and xenon were ±3.36%, ±3.66%, ±1.32%, ±2.99%, and ±6.96%, respectively, and the relative deviations of the noble gases $^3He/^4He$, $^{20}Ne/^{22}Ne$, $^{21}Ne/^{22}Ne$, $^{40}Ar/^{36}Ar$, $^{38}Ar/^{36}Ar$, $^{129}Xe/^{130}Xe$, and $^{132}Xe/^{130}Xe$ isotopes were ±4.50%, ±1.32%, ±1.27%, ±1.39%, ±1.63%, ±1.84%, and ±2.13%, respectively.
(3) A natural gas carbon isotope analysis was performed using the Finnigan MAT Delta PLUS GC/C/IRMS isotope mass spectrometer. The gas was separated by gas chromatography, converted to CO_2, and then fed into IRMS for carbon isotope determination. The chromatographic column was PLOT Q (30 m × 0.32 mm × 20 mm). The temperature rise procedure of chromatographic column was heating up from 35°C to 80°C at the rate of 8°C/min, then to 260°C at the rate of 5°C/min, and finally, kept at a constant temperature for 10 min. Each sample was analyzed three times, and the analytical standard was VPDB with an analytical accuracy of ±0.5%.
(4) The chromatographic separation column was HP PLOT Q (30 m × 0.32 mm × 20 mm). The carrier gas was helium, and the temperature rise procedure of methane column was heating up from 35°C to 80°C at the rate of 8°C/min, then to 260°C at the rate of 5°C/min, and finally, kept at a constant temperature for 10 min. Each sample was analyzed three times, and the analytical standard was SMOW with an analytical accuracy of ±0.5%.

TABLE 1 | Data table of hydrocarbon, non-hydrocarbon, and noble gas components in some natural gas samples from the Sulige gas field in Ordos Basin.

Gas field	Well	Strata	CH_4 (%)	CO_2 (%)	N_2 (%)	H_2S (%)	C_2 (%)	C_3 (%)	C_1/C_{1+}	He (10^{-6})	Ne (10^{-6})	Ar (10^{-6})	Kr (10^{-6})	Xe (10^{-8})	References
Sulige	S47-44-37	P	89.07	2.76	6.72	0	1.12	0.19	0.984	679.3	7.68	80.3	0.0411	1.0567	This study
	S48-5-84	P	93.01	1.22	1.48	0	3.06	0.43	0.956	540.7	6.17	50.8	0.0211	0.6728	
	S53	P	83.77	1.12	0.78	0	7.93	2.45	0.854	563.3	6.59	62.4	0.0455	0.6714	
	S10-42-48	P	91.76	1.04	1.04	0	4.57	0.86	0.937	679.8	7.74	69.0	0.0278	0.9198	
	S20-16-16	P	91.4	0.74	1.05	0	4.85	0.95	0.931	371.9	4.69	43.5	0.0161	0.4743	
	S20-15-14	P	91.47	0.71	1.04	0	4.94	0.98	0.931	395.2	5.01	48.6	0.0187	0.5868	
	S14-5-52	P	93.8	1.46	0.69	0	3.24	0.48	0.959	360.1	4.32	51.7	0.0203	0.4780	
	S14-4-47	P	91.98	1.01	0.94	0	4.41	0.84	0.938	345.3	4.45	38.3	0.0164	0.4924	
	S14-17-34	P	92.96	0.93	0.96	0	3.92	0.64	0.948	345.0	4.71	40.6	0.0163	0.5432	
	S14-16-42	P	93.6	0.89	0.85	0	3.5	0.64	0.953	331.2	4.27	73.1	0.0239	0.5347	
	T7-9-2	P	87.95	1.02	5.6	0	4.03	0.76	0.942	228.8	4.30	29.9	0.0148	0.4513	
	S11-18-36	P	89.93	1.56	0.92	0	5.48	1.14	0.922	688.6	7.49	69.1	0.0210	0.7117	
	Z61	P	89.86	0.99	0.96	0	5.84	1.29	0.916	762.5	7.84	56.4	0.0224	0.9563	
	S75-79-34H	P	92.33	1.62	1.16	0	3.7	0.62	0.950	719.7	7.24	71.5	0.0212	0.7850	
	S38	P	89.89	2.09	1.64	0	4.66	0.88	0.934	465.4	5.57	47.3	0.0158	0.5721	
	S75	P	90.19	1.59	0.94	0	5.42	1.06	0.925	691.5	7.20	81.4	0.0235	0.7887	
	S95	P	92.3	1.67	1.08	0	3.77	0.62	0.949	675.9	7.05	76.3	0.0213	0.7238	
	S55	P	90.41	0.62	0.88	0	6.2	1.14	0.918	515.7	5.66	46.4	0.0149	0.5510	
	S148	P	93.46	2.17	2.79	0	1.31	0.15	0.983	742.7	7.57	67.2	0.0204	0.7767	
	S5-15-8	P	90.97	0.86	0.92	0	5.31	1.09	0.926	417.2	4.99	41.7	0.0135	0.5173	
	S76	P	85.95	0.16	1.3	0	8.3	2.42	0.872	718.6	7.74	20.2	0.0558	1.2329	
	S54-24-65	P	93.32	2.23	1.43	0	2.43	0.32	0.969	681.3	6.92	102.3	0.0363	0.9511	
	S77-4-6	P	93.87	0.30	1.2	0	3.48	0.6	0.953	512.7	5.71	50.5	0.0166	0.6699	
	S120-79-69	P	91.71	2.97	2.72	0	1.84	0.29	0.972	674.6	6.94	108.8	0.0371	0.8943	
	S48-2-86	P	93.06	1.47	0.6	0	3.79	0.61	0.950	401.9	4.80	26.5	0.0103	0.4313	
	S109	P	91.93	2.29	3.16	0	1.91	0.42	0.972	666.8	7.20	213.4	0.0496	0.9983	
	S77-70-13	P	91.11	1.75	0.9	0	4.74	0.86	0.936	651.0	6.56	60.3	0.0176	0.5690	
Changshen and Xushen	XS 6	K	95.22	0.40	1.30	0	2.39	0.38	0.968	151	3.1291	48.102	0.0149	0.2783	Wang et al. (2013); Dai, (2014)
	CS 2	K	1.57	97.45	0.71	0	0.01	0	0.9936	195	4.144	74.027	0.0154	0.4418	
	CSP 2	K	68.16	25.65	4.7	0	1.12	0.05	0.9831	453	6.179	331.313	0.0404	0.8632	
Weiyuan	W 2	Z	85.07	4.66	8.33	1.31	0.11	0	0.9987	2500		530			Dai et al. (2003)
	W 5	P	94.28		3.36		0.21	0.01	0.9976	1080		480			
	W 23	C-Z	85.44	4.75	8.14	1.25	0.15	0	0.9982	2620		150			
	W 27	Z	85.85	4.7	7.81	1.20	0.17	0	0.9980	2180		480			
	W 28	Pt	67.03	1.23	26.7	0.01	0.21	0.03	0.9964	2480		2050			
	W 30	Z	86.57	4.40	7.55	0.95	0.14	0	0.9984	3,420		460			
	W 39	Z	86.74	4.53	7.08	1.22	0.12	0	0.9986	2730		710			
	W 46	Z	85.66	4.66	8.11	1.17	0.11	0	0.9987	2520		490			
	W 100	Z	86.80	5.07	6.47	1.18	0.13	0	0.9985	2980		460			
	W 106	Z	86.54	4.82	6.26	1.32	0.07	0	0.9992	3,150		430			

(5) A natural gas light hydrocarbon analysis was performed by using a gas chromatograph 7890A with helium as the carrier gas and the PLOT Al_2O_3 capillary column of 50 m × 0.20 mm × 0.5 μm for component separation, the inlet temperature was 120°C, and liquid nitrogen cold trap for light hydrocarbon enrichment for 5 min. FID detector temperature was 320°C, the injection volume was 10–15 ml, and the ramp-up program was initiated. The initial temperature of the rise procedure was 30°C, kept at constant temperature for 15 min, and then the rise procedure was 1.5°C/min to 70°C, 3°C/min to 160°C and 5°C/min to 300°C and kept at a constant temperature for 20 min, respectively.

4 RESULTS AND DISCUSSIONS

4.1 Distribution Characteristics of Alkane, Non-Hydrocarbon, and Noble Gas Components in the Sulige Gas Field

The alkane gas content of natural gas in the Sulige gas field occupies an absolute advantage. The highest methane content is mostly distributed in the range of 83.77%–93.87%, with an average of 91.15%. Ethane is mostly distributed in the range of 1.12%–8.30%, with an average of 4.21%. Propane is mostly distributed in the range of 0.15%–2.45%, with an average of

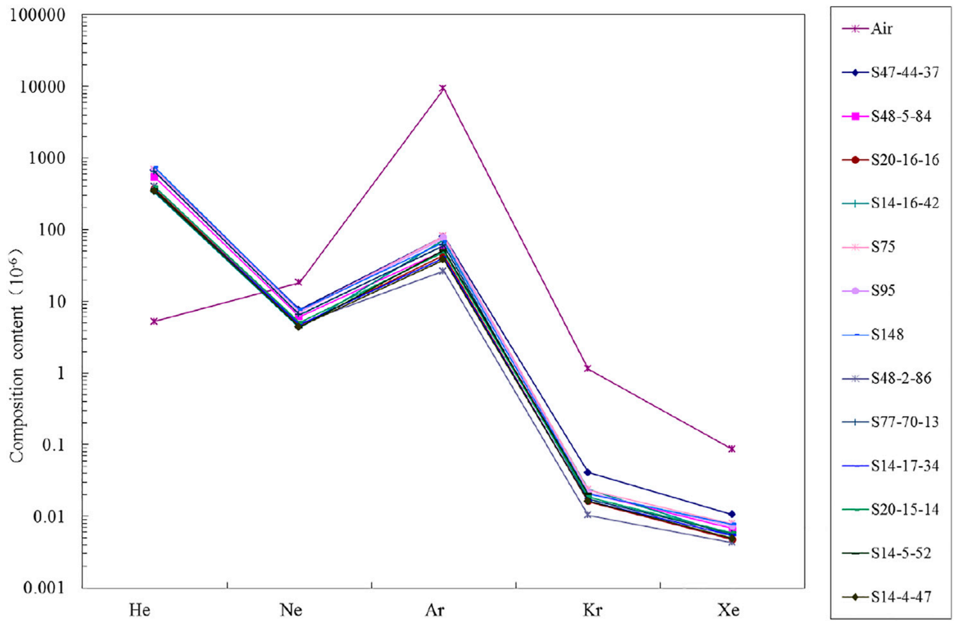

FIGURE 2 | Components abundance of noble gas He, Ne, Ar, Kr, and Xe of natural gases in the Sulige gas field in Ordos Basin.

0.84%. The drying factor (C_1/C_{1+}) is mostly distributed in the range of 0.854–0.984, with an average of 0.940 (**Table 1**). The non-hydrocarbon gas nitrogen (N_2) content was mostly distributed in the range of 0.60%–6.72%, with an average value of 1.62%, and carbon dioxide (CO_2) content was mostly distributed in the range of 0.30%–2.97%, with an average value of 1.38%, without H_2S.

Compared with hydrocarbon and non-hydrocarbon gases, the contents of noble gases in natural gas are relatively low. In general, the distribution of noble gas components in natural gases from the Sulige field in Ordos Basin has many characteristics (**Table 1**; **Figure 2**). 1) The helium content of natural gas samples is mostly distributed around (228.8–762.5)$\times10^{-6}$, with an average value of 549.1$\times10^{-6}$, which is about two orders of magnitude higher than the atmospheric value of 5.24$\times10^{-6}$ in general. 2) The neon content in natural gas samples is around (4.27–7.84) $\times10^{-6}$, with an average value of 6.16$\times10^{-6}$, which is about 1/3 of the atmospheric value of 18.18$\times10^{-6}$. 3) The argon content in natural gas samples is around (20.2–213.4)$\times10^{-6}$, with an average value of 64.0$\times10^{-6}$, which is about two orders of magnitude lower than the atmospheric value of 0.934%. 4) The krypton content in natural gas samples is around (0.0103–0.0558)$\times10^{-6}$, with an average value of 0.0244$\times10^{-6}$, which is about two orders of magnitude lower than the atmospheric content value of 1.14$\times10^{-6}$. 5) The xenon content in natural gas samples is around (0.0043–0.0123)$\times10^{-6}$, with an average value of 0.0070$\times10^{-6}$, which is about one order of magnitude lower than the atmospheric content value of 0.078$\times10^{-6}$. Compared with noble gases data of previous researchers on the Weiyuan gas field in Sichuan Basin and Changshen and Xushen gas fields in Songliao Basin (**Table 1**), the helium content of Sulige is obviously less than that of Weiyuan and relatively larger than Changshen and Xushen, while the argon content in Weiyuan is relatively larger than that of Sulige, Changshen, and Xushen.

4.2 Carbon Isotope Distribution Characteristics and Genesis Identifications of Alkane Gases and Carbon Dioxide in the Sulige Gas Field

The carbon isotope analysis data of alkane gas components in the Sulige gas field are shown in **Table 2**. The methane carbon isotope $\delta^{13}C_1$ values of natural gas samples are distributed in the range of −36.7‰ ~ −27.6‰, with main values ranging from −35‰ ~ −30‰ and an average value of −32.3‰. Ethane carbon isotope $\delta^{13}C_2$ values are distributed in the range of −27.9‰ ~ −22.4‰, with main values ranging from −26‰ ~ −23‰ and an average value of −24.1‰. Propane carbon isotope $\delta^{13}C_3$ values are distributed in the range of −28.9‰ ~ −21.9‰, with the main values ranging from −28‰ ~ −23‰ and an average value of −24.7‰. The *n*-butane carbon isotope δ^{13} nC_4 values are distributed in the range of −24.1‰ ~ −19.7‰, with main values ranging from −24‰ ~ −21‰ and an average value of −22.5‰. The isobutane carbon isotope δ^{13} iC_4 values are distributed in range of −24.2‰ ~ −20‰, with main values ranging from −23‰ ~ −20‰ and an average value of −21.6‰. The carbon dioxide isotope values of $\delta^{13}C_{CO2}$ in the Sulige gas field are mostly distributed in the range of −15.3‰ ~ −5‰, with an average value of about −10.3‰. For the hydrogen isotopes of alkane gases in the Sulige gas field, the δ^2H_1 values are generally distributed in the range of −203‰ ~ −171‰, with an average of −185.3‰; the δ^2H_2 values are usually distributed in the

TABLE 2 | Carbon isotope data of alkane gas components in some natural gas of the Sulige gas field.

Well	Strata	Hydrocarbon isotope value $\delta^{13}C$‰ (VPDB)					
		CO_2	C_1	C_2	C_3	iC_4	nC_4
S47-12-65	P_1s		−30.2	−24.8	−27.6		
S47-44-37	P_2x		−28.4	−24.7	−24.6		
S48-5-84	P_2x		−30.8	−22.8	−21.9	−20.2	−19.7
S53	P_1s-P_2x		−34.7	−24.5	−23.2	−23.3	−22.2
S20-16-16	P_1s-P_2x		−33.5	−23.8	−24.4	−21.7	−22.2
S20-15-14	P_1s-P_2x		−32.9	−23.2	−24.3	−21.5	−22.8
S14-5-52	P_1s		−32.5	−23.2	−26.0	−22.9	−24.1
S14-17-34	P_2x		−33.2	−24.3	−24.5	−21.9	−22.8
S14-16-42	P_2x		−33.1	−24.5	−24.7	−21.9	−22.7
T2-2-12	P_1s	−10.0	−33.0	−23.4	−24.3	−20.5	−22.8
T7-16-22X4	P_2x	−10.5	−34.0	−23.2	−23.7	−21.6	−22.3
S120-35-81C3	P_1s	−11.5	−32.9	−22.9	−23.3	−20.0	−21.4
T2-26-16	P_1s-P_2x	−11.5	−30.0	−24.4	−25.5	−20.6	−23.4
T2-26-2	P_2x	−10.4	−30.2	−23.0	−25.4	−21.1	−23.5
S47-17-61	P_2x	−8.4	−30.0	−27.9	−28.9		
S120-97-87	P_1s	−8.9	−27.6	−27.3	−27.4		
S47-36-43	P_1s	−5.0	−28.0	−24.2	−26.8		
S48-17-74	P_1s	−8.9	−30.3	−23.4	−24.5	−20.3	−22.0
S14-6-11H	P_2x	−8.6	−32.5	−23.8	−24.3	−21.2	−22.2
SD47-38	P_2x	−5.7	−34.8	−25.3	−25.6		
SD 47-35	P_1s	−10.4	−34.4	−24.2	−23.5	−21.1	−21.9
S5-9-8	P_1s	−11.5	−33.8	−22.4	−23.3	−21.6	−22.1
S63-09	P_2x	−13.7	−33.6	−23.8	−24.7	−22.0	−22.9
S66-1C1	P_1s	−10.6	−34.2	−23.7	−23.9	−21.5	−22.7
S77-32-37	P_1t	−15.3	−36.7	−26.5	−24.9	−24.2	−23.6
S36-16-15	P_1s	−8.1	−34.4	−23.5	−24.1	−21.9	−23.1
S36-16-12	P_2x	−11.8	−33.7	−24.5	−24.4	−21.9	−23.2
S54-35-113H2	P_2x	−13.6	−31.8	−23.6	−24.2	−21.3	−22.4
S54-22-80	P_2x	−11.9	−31.5	−23.2	−24.2	−21.9	−22.4
S11-17-35	P_1s	−10.1	−32.6	−23.7	−23.2	−21.8	−21.6

range of −168‰ ~ −151‰, with an average of −159.6‰; and the δ^2H_3 values are commonly distributed in the range of −165‰ ~ −150‰, with an average of −157.6‰ (**Table 3**).

The carbon isotopes of alkane gases of organic origin have the characteristic of increasing the $\delta^{13}C$ value with increasing carbon number ($\delta^{13}C_1<\delta^{13}C_2<\delta^{13}C_3<\delta^{13}C_4$), which is called a positive carbon isotope sequence of alkane gases; when the carbon isotope arrangement of alkane gases appears chaotic, it is called carbon isotope inversion; when $\delta^{13}C_i>\delta^{13}C_{i+1}$, it is called partial inversion of carbon isotope sequence; and when $\delta^{13}C_1>\delta^{13}C_2>\delta^{13}C_3>\delta^{13}C_4$ appears completely, it is called negative carbon isotope sequence of alkane gas (Dai, 2014). The carbon isotope sequence of alkane gases in Sulige gas samples generally show positive carbon isotope sequence of $\delta^{13}C_1<\delta^{13}C_2<\delta^{13}C_3<\delta^{13}C_4$, and some samples show $\delta^{13}C_2>\delta^{13}C_3$ inversion of carbon isotope sequence (**Figure 3A**).

Similarly, the hydrogen isotopes of alkane gas of organic origin also have the characteristics of increasing the δ^2H value with increasing carbon number ($\delta^2H_{CH4}<\delta^2H_{C2H6}<\delta^2H_{C3H8}$), which is called positive hydrogen isotope sequence of alkane gas; when the carbon isotope arrangement of alkane gas appears chaotic, it is called hydrogen isotope inversion; when $\delta^2H_{CiHi+2}>\delta^2H_{Ci+1\ H2i+4}$ appears, it is called partial inversion of hydrogen isotope sequence; and when $^2H_{CH4}>\delta^2H_{C2H6}>\delta^2H_{C3H8}$ appears completely, it is called negative carbon isotope sequence of alkane gas (Dai, 2014). The hydrogen isotopes of alkane in natural gas samples from the Sulige field generally show the characteristics of $\delta^2H_{CH4}<\delta^2H_{C2H6}<\delta^2H_{C3H8}$ positive hydrogen isotope sequence, and some of samples show $\delta^{13}C_2>\delta^{13}C_3$ inversion of alkane gas hydrogen isotopes (**Figure 3B**).

Stable carbon isotope values are the most reliable and commonly used methods for natural gas genesis, with methane isotope values generally ranging of −55‰ ~ −35‰ for oil-typed gas and −35‰ ~ −22‰ for coal-formed gas. Since methane carbon isotopes are strongly influenced by the degree of thermal evolution of source rocks, ethane isotopes are usually little influenced by the maturity of thermal evolution of source rocks and have good parent material inheritance effects, so the ethane carbon isotope value of $\delta^{13}C_2$ = −28.5‰ is generally used as the threshold for determining oil-typed gas and coal-formed gas (Dai et al., 1989; Dai, 1992, 2014). The $\delta^{13}C_2$ values of natural gas from the Sulige gas field in Ordos Basin are distributed in the range of −27.9‰ ~ −22.4‰, with main values ranging from −26‰ ~ −23‰ and an average value of −24.1‰. The natural gas $\delta^{13}C_2$ is significantly heavier than −28.5‰, which is significantly different from oil-typed gas, and can be identified as typical coal-formed gas based on ethane carbon isotopes. According to carbon isotopes of methane, ethane, and propane proposed by Dai (1993), the majority of the carbon isotope data points of the gas samples distribute in coal-formed gas area, and some of points fall in or near mixed carbon isotope inversion series area (**Figure 4**), indicating that natural gases from the Sulige gas field is generally coal-formed gas.

CO_2 is an important non-hydrocarbon gas component in natural gases. The CO_2 content in the Sulige gas field mostly distributes in the range of 0.30%–2.97%, and the average value is 1.38%. The $\delta^{13}C_{CO2}$ is mostly distributed in the range of −15.3‰ ~ −5‰, and the average value is −10.3‰. 1) When the CO_2 content is less than 15%, $\delta^{13}C_{CO2}$ < −10‰ is organic CO_2; 2) when $\delta^{13}C_{CO2} \geq$ −8‰, all are inorganic CO_2; and 3) when the CO_2 content is greater than 60%, all are inorganic CO_2 (Dai et al., 1989; Dai, 1992). According to the identification chart of different CO_2 genesis by Dai et al. (1992) (**Figure 5**), the CO_2 in the Sulige gas field is mostly organic in general with small part inorganic.

4.3 Noble Gases Isotope Distribution Characteristics and Genesis Identifications of the Sulige Gas Field

(1) The $^3He/^4He$ values (R) of He in natural gas samples from the Sulige gas field mostly distribute around (2.89–7.30)×10^{-8} (0.021–0.052Ra), with an average of 4.81×10^{-8} (0.034Ra). From the He-R/Ra genesis identifications of noble gases in natural gas, it can be found that the $^3He/^4He$ values of natural gas in the Sulige gas field generally distribute between 0.01<R/Ra<0.10, and the sample points all fall in a typical crust genesis area (**Table 4**; **Figure 6**). It indicates that noble gas helium in natural gas from the Sulige gas field is mostly of typical crustal genesis, and the proportion of crustal genesis helium is greater than 98.8%, originating from the decay of crustal genesis radioactive elements U and Th.

(2) From the noble gas Ne-$^{20}Ne/^{22}Ne$, $^{20}Ne/^{22}Ne$-$^{21}Ne/^{22}Ne$, and $^3He/^4He$-$^{20}Ne/^{22}Ne$ genesis identifications (**Figure 7**),

TABLE 3 | Hydrogen isotope data of alkane gas in some natural gas of the Sulige gas field.

Well	Strata	$\delta^{13}C$ (‰,VPDB)				$\delta^{2}H$ (‰, VSMOW)			References
		CH_4	C_2H_6	C_3H_8	C_4H_{10}	CH_4	C_2H_6	C_3H_8	
S11-18-36	P_2x	−33	−23.3	−22.3	−22.9	−180	−152	−152	Yu et al. (2013)
S120-42-84	P_1s-P_2x	−31.9	−23.6	−24.7	−22.7	−174	−152	−158	
S120-52-82	P_1s-P_2x	−31.1	−23.3	−25.6	−23.6	−176	−163	−164	
S139	P_1s-P_2x	−30.4	−24.2	−26.8	−23.7	−176	−165	−165	
S14-0-31	P_1s-P_2x	−32	−23.8	−24.7	−22	−180	−155	−157	
S14-11-09	P_2x	−31.6	−24	−24.2	−22.6	−172	−154	−158	
S336	P_1s-P_2x	−28.7	−22.6	−25.1	—	−173	−156	−153	
S48-14-76	P_1s-P_2x	−33.5	−22.8	−24.2	−22.2	−176	−159	−156	
S48-15-68	P_2x	−29.8	−23.4	−25	−22.6	−179	−157	−157	
S48-2-86	P_1s	−31.7	−23.2	−24.3	−22.3	−174	−159	−155	
S55	P_2x	−35.1	−24.6	−24.1	−24.8	−186	−151	−158	
S76-1-4	P_1s	−32.7	−23.6	−22.9	−23	−182	−155	−150	
S76-15-18	P_1s-P_2x	−35.7	−25.3	−24.8	−24.8	−189	−151	−152	
SN3-45	P_1s-P_2x	−31.3	−22.1	−22.8	−20.7	−172	−153	−154	
S48-13–79C3	P_1s-P_2x	−30.2	−22.9	−23.4	−21.9	−171	−158	−153	
S21	P_2x	−33.4	−23.4	−23.8	−22.7	−194	−167	−−163	Dai, 2014
S53	P_1s-P_2x	−35.6	−25.3	−23.7	−23.9	−202	−165	−160	
S75	P_1s-P_2x	−33.2	−23.8	−23.4	−22.4	−194	−−163	−157	
S95	P_1s-P_2x	−32.5	−23.9	−24	−22.7	−193	−167	−160	
S76	P_1s-P_2x	−35.1	−24.6	−24.4	−24.4	−203	−165	−161	
S53-78-46H	P_2x	−33.9	−23.9	−23	−23.2	−198	−165	−156	
S75-64-%X	P_1s-P_2x	−33.5	−24	−23.3	−22.8	−199	−167	−159	
S77-2-5	P_1s-P_2x	−30.8	−22.7	−23.3	−22.9	−−194	−168	−164	
S77-6-8	P_1s-P_2x	−33.6	−23.9	−24.1	−23.5	−201	−165	−165	
Z61	P_1s	−33.2	−23.5	−23.3	−23.2	−194	−159	−154	

it can be found that the $^{20}Ne/^{22}Ne$ values of natural gas samples from the Sulige gas field are mostly distributed in the range of 9.56–9.74, and $^{21}Ne/^{22}Ne$ values are mostly distributed in the range of 0.0295–0.0315. The ^{20}Ne is relatively deficient, while the ^{21}Ne and ^{22}Ne are relative excess. It indicates that neon in natural gas from the Sulige gas field is dominated by typical crustal genesis, which is a result of variable depositional environments of uranium, thorium, oxygen, and fluorine elements in crust. The $^{20}Ne/^{22}Ne$ ratios of Changshen and Xushen gas fields are larger than 9.8, reflecting the relative atmospheric excess of ^{20}Ne. It indicates that neon in natural gas from Changshen and Xushen fields has significant mantle genesis neon mixing characteristics.

(3) The $^{40}Ar/^{36}Ar$ values of natural gas samples from the Sulige gas field are relatively high and widely distributed, mostly in the range of 506–1940, with an average value of 1148, while the $^{38}Ar/^{36}Ar$ values are mostly distributed in range of 0.1709–0.1959, with an average value of 0.1909 (**Table 4**). According to the $^{3}He/^{4}He$–$^{40}Ar/^{36}Ar$ genesis identification chart, it can be found that the noble gas $^{3}He/^{4}He$ values in the Sulige gas field generally distribute around $(2.89–7.30)\times10^{-8}$, with an average value of 4.81×10^{-8}, and the $^{3}He/^{4}He$ and $^{40}Ar/^{36}Ar$ have an overall negative correlation growth. It shows that helium and argon in natural gases from the Sulige gas field have the characteristics of typical crustal origin genesis (**Figure 8**). In contrast to Yingcheng Formation in Changshen and Xushen gas fields, the $^{3}He/^{4}He$ is larger than 1.4×10^{-6} and the $^{3}He/^{4}He$ is positively correlated with $^{40}Ar/^{36}Ar$, indicating that the noble gases have significant mantle genesis mixing characteristics.

(4) The $^{129}Xe/^{130}Xe$ values of natural gas in the Sulige gas field are mostly distributed in the range of 6.362–6.479 with an average value of 6.436, while the $^{132}Xe/^{130}Xe$ values are mostly distributed in the range of 6.629–6.731 with an average value of 6.670 (**Table 4**). According to the noble gas $^{129}Xe/^{130}Xe$–$^{132}Xe/^{130}Xe$ genesis identification, it can found that the ^{129}Xe in the Sulige gas field is generally deficient relative to atmosphere and the ^{132}Xe is excess relative to atmosphere, and the gas sample points fall into a crustal genesis region (**Figure 9**), indicating that the noble gas xenon in natural gas is mostly of crustal genesis. While the ^{129}Xe in Changshen and Xushen gas fields in Songliao Basin is excess relative to the atmosphere, which has the characteristics of significant mantle genesis mixing.

4.4 Distribution Characteristics and Genesis Identifications of C_7 and C_8 Light Hydrocarbons

Light hydrocarbons are $C_{5–10}$ hydrocarbon compounds with a boiling point less than 200°C, including *n*-alkanes, isoalkanes, cycloalkanes, and aromatic compounds, which are very important components of natural gas and crude oil and contain rich geochemical information. The geochemistry of light hydrocarbons can be used to study the parent material types and genesis, depositional environments, maturities, and secondary migration of natural gases. Some researchers have established genetic identification indexes and charts for the composition of C_7 light hydrocarbon series (*n*-heptane, methylcyclohexane, and dimethylcyclopentane), $C_{5–7}$ aliphatic compositions (*n*-alkanes, isoalkanes, and cycloalkanes), light hydrocarbon monomer hydrocarbon carbon isotope ratios (benzene, toluene, cyclohexane, and methylcyclohexane), $C_{6–7}$ aromatic and branched alkane combinations, benzene and toluene content, methylcyclohexane, and aromatic alkane—paraffin—to identify coal-formed and oil-typed gas (Leythauser et al., 1979; Leythauser et al., 1980; Hu et al., 1990; Dai, 1992; Dai, 1993; Jiang et al., 1999; Li et al., 2001; Li et al., 2003; Hu et al., 2007; Hu et al., 2010; Hu et al., 2012). At present, the studies of light hydrocarbons in natural gases have become a hot spot in the field of oil and gas geology. $C_{5–7}$ and C_7 light hydrocarbon compositions have been widely used in oil and gas exploration and geological studies; however, few researches have been conducted on C_8 light hydrocarbon compounds. There is a lack of methods to identify the genesis and sources of coal-formed gas and oil-typed gas by using C_8 light hydrocarbon parameters. In this study, based on the researches of $C_{5–7}$ and C_7 light hydrocarbons, the distribution characteristics and genetic identifications of C_8 light hydrocarbons were carried out.

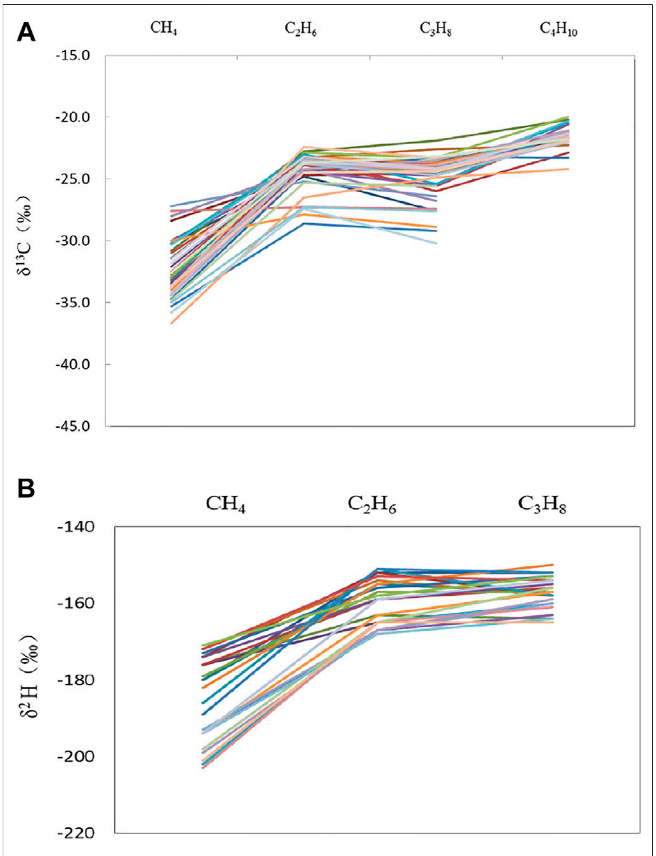

FIGURE 3 | Folding line of carbon **(A)** and hydrogen **(B)** isotope series of natural gas alkane from the Sulige gas field in Ordos Basin.

4.4.1 Distribution Characteristics and Genesis Identifications of C_7 Light Hydrocarbon Compounds

The C_7 light hydrocarbon compounds include *n*-heptane, methylcyclohexane, and dimethylcyclopentane. The *n*-heptane is mostly from algae and bacteria, which is a good maturity indicator. The methylcyclohexane is mostly from higher plant lignin and cellulose, which is a good parameter to reflect the type of terrestrial parent material. The dimethylcyclopentane is mostly from lipid-like compounds of aquatic organisms, which is a characteristic of oil-typed gas light hydrocarbons (Chung et al., 1998; Chung et al., 1991; Whiticar and Snowdon, 1999). Hu et al. (2007) analyzed 173 coal-formed gas samples in China and found that 92% of the gas samples had a methylcyclohexane volume fraction greater than 50%. Therefore, the dominant distribution of methylcyclohexane in a C_7 light hydrocarbon system is a major characteristic of coal-formed light hydrocarbons. According to the ternary diagram of relative content of the C_7 light hydrocarbon compounds system of natural gases from the Sulige gas field (**Figure 10A**), the methylcyclohexane is distributed in the range of 53.9%–77%, with an average of 66%; the *n*-heptane is distributed in the range of 9.6%–28.6%, with an average of 15%; the dimethylcyclopentane is distributed in the range of 11.5%–30%, with an average of 19%. All gas samples of the Sulige gas field is distributed in the lower left corner and show that methylcyclohexane content is dominant. So, the natural gases from the Sulige gas field are typical coal-formed gas mostly generated from humic parent material.

4.4.2 Distribution Characteristics and Genesis Identifications of $C_{5–7}$ Light Hydrocarbon Aliphatic Compositions

The ternary diagram of relative contents of $C_{5–7}$ aliphatic composition of *n*-alkanes, isoalkanes, and cycloalkanes is also commonly used to identify different types of natural gases. According to the ternary diagram of $C_{5–7}$ aliphatic composition of *n*-alkanes, isoalkanes, and cycloalkanes of natural gases from the Sulige gas field (**Figure 10B**), it can be seen that *n*-alkane content of Sulige natural gas samples in the range of 12.2%–25.9%, with an average of 19.2%; isoalkane content in the range of 21%–62.1%, with an average of 49%; and cycloalkane content in the range of 14.5%–68%, with an average of 31.8%. The $C_{5–7}$ aliphatic compositions show that natural gases from the Sulige gas field are rich in isoparaffins and cycloalkanes, indicating that natural gases from the Sulige gas field are typical coal-formed gases mostly originated from humic parent material.

4.4.3 Distribution Characteristics and Genesis Identifications of C_8 Light Hydrocarbon Aliphatic Compositions

Previous studies suggest that the light hydrocarbon fraction of humic parent material is rich in isoalkanes and aromatics (Leythaeuser et al., 1979; Leythaeuser et al., 1980), and the light hydrocarbon fraction of condensate from terrestrial parent material is rich in

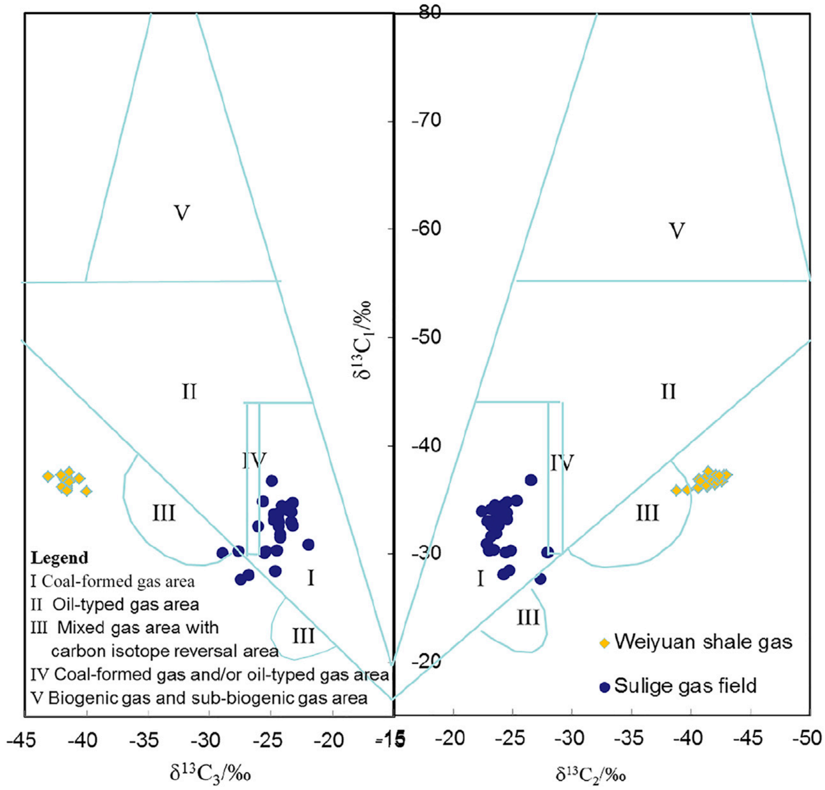

FIGURE 4 | Identification of $\delta^{13}C_1$-$\delta^{13}C_2$-$\delta^{13}C_3$ alkane gas genesis in the Sulige gas field, Ordos Basin (plate According to Dai, 1993; Modified).

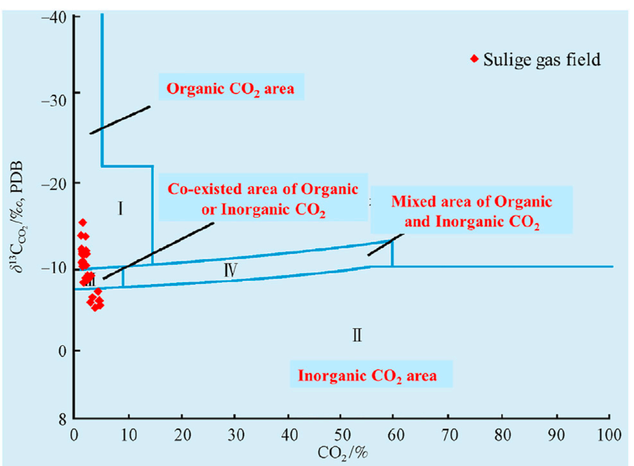

FIGURE 5 | Identification of organic and inorganic carbon dioxide in natural gases from the Sulige gas field (plate according to Dai, 1989).

cycloalkanes (Snowdon and Powell, 1982). Therefore, the relative content of C_8 aliphatic compositions of *n*-alkanes, isoalkanes, and cycloalkanes can also be used to identify natural gas genesis. In this study, based on a large number of experimental analysis and statistics of light hydrocarbons, the following indicators and ternary diagram were established for the identification of coal-formed gas and oil-typed gas by the relative contents of C_8 light hydrocarbon *n*-alkane, isoalkane, and cycloalkane compositions. 1) When $0 \le C_8$ *n*-alkane relative content $\le 20\%$, $20\% \le C_8$ cycloalkane relative content $\le 100\%$, and $0\% \le C_8$ isomeric alkane relative content $\le 70\%$, natural gas is generally coal-formed gas. 2) When $20\% \le C_8$ n-alkane relative content $\le 100\%$, $0 \le C_8$ cycloalkane relative content $\le 50\%$, and $20\% \le C_8$ isoalkane relative content $\le 60\%$, natural gas is generally oil-typed gas. According to the ternary diagram of C_8 aliphatic compositions for natural gases from the Sulige gas field (**Figure 11A**), *n*-alkane content of natural gas samples is in the range of 4%–18.2%, with an average of 7.9%; isoalkane content in the range of 10%–64.9%, with an average of 52.6%; and cycloalkane content in the range of 21.4%–86.3%, with an average of 39.5%. The C_8 aliphatic compositions show that natural gases from the Sulige gas field is rich in isoalkanes and cycloalkanes, indicating that natural gases from the Sulige gas field are typical coal-formed gases generated from humic parent material.

4.4.4 Distribution Characteristics and Genesis Identifications of C_8 Light Hydrocarbon Compounds

C_8 light hydrocarbon system compounds include cis-1,3-dimethylcyclohexane, *n*-octane, and 2-methylheptane. Dimethylcyclohexane is mostly derived from lignin and cellulose of higher plants, reflecting the characteristics of terrestrial parent material. While *n*-octane is mostly from algae and bacteria, 2-methylheptane is mostly from lipid-like compounds of aquatic

TABLE 4 | Some isotopic ratios of noble gases of natural gases in the Sulige gas field, Ordos Basin.

Gas field	Well	Strata	$^3He/^4He$ (10^{-8})	R/Ra	$^{20}Ne/^{22}Ne$	$^{21}Ne/^{22}Ne$	$^{40}Ar/^{36}Ar$	$^{38}Ar/^{36}Ar$	$^{129}Xe/^{130}Xe$	$^{132}Xe/^{130}Xe$
Sulige	S47-44-37	P	7.30	0.052	9.74	0.0295	610	0.1923		6.674
	S48-5-84	P	6.08	0.043	9.63	0.0309	1018	0.1959		6.639
	S53	P	7.12	0.051	9.66	0.0304	872	0.1908	6.460	6.694
	S10-42-48	P	4.55	0.032	9.68	0.0298	1093	0.1921	6.404	6.671
	S20-16-16	P	5.07	0.036	9.63		1190	0.1961		6.678
	S20-15-14	P	5.15	0.037	9.69	0.0301	1062	0.1935	6.362	6.696
	S14-5-52	P	4.37	0.031	9.56	0.0299	630	0.1919	6.437	6.642
	S14-4-47	P	4.48	0.032	9.64	0.0295	696	0.1904	6.404	6.660
	S14-17-34	P	5.52	0.039	9.62	0.0298	1163	0.1923	6.356	6.669
	S14-16-42	P	4.91	0.035			518	0.1890	6.469	6.652
	S11-18-36	P	6.74	0.048	9.63	0.0297	1197	0.1981	6.407	6.685
	Z61	P	4.48	0.032	9.67	0.0315	964	0.1918	6.462	6.638
	S75-79-34H	P	4.69	0.034	9.70		1786	0.1900	6.424	6.646
	S38	P	5.17	0.037			1391	0.1883	6.461	6.729
	S75	P	4.84	0.035			1030	0.1904	6.469	6.660
	S95	P	4.96	0.035			1195	0.1896	6.426	6.679
	S55	P	2.94	0.021			1289	0.1882	6.440	6.695
	S148	P	3.84	0.027			1940	0.1939	6.447	6.668
	S5-15-8	P	2.89	0.021			1327	0.1897	6.479	6.663
	S76	P	3.13	0.022			602	0.1836	6.422	6.649
	S54-24-65	P	4.59	0.033			1928	0.1926	6.477	6.642
	S77-4-6	P	3.06	0.022			1765	0.1923	6.431	6.629
	S120-79-69	P	5.58	0.040			1403	0.1959	6.417	6.634
	S48-2-86	P	3.72	0.027			1350	0.1918	6.464	6.731
	S109	P	5.28	0.038			506	0.1707	6.440	6.674
	S77-70-13	P	4.48	0.032			1316	0.1919	6.471	6.711
	T7-9-2	P	5.05	0.036						

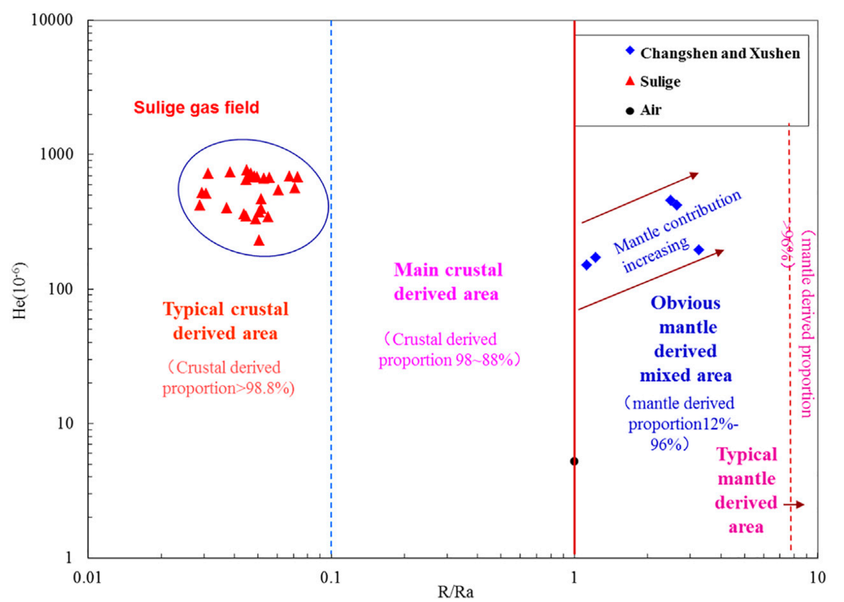

FIGURE 6 | Genesis identification chart of He and R/Ra of natural gases in the Sulige gas field, Ordos Basin (Plate according to Wang et al. 2013, Wang et al. 2016).

organisms, both reflecting the characteristics of oil-typed gas light hydrocarbons. Similarly, we also established a ternary diagram to identify coal-formed gas and oil-typed gas by the relative contents of cis-1,3-dimethylcyclohexane, n-octane, and 2-methylheptane of C_8 light hydrocarbon system compounds: 1) when 40% ≤ cis-1,3-dimethylcyclohexane relative content ≤ 100%, n-octane relative percentage content ≤ 40%, and 2-methylheptane relative percentage content ≤ 40%, natural gas is generally coal-formed gas. 2) When 0 ≤ cis-1,3-dimethylcyclohexane relative content ≤ 40%, 30% ≤ n-octane relative percentage ≤ 100%, and 0 ≤ 2-

methylheptane relative percentage ≤ 50%, natural gas is generally oil-typed gas. According to the ternary diagram of C_8 light hydrocarbon cis-1,3-dimethylcyclohexane, *n*-octane, and 2-methylheptane in the Sulige gas field, the relative contents of cis-1,3-dimethylcyclohexane, *n*-octane, and 2-methylheptane are 31.1%–92.2%, with an average of 56.6%; 2-methylheptane is 3.3%–36.3%, with an average of 20.1%; *n*-octane is 4.5%–46.9%, with an average of 23.3%; and *n*-octane is 4.5%–46.9%, with an average of 20.1%. The relative contents of 2-methylheptane is in the range of 3.3%–36.3%, with an average of 20.1%; the relative contents of *n*-octane is in the range of 4.5%–46.9%, with an average of 23.3%. Therefore, the aliphatic compositions of the C_8 light hydrocarbon compounds in the Sulige gas field is rich in cis-1,3-dimethylcyclohexane. The gas samples from the Upper Paleozoic tight gas field in the Sulige gas field fall into the coal-formed gas region (**Figure 11B**) and show typical coal-formed gas generated from humic parent material.

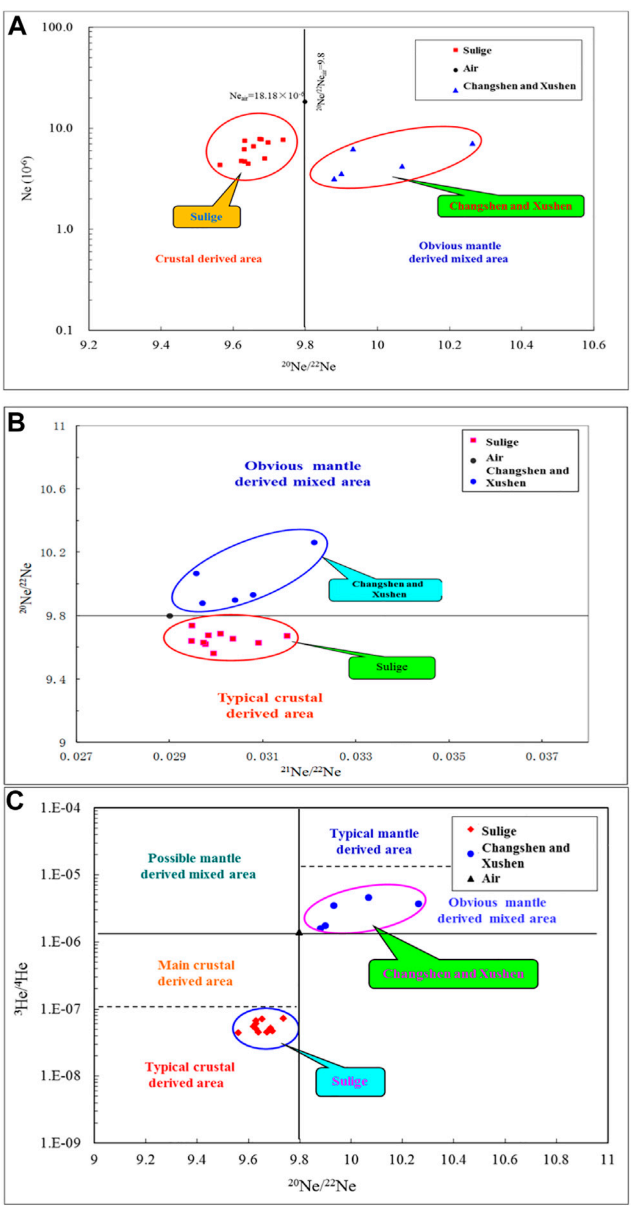

FIGURE 7 | Genesis identification chart of Ne and $^{20}Ne/^{22}Ne$ **(A)**, $^{20}Ne/^{22}Ne$ and $^{21}Ne/^{22}Ne$ **(B)**, and $^{3}He/^{4}He$ and $^{20}Ne/^{22}Ne$ **(C)** of natural gases in the Sulige gas field in Ordos Basin (Plate according to Wang et al. 2013, Wang et al. 2016).

5 GAS SOURCE CORRELATIONS OF THE SULIGE GAS FIELD IN ORDOS BASIN

5.1 Conventional Gas Source Correlations of the Sulige Gas Field

In total, two sets of source rocks are developed in Paleozoic of Ordos Basin: Carboniferous-Permian coal measure source rocks of Upper Paleozoic and marine carbonate source rocks of Lower Paleozoic. The Carboniferous-Permian coal measure source rocks are considered as the main source rocks in Ordos Basin (Chen et al., 1993; Dai et al., 1996; Dai et al., 2003; Dai et al., 2005). The coal measure source rocks of Carboniferous-Permian in Ordos Basin are widely distributed throughout the basin, mainly including coals and dark mudstones in Shanxi Formation, Taiyuan Formation, and Benxi Formation. The coals of Benxi, Taiyuan, and Shanxi Formations basically cover the whole basin and are main source rocks of Upper Paleozoic. The cumulative thickness of coals ranges from 4 to 40 m. Dark mudstones are developed in Benxi, Taiyuan, and Shanxi Formations, mainly in Taiyuan Formation and Shan2 member of Shanxi Formation, and widely distribute in upper Paleozoic with total thickness from 20 to 80 m. In Yan'an-Wuqi area in the southern part of Ordos Basin, the highest thermal evolution maturity of source rocks reach 2.8%, decreasing in a ring-like pattern toward north and south and basin margin. The thermal evolution maturity of most parts of the basin are above 1.5%, generally entering high mature to over mature stage.

According to the genetic identification, the natural gases of the Sulige gas field are typical coal-formed gas, mainly from coal measure source rocks of humic parent material. Combing the typical characteristics of existed Paleozoic source rocks in Ordos Basin, it is concluded that the natural gases of the Sulige gas field originate from Carboniferous-Permian coal measure source rocks. Furthermore, by calculating the natural gas maturity of the Sulige gas field, the calculated Ro values of natural gas maturity are mainly distributed from 1.0% to 2.0%, and the maturity of natural gas in southern part of the Sulige gas field is relative higher, while the maturity of natural gas in northern part is relative lower, having a similar trend with maturity of Carboniferous-Permian coal measure source rocks in Ordos Basin. It indicates that the natural gas in the Sulige gas field mainly originates from Carboniferous-Permian coal measure source rocks and also reflects that the natural gas in the Sulige gas field is mainly generated from near source rocks. The coal measure source rocks of Carboniferous-Permian in Ordos are characterized by widespread hydrocarbon generation, and the area with hydrocarbon generation intensity greater than $12{\times}10^8$ m^3/km^2 accounting for 71.6% of the total basin. The source rocks of the Sulige gas field and its nearby area have a

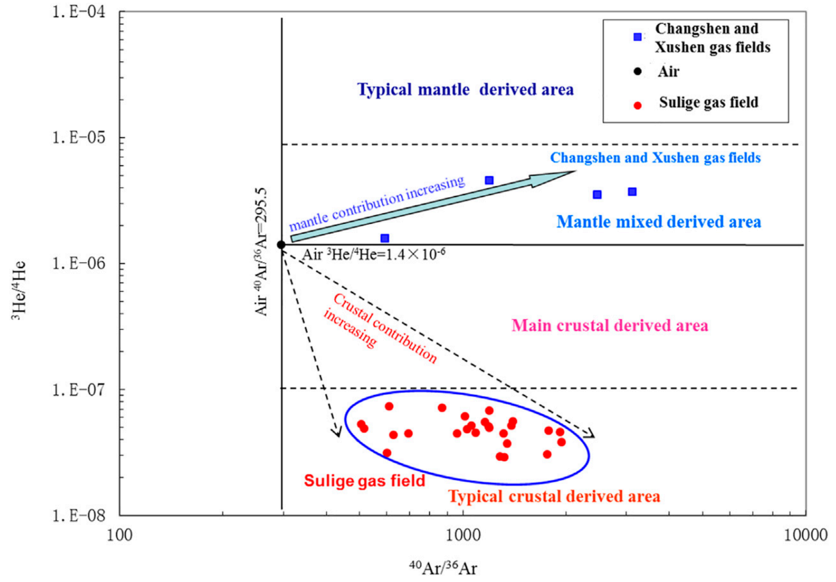

FIGURE 8 | Genesis identification chart of $^{3}He/^{4}He$ and $^{40}Ar/^{36}Ar$ of natural gases in the Sulige gas field in Ordos Basin (Plate according to Wang et al. 2013, Wang et al. 2016).

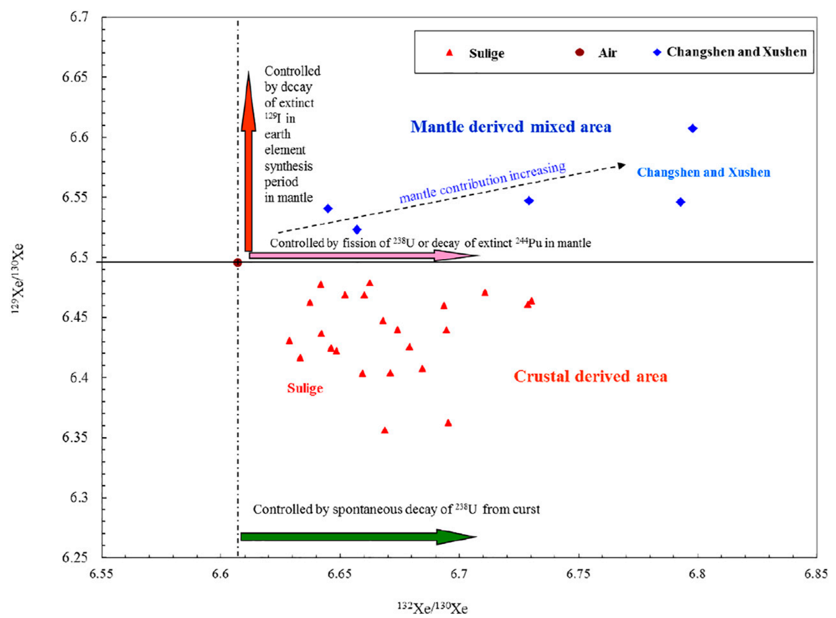

FIGURE 9 | Genesis identification chart of $^{129}Xe/^{130}Xe$ and $^{132}Xe/^{130}Xe$ of natural gases in the Sulige gas field in Ordos Basin (Plate according to Wang et al. 2013, Wang et al. 2016).

gas generation intensity of $(12–30)\times10^{8}$ m^3/km^2 (Li et al., 2012; Zhao et al., 2013a).

Therefore, the natural gases from the Sulige gas field are typical coal-formed gas in high mature to over mature stage, mainly generating from coal measure source rocks of Upper Paleozoic Carboniferous-Permian in Ordos Basin.

5.2 Noble Gas Sources Correlation of the Sulige Gas Field

Based on the noble gas isotope analysis, the gas source comparison study of Ordos Sulige gas was further conducted by using helium and argon isotopes and argon isotope age accumulation effect. The $^{3}He/^{4}He$ values (R) of

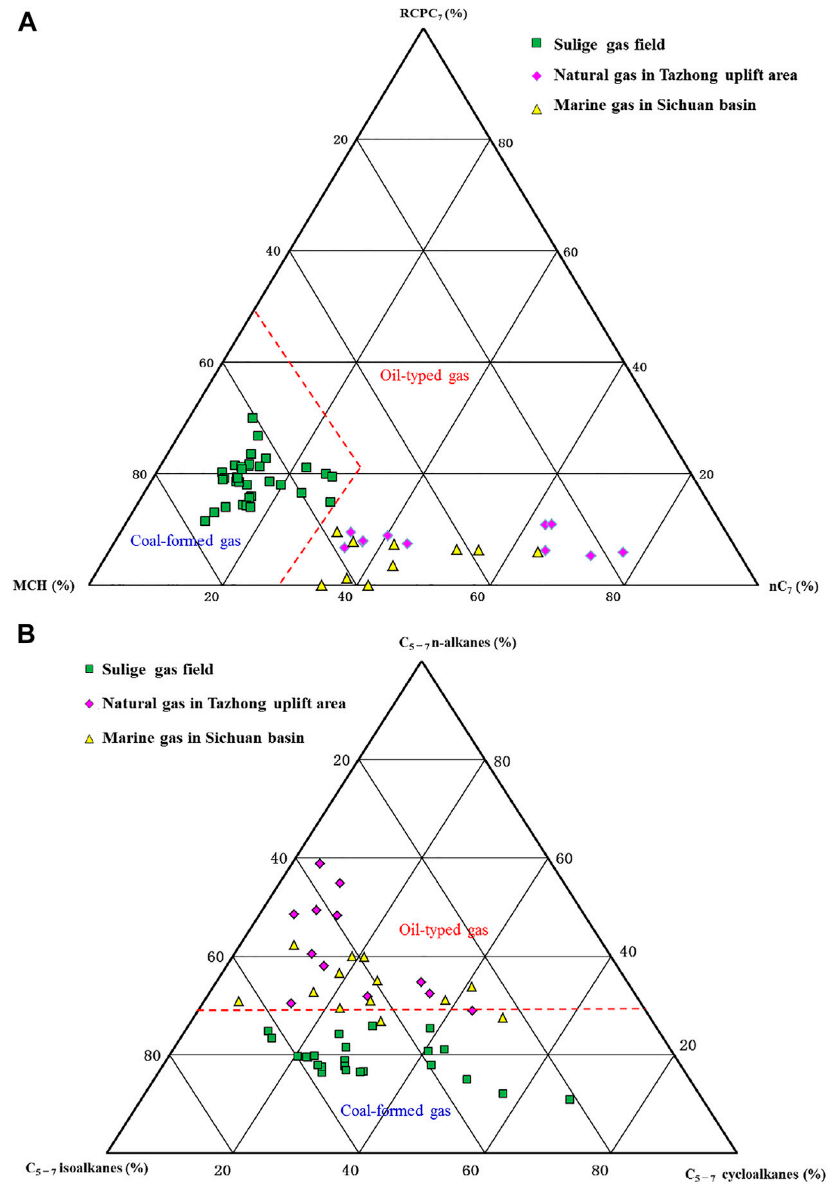

FIGURE 10 | Ternary diagram of the C_7 compositions **(A)** and C_{5-7} **(B)** light hydrocarbons system in the Sulige gas field (Plate according to Hu et al., 2007)

gas samples from the Sulige gas field mainly distribute around $(2.89–7.30)\times10^{-8}$ $(0.021\sim0.052R_a)$,with an average value of 4.81×10^{-8} $(0.034R_a)$, which is in the order of 10^{-8} with an overall distribution in $0.01<R/R_a<0.10$. It shows that noble gas helium in natural gas of the Sulige gas field is a typical crustal source genesis, and the proportion of crustal source contribution is more than 98.8%, mainly coming from decay of crustal source radioactive elements U and Th. Therefore, comparative gas source studies can be performed by using natural gas argon isotope age accumulation effect. The $^{40}Ar/^{36}Ar$ values of gas samples from the Sulige gas field are relatively high and widely distributed, mainly in the range of 506–1940, with an average value of 1148. The average value is in the range of 920–1450 for natural gas generated from Carboniferous-Permian source rocks (Shen et al., 1995). Furthermore, based on the relationship between argon isotopes of clastic strata and parent source stratigraphic age proposed by Xu et al. (1996), parent source stratigraphic age of natural gases in the Sulige gas field is estimated to be about 299 Ma, which is in the range of 250~355 Ma for Carboniferous-Permian coal measure source rocks in Upper Paleozoic of Ordos Basin. Therefore, the natural gases in the Sulige gas field mainly originate from Carboniferous-Permian coal measure source rocks of Upper Paleozoic. The comparative study of noble gas source correlations in the Sulige gas field by using age accumulation effect of argon isotopes of natural gas provides evidences for gas source correlation from a noble gas point of view, which is

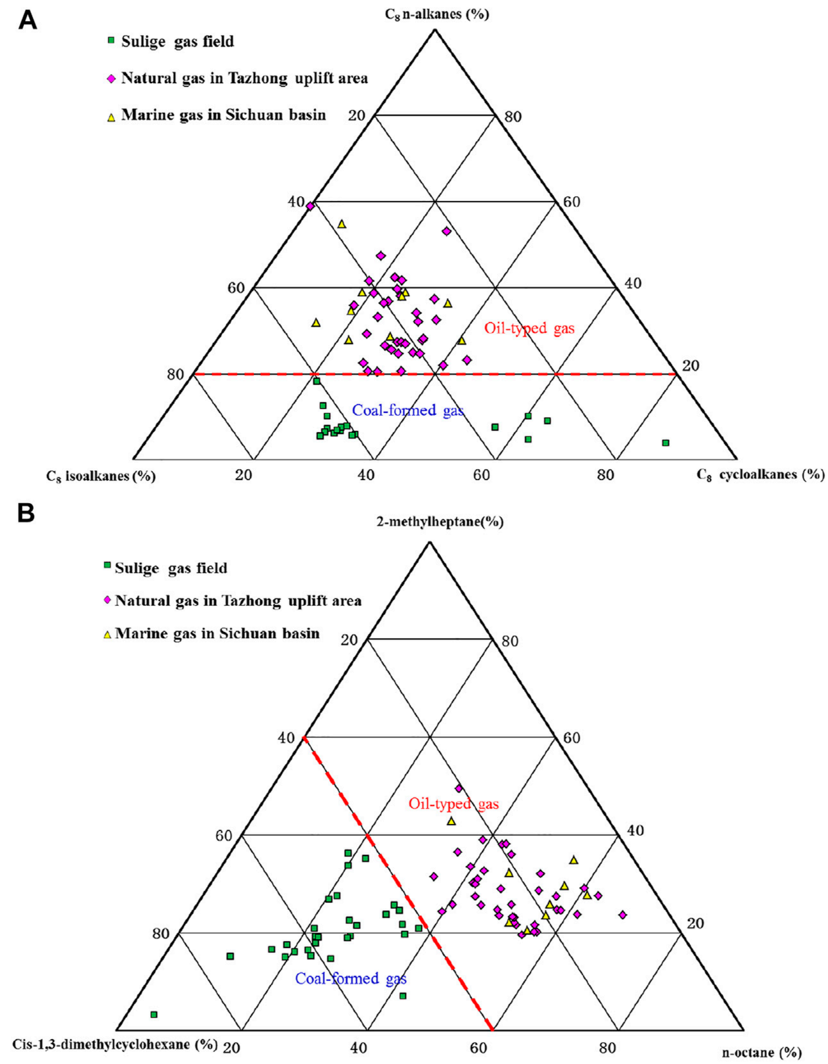

FIGURE 11 | Ternary diagram of C_8 light hydrocarbon aliphatic **(A)** and C_8 light hydrocarbon compounds **(B)** in natural gases from the Sulige gas field.

consistent with the results obtained by conventional gas source correlation methods by alkane gas isotopes, further deepening the understandings of gas sources in the Sulige gas field.

5.3 Quantitative Evaluation of Gas Sources Contributions From Noble Gas and Alkane Gas Isotopes for the Sulige Gas Field

Since coal measure source rocks usually contain coals and mudstones, the average content of K in Chinese coals is about 0.214% and the average content of potassium in Chinese mudstones is about 2.67%. Due to great differences on potassium content between coals and mudstones, the $^{40}Ar/^{36}Ar$ values in natural gas produced from coals is relatively low and in natural gas produced from mudstones in relatively high (Liu and Xu, 1987, 1993; Zhang et al., 2005). Therefore, the source contribution proportions of coals and mudstones can be quantitatively studied by the noble gas argon isotope $^{40}Ar/^{36}Ar$. The distribution of $^{40}Ar/^{36}Ar$ in the natural gases of the Sulige gas field ranges from 506 to 1940, and the average value is about 1148. The minimum value of $^{40}Ar/^{36}Ar$ in Upper Paleozoic natural gas samples of the Sulige gas field is approximated as the end element value of natural gas generated from coals, and the maximum value of $^{40}Ar/^{36}Ar$ in Upper Paleozoic natural gas samples of the Sulige gas field is approximated as the end element value of natural gas generated from mudstones. So, the source contribution proportions of coals and mudstones in Carboniferous-Permian coal measure source rocks are calculated as 55% and 45%, respectively, by using noble gas argon isotope of the $^{40}Ar/^{36}Ar$ method.

In addition, the contribution proportions of coals and mudstones in the coal measure source rocks of the Sulige gas field was also quantitatively investigated by using the alkane gas methane carbon isotope method. By comparing the data with previous analyses, the heaviest methane carbon isotope $\delta^{13}C_1$ of the positive sequence of alkane gases carbon isotopes in the Sulige gas field analyzed here is –28.4‰, which can be approximated as the methane carbon isotope end element value of natural gas from coals. The lightest methane carbon isotope $\delta^{13}C_1$ with positive sequence of alkane gas isotope in Upper Paleozoic of Ordos Basin was found to be −38.1‰ (Dai, 2014), which can be used as the approximate end element value of methane carbon isotope for natural gas production from mudstones. In this study, the average value of methane carbon isotope $\delta^{13}C_1$ in the Sulige gas field is about −32.3‰, and the contribution of coals and mudstones in Carboniferous-Permian coal measure source rocks is calculated as 60% and 40% separately from the alkanes carbon isotope quantification.

Therefore, the Sulige gas field in Ordos Basin is mainly derived from coal measure source rocks of Carboniferous-Permian, and the contribution of coals mainly accounting for 55–60% and the contribution of mudstones accounting for 40–45%.

6 CONCLUSION

1. The Sulige gas field is mainly alkane gases with high methane content; low heavy hydrocarbon contents; high drying coefficient; low non-hydrocarbon gases contents, mainly CO_2 and N_2; and relatively low noble gas contents in general. The helium content is slightly higher than atmospheric content by about two orders of magnitude, while neon, argon, krypton, and exon contents are relatively lower than atmospheric values by about 1,2,2,1 orders of magnitude.
2. Natural gas alkane carbon and hydrogen isotopes are generally distributed in positive sequences, with a few parts inversion. The $^3He/^4He$ values are mainly distributed around $(2.89–7.30)\times10^{-8}$, with an average of 4.81×10^{-8}. The $^{40}Ar/^{36}Ar$ values are distributed in a wide range, from 506 to 1940, with an average value of 1148. The ^{129}Xe is relatively deficient, while the ^{132}Xe is relatively surplus.
3. The carbon isotopes of alkanes in the Sulige gas field indicate that the natural gases are high mature to over mature coal-formed gas. The genetic identification of C_7 and C_8 light hydrocarbons reflect that the natural gases are typical coal-formed gases, mainly originating from humic source rocks. The comprehensive identification of noble gas isotopes indicate that noble gases are typical crustal genesis, mainly generated from radioactive decay of U, Th, and other elements of crust. The CO_2 in the natural gases are mainly organic in general and partly inorganic.
4. The gas source correlations of noble gases and alkane gases and quantitative evaluation of gas source contributions show that the natural gases in the Sulige gas field mainly come from Carboniferous-Permian coal measure source rocks in Ordos Basin, where the contribution proportion of coals accounts for 55–60%, while the contribution proportion of mudstones accounts for 40–45%. The evaluation results of noble gas argon isotope and alkane gas carbon isotope are basically compatible, further deepening the understandings on gas sources of natural gases in the Sulige gas field.

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 authors.

AUTHOR CONTRIBUTIONS

All authors listed have made a substantial, direct, and intellectual contribution to the work and approved it for publication.

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