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Geochemical Characteristics of Helium in Natural Gas From the Daniudi Gas Field, Ordos Basin, Central China

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Helium-bearing gas is accumulated in the Lower Ordovician, Upper Carboniferous, and Lower Permian reservoirs of the Daniudi gas field in Ordos Basin, and the helium concentrations and isotopic compositions are investigated in order to reveal the abundance and origin of helium. Geochemical characteristics indicate that the natural gas from the Daniudi gas field has helium concentrations of 0.0271–0.1273%, with R/Ra ratios of 0.007–0.072. The $^{4}He/^{20}Ne$ ratios range from 848 to 17,000, which are substantially higher than the ratio of air or air saturated water. The proved helium reserves of the Daniudi gas field exceed $100 \times 10^{6}\ m^{3}$, suggesting an extra-large helium gas field. Helium in the field is of crustal origin and derived from the radioactive decay of U and Th in the rocks and minerals, with no significant contribution by atmospheric or mantle-derived helium. The natural gas in the Daniudi gas field displays the characteristics of typical crustal helium, which is consistent with the gases from cratonic basins (Ordos, Sichuan, and Tarim) in China, whereas the gases from rift basins (Songliao, Bohai Bay, and Subei) have experienced a significant addition of mantle-derived helium.

Keywords: helium, concentrations, isotopic compositions, Daniudi gas field, Ordos Basin

INTRODUCTION

Helium (He) is an exhaustible natural resource with strategic values, and its unique physiochemical property leads it to an irreplaceable role in high-tech fields (Xu et al., 1998; Anderson, 2018). The development of science and technology made the application fields of helium become more and more extensive, causing the global demand of helium gas to increase annually by 4–6% (Zhao et al., 2012), and thus the shortage of helium supply existed for a long time. The leading locations for estimated helium resources in the world are the United States, Qatar, Algeria, and Russia, and their helium resources account for >90% of the world's total amount (Anderson, 2018). Helium resources in China are rare, with the supply basically relying on imports, and they have been poorly studied and explored to date. Therefore, the amounts of helium resources and reserves urgently need to be further evaluated (Tao et al., 2019; Chen et al., 2021).

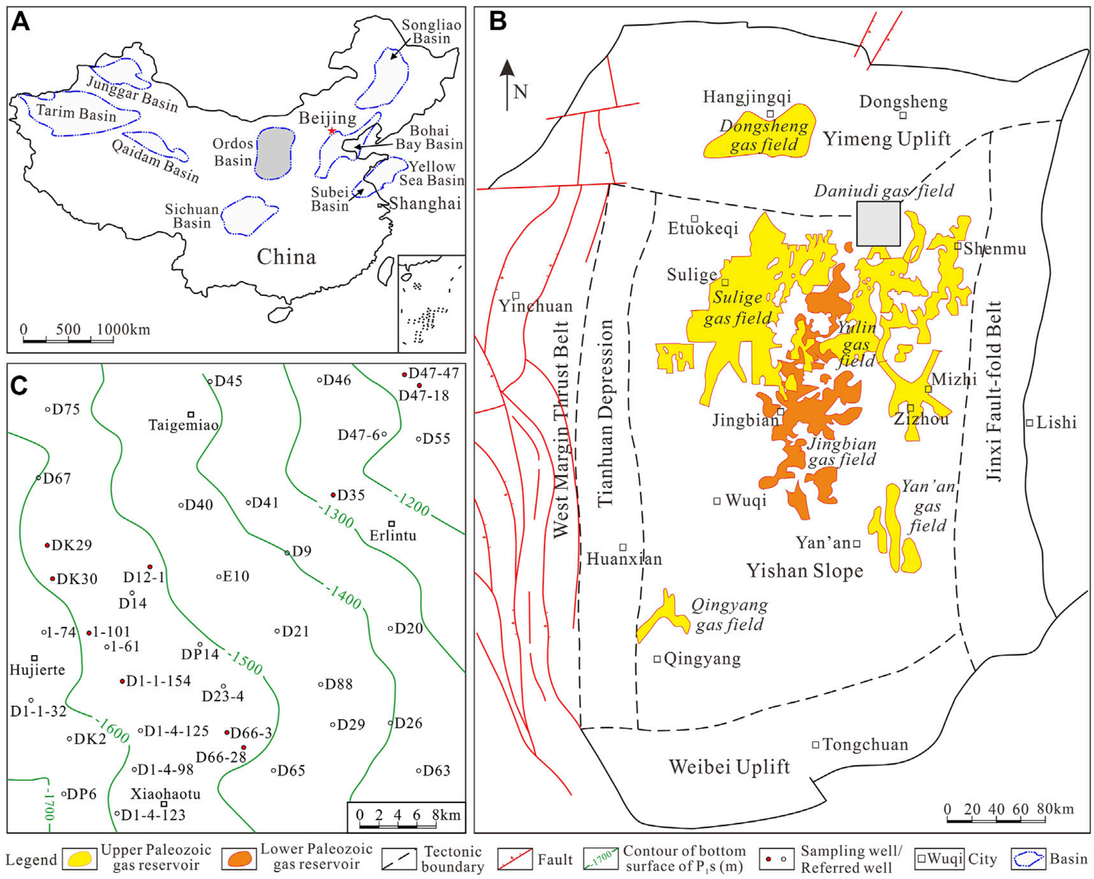

FIGURE 1 | The location of Ordos Basin in China **(A)**, location of the Daniudi gas field in the basin **(B)**, and distribution of gas wells in the gas field **(C)**.

Helium in natural gas has three different sources, including (1) atmospheric helium—being the helium in the air with a concentration of 5.24×10^{-6}, it was mainly produced from volcanic eruption, magma degassing, and rock weathering (Porcelli et al., 2002; Wang et al., 2020; Chen et al., 2021); (2) crustal helium—which is being generated by the radioactive decay of uranium (U) and thorium (Th) in crustal rocks and minerals (Oxburgh et al., 1986; Xu et al., 1998; Porcelli et al., 2002), and the production of crustal helium depends on the U and Th concentrations as well as the history of the rocks and minerals (Chen et al., 2021); and (3) mantle-derived helium—which is from mantle-derived volatiles and released into the sedimentary strata through magmatism and fault activities (O'Nions and Oxburgh, 1983; Poreda et al., 1986; Poreda et al., 1988; Xu et al., 1997; Anderson, 2000).

Helium commonly has two isotopes, *i.e.*, ^{3}He and ^{4}He, and the isotopic compositions of helium from different sources have certain differences. The ^{3}He/^{4}He ratio of air (Ra) is 1.4×10^{-6} (Mamyrin et al., 1970), whereas the ratios of crustal and mantle-derived helium are 2×10^{-8} and 1.1×10^{-5}, respectively (Lupton, 1983; Xu, 1996). The R/Ra ratios (R refers to the ^{3}He/^{4}He ratio of the samples) were commonly used to characterize helium isotopic compositions (Xu et al., 1995a; Ni et al., 2014), and they have played an important role in revealing mantle-derived magmatism (Sano et al., 1984; Poreda et al., 1988; Marty et al., 1989) and tectonic background (Xu et al., 1995a; Xu, 1997; Polyak et al., 2000; Ding et al., 2005) and tracing the origin and source of fluids in the petroleum system (Xu et al., 1995b; Dai et al., 2008; Ni et al., 2014; Dai et al., 2017; Zhang et al., 2019; Cao et al., 2020).

The only profitable way to produce helium at present is to extract from helium-bearing natural gas (Anderson, 2018; Tao et al., 2019), and the helium in natural gas pools is mainly crustal and mantle-derived (Xu et al., 1995a; Chen et al., 2021). Ordos Basin is a crucial petroliferous basin in China with an annual gas production of 46.496×10^9 m^3 in 2018, and it becomes the first Chinese basin with an annual gas production of over 40×10^9 m^3 (Dai et al., 2019). Helium-bearing gas with He% ≥0.05%, having potential industrial helium values, has been confirmed in the Sulige gas field in the basin (Dai et al., 2017). The Daniudi gas field is located in northern Ordos Basin with the proved gas reserves of 454.563×10^9 m^3 (Yang and Liu, 2014). Previous studies on the natural gas of the Daniudi gas field principally focused on genetic types and filling patterns (Liu et al., 2015; Wu et al., 2017a; Wu et al., 2017b), and less attention has been paid to helium. Therefore, the geochemical characteristics of helium for the Daniudi gas field were studied in this work based on the concentrations and isotopic compositions of helium and its relationship with neon and argon isotopes as well as the

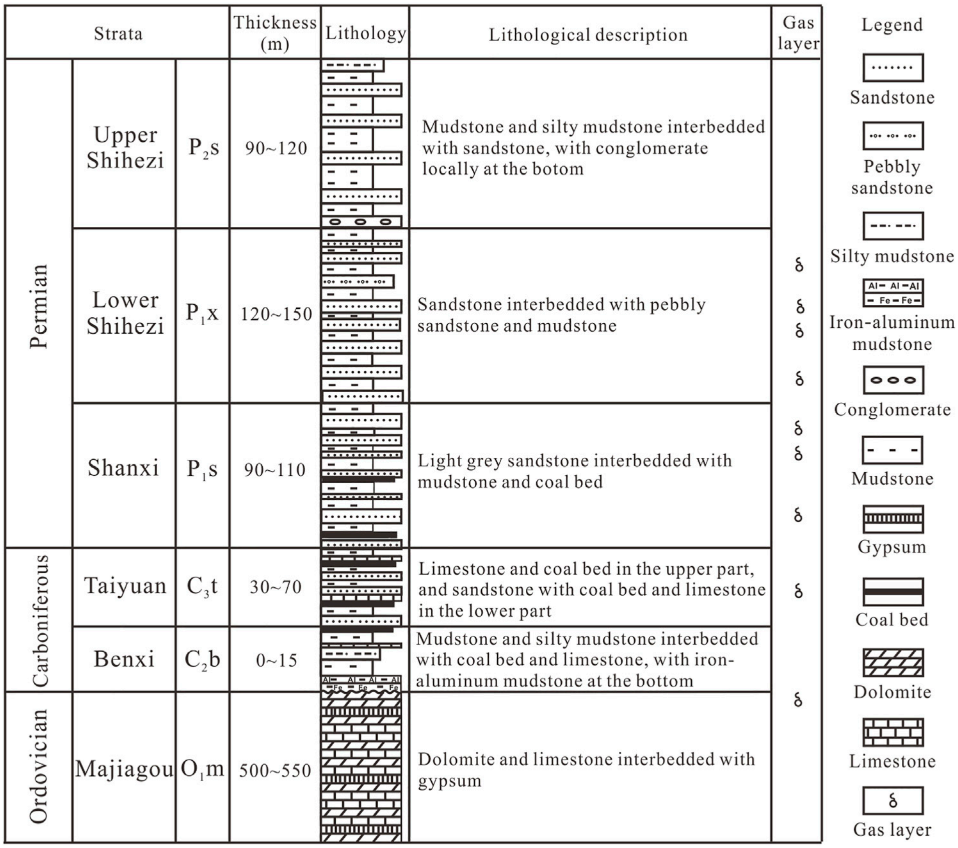

FIGURE 2 | Stratigraphic column of the Daniudi gas field, Ordos Basin.

relationship with CH_4 and CO_2 contents. The abundance and origin of helium were further investigated, which would provide scientific proofs for revealing helium enrichment mechanisms and evaluating the resource potential of helium.

GEOLOGICAL SETTING

Ordos Basin, a multicycle cratonic basin in central China (**Figure 1A**), tectonically belongs to the western margin of the North China Block, and it is the second largest sedimentary basin in China, covering an area of 37×10^4 km^2 (Yang and Pei, 1996). The basin is commonly divided into six secondary structural units (Yang et al., 2005), *i.e.*, Weibei Uplift, Yishan Slope, Yimeng Uplift, Tianhuan Depression, West Margin Thrust Belt, and Jinxi Fault–Fold Belt (**Figure 1B**). The exploration fields of conventional natural gas in Ordos Basin mainly include the Upper Paleozoic Carboniferous–Permian and Lower Paleozoic Ordovician strata. The natural gas of the discovered large gas fields is accumulated in the Carboniferous–Permian tight sandstone reservoirs in majority, except that the gas from the Jingbian gas field is mainly reservoired in marine carbonate rocks in the Lower Ordovician Majiagou Formation (Dai et al., 2005a; Liu et al., 2009; Liu et al., 2015; Dai, 2016).

The Daniudi gas field is located in the north margin of the Yishan Slope (**Figure 1B**), covering an area of 2,003.71 km^2 (Dai, 2016), and the strata dip gently to the southwest (**Figure 1C**). Natural gas in the field is mainly accumulated in the Upper Paleozoic tight sandstone reservoirs in the Carboniferous Taiyuan (C_3t), Lower Permian Shanxi (P_1s), and Lower Shihezi (P_1x) formations, and natural gas has also been discovered in carbonate reservoirs in the Lower Ordovician Majiagou Formation (O_1m) (**Figure 2**). The Upper Paleozoic (C_3t, P_1s, and P_1x) gas is mainly coal-derived gas and sourced from the C_3t-P_1s coal measures, and the regional caprocks of the gas pools are the stably distributed lacustrine mudstone and silty mudstone in the Upper Shihezi Formation (P_2s) (Liu et al., 2015; Wu et al., 2017a). However, the Lower Paleozoic (O_1m) gas is mixed by the Upper Paleozoic coal-derived gas and oil-associated gas from the O_1m source rocks (Wu et al., 2017b; Liu et al., 2015), and the main caprocks of the gas pools are the mudstone and iron–aluminum mudstone in the Benxi Formation (C_2b) (**Figure 2**).

SAMPLES AND ANALYTICAL METHODS

In this study, 20 gas samples from the O_1m, C_3t, P_1s, and P_1x reservoirs in the Daniudi gas field were collected directly from well heads. Double-ended stainless steel cylinders (5-cm radius, ~ 7,000-cm^3 volume) were used to collect the gas samples, and air

TABLE 1 | Noble gas concentrations and isotopic compositions of natural gas in the Daniudi gas field, Ordos Basin.

Well	Strata	CH_4 (%)	C_{2+} (%)	CO_2 (%)	^4He ($\times10^{-6}$)	^{20}Ne ($\times10^{-6}$)	^{40}Ar ($\times10^{-6}$)	$^3He/^4He$ ($\times10^{-8}$)	R/Ra	$^4He/^{20}Ne$	$^{40}Ar/^{36}Ar$	$CH_4/^3He$ ($\times10^9$)	$CO_2/^3He$ ($\times10^9$)
1-101	P_1x	97.19	2.55	0.25	586	0.122	52.9	3.976	0.028	4,803	797.0	41.7	0.107
2-1	P_1x	94.47	4.92	0.41	1,273	0.340	85.4	2.856	0.020	3,744	654.5	26.0	0.112
D66-28	P_1x	88.90	10.66	0.42	450	0.092	74.8	5.180	0.037	4,891	616.1	38.1	0.180
DK30	P_1x	94.39	5.17	0.42	459	0.027	52.4	6.440	0.046	17,000	1,163.9	31.9	0.141
D66-3	P_1x	97.52	2.38	0.09	421	0.158	66.5	2.660	0.019	2,665	514.0	87.1	0.081
DP14	P_1x	87.92	11.55	0.43	390	0.460	234.0	5.012	0.036	848	368.7	45.0	0.221
DK29	P_1x	88.70	10.57	0.55	371	ND	70.1	0.913	0.007	ND	537.5	261.9	1.621
D1-1-154	P_1x	90.54	8.30	1.14	394	0.161	90.2	2.618	0.019	2,447	547.5	87.8	1.108
2-47	P_1s	90.73	6.87	2.38	316	0.180	24.7	3.598	0.026	1756	1062.8	79.8	2.097
D1-4-107	P_1s	89.33	9.99	0.65	342	0.095	31.0	1.005	0.007	3,600	937.0	259.8	1.897
2-21	P_1s	89.15	9.93	0.90	376	ND	39.9	10.115	0.072	ND	2328.2	23.4	0.237
1-80	P_1s	88.92	10.26	0.80	381	0.089	48.3	2.688	0.019	4,281	945.1	86.8	0.783
2-45	P_1s	87.98	11.22	0.78	372	ND	44.1	1.226	0.009	ND	901.7	192.8	1.711
D12-1	P_1s	89.95	9.38	0.65	415	0.050	56.5	1.764	0.013	8,300	1038.4	122.9	0.889
D35	C_3t	90.33	7.56	1.98	287	0.164	69.5	2.940	0.021	1750	376.8	107.1	2.352
D23-4	C_3t	90.58	6.86	2.56	271	0.076	36.8	2.394	0.017	3,566	451.1	139.6	3.941
D47-18	C_3t	91.09	8.04	0.87	452	0.080	66.7	8.016	0.057	5,650	500.2	25.1	0.240
D35-22	C_3t	90.43	7.37	2.11	303	0.240	67.3	4.424	0.032	1,263	376.6	67.5	1.577
D47-47	C_3t	90.93	8.10	0.73	401	0.052	43.9	6.440	0.046	7,712	930.6	35.2	0.284
D66-38	O_1m	91.66	7.26	1.08	232	0.155	52.1	3.612	0.026	1,497	396.5	109.4	1.293

ND, no data.

contamination was removed by flushing the lines first for 15–20 min. The chemical composition of the main components and the stable carbon isotopic data for alkane gas were available from Liu *et al.* (2015) and Wu *et al.* (2017a). Noble gas concentrations and isotopic ratios were measured using a Noblesse SFT noble gas mass spectrometer at the Key Laboratory of Petroleum Resources Research, Chinese Academy of Sciences, in Lanzhou.

The gas was purified using a spongy titanium furnace at 800°C to remove active gases (C_1–C_4, N_2, CO_2, *etc.*) and then separated at a temperature ranging from 8 to 100 K using a cryogenic trap filled with activated charcoal. Noble gases were adsorbed in the trap at 8 K for 20 min, and He, Ne, and Ar gases were released for analysis at the temperature of 15, 50, and 100 K, respectively. ^4He isotope was analyzed using a Faraday collector, and ^3He was analyzed using an electric multiple detector. ^{20}Ne isotope was analyzed using a Faraday collector. ^{40}Ar and ^{36}Ar isotopes were analyzed using a Faraday cup. The volume of He, Ne, and Ar, respectively, was determined for the calibration of their concentration. The detailed analytical procedure can be seen in Cao *et al.* (2018).

The authors have also collected noble gas data from the Sichuan (Fan, 1999; Wu et al., 2013; Ni et al., 2014; Dai et al., 2017), Bohai Bay (Dai et al., 2005b; Zhang et al., 2008), Songliao (Liu et al., 2016), Subei (Xu et al., 1997; Liu et al., 2017), Tarim (Liu et al., 2018), and Ordos (Dai et al., 2017) basins for comparative analysis.

RESULTS

The He, Ne, and Ar concentrations and isotopic ratios of natural gas from the Daniudi gas field in Ordos Basin are listed in **Table 1**.

Helium Concentrations

The helium concentrations of natural gas from the Upper Paleozoic C_3t, P_1s, and P_1x reservoirs of the Daniudi gas field are in the ranges of 0.0271–0.0452, 0.0316–0.0415, and 0.0371–0.1273%, respectively, with the corresponding average values of 0.034% ($N = 5$), 0.0367% ($N = 6$), and 0.054% ($N = 8$) (**Table 1**; **Figure 3A**). One gas sample (Well D66-38) from the Lower Paleozoic O_1m reservoir displays a lower helium concentration of 0.0232% (**Table 1**; **Figure 3A**).

Helium Isotopic Compositions

The $^3He/^4He$ ratio of air (Ra) is generally considered as 1.4×10^{-6} (Mamyrin et al., 1970), whereas the $^3He/^4He$ ratios (R) of natural gas in the Daniudi gas field are in the range of $(0.9128–10.115) \times 10^{-8}$, and thus the calculated R/Ra ratios of the gas range from 0.007 to 0.072 (**Table 1**). The R/Ra ratios of the C_3t, P_1s, and P_1x gases are in the ranges of 0.017–0.057, 0.007–0.072, and 0.007–0.046, with average values of 0.035, 0.024, and 0.026, respectively (**Table 1**). The O_1m gas sample has a R/Ra ratio of 0.026 (**Table 1**).

$^4He/^{20}Ne$ and $^{40}Ar/^{36}Ar$ Ratios

The $^4He/^{20}Ne$ ratios of the C_3t, P_1s, and P_1x gases from the Daniudi gas field are in the ranges of 1,263–7,712, 1,756–8,300, and 848–17,000, respectively, and the corresponding $^{40}Ar/^{36}Ar$ ratios are in the ranges of 376.6–930.6, 901.7–2,328.2, and 368.7–1,163.9, respectively (**Table 1**). The O_1m gas sample has $^4He/^{20}Ne$ and $^{40}Ar/^{36}Ar$ ratios of 1,499 and 396.5, respectively (**Table 1**).

$CH_4/^3He$ and $CO_2/^3He$ Ratios

The $CH_4/^3He$ ratios of the P_1x, P_1s, and C_3t gases from the Daniudi gas field are in the ranges of $(26.0–261.9) \times 10^9$,

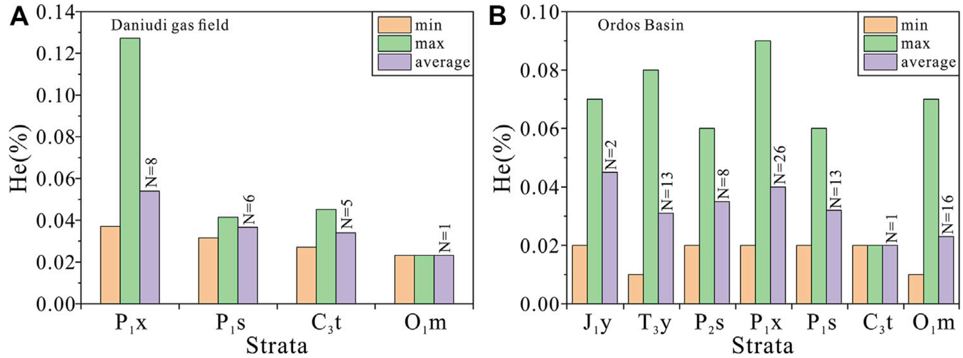

FIGURE 3 | Distribution of helium concentrations in natural gas from different strata in the Daniudi gas field **(A)** and Ordos Basin **(B)**. The data in **(B)** are from Dai *et al*. (2017).

(23.4–259.8) × 10^9, and (25.1–139.6) × 10^9, respectively, with corresponding average values of 77.4 × 10^9 (N = 8), 127.6 × 10^9 (N = 6), and 74.9 × 10^9 (N = 5) (**Table 1**). The $CO_2/^3He$ ratios of these gases are in the ranges of (0.081–1.621) × 10^9, (0.237–2.097) × 10^9, and (0.240–3.941) × 10^9, respectively, with corresponding average values of 0.446 × 10^9, 1.269 × 10^9, and 1.679 × 10^9 (**Table 1**). The O_1m gas sample has $CH_4/^3He$ and $CO_2/^3He$ ratios of 109.4 × 10^9 and 1.293 × 10^9, respectively (**Table 1**).

DISCUSSION

Helium Abundance in Natural Gas

Dai et al. (2017) have classified helium-bearing gas into five categories based on helium concentration, *i.e*., helium extremely depleted (He% <0.005%), helium depleted (0.005% ≤He% <0.050%), helium general (0.050% ≤He% <0.150%), helium rich (0.150% ≤He% <0.500%), and helium extremely rich (He % ≥0.500%) gases. The natural gas from the Panhandle–Hugoton field in the US is helium-rich gas with an average helium concentration of 0.586% (Brown, 2019), and the gas from the Weiyuan gas field in Sichuan Basin in China is also helium-rich gas with an average helium concentration of 0.251% for 215 gas samples (Dai et al., 2017).

The natural gas in Ordos Basin is mainly accumulated in the P_1x, P_1s, C_3t, and O_1m reservoirs, and the statistics conducted by Dai *et al*. (2017) indicate that the average helium concentrations are 0.04, 0.032, 0.02, and 0.023%, respectively (**Figure 3B**). The gas is mainly helium-depleted gas with 0.005% ≤He% <0.050%, except for a small amount of gas samples being general gas with 0.050% ≤He% <0.150%. The helium concentrations of 20 gas samples from the Daniudi gas field are in the range of 0.0271–0.1273% (**Table 1**), in which 18 gas samples are helium depleted (0.005% ≤He% <0.050%), and the other two samples from the P_1x reservoirs (wells 1-101 and 2-1) are helium general (0.050% ≤He% <0.150%). The average helium concentration (0.0425%; **Table 1**) is consistent with the standard of helium-depleted gas, with no samples being helium rich or extremely rich.

According to the helium amount in the proved gas reserves, the helium-bearing gas fields can be classified into very small, small, medium, large, and extra-large gas fields, with helium reserves of <5 × 10^6 m^3, (5–25) × 10^6 m^3, (25–50) × 10^6 m^3, (50–100) × 10^6 m^3, and ≥100 × 10^6 m^3, respectively (Dai et al., 2017). The helium reserves of the Panhandle–Hugoton field in the US were 18 × 10^9 m^3 at the time of discovery (Brown, 2019), and the proved helium reserves of the Weiyuan gas field in Sichuan Basin and the Hetianhe gas field in Tarim Basin in China are 80 × 10^6 and 195.91 × 10^6 m^3, respectively (Tao et al., 2019), which imply that these three fields meet the standard of an extra-large helium gas field. The proved gas reserves of the Daniudi gas field are 454.563 × 10^9 m^3 (Yang and Liu, 2014), and the proved helium reserves are calculated as 193.19 × 10^6 m^3 according to the average helium concentration of 0.0425% (**Table 1**), which also meet the standard of an extra-large helium gas field.

The only profitable way to produce helium is believed to be by extracting from helium-bearing natural gas, and it was previously considered that helium concentrations were required to reach 0.1% (Tao et al., 2019; Chen et al., 2021). Helium was profitably produced as a byproduct of liquefied natural gas in Qatar, and the economically required helium concentrations can be as low as 0.04% (Anderson, 2018). The average helium concentration in natural gas from the Daniudi gas field is 0.0425% (**Table 1**), which meets the commercial requirement of helium production through the above-mentioned mechanism.

Origin of Helium

Helium in natural gas has three sources, and the typical $^3He/^4He$ ratios of atmospheric, crustal, and mantle-derived helium are 1.4 × 10^{-6} (Mamyrin et al., 1970), 2 × 10^{-8}, and 1.1 × 10^{-5} (Lupton, 1983; Xu, 1996), respectively. The calculated typical R/Ra ratios for crustal and mantle-derived helium are 0.014 and 7.9, respectively. Helium isotopic compositions have been widely used to trace the mantle-derived volatiles (Wakita and Sano, 1983; Oxburgh et al., 1986; Poreda et al., 1988; Xu et al., 1997). It is commonly considered that R/Ra >1 suggests the input of considerable mantle-derived helium, whereas R/Ra ≤0.1

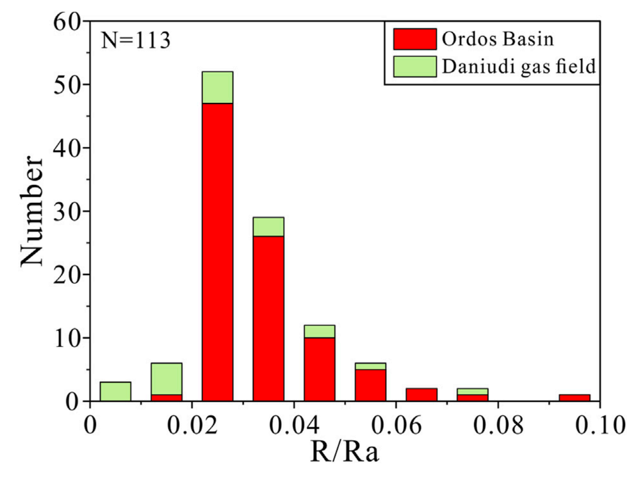

FIGURE 4 | Distribution of the R/Ra ratios of natural gas in Ordos Basin (Dai et al., 2017) and the Daniudi gas field.

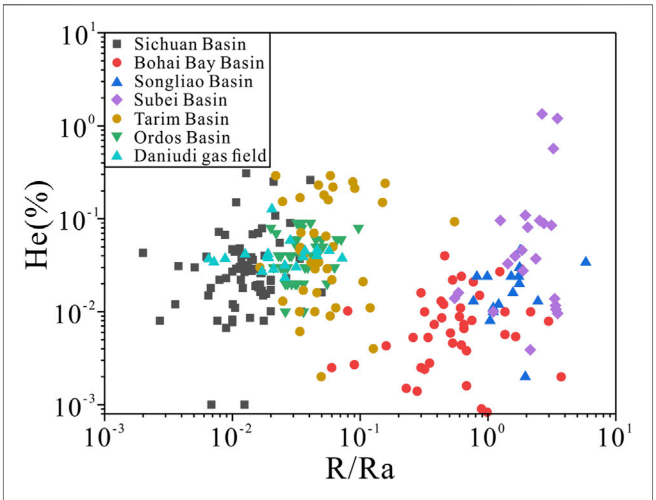

FIGURE 5 | Correlation of He (%) *versus* R/Ra of natural gas in the Daniudi gas field and typical petroliferous basins in China.

implies typical crustal helium (Xu et al., 1998; Li et al., 2017; Chen et al., 2021).

The study on helium isotopic compositions of natural gas in Chinese sedimentary basins indicated that the distribution pattern of $^3He/^4He$ ratios was controlled by the tectonic environment (Xu et al., 1995a). Both the Ordos and Sichuan basins belong to the central tectonic domain in China, with almost pure crustal helium (Xu et al., 1995a). Helium gas from the Sinian to Jurassic reservoirs in Sichuan Basin is of typical crustal origin (Ni et al., 2014). The R/Ra ratios of 113 gas samples from the basin are in the range of 0.002–0.05, with an average of 0.015, and a correlation between the R/Ra ratios and He concentrations is not observed (Wang et al., 2020).

The 93 gas samples from the Lower Ordovician Majiagou to Lower Jurassic Yan'an formations in Ordos Basin have R/Ra ratios of 0.0148–0.0974, with an average of 0.0334 (Dai et al., 2017). The R/Ra ratios for the Daniudi gas field range from 0.007 to 0.072 (**Table 1**), which are consistent with those for other areas of Ordos Basin (**Figure 4**). The R/Ra ratios for the different gas fields, including the Daniudi gas field, in the basin are lower than 0.1 (**Figure 4**), suggesting typical crustal origin with few mantle-derived helium, and the R/Ra ratios and He concentrations display few correlations (**Figure 5**). Therefore, helium in natural gas from the Daniudi gas field is supposed to be derived from the radioactive decay of U and Th in the rocks and minerals. Since the rocks and minerals in different strata, including P_1x, P_1s, and C_3t, contain various contents of U and Th, the radioactive decay of these U and Th could generate different concentrations of helium, and the generated helium could mix and accumulate in the reservoirs. Moreover, it is much easier for helium to migrate than alkane gas due to its smaller molecular. Therefore, it seems impossible to figure out where and which strata the rocks and minerals are from. Ordos Basin is a cratonic basin (Xu et al., 1995a; Dai et al., 2017) characterized by gentle structure and stable subsidence with a few faults or magmatism (Wu et al., 2017a). Natural gas from cratonic basins (*e.g.*, Sichuan, Ordos) in China has significantly lower R/Ra ratios than that from rift basins (*e.g.*, Bohai Bay) (**Figure 5**), indicating a few inputs of mantle-derived helium in the cratonic basins (Dai et al., 2017).

The He concentrations and R/Ra ratios of natural gas from different petroliferous basins in China are uncorrelated (**Figure 5**). The natural gas from Bohai Bay Basin has been contributed by a small amount of mantle-derived components, with R/Ra ratios and He concentrations commonly higher than 0.1 and lower than 0.1%, respectively (**Figure 5**). Although gases from the Subei and Songliao basins display a substantial contribution of mantle-derived helium with R/Ra ≥0.5, the He concentrations are generally lower than 0.1 (**Figure 5**). The natural gases from the Sichuan, Ordos, and Tarim basins are mainly of crustal origin, with R/Ra <0.1, and the He concentrations are principally lower than 0.1%, suggesting few differences with those for the Subei and Songliao basins (**Figure 5**). Therefore, the contribution of mantle-derived helium does not necessarily lead to the increase of He concentrations (**Figure 5**).

The natural gases from the Sinian and pre-Sinian strata in southern Sichuan Basin have high helium abundance compared to those from the other areas of the basin, suggesting an increase of crustal helium with time (Ni et al., 2014). However, the helium abundance of natural gas from different strata of the Daniudi gas field and other areas of Ordos Basin displays a few increase with time (**Figure 3B**), which indicates a little accumulation effect of crustal helium with time.

Helium Relationship With Argon and Neon Isotopes

Li *et al.* (2017) have proposed a diagram of $^3He/^4He$ *versus* $^{40}Ar/^{36}Ar$ to identify gases of crustal and mantle origins, which determines different participation degrees of mantle-derived components according to the $^3He/^4He$ ratios (**Figure 6**). The natural gas from Subei Basin generally has a higher $^3He/^4He$ ratio

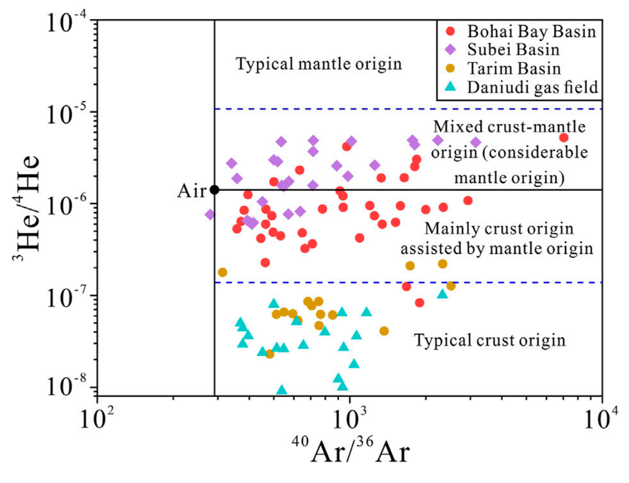

FIGURE 6 | Correlation of $^{3}He/^{4}He$ *vs*. $^{40}Ar/^{36}Ar$ of natural gas in the Daniudi gas field and typical petroliferous basins in China [modified after Li *et al*. (2017)].

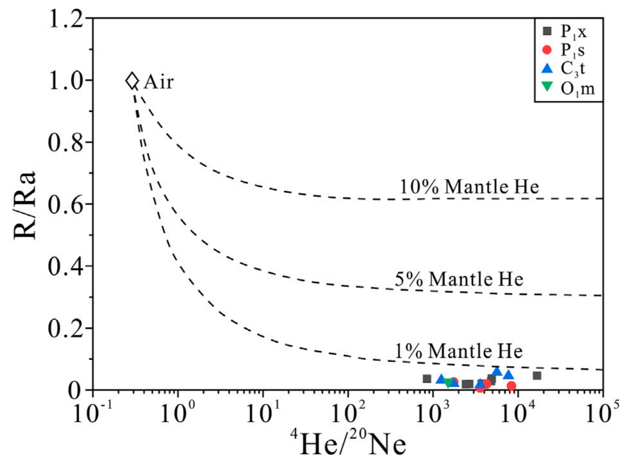

FIGURE 7 | Correlation of R/Ra *versus* $^{4}He/^{20}Ne$ of natural gas in the Daniudi gas field [modified after Zhang *et al*. (2019)].

than the air, *i.e.*, R/Ra >1, suggesting the mixed crust–mantle origin with considerable mantle helium (**Figure 6**). The natural gases from Bohai Bay Basin are mainly of crust origin assisted by mantle origin with R/Ra >0.1, and some of the gases are of mixed crust–mantle origin (with considerable mantle origin) with R/Ra >1 (**Figure 6**). The $^{40}Ar/^{36}Ar$ ratios for the Daniudi gas field in Ordos Basin range from 368.7 to 2,328.2 with R/Ra <0.1 (**Table 1**), suggesting a typical crust origin, and the $^{3}He/^{4}He$ and $^{40}Ar/^{36}Ar$ ratios are uncorrelated (**Figure 6**).

The $^{4}He/^{20}Ne$ ratios of natural gas from the Daniudi gas field range from 848 to 17,000 (**Table 1**), which are substantially higher than the ratio of air (0.318; Sano et al., 2013) or air-saturated water (ASW) (0.288, Kipfer et al., 2002). Therefore, atmospheric or ASW-derived helium has little contribution to natural gas in the field. The proportion of mantle-derived helium in the gas is less than 1% as indicated in the correlation diagram between R/Ra and $^{4}He/^{20}Ne$ ratios, which suggests the typical characteristics of crustal noble gas (**Figure 7**). Zhang *et al.* (2019) have used noble gases to trace ground water evolution and assessed helium accumulation in Weihe Basin in China, which provides an example for us to study the role of water in the migration of helium. It is a pity that we have not conducted such studies in the Daniudi gas field, and the present work only focused on the geochemical characteristics of helium in natural gas.

Helium Relationship With CH_4 and CO_2 Abundances

The CH_4 content of natural gas from the Daniudi gas field is in the range of 87.98–97.52%, with an average of 91.04%, whereas the CO_2 content is in the range of 0.09–11.22%, with an average of 0.96% (**Table 1**). Both the CH_4 and CO_2 contents have an insignificant correlation with the R/Ra ratios (**Figures 8A,B**), indicating that the origins of CH_4 and CO_2 are uncorrelated with the origin of He. Helium in natural gas from the Daniudi gas field is of crustal origin (**Figure 6, Figure 7**) and derived from the radioactive decay of U and Th in crustal rocks and minerals. The genetic identification of natural gas indicates that alkane gases, including CH_4, in the field are thermogenic and sourced from the direct or indirect thermal cracking of organic matters (Liu et al., 2015; Wu et al., 2017a; Wu et al., 2017b). CO_2 in natural gas from the Chinese cratonic basins (Sichuan and Ordos) commonly has contents lower than 5% (**Figure 8B**) and associated with the hydrocarbon generation process and decomposition of carbonates (Dai et al., 2017). However, natural gas from the rift basins (Bohai Bay, Subei, and Songliao) may have higher CO_2 contents of up to 100% (**Figure 8B**), and this CO_2 is of magmatic or mantle-derived origin (Dai et al., 2017). Therefore, the CO_2 contents and R/Ra ratios of natural gas from Chinese sedimentary basins generally have few correlations due to the different origins of CO_2 and He, except that only a few gas samples from rift basins (Bohai Bay, Subei, and Songliao) with both high CO_2 contents and R/Ra ratios were directly from magmatism or mantle degassing (**Figure 8B**).

The cross-plot of $CH_4/^{3}He$ *versus* R/Ra is conducive to constrain the possible crustal and magmatic source of natural gas (Poreda et al., 1986; Jenden et al., 1993; Ni et al., 2014; Dai et al., 2017). Ni *et al.* (2014) and Dai *et al.* (2017) have demonstrated that most gases from cratonic basins (Ordos, Sichuan, and Tarim) in China have $CH_4/^{3}He$ ratios mainly between 10^9 and 10^{12}, with R/Ra <0.1, suggesting a typical crustal origin, whereas the gases from rift basins (Bohai Bay, Songliao, and Subei) commonly display $CH_4/^{3}He$ ratios between 10^6 and 10^{11}, with R/Ra >0.1, indicating the incorporation of magmatic components (**Figure 9A**). The $CH_4/^{3}He$ ratios of natural gas from the Daniudi gas field are in the range of $(23.4–261.9) \times 10^9$ (**Table 1**), which are consistent with those from other areas of Ordos Basin (**Figure 9A**).

The $CO_2/^{3}He$ ratios of natural gas from magmatic systems are within a narrow range (10^9–10^{10}) compared with those of crustal fluids (10^5–10^{13}) (Ballentine et al., 2000). In the plot of $CO_2/^{3}He$ *versus* R/Ra, natural gas from active continental margins was explained by a two-component mixing between a crustal high-$CO_2/^{3}He$, low-R/Ra end-member and a magmatic low-$CO_2/^{3}He$, high-R/Ra end-member (Poreda et al., 1988). The varying $CO_2/^{3}He$ (3.0×10^7–1.3×10^{10}) and low R/Ra (0.002–0.035) ratios of natural

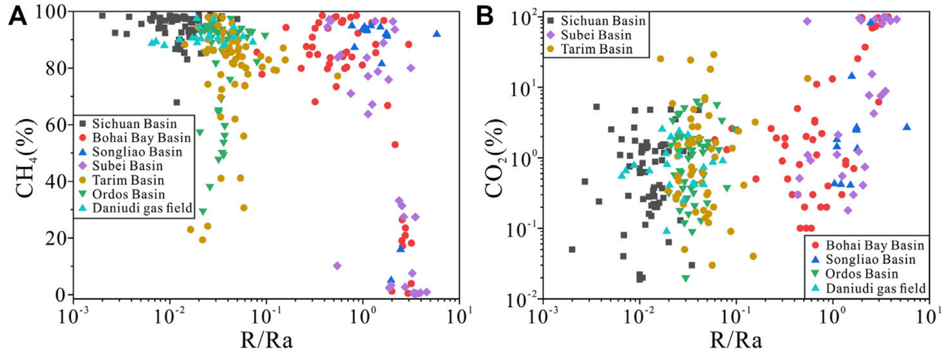

FIGURE 8 | Cross-plots of CH_4 (%) **(A)** and CO_2 (%) **(B)** *vs*. R/Ra of natural gas in the Daniudi gas field and typical petroliferous basins in China.

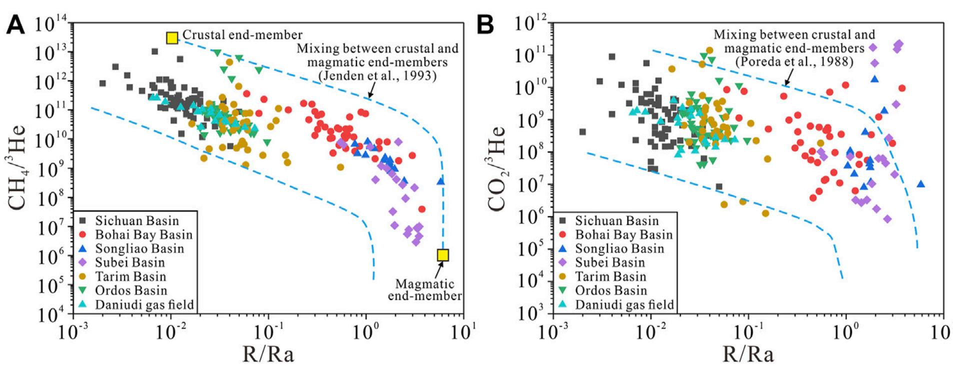

FIGURE 9 | Cross-plots of $CH_4/^3He$ *versus* R/Ra **(A)** and $CO_2/^3He$ *versus* R/Ra **(B)** of natural gas in the Daniudi gas field and typical petroliferous basins in China. Crustal end-member, magmatic end-member, and the mixing line in **Figure 1** are from Jenden *et al*. (1993), and the mixing line in **Figure 1** is from Poreda *et al*. (1988).

gas from eastern Sichuan Basin suggest a typical crustal origin (Wu et al., 2013). The $CO_2/^3He$ ratios of natural gas from the Daniudi gas field range from 0.081×10^9 to 3.941×10^9, with R/Ra <0.1 (**Table 1**), which are consistent with those from other areas in Ordos Basin (**Figure 9B**). The natural gases from cratonic basins (Ordos, Sichuan, and Tarim) in China display crustal characteristics of both $CO_2/^3He$ and R/Ra ratios, whereas those from rift basins (Songliao, Bohai Bay, and Subei) display the contribution of magmatic end-member (**Figure 9B**).

Moreover, it is noteworthy that the $CH_4/^3He$ ratio does not necessarily mean that the hydrocarbon gas is organic or inorganic since the CH_4 and He might be derived from the crust and the mantle, respectively (Dai et al., 2017). The $CO_2/^3He$ ratio may display similar features. Therefore, great attention has to be paid to only use these two parameters, and the identification of natural gas origin needs to be conducted based on comprehensive analyses of more parameters.

Future Prospects of Helium in Natural Gas in China

Helium is considered as an exhaustible natural resource with strategic values, and the replaceable role and wide application of helium determine that helium gas exploration needs more input and investment. The relevant studies on helium in natural gas in China mainly focused on the helium contents and isotopic compositions. However, the migration and accumulation mechanisms are weakly studied. The understandings of both origin and source of helium will be conducive to reveal the migration pathways and accumulation model. To accelerate the studies on the enrichment mechanisms of helium is an effective approach to find more helium resources.

CONCLUSION

The helium concentrations of natural gas from the Daniudi gas field in Ordos Basin are in the range of 0.0271–0.1273%, with an average helium concentration of 0.0425%. The $^3He/^4He$ ratios of the gas are in the range of $(0.9128–10.115) \times 10^{-8}$, and the calculated R/Ra ratios range from 0.007 to 0.072. The $^4He/^{20}Ne$ ratios are in the range of 848–17,000, with the $^{40}Ar/^{36}Ar$ ratios ranging from 368.7 to 2,328.2. The $CH_4/^3He$ and $CO_2/^3He$ ratios of the gas are in the ranges of $(23.4–261.9) \times 10^9$ and $(0.081–3.941) \times 10^9$, respectively.

The Daniudi gas field is an extra-large helium gas field with proved helium reserves of over 100×10^6 m^3. Helium from the different strata of the field is of crustal origin, with R/Ra <0.1, and it is supposed to be derived from the radioactive decay of U and Th in the rocks and minerals, with little contribution by atmospheric or mantle-derived helium. The natural gases from cratonic basins (Ordos, Sichuan, and Tarim) in China display crustal helium characteristics, whereas those from rift basins (Songliao, Bohai Bay, and Subei) display the contribution of mantle-derived helium.

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/supplementary material. Further inquiries can be directed to the corresponding author.

AUTHOR CONTRIBUTIONS

QL contributed to conceptualization, data curation, and writing. XW contributed to data curation and writing. HJ contributed to conceptualization. CN contributed to data curation and methodology. JZ contributed to methodology and investigation. JM contributed to investigation. DZ contributed to methodology. QM contributed to investigation. WP contributed to data curation. HX contributed to investigation.

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Conflict of Interest: QL, XW, HJ, CN, JZ, JM, DZ, QM, WP, and HX were employed by the company SINOPEC.

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