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Characteristics and main controlling factors of helium resources in the main petroliferous basins of the North China Craton

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Abstract

At present, the main controlling factors of helium accumulation is one of the key scientific problems restricting the exploration and development of helium reservoir. In this paper, based on the calculation results of He generation rate and the geochemical characteristics of the produced gas, both the similarities and differences between natural gas and He resources in the Bohai Bay, Ordos and the surrounding Songliao Basin are compared and analyzed, discussing the main controlling factors of helium resources in the three main petroliferous basins of the North China Craton. It is found that the three basins of Bohai Bay, Ordos and Songliao have similar characteristics of source rocks, reservoirs and cap rocks, that's why their methane resource characteristics are essentially the same. The calculated ^{4}He generation per cubic metamorphic crystalline basement in the three basins is roughly equivalent, which is consistent with the measured He resources, and it is believed that the ^{4}He of radiogenic from the crust is the main factor controlling the overall He accumulation in the three basins; there is almost no contribution of the mantle-derived CH_4, which suggests that the transport and uplift of mantle-derived ^{3}He carried by the present-day magmatic activities along the deep-large faults is not the main reason for the mantle-derived ^{3}He mixing in the basins. Combined with the results of regional volcanic and geophysical studies, it is concluded that under the background of the destruction of North China Craton, magma intrusion carried a large amount of mantle-derived material and formed basic volcanic rocks in the Bohai Bay Basin and Songliao Basin, which replenished mantle-derived ^{3}He for the interior of the basins, and that strong seismic activities in and around the basins also promoted the upward migration of mantle source ^{3}He. This study suggests that the tectonic zone with dense volcanic rocks in the Cenozoic era and a high incidence of historical strong earthquakes history may be a potential area for helium resource exploration.

Key words: Helium, Bohai Bay Basin, destruction of the North China Craton, fault, Cenozoic volcanic rocks, strong earthquake activity

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1 Introduction

In the universe, the concentration of helium is about 23%, second only to hydrogen (Qin and Li, 2021). However, helium is a very scarce resource on Earth. Due to its unique physical and chemical properties, helium is widely used in basic science and high-tech frontier research fields, such as national defense, aerospace, and earthquake disaster prediction (Wang et al., 2019; Anderson, 2018; Provornaya et al., 2022; Virk and Walia, 2001). However, China is a "helium-poor" country, and the degree of helium exploration is still very low, with more than 95% of helium dependent on imports (Li et al., 2022b). In order to avoid shortfalls in helium reserves, the most direct and effective way should be to improve our scientific understanding of the accumulation and exploration and development of helium resources in China.

At present, the exploration and development of helium resources is mainly concentrated in petroliferous basins, and many countries in the world have extracted high-quality helium resources from petroliferous reservoirs, such as the Four Corners region of the United States (the confluence of Utah, Colorado, New Mexico, and Arizona), where the helium concentration in natural gas can be as high as 10% (Jia et al., 2022a). There are many sedimentary basins in China, and they are mainly small and medium-sized basins. Although helium has also been detected in most of China's petroliferous basins, such as the Songliao Basin, Hailar Basin, Bohai Bay Basin, Subei Basin, Sichuan Basin, Ordos Basin, Qaidam Basin, and Tarim Basin (Feng et al., 2001; Ni et al., 2014, 2022; Liu et al., 2022b; Zhao et al., 2023; Wang et al., 2008, 2023; Tao et al., 1997a; Chao et al., 2022; Xu et al., 2017; Zhang et al., 2016) , the actual exploration in most areas has little economic benefit due to the low concentrations and reserves. Helium is mainly deposited in natural gas reservoirs as trace

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components (Lu, 2022; You et al., 2023; Li et al., 2017). There are three main types of natural gas genesis: inorganic, organic, and mixed genesis (Dai et al., 1992). Among which there are two main sources of inorganic gases, namely the metamorphism of carbonate rocks at high temperatures and underground magmatic activity (Song and Xu, 2005). And there are three sources of organic gase: the biochemical action of anaerobic bacteria in the immature stage; the thermal catalytic degradation during mature and high maturity evolution stages; high-temperature cracking (Song and Xu, 2005). However, unlike other natural gas components, helium has three main sources, namely, atmospheric sources: introduction into the earth's crust through groundwater recharge; mantle source: from magmatic activity area; crustal source: Gases produced in the earth's crust by radioactive decay (Mamyrin and Tolstikhin, 2013; White, 2023). Therefore, the accumulation mechanism of helium may be different from that of other natural gas components. First, crust-derived helium from radioactive source is highly likely to be evenly deposited in the rock medium in the form of dispersed aggregation due to the uniform distribution of its source rocks in the lithosphere, while mantle-derived helium is mainly blocked by the thick lithosphere and mainly concentrated in the deep melt of the earth. So it is likely that these helium gases migrate and accumulate from rock media or deep melts to sedimentary basins mainly through volcanic, deep-large fault zones and block boundaries (Zhang et al., 2021). Therefore, regional tectonic movement and lithospheric destruction may play a very important role in helium accumulation. However, there are relatively few studies on this aspect.

As one of the oldest cratons in China, the North China Craton was destructed 200–250 million years ago, bounded by the central orogenic belt, and the eastern massif suffered destruction (Zhu et al., 2011, 2012; Xu et al., 2009; Wu et al., 2019), resulting in two strong earthquakes around the margin of the deep-large fault zone such as the Tan-Lu fault zone in the region, namely the famous Haicheng 7.3 magnitude earthquake and Tangshan 7.8 magnitude earthquake (Cui and Xie, 2001; Feng et al., 2017), while the western massif remained relatively stable (Zhu et al., 2012). In addition, there are three large sedimentary basins in and around the North China Craton area, including the Bohai Bay Basin, the Songliao Basin, which are very important natural gas reserves in China. In addition, researchers have detected lower concentrations of helium in petroliferous reservoirs in these basins, and there are differences in helium concentrations and sources (Feng et al., 2001; Ni et al., 2022; Liu et al., 2022a; Zhao et al., 2023; Wang et al., 2023). Due to the local characteristics of the destruction and the obvious differences in helium concentration and sources in the sedimentary basins in the region, the North China Craton has become an excellent natural experimental site for further exploring the role of regional tectonic movement and lithospheric destruction in helium accumulation. Therefore, this paper analyzes the impact of the destruction of the North China Craton on helium accumulation by analyzing and comparing the geochemistry features of natural gas in the petroliferous reservoirs in the Bohai Bay Basin, Ordos Basin within the North China Craton and Songliao Basin within its periphery (Fig. 1a), combined with the research results of regional petroleum geology, geophysics and seismology, which may provide possible directions or ideas for the exploration and development of helium resources in China.

2 Geological and tectonic background

The North China Craton is located at the eastern margin of the Eurasian Plate, at the junction of the Tethys, the Paleo-Asian Ocean, and the Pacific Ocean, bounded by the Solonker-Linxi suture zone to the north and the Siberian plate, the Qilian Mountains to the west, the Qinling-Dabie orogenic belt to the south, and the Pacific subduction zone to the east (Chen, 2010). At present, the Precambrian basement of the North China Craton can be divided into "two belts", namely the eastern massif, the western massif, and the central orogenic belt (Zhao et al., 2005).

The Songliao Basin is located in northeastern China, west of the Yilan-Yitong fault and east of the Greater Khingan Mountains (Fig. 1b), within the complex tectonic evolution zone of the Sino-Korean and Siberian plates, and is influenced by the Tan-Lu fault, the Central Asian orogenic belt, and the subduction of the Pacific plate (Zhao et al., 2023). The deep-large faults in the basement of the Songliao Basin are extremely developed, mainly NNE-trending faults, which control the formation and development of the basin. The main faults include Nenjiang-Baicheng fault, Sunwu-Shuangliao fault and Harbin-Changchun fault. NW-trending deep-large faults are generally crustal faults or basal faults, with deep cuts and large scales (Ma, 2007). The staggered faults divide the basin into different regions, showing the spatial characteristics of east-west zones and north-south zones (Tian and Han, 1993). According to the basin boundary faults and basement morphology, the Songliao Basin can be divided into six tectonic units: Northern Pitching End, Western Slope, Northeast Uplift, Central Sag Zone, Southwest Uplift and Southeast Uplift (Ma, 2007).

Located in central China (Fig. 1c), the Ordos Basin is a typical craton depression basin in the western part of the North China Craton, with an area of about 250×10^3 km^2 during the Paleozoic Era, and is one of the most structurally stable basins in China (Dai et al., 2017; Zhao et al., 2012). Six sub-structures are developed in the basin, namely the Yishan Slope, the Tianhuan Depression, the Yimeng Uplift, the Weibei Uplift, the Jinxi Flextural Belt and the West Margin Thrusting Belt. The main body of the basin belongs to a large-scale westerly dipping gentle Yishan Slope. Although the Ordos Basin has undergone several tectonic movements, it is dominated by overall uplift, with stable internal structure and undeveloped faults (Ma et al., 2013; Fu et al., 2021). The Paleozoic Basin is conducive to natural gas accumulation, and the gas fields are mainly distributed in the northern part of the basin. The Mesozoic Basin is conducive to oil accumulation, and the oilfields are mainly distributed in the southern part of the basin (Dai et al., 2017; Fu et al., 2019).

The Bohai Bay Basin, located in eastern China (Fig. 1d), it is a typical inland rift basin within the eastern Craton massif in North China and one of the most important petroliferous basins in China. The basin covers an area of about 200×10^3 km^2, and is bounded by the Tancheng-Lujiang fault zone in the east, the eastern foothill of Taihang Mountain fault zone in the west, the Yanshan orogenic belt in the north, and the Luxi massif in the south (Li et al., 2010). The tectonic evolution of the basin has undergone three large-scale tectonic movements: the Indosinian, Yanshan and Xishan periods. The tectonic movements of the Indosinian and Yanshan periods overlapped to form the basement of the basin, and the Indosinian tectonic movements were distributed in the central and northern parts of the basin in an east-west direction. The Yanshan tectonic movement was NEE, mainly affecting the western and eastern parts of the basin. Since the Himalayan orogeny, the differential movement of oceanic plates has led to the upward arching of the lower mantle in the Bohai bay area, which flows along the NW-SE direction, which in turn drives the NW-SE extension of the upper crust. This extensional effect controls the evolution characteristics of the Bohai Bay Basin, which mainly forms the Paleogene faulted lake basin (Liu et al., 2022a). The tectonic movement is very active, the

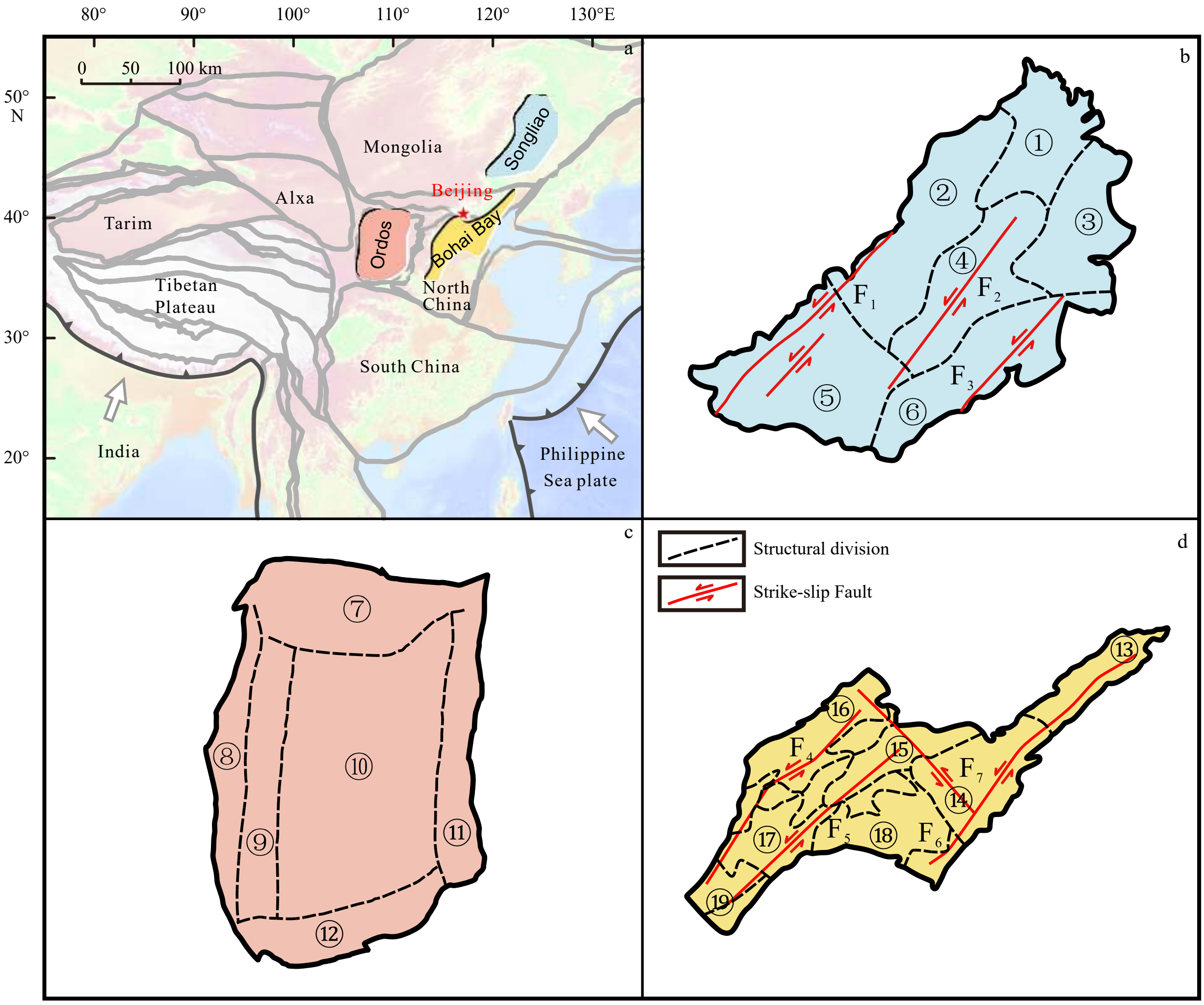

Fig. 1. Geographic location (a) and tectonic units of Songliao Basin(b), Ordos Basin(c) and Bohai Bay Basin(d) (modified from Song and Xu, 2005; Mamyrin and Tolstikhin, 2013; White, 2023). The numbers in the figure represent, 1: Northern Pitching End; 2: Western Slope; 3: Northeast Uplift; 4: Central Sag Zone; 5: Southwest Uplift; 6: Southeast Uplift; 7: Yimeng Uplift; 8: West Margin Thrusting Belt; 9: Tianhuan Depression; 10: Yishan Slope; 11: Jinxi Flextural Belt; 12: Weibei Uplift; 13: Liaohe Depression; 14: Bozhong Depression; 15: Huanghua Depression; 16: Jizhong Depression; 17: Linqing Depression; 18: Jiyang Depression; 19: Dongpu Sag; F_1: Nenjiang-Baicheng fault; F_2: Sunwu-Shuangliao fault; F_3: Harbin-Changchun fault; F_4: Baxian-Tangyin fault; F_5: Huanghua-Dongpu fault; F_6: Tan-Lu fault (Bohai section); F_7: Zhangjiakou-Penglai fault.

faults are well developed, and there are four strike-slip fault zones in the basin, including the Tan-Lu fault zone, the Zhangjiakou-Penglai fault zone, the Huanghua-Dongpu fault zone and the Baxian-Tangyin fault zone. There are four uplifts in the basin, namely the Xingheng, Cangxian, Chengning, and Neihuang Uplifts, and seven depressions, namely the Liaohe, Bozhong, Huanghua, Jizhong, Linqing, Dongpu, and Jiyang depressions, around which petroliferous fields are widely distributed (Teng et al., 2014; Dai et al., 2017).

3 Geochemical characteristics of natural gas

The average value of natural gas geochemical data in the Bohai Bay Basin, Ordos Basin and Songliao Basin is shown in Table 1 (Dai et al., 1995, 2014, 2017; Xu et al., 1995a, 1995b; Tao et al., 1997b; Liu et al., 2001, 2002, 2007, 2011, 2016; Zheng et al., 2001; Zhang et al., 2004; Wang et al., 2006a; Ni et al., 2009; Yang et al., 2014; Dai, 2016), and the detailed data are shown in Table S1, and the chemical components of natural gas in the three petroliferous basins mainly include CH_4, CO_2 and N_2, and there is a certain amount of He. In the Bohai Bay Basin, the CH_4 concentration was 0.14%–98.79%, and the average value was 49.81%. The CO_2 concentration was 0.02%–98.61%, and the average value was 38.09%. The concentration of N_2 was 0.11%–18.4%, and the average value was 1.66%. The concentration of He was 0.001 1%–0.1%, and the average value was 0.180%. The $^3He/^4He$ value was 0.03–3.90 Ra, and the average value was 1.64Ra. The concentration of CH_4 in Ordos Basin ranges from 29.58% to 97.56%, with an average of 87.41%. The concentration of CO_2 ranged from 0.02% to 8.87%, with an average of 1.77%. N_2 concentration ranged from 0.08% to 67.66%, with an average of 3.86%. He concentration ranged from 0.01% to 0.09%, with an average value of 0.035%. $^3He/^4He$ ranged from 0.02 Ra to 0.097 Ra, with an average value of 0.033 Ra. The concentration of CH_4 in Songliao Basin ranges from 0.52% to 97.8%, with an average of 77.80%. The concentration of CO_2 ranged from 0.02% to 99.48%, with an average of 18.56%. N_2 concentration ranged from 0.12% to 29.5%, with an average of 3.04%. He concentration ranged from 0.002% to 0.45%, with an average of 0.047%. $^3He/^4He$ ranges from 0.088 Ra to 5.84

Table 1. The mean values of geochemical data of natural gas in Songliao Basin, Bohai Bay Basin and Ordos Basin (refer to Dai et al., 1995, 2014, 2017; Xu et al., 1995a, 1995b; Tao et al., 1997b; Liu et al., 2001, 2002, 2007, 2011, 2016; Zheng et al., 2001; Zhang et al., 2004; Wang et al., 2006a; Ni et al., 2009; Yang et al., 2014; Dai, 2016)

Basin name	Field name	CO_2/%	N_2/%	CH_4/%	He/%	$\delta^{13}C_{CO_2}$/‰	$\delta^{13}C_{CH_4}$/‰	$^4He/^{20}Ne$	$^3He/^4He$	Rc/Ra
Songliao Basin	Wankinta	98.60	2.05	1.02	0.13	−4.53	−41.10	3 516.24	6.49×10^{-6}	4.64
	Jilin	8.44	9.52	81.43	–	–	–	5 276.62	1.97×10^{-6}	1.41
	Shuangcheng-Taipingchuan	0.36	–	93.33	0.10	10.41	−31.75	1 775.20	2.88×10^{-7}	0.21
	Changling	37.27	3.45	57.71	0.01	–	–	–	2.90×10^{-6}	2.07
	Daqing	–	–	–	0.03	–		7 471.43	2.28×10^{-6}	1.63
	Xujiaweizi	14.66	2.42	88.31	0.02	−18.38	−36.13	2 310.56	2.11×10^{-6}	1.44
Bohai Bay Basin	Dagang	15.30	0.93	75.33	0.01	−3.80	−28.60	663.78	1.84×10^{-6}	1.31
	Liaohe	0.62	18.40	78.56	0.03	–	−29.00	1 177.27	2.27×10^{-6}	1.62
	Pingfangwang	74.01	0.71	21.65	0.02	−4.45	−51.97	–	3.91×10^{-6}	2.79
	Pingnan	81.68	0.64	14.30	0.03	−2.80	−49.77	–	3.53×10^{-6}	2.52
	Shengli	32.78		–		–		1 642.94	1.83×10^{-6}	1.33
Ordos Basin	Daniudi	0.96	–	91.04	–	–	–	4 457.24	3.89×10^{-6}	0.03
	Shaanxi-Gansu-Ningxia	3.45		–	0.02	–		6 138.32	3.75×10^{-8}	0.03
	Central Ordos	–			–	–		–	4.32×10^{-8}	0.03
	Sulige	1.31	3.17	87.78	0.02	−13.98	−34.38	–	3.12×10^{-8}	0.02

Note: "–" indicates no data.

Ra, with an average value of 1.40 Ra. The Pearson correlation coefficients between He concentration and CO_2 concentration were 0.76, −0.15 and −0.06, and the Pearson correlation coefficients between He concentration and CH_4 concentration were −0.65, 0.07 and 0.02, respectively. The Pearson correlation coefficients between He concentration and N_2 concentration were 0.33, 0.02 and 0.29, and the Pearson correlation coefficients between He concentration and $^3He/^4He$ were 0.45, 0.30 and −0.43 Ra, respectively. He concentration in Bohai Bay Basin has a positive correlation with CO_2 and N_2 concentration, but a weak negative correlation with CH_4 concentration. However, there is no significant correlation between He concentration and other component concentrations in the Ordos Basin and Songliao Basin (Fig. 2).

4 Discussion

4.1 *Source of CH_4 in three petroliferous basins*

It can be observed from Fig. 2 and Fig. 3 that there is no significant difference in CH_4 concentration in Bohai Bay, Ordos and Songliao basins as a whole, ranging from 0.14% to 98.79%, 29.58% to 97.56% and 0.52% to 97.8%, respectively, and CH_4 concentration in most petroliferous fields is above 80% (Table 1). In addition, the CH_4 sources of the three petroliferous Basins are not significantly different (Figs 3 and 4), and the $\delta^{13}C_{CH_4}$ (‰) values range from −54.4‰ to −28.6‰, −59.7‰ to −24.1‰, and −42.1‰ to −17.4‰, respectively (Table 1). In general, the $\delta^{13}C_{CH_4}$ of different genetic types of natural gas CH_4 has obvious differences: The δC_1 value of biogenic gas is −100‰ – −55‰, the δC_1 value of associated gas is −55‰ – −35‰, the δC_1 value of pyrolysis gas is >−35‰, the δC_1 value of coal-derived gas is −35‰ – −22‰, and the δC_1 value of deep mantle source methane gas is about −20‰ (Cao and Ni, 2005). It can be seen from Fig. 4 that CH_4 in the three petroliferous basins mainly comes from coal-derived gas and associated gas, and there is almost no contribution from mantle-derived and biological source of CH_4.

The source rocks of the three petroliferous basins are all coal rocks, the source rocks of Bohai Bay and Ordos basins are mainly carboniferous and Permian coal measures, while the source rocks of Songliao Basin are mainly Cretaceous Shahezi Forma-

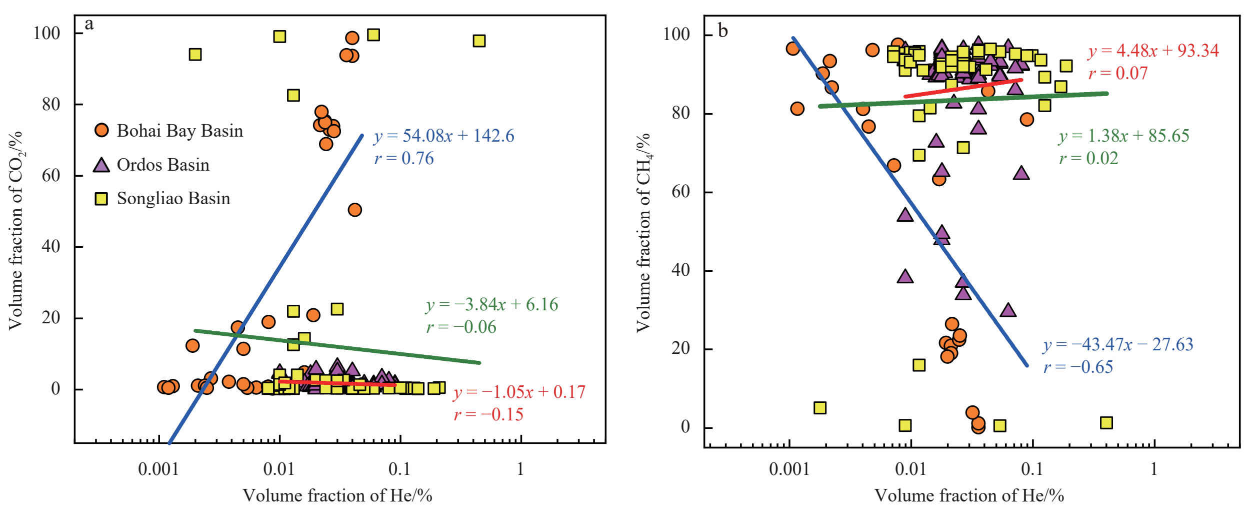

Fig. 2.

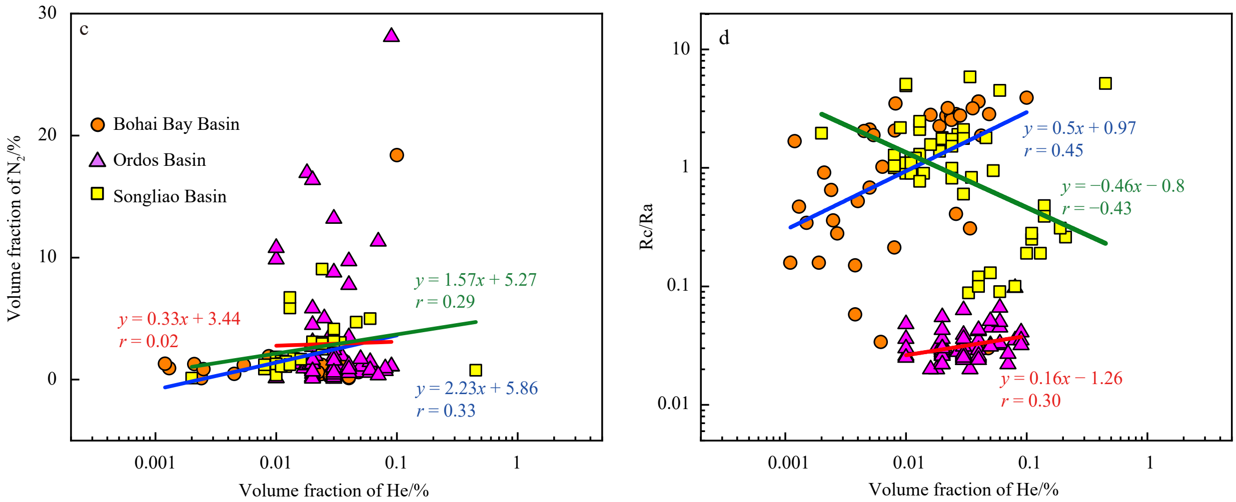

Fig. 2. Correlation diagram between He and CO_2(a), CH_4(b), N_2(c) and Rc/Ra(d) from the Bohai Bay Basin, Ordos Basin and Songliao Basin. The blue, red and green lines in the figure represent the fitted curves between He and other components and Rc/Ra for the Bohai Bay Basin, Ordos Basin and Songliao Basin, respectively.

tion and Jurassic coal measures (Dai et al., 2017; Zhang and Zhao, 2005; Zheng and Hu, 2006; Xu et al., 2012; Tang, 1992). The reservoirs in Bohai Bay Basin are mainly in the upper and lower Paleozoic Shihezi Formation, which are dominated by arkose, lithic arkose or quartz-arkose with low permeability, while bauxite and aluminaceous mudstone are important cap beds (Zhang and Zhao, 2005; Zheng and Hu, 2006; Xu et al., 2012). The reservoirs in the Ordos Basin are mainly upper Paleozoic Carboniferous surface shallow marine sandstone and limestone, mainly clastic rock and carbonate rock, with relatively low permeability, and the cap layer is composed of mudstone and silty mudstone (Zhang and Zhao, 2005; Zheng and Hu, 2006). The reservoir in Songliao Basin mainly includes the sandstone segment and bedrock weathering crust of Quantou Formation and its overlying strata, and the cover layer is mainly argillaceous rock (Zhao et al., 2023; Zhong, 2017). In conclusion, the similar source rocks and favorable reservoir-cap conditions in Bohai Bay, Ordos and Songliao basins may be one of the main reasons that the concentration and source of CH_4 in the three petroliferous basins are not significantly different on the whole.

4.2 *Source of He in three petroliferous basins*

4.2.1 *The controlling factors of ^{4}He in natural gas reservoir of three petroliferous basins*

Among the sources of helium, the contribution of atmospheric He to He in petroliferous fields is basically negligible, while ^{4}He produced by the radioactive decay of U-rich and Th-rich helium

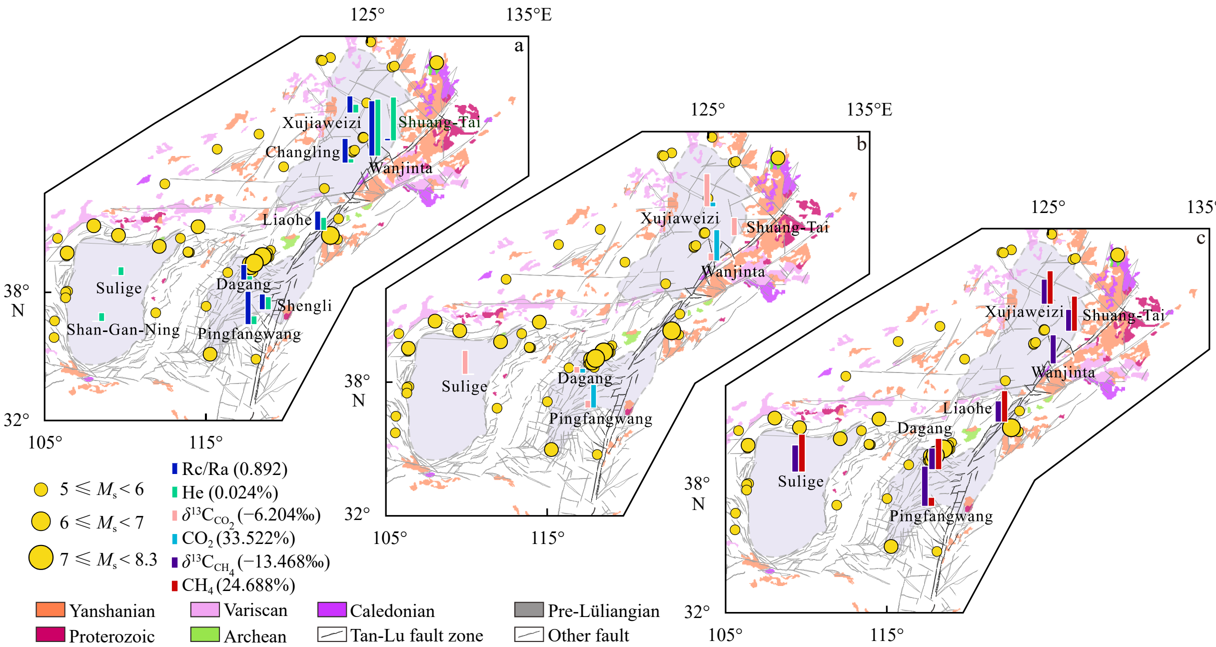

Fig. 3. Distribution of Rc/Ra and concentration of He of natural gas from the Bohai Bay Basin, Ordos Basin and Songliao Basin (a). Distribution of $\delta^{13}C_{CO_2}$ and concentration of CO_2 of natural gas from Bohai Bay Basin, Ordos Basin and Songliao Basin (b). Distribution of $\delta^{13}C_{CH_4}$ and concentration of CH_4 of natural gas from Bohai Bay Basin, Ordos Basin and Songliao Basin (c).

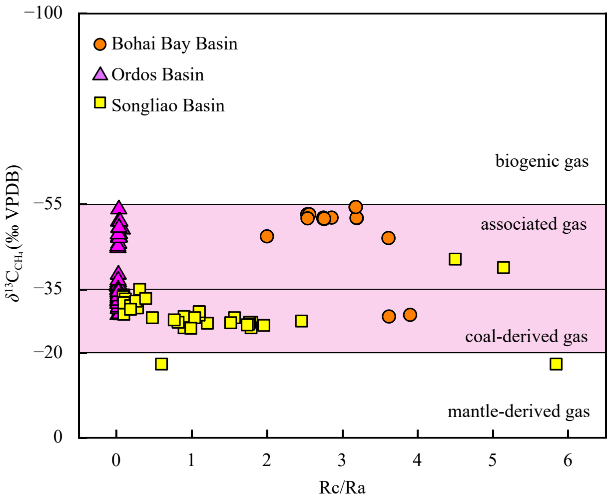

Fig. 4. The relationship between $\delta^{13}C_{CH4}$ and Rc/Ra of natural gas from the Bohai Bay Basin, Ordos Basin and Songliao Basin.

source rocks in the crustal strata and mantle-derived ^{3}He from deep mantle magmatic activity are usually the main helium sources of most helium reservoirs (Tao et al., 1997b; Mamyrin and Tolstikhin, 2013; Ballentine and Burnard, 2002). The relationship between $^3He/^4He$ and $^4He/^{20}Ne$ in Fig. 5 shows the three-terminal distribution of He in natural gas in the study area. It can be seen that He in natural gas in Ordos Basin is crust source, while in Songliao Basin and Ordos Basin, in addition to crust source radioactive source, there are 1%–50% mantle source recharge of He. Figure 5 shows the three-terminal element distribution of natural gas He in the study area through the relationship between $^3He/^4He$ and $^4He/^{20}Ne$. It can be seen that He in natural gas in the Ordos Basin is a crust source, while He in natural gas in the Songliao Basin and the Ordos Basin has 1%–50% mantle source supply in addition to crust source radioactivity. However, it can be seen from Table 1 that the measured concentrations of natural gas samples in the three basins do not show significant differences except for individual differences. Therefore, it is necessary to discuss the specific causes of crust-derived ^{4}He and mantle-derived ^{3}He respectively, to have a general understanding of the causes of He resources in the study area.

To study the crust-derived ^{4}He, we must first understand the endowment of radioactive U and Th elements in regional rocks. It is generally believed that the content of U and Th in volcanic rocks and granite is the highest, followed by metamorphic rocks, sedimentary rocks are the lowest, and the igneous rocks are characterized by acidic rock > neutral rock > basic rock > ultrabasic rock (Meng et al., 2021). However, the content of U and Th in ordinary shale in sedimentary rocks is equal to or even higher than that in granite, and slightly lower in sandstone (Fig. 6) (Brown, 2010). Organic-rich shales are formed in hydrostatic deposition in a strong reduction environment, and a lot of organic matter is preserved in the deposition process. After U^{6+} is reduced to U^{4+} in the reducing environment, it is adsorbed on organic matter and finally reacts with SiO_2 in water to produce uranium minerals (Chen and Guo, 2007). However, although organic-rich shales can not only produce more ^{4}He, they are also good source rocks in gas reservoirs. Danabalan (2017) calculated that the amount of methane gas produced by organic shale rocks with a potential hydrocarbon generation of 2 mg HC/g rock after maturity was 3 000 times that of the total amount of helium produced in 1 billion years. As a result, the helium gas produced by the source rock is greatly diluted by methane and cannot be enriched. Therefore, basement granites rich in U and Th but poor in organic matter are often considered as effective helium source rocks (Li et al., 2022a). Previous studies have shown that since the late Mesozoic in Bohai Bay Basin, the lithosphere has been strongly thinning and accompanied by strong magmatic activity, resulting in the development of a large number of granites, among which late Mesozoic granites are mainly distributed in the eastern part of the basin in the Early Cretaceous, while Cenozoic granites are distributed in all depressions (Zhao and Li, 2016; Wan et al., 2017). The granites in Ordos Basin are basically distributed in the northern part of the basin, and the high He concentration of Dongsheng gas field in Hangjinqi area has been attributed to the radioactive decay of basement Archeoeo-Proterozoic metamorphic-granite series (He et al., 2022). In the Songliao Basin, a series of granites and volcanic rocks are formed along the peripheral orogenic belts of the Greater Khingan Mountains,

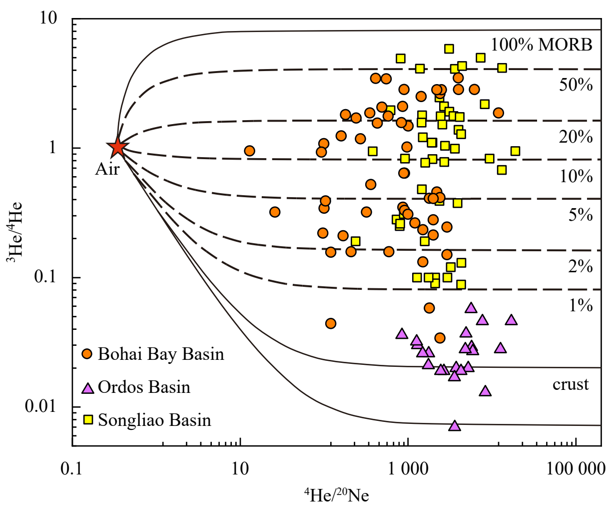

Fig. 5. The relationship between $^3He/^4He$ and $^4He/^{20}Ne$ of natural gas from the Bohai Bay Basin, Ordos Basin and Songliao Basin. MORB: Mid-Ocean Ridge Basalts.

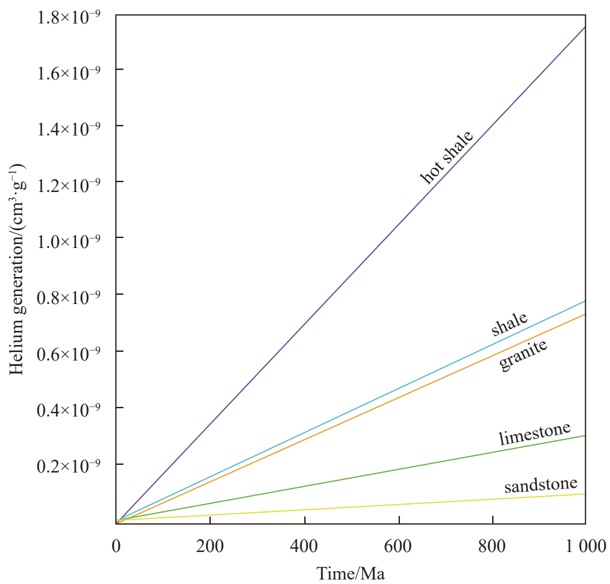

Fig. 6. Comparison of helium yields of different types of rocks (refer to Danabalan, 2017).

Lesser Khingan Mountains and Zhangguangcai Mountains, which are distributed zonally along the periphery of the basin. In the basin, large areas of volcanic rocks and granites are exposed along the deep-large faults (Zhao et al., 2023). According to the calculation of Zhao et al. (2023) the shallow granites in the southern and northern parts of Songliao Basin have better helium generating capacity.

According to the absolute geologic time formula, ^{235}U decay formula and ^{232}Th decay formula, the amount of helium produced by ^{235}U and ^{232}Th can be calculated as follows (Chao et al., 2022):

$$V_{\rm U} = \frac{156.88}{235}(e^{\lambda t}-1)\cdot v\cdot \rho \cdot x,$$
$$V_{\rm Th} = \frac{134.47}{232}(e^{\lambda t}-1)\cdot v\cdot \rho \cdot y,$$

where λ in ^{235}U decay formula and ^{232}Th decay formula are 9.848 5 × 10^{-10} (a^{-1}) and 4.947 5×10^{-10} (a^{-1}) respectively. t is the absolute age of helium source rock formation (a); v is the volume of helium source rock (km^3); ρ is the density of helium source rock (g/cm^3). x is the content of uranium in the helium source rock (10^{-6}); y is the content of thorium in the helium source rock (10^{-6}).

Since the basement granite is the main helium source rock, based on the distribution characteristics of the basement rocks of the above formula and the Bohai Bay Basin, Ordos Basin and Songliao Basin (Table 2), the helium production per cubic meter of the basement of the basin can be estimated. The calculation results show that the ^{4}He production in the basement of Bohai Bay, Ordos and Songliao basins is similar since formation, which is 0.055, 0.062 and 0.071 m^3, respectively Table 2. In addition, there is little difference in the area of Bohai Bay Basin, Ordos Basin and Songliao Basin, which are 2.0 × 10^5 km^2, 2.5 × 10^5 km^2 and 2.6 × 10^5 km^2 respectively. Therefore, if He in the three basins is of radiological source from crust, the He concentration in the gas reservoirs should be similar. The measured data support this view, indicating that the generation of ^{4}He is the main factor controlling the helium resource characteristics in the study area. However, this still cannot explain the significant difference in Rc/Ra values between Songliao Basin and Bohai Bay Basin and Ordos, so it is still necessary to discuss the ^{3}He derived from the mantle.

4.2.2 *The controlling factors of ^{3}He in natural gas reservoir of three petroliferous Basin*

Previous studies generally believe that the development of deep-large faults and strong regional tectonic activity are the necessary conditions for mantle-derived ^{3}He migration and enrichment (Feng et al., 2001; Ni et al., 2022; Liu et al., 2022b; Zhao et al., 2023). Previous geophysical studies have found that there are obvious low-speed abnormal channels under the Tan-Lu fault zone in the eastern part of the North China Craton, indicating that there may be upthrust of deep fluids along the fault zone (Guo et al., 2014; Jia et al., 2022b). In addition, in recent years, there have been many strong earthquakes along the Tan-Lu fault zone, such as the 7.3 earthquake in Haicheng in 1973 and the 7.8 earthquake in Tangshan in 1976. However, most petroliferous fields in Songliao Basin and Bohai Bay Basin are not located above the Tan-Lu fault zone (Fig. 3). Therefore, the upwelling of ^{3}He from the mantle along the Tan-Lu fault zone should not be the main source of ^{3}He supply for petroliferous reservoirs in the Songliao Basin and Bohai Bay Basin, which is also contrary to the absence of mantle source CH_4 detected in the reservoirs of the two basins (Fig. 4), so there must be another source of ^{3}He recharge to the basin reservoirs.

In addition, previous studies on regional magmatic rocks have revealed that the distribution of helium in the crust in areas without deep major faults and active tectonic movements may be close to the characteristics of mantle-derived materials, especially near basic rocks (such as basalt and diabase) that may contain mantle-derived material intrusion, and ^{3}He may be enriched (Zhang et al., 2018). Since Cenozoic, the lithosphere of the North China Craton has undergone obvious transformation and destruction due to the northeast extrusion of the Tibetan Plateau and westward subduction of the Pacific Plate. Rock and geochemical studies have revealed that the North China Craton has lost its lithospheric roots at a depth of 100 km since about 250 million years ago, and asthenosphere upsurges, accompanied by massive magma intrusion and strong seismic activity (Zhu et al., 2012). Therefore, by combining the distribution of regional volcanic rocks with the spatial distribution of Rc/Ra in Figure 7, it is found that Cenozoic volcanic rocks are developed to a large extent in and around Songliao Basin and Bohai Bay Basin, indicating that the Cenozoic magmatic activity in the region spatially controls the mixing of mantle-derived materials. In the eastern part of North China, especially in the Kuandian and Wudalianchi areas of the Songliao Basin, a large number of pyroxene megacrysts and mantle peridotite inclusions are exposed. Among them, Wu et al. (2003) found that the pyroxene megacrysts in the Kuandian area have the highest ^{3}He/^{4}He value, which is nearly 10 times that of the atmosphere (9.98 Ra), which is similar to the results determined by Li et al. (2002), reflecting the characteristics of the Mid-Ocean Ridge Basalt (MORB) source area. The He isotopic composition of lherzolite xenoliths (2.59–4.53 Ra) is much lower than that of pyroxene megacrysts, but it also shows higher mantle source characteristics. At the same time, some researchers believe that in most cases, the isotopic composition of mantle xenoliths essentially reflects the isotopic composition of the source area of the host rock basalt (Dunai and Baur, 1995; Xu and Liu, 1997), indicating that the existence of basalt in eastern North China and its pyroxene megacrysts and mantle peridotite xenoliths may lead to the local enrichment of ^{3}He. Figure 3 shows that seismic activity corresponds to the space of high Rc/Ra values. It shows that large-scale seismic activity may accelerate the release of mantle-derived ^{3}He carried by regional basic magmat-

Table 2. Helium production per cubic meter of basin basement in the three Basin(refer to Meng et al., 2022; Xing et al., 2022; Wang et al., 2006b)

Basin	Average U content/10^{-6}	Average Th content/10^{-6}	Rock density/($g \cdot cm^{-3}$)	Average geological age/Ma	Area/ (10^5 km^2)	^{4}He generation/m^3	He mean content/%
Bohai Bay Basin	611.9	180.2	2.7	1 760.9	2.0	0.055	0.018
Ordos Basin	580.31	63.2	2.7	1 940.5	2.5	0.062	0.034
Songliao Basin	784.1	405.4	2.7	1 717.1	2.6	0.071	0.046

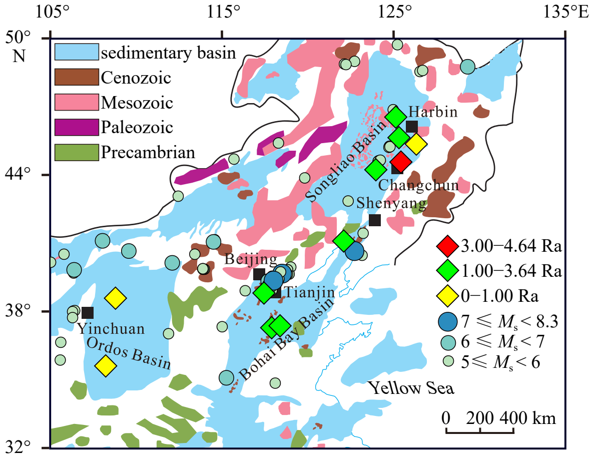

Fig. 7. The relationship between the distribution of volcanic rocks and Rc/Ra from the Bohai Bay Basin, Ordos Basin and Songliao Basin (refer to Zou et al., 2010).

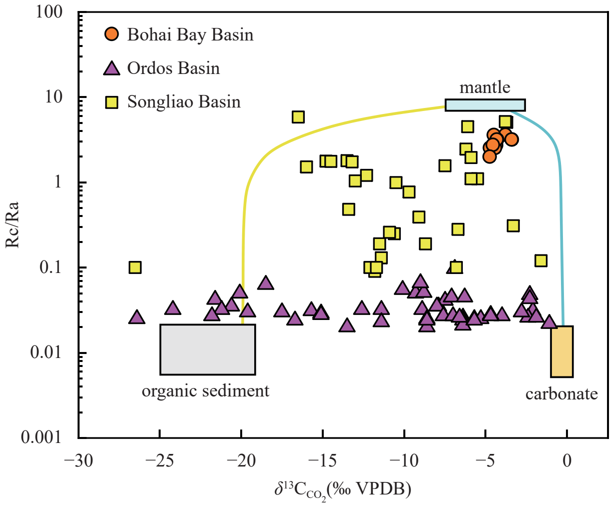

Fig. 8. He-C isotope systematics for gas samples in the Bohai Bay Basin, Ordos Basin and Songliao Basin (modified fromHoefs and Frey, 1976; Zhao et al., 2022).

ic rocks and their inclusions, which should be the reason for the high mantle-derived mixing of He in Songliao Basin and Bohai Bay Basin. This also indicates that the tectonic belt with dense Cenozoic volcanic rocks and strong earthquakes may be a potential area for He accumulation. At the same time, due to the destruction of the craton, extensional bedrock faults are developed in the basin, which should be an important gas transmission fault for regional basic magmatic rocks (Wang, 2005).

Moreover, compared with the Ordos Basin, higher CO_2 concentrations were also detected in the Bohai Bay Basin and the Songliao Basin (Fig. 3). Among them, the average CO_2 concentration in the Bohai Bay Basin was 38.14%, and the average CO_2 concentration in the Songliao Basin was 18.56%, which was much higher than the average CO_2 concentration in the Ordos Basin 1.77% (Table 1). In addition, the $\delta^{13}C_{CO_2}$-R/Ra relationship shows that (Fig. 8), the CO_2 in the Ordos Basin is mainly organic and carbonatite source, while the CO_2 in the Bohai Bay Basin and the Songliao Basin in addition to organic and carbonatite source, there is a certain amount of mantle-derived CO_2 mixed, combined with the good positive correlation between He and CO_2 in the Bohai Bay Basin in Fig. 2, indicating that these mantle-derived CO_2 may also be the same as the mantle-derived ^{3}He, from the regional Cenozoic magma intrusion. Wang et al. (2004) studied the mantle-derived inclusions such as pyroxene and olidine in the volcanic rocks of Jiyang Depression and found that the concentration of the mantle-derived inclusions in basalt is mainly CO_2, accounting for 60% to 70%, and the concentration of the liquid phase components is mostly CO_2, except water vapor (H_2O). It shows that the magma from mantle source carries a large amount of gaseous volatiles, mainly CO_2, during its formation, which is also the main gas source of CO_2 in the study area, which further supports our view.

If the ^{3}He recharge of the Songliao Basin and Bohai Bay Basin comes from regional volcanic rocks, then comparing the ^{3}He concentrations of the two with those of the Ordos Basin (Table 3), which can obtain the ^{3}He recharge amount of the regional volcanic rocks to the helium reservoir. Subtracting them indicates that the natural gas supply of the Songliao Basin and Bohai Bay Basin from the ^{3}He recharge of the Cenozoic volcanic rocks should be in the order of 10^{-8} to 10^{-7}, this also indicates that the supply of ^{3}He has a relatively small impact on the overall He abundance in the basin.

5 Conclusions

(1) According to the absolute geologic time formula, ^{235}U decay formula and ^{232}Th decay formula, the ^{4}He production per cubic meter of Archean and Paleoproterozoic metamorphic crystal-

Table 3. The endowment of ^{3}He in Songliao Basin, Bohai Bay Basin and Ordos Basin (refer to Dai et al., 1995, 2014, 2017; Xu et al., 1995a, 1995b; Tao et al., 1997b; Liu et al., 2001, 2002, 2007, 2011, 2016; Zheng et al., 2001; Zhang et al., 2004; Wang et al., 2006a; Ni et al., 2009; Yang et al., 2014; Dai, 2016)

Basin name	Petroliferous field	^{3}He concentration	^{3}He mean concentration
Songliao Basin	Wankinta oilfield	8.44×10^{-7}	2.02×10^{-7}
	Shuangcheng-taipingchuan oilfield	2.88×10^{-8}	
	Changling fault depression	2.90×10^{-8}	
	Daqing area	6.84×10^{-8}	
	Xujiaweizi fault depression	4.22×10^{-8}	
Bohai Bay Basin	Dagang oilfield	1.84×10^{-8}	6.51×10^{-8}
	Liaohe oilfield	6.81×10^{-8}	
	Square king oilfield	7.82×10^{-8}	
	Pingnan oilfield	1.06×10^{-7}	
	Shengli oilfield	5.49×10^{-8}	
Ordos Basin	Shaanxi-Gansu-Ningxia region	7.50×10^{-10}	6.87×10^{-10}
	Sulige oilfield	6.24×10^{-10}	

line basement in Bohai Bay Basin, Ordos Basin and Songliao Basin is similar to the measured value. It is concluded that the ^{4}He derived from crust-source radioactivity controls the main source of He storage in the three basins.

(2) The characteristics of source rocks, reservoirs and caps in Bohai Bay Basin, Ordos Basin and Songliao Basin are basically similar, which may be one of the important reasons for the consistency of CH_4 concentration and source characteristics in these three basins. Since there is almost no mantle-derived CH_4, it is suggested that the current magmatic activity carrying mantle-derived ^{3}He to the gas reservoir along the deep-large fault migration is not the main reason for the high mixing of mantle-derived ^{3}He in Songliao Basin and Bohai Bay Basin.

(3) In the background of the destruction of the North China Craton, the Cenozoic magma carried mantle-derived ^{3}He upwelling and intruded into the sedimentary crust, and the frequent strong seismic activities in and around the basin may further promote the release of mantle-derived ^{3}He from the basic volcanic rocks, which is the possible reason for the difference between the Songliao Basin and the Bohai Bay Basin and the Ordos. It is also indicates that the tectonic belt with dense Cenozoic volcanic rocks and strong earthquakes history may be the potential area for He accumulation.

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Supplementary information

Table S1. Natural gas geochemical data of Songliao Basin, Bohai Bay Basin and Ordos Basin.

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