Reassessment of the Distribution of Mantle CO₂ in the Bohai Sea, China: The Perspective from the Source and Pathway System



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Abstract: Research on the distribution of mantle CO_2 should involve comprehensive analysis from CO_2 source to accumulation. The crust-mantle pathway system is the key controlling factor of the distribution of mantle CO_2 , but has received little attention. The pathway system and controlling factors of CO_2 distribution in the Bohai Sea are analyzed using data on fault styles and information on the mantle and lithosphere. The relation between volcanic rocks and the distribution of mantle CO_2 is reassessed using age data for CO_2 accumulations. The distribution of mantle CO_2 is controlled by uplift of the asthenosphere and upper mantle, magma conduits in the mantle and fault systems in the crust. Uplifted regions of the asthenosphere are accumulation areas for CO_2 . The area with uplift of the Moho exhibits accumulation of mantle CO_2 at depth. CO_2 was mainly derived from vertical migration through the upper mantle and lower crust. The fault style in the upper crust controls the distance of horizontal migration and the locations of CO_2 concentrations. The distribution of mantle CO_2 in the Bohai Sea is concentrated in the Bozhong sag and the surrounding area, particularly in a trap that formed before 5.1 Ma and is connected to crustal faults (the Bozhang faults) and lithospheric faults (the Tanlu faults).

Key words: mantle CO2, source area, pathway system, fault, Moho, Bohai Sea

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1 Introduction

A large number of reservoirs with high levels of CO₂ of inorganic origin and CO₂ gas pools of inorganic origin occur in the Songliao Basin, Bohai Bay Basin, Subei Basin, Sanshui Basin, Pearl River Mouth Basin and Qiongdongnan Basin (Li M J et al., 2008; Dai et al., 2009). High-purity CO_2 gas reservoirs have been exploited as commercial raw materials and used for CO₂ gas flooding to improve the recovery efficiency of reservoirs (Tang, 1983). With the recognition that the increased atmospheric CO₂ concentration has significantly promoted global warming and the current economic model of achieving low carbon emissions, reduction in CO₂ emissions and CO₂ storage have become of greater interest (Bachu and Shaw, 2003; Vincent et al., 2009; Poulsen et al., 2011; Zatsepina and Darvish, 2011; Tasianas and Koulouzas, 2016; Lee et al., 2017). In terms of oil and gas exploration, previous studies have considered the characteristics of faults, volcanic rocks and the basement structure to avoid CO₂ accumulation areas (Imbus et al., 1998; Huang et al., 2002; Shen et al., 2007; Li Z S et al., 2008). By comprehensively analyzing the CO₂ source, transport pathways and accumulation period of CO₂ gas pools and high-content CO₂ reservoirs, we reconsider the distribution characteristics of mantle CO₂, which is important reference information for assessing the spatial extent of CO₂ accumulation areas, effectively avoiding CO₂ risk, and solving such problems faced by the petroleum industry globally.

Mantle CO_2 is mainly abiogenic in origin, as inferred from analysis of carbon and helium isotopes (Guan, 1990; Dai et al., 1994; Guo et al., 2000). There are two main sources of mantle CO_2 : magmatic degassing (Frey and Lange, 2011; Benvenuti et al., 2013; Pichavant et al., 2013) and thermal decomposition of carbonate minerals in the lithosphere (Aiuppa et al., 2004; Carter and Dasgupta, 2015). Magmatic degassing is the dominant process, and may occur during ascent of material caused by mantle plume convection, volcanic eruptions and magma intrusion (Lin, 2005; Shinohara, 2008; Girona et al., 2014; Kotarba et al., 2014). The source area of mantle CO_2 is

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associated with asthenospheric uplift. As deformation of the Moho is consistent with that of the asthenosphere, it is hypothesized that areas of CO_2 accumulation occur in the mantle underlying areas of Moho uplift.

There are two migration conditions for CO_2 : tensional environments and regional deep faults. In areas that currently emit CO_2 , development of volcanic activity, mud volcanoes and flower-like faults indicate the existence of an effective migration path (Bigi et al., 2014; Imbus et al., 1998). The flower-like and negative flower-like structures of the Tanlu and Bozhang fault belts (large-scale strikeslip faults) in the Bohai Sea are closely related to CO_2 accumulation (Zhang, 1991; Hou et al., 1996; Qiu and Wang, 2006; Li et al., 2012).

The accumulation period of CO₂ is also important. From a global perspective, mantle CO₂ may be produced during degassing and supply of persistent slow type and rapid mutant type (Schwartzman, 1978; Hart and Hogan, 1978). In terms of oil and gas exploration, attention has been paid to mantle CO₂ that has become concentrated in traps. In hydrocarbon reservoirs, mantle CO₂ and hydrocarbon gas are charged simultaneously or mantle CO₂ is charged after hydrocarbon injection (Jiang and Wang, 2010; Yang et al., 2011; Tian et al., 2013). The accumulation period of mantle carbon dioxide in the 1st and 2nd members of the Shahejie Formation in the QHD29-2-3 well was from approximately 5.1-3 Ma to the present day (Li et al., 2012; Zhou et al., 2012), which coincided with the period of hydrocarbon filling (Du and Wang, 2007; Chen et al., 2021).

Previous research has provided a basis for clarifying the distribution pattern of CO_2 and avoiding the risk of CO_2 exploration. Mantle CO_2 occurs in the basin areas that are closely associated with volcanic rocks and deep fault zones. However, there is still a lack of comprehensive analysis of mantle CO_2 distribution patterns in terms of the source, pathway system and accumulation area, which hinders full understanding of this matter. To study mantle CO_2 in the Bohai Sea, we carried out comprehensive analysis of the deep structure and crust-mantle pathway system in this area. We reanalyzed the relationship between the occurrence of volcanic rocks and the accumulation period of CO_2 , to provide a basis for deep understanding of CO_2 distribution patterns in the Bohai Sea.

2 Geological Setting

Drilling into mantle carbon dioxide reservoirs has been conducted in 16 structural areas in the Bohai Sea. Data on carbon and helium isotopes indicate that the CO₂ is mainly abiogenic gas, as is the case in the Jiyang depression and the Huanghua depression (He et al., 1997; Zheng et al., 1997; Yang, 2004; Du et al., 2006; Zhang T W et al., 2008; Hu et al., 2009). Reservoirs with high levels of CO₂ mainly occur around the Bozhong sag (Fig. 1, Table 1); those with low CO_2 levels (< 1%) are found in Liaodong Bay. Around the Bozhong sag, mantle carbon dioxide is mainly present in the Shahejie and Dongying formations, with small amounts in the Minghuazhen, Guantao and Kongdian formations. Mantle carbon dioxide in the 1st and 2nd members of the Shahejie Formation in the QHD29-2-3 well accumulated from approximately 5.1 Ma until the present day (Li et al., 2012; Zhou et al., 2012; Tian et al., 2013). CO₂ occurrence is mostly associated with flower-like, negative flower-like and listric normal



Fig. 1. Map of CO_2 distribution in the Bohai Sea (CO_2 distribution data are from Li et al., 2012).

Notes: LXS-Liaoxi sag, LXU-Liaoxi uplift, LZS-Liaozhong sag, LDU-Liaodong uplift, LDS-Liaodong sag, LXNU-Liaoxinan uplift, QNU-Qinnan uplift, QNS-Qinnan sag, NBS-Nanbao sag, QKS-Qikou sag, SLTU-Saleitian uplift, SJTU-Shijiutuo uplift, BDLU-Bodong low uplift, BDS-Bodong uag, SNS-Shanan uag, CZKU-Chengzikou uplift, CBLU-Chengbei low uplift, CBLS-Chengbei low sag, BZS-Bozhong sag, BNLU -Bonan low uplift, MXBU-Miaoxibei uplift, MXNU-Miaoxinan uplift, HHKS-Huanghekou sag, LBU-Laibei uplift, MXS-Miaoxi sag, KDU-Kendong uplift, HBU-Huaibei uplift.







Fig. 2. Fault styles associated with typical CO₂-bearing gas pools, Bohai Sea.

faults (Fig. 2). Lithospheric faults in the extended tectonic environment have strong effects on mantle degasification (Tao et al., 2005); the Tanlu faults are important conduits for rising CO_2 (Zhang, 1991; Hou et al., 1996; Qiu and Wang, 2006).

3 Materials and Methods

We obtained data from previous studies and used those data to interpret the fault system of the study area using corrections from more than 40 seismic sections to determine seismic horizons on the basis of drilling calibration, and to analyze the fault system by using information on the Moho and regional deep faults to extend seismic identification of fractures to depth. The free-space gravity data used to determine the Moho were derived from the global satellite gravity anomaly database (Smith and Sandwell, 1997; Sandwell et al., 2014). The database is maintained by the Scripps Marine Association of the University of California and the Satellite Altimetry Laboratory of the National Oceanic and Atmospheric Administration, in which the gravity data (V23.1) consists of 1' \times 1' grids. The data grid size is 1.402–1.484 km latitudinally and 1.850-1.851 km longitudinally. The overall accuracy of the gravity data can reach 3.03×10^{-5} m/s², and can be 1.8×10^{-5} m/s² locally. Bathymetric data (V18.1) were used as depth data to correct gravity data; the source and parameters of these data are the same as those of the gravity data. Data for Cenozoic sedimentary rocks were obtained from the literature (Zhu and Mi, 2010). Inversion of Moho depth was calculated using the Parker method, and was used to calculate the depth of the Moho.

4 Results

4.1 Crust-mantle fault system

Four types of fault are developed in the Bohai Sea: lithospheric, crustal, basement and cap-rock faults. These faults constitute the main conduit system for migration of mantle CO_2 from depth to shallow regions in the Bohai Sea. The characteristics of the various fault types are described below.

A lithospheric fault occurs in the Yingkou-Weifang fault zone (the middle-segment of the Tanlu fault zone). This fault is a large NE strike-slip fault (Fig. 3). The fault was active from the Mesozoic to the Ouaternary. From the distribution of Cenozoic basic rocks and data from the Donggou-Dongwu geological transect, the fault cuts through the deep part of the lithosphere. The fault exhibits a negative flower structure in profile, reflecting tensile-slip stress. However, there is a significant difference between the south and the north of the Bohai Sea. In the north, the negative flower-like faults in the Liaodong uplift, Liaodong Bay, cut the top boundary of the Mesozoic strata, have some impact on the lower Cenozoic strata, and do not affect the Neogene Minghuazhen Formation. The normal faults in the Liaodong uplift affect the strata from the Shaheije Formation to the Pingvuan Formation (Fig. 4a). In the area of the Miaoxi sag and Miaoxinan uplift in the south, negative flower-like faults cut Mesozoic and Cenozoic strata. The major fracture was inherited, and the fault activity lasted until the Quaternary (Fig. 4b).

Crustal faults (the Bozhang fault zone) also occur in the study area. This zone strikes northwest and nearly eastwest and consists of more than ten segments, including the Qinnan No. 2 fault, Shijiutuo No. 1 fault and Bonan No. 1 fault. From deep reflection seismic profiles (e.g. profile J003), the faults penetrate to the middle-lower crust, and exhibit listric fault characteristics. A negative flower structure occurs in Cenozoic strata (Yang et al., 2009), indicating that the faults formed in a tensional environment; the faults display local tensile slip.

Basement faults in the study area can be divided into NE-trending, nearly EW-trending and NW-trending types. Although most of the fractures only cut the upper part of the basement, they control the thickness of the Paleogene strata to some extent.

In the study area, thick mudstone is developed in the lower part of the Minghuazhen Formation. This mudstone



Fig. 3. Fault systems of the Bohai Sea area (after Qi et al., 2010; Zhu et al., 2010; Ren et al., 2013).

is a regional cap rock for shallow secondary hydrocarbon accumulations. The smearing effect of faulting makes the mudstone a better cap rock. The cap-rock faults run nearly EW to ENE. In most cases, the planar extension of the fault is less than 10 km, and the fault often runs oblique to a deep fault. Most cap-rock faults only cut the Minghuazhen and Pingyuan formations, and the fault displacement is small.

4.2 Deep structural characteristics

In this section, the Moho depth and the characteristics of the lithosphere and thermal structure, which are closely related to mantle CO_2 , are considered.

The depth of the Moho discontinuity is between 23 and

35 km in the study area. The average depth is 29 km, which is the shallowest Moho depth in the North China platform, and is typical of continental crust in general. The Moho depth is shallower in the southern Bohai Sea than in the northern part of the area. There is a local area of uplift in the Bozhong region, where the shallowest Moho depth is 23 km. This area is approximately 80 km wide and approximately 180 km long. The trend of this area, like that of the Tanlu fault, is NE. There is a local area of uplift trending nearly EW in the western part of the area, with a minimum depth of 26–29 km (Fig. 5). On the basis of data from the Donggou–Dongwu geological transect, in the lower reaches of the Liaohe fault depression, the depth of the top boundary upwarp of the asthenosphere is 95–100





Qp-Pingyuan Formation, $N_{1-2}m$ -Minghuazhen Formation, N_{1g} -Guantao Formation, E_{3d} -Dongying Formation, E_{2-3s} -Shahejie Formation, E_{1-2k} -Kongdian Formation, Mz-Mesozoic, Pz-Paleozoic, Pz-Proterozoic, Ar-Archean. (a) Guoliaoxi uplift-Qinnan sag-Liaoxi Uplift-Liaozhong sag-Liaodong sag-Bodong sag profile (Fig. 1); (b) Guoqinnan sag-Shijiutuo uplift-Bozhong sag-Miaoxi sag profile (Fig. 1).

km (Lu, 1992), and the depth of the Moho upwarp is 30 km. The Moho depth in the Bozhong sag is shallower, and the depth of the underside of the lithosphere should be less than that in the lower reaches of the Liaohe fault depression.

The study area contains high-conductivity layers in the upper mantle and the middle-lower crust, with depths of 17 and 45–50 km (Ma et al., 1984). Therefore, we hypothesize that there is a magma chamber in this area. The average value of heat flow in the study area is 63.2 mW/m², markedly lower than that in the Bozhong sag (98.9 mW/m²) (Wu et al., 1988). The ratio of mantle heat

flow to surface heat flow is about 60%, and the geothermal structure of the lithosphere is of a hot mantle and cold crust (Peng and Zou, 2013). This information indicates that the Bozhong area is a hot active area with strong thermal convection, which is conducive to the transport of mantle materials (e.g., CO_2) to the crust.

5 Discussion

To study the distribution pattern of mantle CO_2 , it is necessary to consider the source, migration path, migration time, traps and other factors. The first two of these factors are the most important, but have not received sufficient attention, as discussed in detail below.

5.1 The source area of mantle CO₂

There are two main sources of mantle CO₂: magmatic degassing (Frey and Lange, 2011; Benvenuti et al., 2013;



Fig. 5. Map of Moho depth in the Bohai Sea.

Pichavant et al., 2013; Girona et al., 2014) and thermal decomposition of carbonate minerals in the lithosphere (Aiuppa et al., 2004; Carter and Dasgupta, 2015). Magmatic degassing appears to be the dominant process. Magmatic degassing is mainly controlled by pressure: pressure decreases as magma rises, and the content of free CO_2 will increase as a result of exsolution when the content of CO_2 in the magma exceeds its solubility. Magma may rise with a mantle plume, be emplaced into the crust, or move upward as part of magma eruption (Tao et al., 1999; Berkesi et al., 2012).

All these processes are accompanied by asthenosphere uplift. Data from the Donggou-Dongwu geological transect indicate that areas of mantle uplift correspond to areas with underlying uplift of the asthenosphere (Lu, 1992), which can be confirmed by identifying areas with uplift of the Moho. As the density of free CO_2 is much lower than the density of magmatic melt, CO_2 will accumulate in relatively high areas, such as the uplifted part of the asthenosphere, under the effect of buoyancy. Therefore, analyses of CO_2 source areas can concentrate on asthenospheric bulges, i.e., areas with Moho uplift (Figs. 5–6).

In the Bohai Sea, the greatest uplift occurs in the Bozhong sag and the surrounding area, as inferred from analysis of the Moho depth. Therefore, the main accumulation area of mantle CO_2 is the deep part of the area; this area can continuously provide free CO_2 , so the hydrocarbon reservoir contains high levels of supercritical CO_2 (Zhu et al., 2017).



Fig. 6. Model of the mantle to crust pathway system and distribution of mantle CO_2 in the Bohai Sea. The structural characteristics of the upper mantle and lower crust were obtained from Lu et al. (2008).

5.2 Pathway system for movement of mantle CO₂

Movement of mantle CO₂ from a source to a sink requires a smooth crust-mantle pathway system (Fu and Song, 2005; Lu et al., 2008; Kotarba et al., 2014). The main components of such pathways are fault systems and magma conduits. CO2 can migrate upwards along subcrustal faults, detachment zones and listric faults (Guo et al., 2006; Li X Q et al., 2008); intersection of multiple groups of faults is more favorable for CO₂ migration (Ding et al., 2004). During neotectonic movement in the Bohai Sea, active faults would have been the main migration path for CO₂ (Li et al., 2004, 2012). Comprehensive analysis of seismic and geoscience data has demonstrated that the deep faults associated with mantle CO₂ in the Bohai Sea are flower-like, negative flower-like and listric normal faults (Fig. 2). Flower-like structures usually exhibit a small dip angle in the top section and a larger dip angle in the lower section. These conduits may be steeper in their lower part and less steep in their upper part, which may facilitate vertical migration at depth and lateral migration in shallower regions. In particular, when listric faults are developed at shallow depths, the horizontal migration distance may be longer, such as in wells QHD29-2a (Fig. 2b) and BZ3-1 (Fig. 6). Therefore, the fault style in the crust-mantle region may directly affect the horizontal migration distance of CO₂, thus influencing the planar distribution of CO₂. In addition, high-angle magma conduits are developed in the upper mantle and lower crust, forming important components of the deep pathway system and playing a role in vertical migration. Large unconformity surfaces in cap rock within the basin and zones of high porosity and permeability in the strata are also important migration conduits for CO_2 and play a major role in horizontal migration.

5.3 Relationship of mantle CO₂ with volcanic rocks

Large amounts of Cenozoic volcanic rocks occur in the Bohai Sea, and are found in almost all areas of sag and uplift. Drilling data reveal that medium-thin-layered basic rocks mainly occur in the Dongying and Guantao formations; these are mainly effusive rocks. The Shahejie and Kongdian formations are dominated by medium-thick -layered basic rocks and intermediate-acidic rocks, mostly of eruptive and effusive facies (Fig. 7). The volcanic rocks are dominated by basalt. The Paleogene basalts in the study area were derived from slightly enriched mantle that was similar in composition to original mantle (Hou et al., 2003), and the magma chamber could have provided mantle CO_2 .

Because of the limited duration of volcanic activity, it is difficult to preserve a large amount of CO_2 that is released in a short time. In addition, for modern volcanoes most of the gases released by volcanic eruptions enter the atmosphere, and only small amounts of gas are preserved in pores or inclusions at low concentrations (Berkesi et al., 2012). Therefore, the existence of a large-scale CO_2 reservoir remains to be confirmed. However, mantle CO_2 migrates upward during intermittent or a later period of volcanic eruption; this CO_2 migration may persist for a



Fig. 7. Cenozoic volcanic rocks in the Bohai Sea.

long time and with a large flux. For example, the CO₂ emission flux of the Julong, Hubin and Jinjiang hot springs in Changbai Mountain, Jilin Province, China, reaches 5.5 \times 10⁴ m³/a (Zhang et al., 2011), the CO₂ emission flux of several measuring points in the Tengchong volcanic area, Yunnan Province, can be up to 13.8×10^4 m³/a (Cheng et al., 2014), and there are insidious CO₂ input points in the Brimstone Basin in Yellowstone National Park (Bergfeld et al., 2012). Comparing the amount of CO₂ produced during volcanic activity with that produced during intermittent or a later period of volcanic eruption, the CO₂ produced during intermittent or a later period of volcanic eruption is more likely to enter a suitable trap and accumulate there. Therefore, it can be inferred that after intermittent or a later period of volcanic eruption, degassing of the magma chamber is the main source of mantle CO₂ in a reservoir.

From the relationship between the timing of volcanic eruption (the latest volcanism having occurred during deposition of the Guantao Formation, > 12.3 Ma) and CO₂ accumulation (5.1 Ma to the present) in the Bohai area, it can be inferred that magma degassing after volcanic eruptions was the main process of accumulation of mantle CO_2 , and was related to the timing of trap formation in the basin. Both volcanic rocks and CO2 are products of magmatic activity, and they are controlled by the same factors rather than one controlling the other. Theoretically, volcanic rocks and CO₂ can share the deep part of the same pathway system. The distribution patterns of basic igneous rock can provide some information on CO₂ distribution (Zheng et al., 2001; Wang et al., 2004; Zhang Y et al., 2008; Liao et al., 2012), but there is not necessarily a relationship between them. The distributions of volcanic rocks and CO₂ can intersect, but in most cases they do not, which may be related to the fact that the fracturing at the top of the pathway system after the volcanic period can extend farther along the strike.

5.4 Exploration significance

The occurrence of mantle CO_2 is mainly constrained by the asthenosphere/mantle uplift area, the crust-mantle fault systems and the associated traps. The former two are the main controlling factors. Therefore, the main factors to be considered to avoid the risk of CO_2 exploration are the characteristics of the CO_2 source area and the pathway system.

In the Bohai Sea, the Bozhong sag and the surrounding area is the main source area of mantle CO_2 (Fig. 1). This area has the shallowest Moho depth of the North China platform, reflecting the presence of asthenospheric uplift and the occurrence of abundant free CO2 after deep magmatism. This area has the most fertile source of mantle CO2 in the Bohai Sea. The area also contains the intersection of the NE-trending Tanlu fault belt and the NW- and EW-trending Bozhang fault belts. These deep faults underwent tensile-slip activity during the Quaternary and can form good transport pathways. Therefore, considering the characteristics of the source area and the pathway system, the Bozhong sag and the surrounding region are high-risk areas for accumulation of mantle CO₂, followed by the Qikou sag and adjacent area. CO_2 accumulations can only form when faults act as longdistance horizontal migration conduits. Other areas lack sources of mantle CO_2 , so the risk of CO_2 accumulation is limited.

Because of the high abundance of deep CO_2 , it can be dissolved in formation water in the migration path, so the formation water becomes acidic. This acidic water facilitates dissolution of acid-labile minerals; for example, feldspar is easily altered by CO_2 , transforming into authigenic dawsonite, kaolinite and quartz. Thus, cavernous-sieve dissolution pores and moldic pores are abundantly developed in sedimentary and volcanic rocks with acid-labile components (Zhou et al., 2012; Li and Li, 2017; Zhu et al., 2017). Dissolution pores or holes may form in limestone. Areas of CO_2 accumulation are often areas with strong secondary dissolution and are favorable for exploration for secondary quality reservoirs in deep strata (Jin et al., 2003; Li et al., 2021).

6 Conclusions

The distribution of mantle CO_2 in the Bohai Sea is mainly constrained by the area of asthenosphere/mantle uplift and the crust-mantle fault system. The area of mantle uplift is the source area for CO_2 accumulation. CO_2 migrates vertically through the upper mantle and lower crust. The fault style in the upper crust controls the horizontal and vertical migration of mantle CO_2 and affects the formation of CO_2 accumulations.

Four types of faults are developed in the Bohai Sea: lithospheric faults (the mid-segment of the Tanlu fault zone), crustal faults (the Bozhang faults), basement faults and cap-rock faults. These faults constitute the main conduit system for movement of mantle CO_2 from depth to shallow regions in the Bohai Sea.

There are two main Moho uplifts in the Bohai Sea: the Bozhong sag and the surrounding area, and the Qikou sag and the surrounding area. The trend of these areas, like that of the Tanlu fault, is NE. Moho uplift reflects uplift of the underlying asthenosphere and is associated with recognition of areas of accumulation of mantle CO_2 at depth.

In the Bohai Sea, the Bozhong sag and the surrounding area contain high concentrations of mantle CO_2 , particularly in a trap that formed prior to 5.1 Ma and is associated with crustal faults (the Bozhang faults) and lithospheric faults (the Tanlu faults).

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