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RECEIVED 22 May 2024 ACCEPTED 28 October 2024 PUBLISHED 07 November 2024

#### CITATION

Wang M, Zeng J, Li C, Qiao J, Wei W, Zhang H and Cui H (2024) The impacts of  $CO_2$  on sandstone reservoirs in different fluid environments: insights from mantle-derived  $CO_2$  gas reservoirs in Dongying Sag, Bohai Bay Basin, China. *Front. Earth Sci.* 12:1436573. doi: 10.3389/feart.2024.1436573

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# The impacts of $CO_2$ on sandstone reservoirs in different fluid environments: insights from mantle-derived $CO_2$ gas reservoirs in Dongying Sag, Bohai Bay Basin, China

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**Introduction:** Mantle-derived  $CO_2$ , as an important component of hydrothermal fluids, is widely distributed in petroliferous basins. While previous experimental studies have suggested that  $CO_2$  can improve sandstone reservoir quality through mineral dissolution in open fluid setting, they have overlooked its nagetive effects to sandstone reservoir quality by carbonate cementation. Additionally, the roles of various fluid environments in  $CO_2$ -reservoir interactions have not been studied in detail.

**Methods:** To systematically investigate the influences of  $CO_2$  on sandstone reservoirs, we examine a typical mantle-derived  $CO_2$  gas reservoir, Bohai Bay Basin, China. This study employs integrated methods, including electron microscopy, scanning electron microscopy, X-ray diffraction, stable C- and O-isotope analysis, and physical property data. The aim is to investigate the evidence and mechanisms by which mantle-derived  $CO_2$  impacts sandstone reservoirs, particularly focusing on its effects in open and closed fluid environments.

**Results and Discussion:** Our findings reveal that dawsonite and ankerite are prevalent within the mantle-derived CO<sub>2</sub> gas reservoir, while isotopic analysis of carbonate cements indicates values ( $\delta^{13}$ C: -9.0% to -1.6%;  $\delta^{18}$ O: -21.7% to -12.7%) consistent with mantle-derived CO<sub>2</sub> and hydrothermal fluids. These pieces of evidence indicate that CO<sub>2</sub>-rich hydrothermal fluids participate in water-rock interactions, thereby significantly influencing the diagenesis of reservoirs. Further, we notice that CO<sub>2</sub> reservoirs adjacent to faults exhibit an open fluid environment, characterized by superior porosity and permeability, more quartz, but fewer feldspar, carbonate, and clay minerals compared to those in closed fluid environments. Notably, kaolinite predominates in open fluid environments, while illite/smectite (I/S) is more common in closed settings. The dual roles of mantle-derived CO<sub>2</sub> are highlighted in our analysis: while it enhances reservoir storage and permeability through mineral dissolution, the carbonate cement generated by CO<sub>2</sub>-water-rock interaction can also adversely affect reservoir quality. In open fluid environments, CO<sub>2</sub> facilitates

the dissolution of feldspar and carbonate minerals, promoting the timely removal of dissolution by-products (clay mineral) and inhibiting carbonate cementation, thereby improving reservoir properties. Conversely, in closed fluid environments, decreasing  $CO_2$  concentrations with depth leads to diminishing dissolution effects and increased carbonate cementation, resulting in reduced reservoir porosity and permeability. Overall, the significance of this study is to correct the deviation in the impacts of  $CO_2$  on sandstone reservoirs at laboratory setting through case study of typical mantle-source  $CO_2$  gas reservoir.

This work can be applied to the studies of reservoir homogeneity and sweet spots in regions with hydrothermal and mantle-derived  $CO_2$  activities. However, due to the limitation of  $CO_2$  content range (about 15%–70%) in the study case, we are unable to investigate the effects of low-concentration  $CO_2$  on sandstone reservoirs, which may affect the generalizability of this work. Besides, the formation temperature and pressure, and salinity of formation water, should be considered when dealing with other cases.

KEYWORDS

 $\rm CO_2\text{-}water\text{-}rock$  interaction, fluid environment, mantle-derived  $\rm CO_2$ , diagenesis, Dongying Sag

## **1** Introduction

Hydrothermal activities have occurred in petroliferous basins during their formation and evolution (Zhang et al., 2023; Zhu et al., 2017). Mantle-derived CO<sub>2</sub>, as a primary component of hydrothermal fluids (Diker et al., 2024; Liu et al., 2024; Ranta et al., 2023; Sun and Dasgupta, 2023), significantly influences reservoir quality, and hydrocarbon expulsion and migration (Dmitriyevskiy et al., 2010; Guo et al., 2011; Zhu, et al., 2017; Liu et al., 2017; Lu et al., 2022; Huang et al., 2021). Furthermore, basins with rift tectonic background frequently experience hydrothermal activities, leading to the development of CO2-rich oil and gas fields, such as The South Atlantic Ocean margin (Anderson et al., 2023; Liu et al., 2022; Wright, 2020), Hailar (Yang et al., 2021; Liu et al., 2009), Songliao (Liu N. et al., 2019; Yang et al., 2009), Bohai Bay (Miao et al., 2020), Subei (Liu J. et al., 2023; Zhou et al., 2020; Liu et al., 2017), Sanshui (Zhu et al., 2017), Yinggehai (Liu Q. et al., 2019; Yang et al., 2023), and other rift basins in eastern China (Zhao et al., 2017). This mantle-derived CO<sub>2</sub> migrates into the reservoirs and dissolves in formation water, generating  $H^+$  and  $CO_3^{2-}$  (Li et al., 2015), where  $H^+$  leads to the dissolution of aluminosilicate and carbonate minerals (Portier and Rochelle, 2005), while CO32- combines with ions like Ca2+, Mg<sup>2+</sup>, and Fe<sup>2+</sup> (Li et al., 2015), forming carbonate minerals that occupy pore space within the reservoirs. Therefore, in regions with hydrothermal activity, as well as high CO2 concentrations, CO<sub>2</sub> is one of the critical factors that cannot be overlooked in contributing to reservoir heterogeneity. However, previous studies on mantle-derived CO2 have mostly emphasized its positive effects on reservoirs (Liu Q. et al., 2023; Yuan et al., 2023; Liu et al., 2016; Zhou et al., 2020; Zhang et al., 2018), with little evidence showing its participation in carbonate cementation that could reduce reservoir quality (Hu, 2016). As a result, the overall impacts of mantle-derived CO<sub>2</sub> on reservoirs has not been comprehensively understood.

Most studies of CO2-water-rock reaction experiments suggest that the dissolution of minerals by CO2 does not always enhance reservoir porosity and permeability. In these experiments, the dissolution of feldspar and carbonate minerals by CO<sub>2</sub> increases the porosity of the core plugs (Wu et al., 2019; Fu et al., 2016; Yu et al., 2016; Fischer et al., 2010), while the permeability typically decreases as the pore throats are blocked by dissolution by-products, such as clay minerals (Pearce et al., 2019; Wu et al., 2019; Aminu et al., 2018). In the geological conditions, due to frequent fault activities in rift basins, reservoirs near faults often have relatively open fluid environments during geological history (Gudmundsson, 2022; Ward et al., 2016; Zhou et al., 2020; Wei et al., 2017; Li et al., 2016). The open fluid environments are conducive to the timely discharge of dissolution by-products, leading to significant improvements in reservoir porosity and permeability (Hu, 2016; Wei et al., 2017; Yuan et al., 2013a). However, most oil and gas reservoirs actually have a stable pressure system, and the fluid environment is relatively closed, making fluid migration and discharge of dissolution by-products relatively difficult (Yuan et al., 2013a; Zhao et al., 2016). Currently, there is a lack of systematic study on the impacts of mantle-derived CO<sub>2</sub> on reservoirs in different fluid environments, which, to some extent, limits the research on the reservoir homogeneity and the prediction of sweet spots in regions with hydrothermal activity, as well as high CO<sub>2</sub> concentrations.

Dongying Sag, located in Bohai Bay Basin, is characterized by numerous anticline oil and gas traps that have been modified by normal faults and are rich in mantle-derived  $CO_2$  (Miao et al., 2020; Li et al., 2016). The mantle-derived  $CO_2$  has unique stable carbon isotope compositions (Dai et al., 2001), which can contribute to confirming if the mantle-derived  $CO_2$  participates in water-rock interactions and affects reservoirs (Liu R. et al., 2019; Hu, 2016; Liu et al., 2016). Therefore, Dongying Sag is an ideal place to research the synergistic impacts of fluid environments and  $CO_2$ on reservoirs.



FIGURE 1

(A) Distribution of rift basins and  $CO_2$  gas fields in eastern China [modified from Zhao et al. (2017)]. (B) Distribution of  $CO_2$  gas fields in Dongying Sag, the top structure,  $CO_2$  content distribution, and migration map of Pingfangwang  $CO_2$  gas reservoir (modified from the Shengli Oilfield). (C) Profile map (see Figure 1B for location) and  $CO_2$  source of study area (modified from Zeng, 2000). (D) Stratigraphy, lithology, tectonic movements, and reservoir distribution in the Dongying Sag [modified from Wei et al. (2017)].

This study examines typical mantle-derived  $CO_2$  gas reservoirs in Dongying Sag as a case study, aiming to better understand the impacts of mantle-derived  $CO_2$  on sandstone reservoirs in open and closed fluid environments using a multi-technique approach of combined mineralogy (Thin section, Scanning electron microscopy, and X-ray diffraction) and geochemistry (stable C- and O-isotopes). The main objectives of the current study are:

- To identify the mineralogical and geochemical evidence for the impacts of CO<sub>2</sub> on reservoirs.
- (2) To investigate the characteristics of diagenetic minerals in mantle-derived CO<sub>2</sub> reservoirs and their impact on the sandstone reservoirs.
- (3) To elucidate the impact mechanisms of CO<sub>2</sub> on sandstone reservoirs, with a focus on the impact laws of CO<sub>2</sub> on reservoirs in different fluid environments.

## 2 Geological setting

Due to the influence of the Pacific Plate's subduction, oil and gas fields rich in mantle-derived  $CO_2$  are widely distributed in the rift basins of eastern China (Zhao et al., 2017) (Figure 1A). The Dongying Sag, located in the southeastern of the Bohai Bay Basin, is a graben of Mesozoic and Cenozoic (Niu et al., 2022). Along the Gaoqing-Pingnan deep fault in the Dongying Sag, there are several  $CO_2$  gas fields (Han et al., 2010) (Figure 1B). These  $CO_2$  mainly migrate from the mantle to sedimentary basins through magmatic activity and deep faults (Figure 1C) (Zhang et al., 2023; Guo et al., 2006), have special stable carbon isotope compositions (with  $\delta^{13}C$  mostly around  $-6\% \pm 2\%$ ) (Dai et al., 2001).

Since the Cenozoic, the Gaoqing-Pingnan deep fault has been active, accompanied by multiple magmatic and mantle-derived  $CO_2$  activity events, forming multiple sets of  $CO_2$ -rich reservoirs (Figure 1D) (Kang et al., 2014; Cheng et al., 2020). According to drilling data, the Cenozoic stratigraphic unit includes the Kongdian Formation (Ek), Shahejie Formation (Es), Dongying Formation (Ed), Guantao Formation (Ng), Minghuazhen Formation (Nm), and Pingyuan Formation (Qp) (Figure 1D). Sediments of the Cenozoic strata in the Dongying Sag include the saline lake-delta-fluvial (the rifting stage) and fluvial (the depression stage) (Niu et al., 2022).

Pingfangwang CO<sub>2</sub> gas field, the typical mantle-derived CO<sub>2</sub> gas reservoir selected in this study, is an anticlinal trap that has been reformed by normal faults (Figure 1B). The target layer is a set of beach-bar sand in the 4th member of Shahejie Formation (Es<sup>4</sup>) (Figure 1D). Its natural gas is mainly composed of CO<sub>2</sub>, with a content of 61.2%–98.6%, CO<sub>2</sub> content decreases from south to north (Figure 1B). The  $\delta^{13}C_{CO2}$  ranges from -5.08‰–-4.32‰. CO<sub>2</sub> from the mantle first migrates vertically to the target layer through the Gaoqing-Pingnan deep fault and then migrates northward through some nearly south-north trend normal faults (Figures 1B, C) (Wang et al., 2004; Zeng, 2000).

## 3 Data and methods

To investigate the characteristics of mantle-derived  $CO_2$  reservoirs in the study area, 1,343 sets of porosity and permeability data and 46 sandstone samples of target layers from 4 wells

(represented as red circles in Figure 1B) were collected from the PEDRI, Shengli Oilfield. Four types of experiments were performed on these sandstone samples. 21 casting thin sections were observed using an optical microscope. Selected typical samples underwent scanning electron microscopy. X-ray diffraction analysis was conducted on 15 groups for whole rock and clay minerals. Additionally, C and O isotope analysis was carried out on a total of 10 sandstone samples and carbonate veins.

### 3.1 Analysis of thin sections

Thin sections were impregnated with blue epoxy resin to highlight the pore space and were dyed with Alizarin Red-S and Kferricyanide for carbonate mineral identification. Calcite was dyed red, ferrocalcite was dyed purplish red, and ankerite was dyed blue-purple. Thin sections were observed using the Leica 4500P polarizing microscope. ImageJ software was used for quantitative analysis of surface porosity.

### 3.2 Scanning electron microscopy (SEM)

The Quanta-200F SEM was used to observe the fresh surface of gold-plated rock samples, facilitating the identification of diagenetic phenomena and minerals. The SEM was operated at an acceleration voltage of 20 kV and an emission current of 3 nA.

### 3.3 Whole rock and clay X-ray diffraction analysis (XRD)

Each group of samples was crushed and mixed evenly. A portion of the powder was used for whole rock XRD analysis, while the remainder was analyzed for clay XRD. This approach was adopted to minimize the effect of lithologic heterogeneity. The D8 Advance X-ray diffractometer was used for whole rock and clay X-ray diffraction analysis. This allowed for quantitative analysis of the types and contents of the whole rock and clay minerals in  $CO_2$  reservoirs.

### 3.4 Stable C- and O-isotope analysis

Eight whole rocks and two calcite vein samples were selected for C- and O-isotope analysis. A ThermoFisher MAT-253 stable isotope mass spectrometer was used to determine the  $\delta^{13}$ C and  $\delta^{18}$ O of carbonate cement.

## 3.5 Calculation of carbonate cement formation temperature

To confirm the correlation between carbonate minerals in  $CO_2$  reservoirs and mantle-derived  $CO_2$  with high crystallization temperature properties, we calculated the precipitation temperatures of carbonate minerals in mantle-derived  $CO_2$  gas

reservoirs using the Formula 1 established by Friedman and O'Neil (1977).

$$1000 * \ln \alpha_{carbonate} - _{water} \approx \delta^{18}O_{carbonate} - \delta^{18}O_{water} = 2.78 * 10^{6}/T^{2} - 2.89$$
(1)

Where  $\alpha_{carbonate\_water}$  is the fractionation coefficient,  $\delta^{18}O_{carbonate}$  is the  $\delta^{18}O_{PDB}$  value of carbonate, The  $\delta^{18}O_{V-SMOW}$  value of pore water in the study area is –2.55‰ (Han et al., 2012), T is the temperature of carbonate precipitation.

### 4 Results

## 4.1 Petrology characteristics and mineral compositions of CO<sub>2</sub> gas reservoirs

The identification results from the thin sections (Table 1) show that the terrigenous detrital components of  $CO_2$  reservoirs consist of quartz (average: 73.0%, range: 58.8%–86.8%), feldspars (average: 13.4%, range: 3.8%–23.7%), and rock fragments (average: 13.7%, range: 4.6%–24.3%). There is a significant difference between the reservoirs near the faults (wells A and B) and the reservoirs far away from the faults (wells C and D). The former has a higher quartz content and a lower feldspar content, with the lithology consisting of feldspathic litharenite and sublitharenite (Li et al., 2024), while the latter mainly consists of lithic arkose (Figure 2).

The XRD results for whole rock and clay minerals (Table 2) show that the mineral composition of the CO<sub>2</sub> sandstone reservoir in the study area is predominantly quartz, with an average of 58.2%, with a relatively low content of feldspar (K-feldspar and plagioclase). Moreover, carbonate minerals are widely developed, comprising an average content of 12.6% calcite, 6.0% ankerite, 3.1% dawsonite, and small amounts of siderite. Among them, Wells A, B, C, and D have average quartz contents of 69.8%, 67.1%, 47.3%, and 45.5%, respectively. The average contents of total carbonate minerals (calcite, ankerite, siderite, and dawsonite) are 10.2%, 11.6%, 36.6%, and 40.3%, respectively. The average contents of clay minerals are 7.1%, 6.9%, 9.0%, and 12.4%, respectively. As for clay minerals, kaolinite dominates with an average of 40.9%, followed by the illite/smectite (I/S) mixed layer, with an average of 35.3%. Among them, the average relative content of kaolinite in Wells A, B, C and D are 52%, 58%, 23%, and 27%, respectively, and the average relative content of I/S is 28%, 21%, 52%, and 42%, respectively.

The mineral composition exhibits a noticeable contrast between reservoirs near and far from faults (Figure 3). The former displays higher quartz content and lower content of calcite, ankerite, and clay minerals compared to the latter. Furthermore, the clay minerals in the reservoirs near faults are predominantly kaolinite, whereas these in the reservoirs far from the faults primarily consist of I/S.

# 4.2 Porosity and permeability of CO<sub>2</sub> gas reservoirs

According to the statistical results of porosity and permeability data (Figure 4), The porosity of  $CO_2$  reservoirs is concentrated

| Location        | Well            | Depth(m)     | The detrit | al grain volum  |                         | Su      | rface poros | sity    | Ratio of |
|-----------------|-----------------|--------------|------------|-----------------|-------------------------|---------|-------------|---------|----------|
|                 |                 |              | Quartz (%) | Feldspar<br>(%) | Rock<br>fragment<br>(%) | PSP (%) | DSP (%)     | TSP (%) | DSP (%)  |
|                 | А               | 1,464.80     | 86.4       | 8.6             | 4.9                     | 6.8     | 20.5        | 27.3    | 75.1     |
|                 | А               | 1,496.90     | 76.9       | 3.8             | 19.3                    | 9.2     | 26.0        | 35.2    | 73.9     |
|                 | А               | 1,531.49     | 85.2       | 9.3             | 5.6                     | 6.5     | 25.0        | 31.5    | 79.4     |
|                 | А               | 1,557.11     | 86.8       | 6.6             | 6.6                     | 7.4     | 22.0        | 29.4    | 74.8     |
|                 | А               | 1,582.64     | 71.4       | 11.9            | 16.7                    | 6.1     | 20.0        | 26.1    | 76.6     |
|                 | Mea             | ns of Well A | 81.4       | 8.0             | 10.6                    | 7.2     | 22.7        | 29.9    | 76.0     |
| Near faults     | В               | 1,442.00     | 76.4       | 9.9             | 13.7                    | 4.4     | 18.0        | 22.4    | 80.4     |
|                 | В               | 1,457.00     | 86.5       | 5.6             | 7.8                     | 5.6     | 20.0        | 25.6    | 78.0     |
|                 | В               | 1,472.26     | 73.3       | 10.0            | 16.7                    | 6.0     | 16.0        | 22.0    | 72.7     |
|                 | В               | 1,496.90     | 70.6       | 9.6             | 19.8                    | 8.5     | 18.4        | 26.9    | 68.5     |
|                 | В               | 1,518.70     | 69.8       | 12.9            | 17.3                    | 5.9     | 16.3        | 22.2    | 73.4     |
|                 | В               | 1,537.62     | 66.3       | 14.1            | 19.6                    | 7.8     | 13.8        | 21.6    | 63.9     |
|                 | Mea             | ns of Well B | 73.8       | 10.4            | 15.8                    | 6.4     | 17.1        | 23.5    | 72.8     |
|                 | С               | 1,503.00     | 75.4       | 20.0            | 4.6                     | 6.0     | 16.4        | 22.4    | 73.2     |
|                 | С               | 1,527.90     | 72.2       | 13.9            | 13.9                    | 4.9     | 11.5        | 16.4    | 70.1     |
|                 | С               | 1,543.40     | 65.7       | 10.0            | 24.3                    | 6.6     | 6.0         | 12.6    | 47.6     |
|                 | С               | 1,569.96     | 63.2       | 23.7            | 13.2                    | 1.3     | 7.0         | 8.3     | 84.3     |
|                 | С               | 1,626.80     | 58.8       | 23.5            | 17.6                    | 0.0     | 4.2         | 4.2     | 100.0    |
| Far from faults | Means of Well C |              | 67.1       | 18.2            | 14.7                    | 3.8     | 9.0         | 12.8    | 75.1     |
| Far from faults | D               | 1,460.23     | 75.0       | 18.8            | 6.3                     | 7.7     | 19.1        | 26.8    | 71.3     |
|                 | D               | 1,495.30     | 75.0       | 12.5            | 12.5                    | 6.0     | 12.5        | 18.5    | 67.6     |
|                 | D               | 1,512.80     | 67.9       | 16.7            | 15.4                    | 6.6     | 8.0         | 14.6    | 54.8     |
|                 | D               | 1,530.45     | 64.9       | 15.6            | 19.5                    | 5.9     | 5.0         | 10.9    | 45.9     |
|                 | D               | 1,556.30     | 64.7       | 23.5            | 11.8                    | 4.5     | 4.0         | 8.5     | 47.1     |
|                 | Mea             | ns of Well D | 69.5       | 17.4            | 13.1                    | 6.1     | 9.7         | 15.9    | 57.3     |
|                 | Means           |              | 73.0       | 13.4            | 13.7                    | 5.9     | 14.7        | 20.6    | 70.4     |

#### TABLE 1 Thin section identification results of CO<sub>2</sub> gas reservoirs in the study area.

Abbreviation: Q-quartz; F-feldspar; RF-rock fragment; PSP-primary surface porosity; DSP-dissolution surface porosity; TSP-total surface porosity.

between 20% and 30% (Figure 4A), with an average of 23.6%. The permeability of  $CO_2$  reservoirs near faults is mainly distributed in the range of 10 ~ 1,000 mD, with an average of 131.7 mD, while  $CO_2$  reservoirs far from faults have permeability mainly distributed in

the range of 1 ~ 100 mD, with an average of 24.3 mD (Figure 4B). As a whole, the physical properties of reservoirs near faults is better than that far from faults (Figure 4). The linear correlations between the porosity and permeability of  $CO_2$  reservoirs are fair (Figure 4C),



suggesting that the pores are the main seepage channels in  $\mathrm{CO}_2$  reservoirs.

### 4.3 Typical diagenesis in CO<sub>2</sub> gas reservoirs

### 4.3.1 Carbonate mineral cementation

Carbonate cementation is the predominant cementation process of  $CO_2$  reservoirs in the study area, mainly developing dawsonite (Figures 5A–C), calcite (Figures 5C–F), ankerite (Figures 5C, F, G), and a small amount of siderite (Figures 5D, H) and dolomite (Figure 51).

#### 4.3.1.1 Dawsonite

Dawsonite, a special carbonate mineral, can stably exist in highconcentration  $CO_2$  environments. It is generated by the continuous reaction of K-feldspar, plagioclase, or kaolinite with  $CO_2$  and the formation water (Ryzhenko, 2006; Johnson et al., 2011). Observation of thin sections and SEM confirms that all  $CO_2$  gas reservoirs have dawsonite, with extremely high recognition. It is filled in the pores of sandstone, forming a hairlike or radial aggregate with a size of approximately 100–200 µm (Figures 5A–C).

#### 4.3.1.2 Calcite

Calcite is the most carbonate mineral in  $CO_2$  reservoirs (Table 1), with pack-pore cementation (Figure 5C) or filling dissolution pores (Figure 5D). It frequently coexists with kaolinite (Figure 5E), indicating that these calcites formed after  $CO_2$  injection into the reservoir. The coexistence of kaolinite and calcite is due to the acidic environment generated by  $CO_2$ , which causes feldspar to dissolve and form kaolinite. Later, the geological environment changes to alkaline, allowing calcite to precipitate within the pre-existing dissolution pores. Notably, some dissolution pores have not yet been incompletely filled (Figure 5D).

#### 4.3.1.3 Ankerite

Ankerite is commonly developed in  $CO_2$  reservoirs, filling dissolution pores and intergranular pores (Figures 5C, F, G). Some ankerites replaced calcite (Figure 5F), indicating that the formation time of ankerite was later than that of calcite.

### 4.3.1.4 Siderite and dolomite

Additionally, the  $CO_2$  reservoir contains minor amounts of authigenic siderite (Figures 5D, H) and dolomite (Figure 51). The dolomite was only observed under the microscope but not detected in the whole rock XRD analysis (Table 2).

#### 4.3.2 Dissolution

The CO<sub>2</sub> reservoir in the study area exhibits intensive dissolution of feldspars, rock fragments, and calcites, forming numerous dissolution pores (Figure 5J-L). Remarkably, certain feldspar particles and calcite have undergone near-complete dissolution, forming mold pores (Figures 5J, K). Although some of the dissolution products, such as kaolinite and authigenic quartz, are filled in the dissolution pores (Figure 5L), and the carbonate minerals in the later stage are partially or completely filled in the dissolution pores (Figures 5C-E), the identification results of CO<sub>2</sub> reservoir thin sections show that the dissolution pores in the CO<sub>2</sub> reservoir still occupy an absolutely dominant position, with an average dissolution surface porosity of 14.7% and an average proportion of 70.4% (Table 1). In addition, the average dissolution porosity in the reservoirs near the faults is 19.4%, while the average dissolution porosity of reservoirs far from the faults is 9.4%. Obviously, the former is much greater than the latter.

# 4.4 The characteristics of carbon and oxygen isotopes in CO<sub>2</sub> gas reservoirs

Analysis results from carbonate veins and whole rock carbon and oxygen isotopes (Table 3) show that the  $\delta^{13}$ C of carbonate minerals in mantle-derived CO<sub>2</sub> gas reservoirs are -9.0%--1.6%, with an average of -5.2%, and the  $\delta^{18}$ O are -21.7%--12.7%, with an average of -17.0%. The average precipitation temperature of carbonate minerals in the study area was 121.9°C, which is much higher than the formation temperature.

## **5** Discussions

## 5.1 Evidence of the impact of mantle-derived CO<sub>2</sub> on reservoirs

 $CO_2$  from the mantle has a unique range of C- and O-isotopes and high-temperature properties of mantle-derived fluids (e.g., Morishita, 2023; Ewa et al., 2012; Jin et al., 2013; Hou et al., 2019), and following prolonged interactions with the formation water and rocks, it ultimately generates carbonate minerals (e.g., Johnson et al., 2011; Harrison et al., 2019; Wei, et al., 2023). Thus, the impact of  $CO_2$  on reservoirs can be confirmed based on the carbonate minerals generated during the mantle-derived  $CO_2$ -water-rock interactions and their isotopic characteristics.

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| Well               |                 | Depth    |        |                | Contei     | nt of whole | Content of whole-rock minerals (%) | als (%)  |           |      | Relat | ive con | tent of | clay mi | Relative content of clay minerals (%) |
|--------------------|-----------------|----------|--------|----------------|------------|-------------|------------------------------------|----------|-----------|------|-------|---------|---------|---------|---------------------------------------|
|                    |                 | Ê        | Quartz | K-<br>feldspar | Plagioclas | Calcite     | Ankerite                           | Siderite | Dawsonite | TCCM | I/S   |         | ×       | υ       | Ratio of<br>I/S                       |
| 7                  | A 1             | 1,464.80 | 81.5   | 8.7            | 0.0        | 0.0         | 1.0                                | 0.0      | 0.2       | 7.1  | 26    | 11      | 59      | 4       | 45                                    |
| 7                  | A 1             | 1,496.90 | 70.6   | 10.2           | 0.0        | 5.6         | 1.6                                | 1.3      | 4.7       | 6.0  | 24    | 16      | 48      | 12      | 50                                    |
| 7                  | A 1             | 1,531.49 | 64.4   | 7.2            | 5.7        | 3.3         | 1.7                                | 1.1      | 8.5       | 8.1  | 30    | 11      | 52      | 2       | 40                                    |
| 7                  | A 1             | 1,557.11 | 62.6   | 9.8            | 8.8        | 8.0         | 2.9                                | 0.7      | 0.0       | 7.2  | 32    | 15      | 49      | 4       | 35                                    |
|                    | Means of Well A | f Well A | 69.8   | 9.0            | 3.6        | 4.2         | 1.8                                | 0.8      | 3.4       | 7.1  | 28    | 13      | 52      | ~       | 43                                    |
| Near faults<br>F   | B               | 1,442.00 | 62.3   | 9.4            | 7.4        | 6.8         | 1.0                                | 6.0      | 4.1       | 8.1  | 32    | 15      | 46      | 2       | 50                                    |
| I                  | B               | 1,457.00 | 75.0   | 8.2            | 8.0        | 3.8         | 0.8                                | 0.0      | 0.0       | 4.2  | 8     | 6       | 82      | 4       | 55                                    |
| I                  | B               | 1,496.90 | 71.1   | 5.1            | 9.5        | 5.9         | 1.8                                | 0.0      | 0.0       | 6.6  | 19    | 14      | 57      | 10      | 35                                    |
| I                  | B               | 1,537.62 | 60.1   | 6.4            | 3.6        | 10.4        | 7.7                                | 0.4      | 2.9       | 8.5  | 24    | 19      | 48      | 6       | 50                                    |
|                    | Means of Well B | f Well B | 67.1   | 7.3            | 7.1        | 6.7         | 2.8                                | 0.3      | 1.8       | 6.9  | 21    | 14      | 58      | 8       | 48                                    |
| )                  | C               | 1,503.00 | 67.1   | 8.7            | 6.0        | 6.3         | 4.3                                | 6.0      | 3.4       | 8.4  | 63    | 10      | 15      | 12      | 50                                    |
| )                  | C 1             | 1,527.90 | 56.3   | 8.4            | 1.2        | 15.8        | 4.6                                | 0.3      | 3.5       | 9.9  | 43    | 14      | 38      | 5       | 45                                    |
| )                  | C               | 1,569.96 | 21.8   | 2.5            | 0.0        | 9.8         | 50.8                               | 1.3      | 6.6       | 7.2  | 52    | 22      | 23      | 3       | 55                                    |
| )                  | C               | 1,626.80 | 43.8   | 6.7            | 0.0        | 35.1        | 3.8                                | 0.0      | 0.0       | 10.6 | 50    | 20      | 14      | 16      | 55                                    |
| Far from<br>faults | Means of Well C | f Well C | 47.3   | 6.6            | 0.5        | 16.8        | 15.9                               | 0.6      | 3.4       | 9.0  | 52    | 17      | 23      | 6       | 51                                    |
| I                  | D 1             | 1,512.80 | 57.8   | 7.6            | 1.3        | 11.6        | 5.4                                | 0.6      | 3.2       | 12.5 | 43    | 25      | 19      | 13      | 45                                    |
| I                  | D 1             | 1,530.45 | 34.7   | 7.4            | 1.1        | 26.8        | 3.2                                | 2.3      | 10.0      | 14.5 | 39    | 23      | 30      | 8       | 45                                    |
| Ι                  | D               | 1,556.30 | 44.1   | 5.4            | 0.0        | 40.3        | 0.0                                | 0.0      | 0.0       | 10.2 | 44    | 17      | 33      | 9       | 55                                    |
|                    | Means of Well D | f Well D | 45.5   | 6.8            | 0.8        | 26.2        | 2.9                                | 1.0      | 4.4       | 12.4 | 42    | 22      | 27      | 6       | 48                                    |
| Me                 | Means           |          | 58.2   | 7.4            | 3.2        | 12.6        | 6.0                                | 0.7      | 3.1       | 8.6  | 35.3  | 15.9    | 40.9    | 8.0     | 47.3                                  |



Mineral compositions of CO<sub>2</sub> gas reservoirs near and far from faults in the study area. (A) Histogram of mineral whole-rock content. (B) Histogram of relative content of clay minerals. TCCM-total contents of clay minerals.



### 5.1.1 Evidence from carbonate minerals

Dawsonite is the product of the long-term reaction between high-concentration mantle-derived  $\mathrm{CO}_2$  and formation water,

as well as various feldspar (Equations 2–4), and is considered a "tracer mineral" for mantle-derived  $CO_2$  fluid migration and accumulation (Ryzhenko, 2006; Liu et al., 2009; Johnson et al.,



#### FIGURE 5

Pictures of carbonate cements and dissolution in CO<sub>2</sub> reservoirs. Picture (A) is derived from observations under cross-polarized light. Pictures (B, E, G) are derived from observations under single polarized light. (A) Dawsonite accompanied with authigenic quartz (B) Dawsonite; (C) Dawsonite, calcite (dyed red) and ankerite (dyed blue-purple) cement; (D) calcite cement (dyed red), dissolution pores filled with calcites and unfilled residual dissolution pore; (E) Paragenesis of calcite and kaolinite; (F) Ankerite and calcite replaced by ankerite; (G) Ankerite and issolution (feldspar, rock fragment, and calcite); (K) Coexistence of dawsonite, kaolinite and intensive dissolution (feldspar and rock fragment), some particles are completely dissolved and formed mold holes; (L) Coexistence of kaolinite; K, kaolinite; DR, dissolution of rock fragment; DF, dissolution of feldspar; C, dissolution of calcite; MP, mold pore.

| Well | Depth (m) | Sample types | δ <sup>13</sup> C <sub>PDB</sub> (‰) | δ <sup>18</sup> Ο <sub>ΡDB</sub> (‰) | FT (°C) | CPT (°C) |
|------|-----------|--------------|--------------------------------------|--------------------------------------|---------|----------|
| А    | 1,496.90  | Whole rock   | -6.4                                 | -17.6                                | 76.9    | 124.5    |
| А    | 1,557.11  | Vein         | -8.0                                 | -19.3                                | 79.2    | 145.3    |
| В    | 1,442.00  | Vein         | -4.8                                 | -12.7                                | 74.8    | 78.5     |
| В    | 1,457.00  | Whole rock   | -1.7                                 | -16.2                                | 75.4    | 109.6    |
| В    | 1,537.62  | Whole rock   | -4.2                                 | -19.3                                | 78.4    | 145.3    |
| С    | 1,527.90  | Whole rock   | -5.0                                 | -13.7                                | 78.1    | 86.6     |
| С    | 1,569.96  | Whole rock   | -1.6                                 | -16.3                                | 79.7    | 110.6    |
| С    | 1,626.80  | Whole rock   | -6.6                                 | -19.4                                | 81.8    | 146.6    |
| D    | 1,530.45  | Whole rock   | -5.0                                 | -14.2                                | 78.2    | 90.9     |
| D    | 1,556.30  | Whole rock   | -9.0                                 | -21.7                                | 79.1    | 181.0    |
|      | Mean      |              | -5.2                                 | -17.0                                | 78.1    | 121.9    |

TABLE 3 The carbon and oxygen isotope data of whole rock and veins and the precipitation temperature of carbonate minerals of CO<sub>2</sub> gas reservoirs.

Abbreviation: GT, formation temperature; CPT, carbonate precipitation temperature.

2011). In the study area, dawsonite cement is generally developed (Figures 5A–C, J, K), while only a small amount of plagioclase and K-feldspar are present (Table 2), suggesting that the reaction between feldspar and mantle-derived  $CO_2$  has generated dawsonite.

$$\begin{split} \mathrm{KAlSi_3O_{8(K-feldspar)} + Na^+ + CO_2 + H_2O &\rightarrow \mathrm{NaAlCO_3(OH)_{2(Dawsonite)}} \\ + \mathrm{3SiO_{2(Quartz)} + K^+} \end{split}$$

$$\begin{split} \text{NaAlSi}_{3}\text{O}_{8(\text{Albite})} + \text{CO}_{2} + \text{H}_{2}\text{O} &\rightarrow \text{NaAlCO}_{3}(\text{OH})_{2(\text{Dawsonite})} \\ &+ 3\text{SiO}_{2(\text{Quartz})} \end{split} \tag{3}$$

$$CaAl_{2}Si_{2}O_{8(Anorthite)} + 2Na^{+} + 3CO_{2} + 3H_{2}O$$
  

$$\rightarrow 2NaAlCO_{3}(OH)_{2(Dawsonite)} + CaCO_{3(Calcite)} + SiO_{2(Quartz)} + 2H^{+}$$
(4)

Moreover, ankerite is generally considered a kind of cement in the middle to late diagenetic stage (e.g., Oluwadebi et al., 2018; Ma et al., 2018; Han et al., 2012), with a high precipitation temperature, and the precipitation temperatures of ankerite in Dongying Sag are about  $110^{\circ}$ C– $135^{\circ}$ C (Han et al., 2012). The ratio of I/S in mantle source CO<sub>2</sub> reservoirs is mainly between 40% and 55% (Table 2), indicating that the current diagenetic evolution is still in the early to middle diagenetic stage. However, the theoretical burial temperature of CO<sub>2</sub> reservoirs is 74.8°C–81.8°C in the study area (Table 3) (Gong et al., 2013), which is much lower than the precipitation temperature of ankerite. That is to say, the formation of ankerite (Figures 5C, F, G) occurs in a hightemperature environment formed by hydrothermal fluids rich in CO<sub>2</sub>. And there is a close genetic relationship between ankerite and mantle-derived CO<sub>2</sub> in the study area.

### 5.1.2 Evidence from carbon and oxygen isotopes

According to the results of C- and O-isotopes analysis (Table 3), the  $\delta^{13}$ C values of carbonate minerals in the mantle-derived CO<sub>2</sub> reservoir range from -9.0‰--1.6‰, with an average of -5.2‰. Which basically fall within the range suggested for mantle-derived CO<sub>2</sub> (-8‰-4‰ VPDB; Morishita, 2023; Dai et al., 2001). Moreover, the precipitation temperature of carbonate minerals is generally much higher than the maximum theoretical formation temperature (Table 3). Furthermore, the carbon and oxygen isotope data were submitted to the genetic chart of carbonate cement in the Bohai Bay Basin established by (Wang and Zhang, 2001), which showed that carbonate cement in mantle-derived CO<sub>2</sub> reservoirs was mainly formed in a transitional environment of low-temperature hydrothermal and high-temperature magmatic-hydrothermal fluids (Figure 6; e.g., Hou et al., 2019).

In summary, evidence from minerals and carbon and oxygen isotopes suggests that mantle-derived  $CO_2$  indeed interacts with formation water and rocks, ultimately generating diverse carbonate cements precipitated in fractures and pores.

## 5.2 The dual effects of mantle-derived CO<sub>2</sub> on sandstone reservoirs

The mechanism of the impact of  $CO_2$  on reservoirs by previous studys mainly comes from  $CO_2$ -water-rock reaction experiments. In these experiments,  $CO_2$  fluids were injected into core plugs to make  $CO_2$ , water, and rock to fully react. And the porosity, permeability, and mineral changes of core plugs were measured preand post-experiment (e.g., Fischer et al., 2010; Aminu et al., 2018; Pearce et al., 2019; Wu et al., 2019; Fu et al., 2016). These studies generally indicate that dissolution is the main reaction between  $CO_2$  and rocks, resulting in an increase in porosity (Wu et al.,



2019; Fu et al., 2016; Yu et al., 2016; Fischer et al., 2010) and a decrease in permeability due to blockage of throats by by-products of dissolution (Pearce et al., 2019; Wu et al., 2019; Aminu et al., 2018). However, the open fluid environment in the formation is relatively limited and generally found near faults (Zhou et al., 2020; Wei et al., 2017). On the contrary, most oil and gas reservoirs feature relatively closed fluid environments, making fluid migration relatively difficult (Yuan et al., 2013a; Zhao et al., 2016). Additionally, various ions can also participate in the water-rock reactions (Ahmat et al., 2022; Portier and Rochelle, 2005; Li et al., 2015). Therefore, the previous works cannot fully elucidate the impact of  $CO_2$  on reservoirs.

In the high-temperature and high-pressure environment of the formation, the  $CO_2$ -water-rock reaction theoretically has dual effects on the sandstone reservoirs (Portier and Rochelle, 2005; Li et al., 2015). On the one hand, A large amount of  $CO_2$  dissolves in the formation water and forms weak acids (H<sup>+</sup>) (Equation 5), which then dissolve the carbonate minerals and aluminosilicate minerals, such as K-feldspar, plagioclase, and so on (Portier and Rochelle, 2005). On the other hand, the  $CO_3^{2-}$  formed in (Equation 5) combines with  $Ca^{2+}$ ,  $Mg^{2+}$ , and  $Fe^{2+}$  in the formation water to generate stable carbonate minerals (Equation 6; Li et al., 2015).

$$\mathrm{CO}_2 + \mathrm{H}_2\mathrm{O} \to \mathrm{CO}_3^{2-} + 2\mathrm{H}^+ \tag{5}$$

$$(Ca, Mg, Fe)^{2+} + CO_3^{2-} \rightarrow (Ca, Mg, Fe) CO_{3(Carbonates minerals)}$$
(6)

## 5.2.1 The impacts of dissolution by mantle-derived $CO_2$ on reservoirs

During the diagenetic evolution of reservoirs, dissolution is the key factor for good physical properties of oil and gas reservoirs (Ehrenberg and Baek, 2019). The dissolution in the mantle-derived  $CO_2$  reservoirs is extremely strong (e.g., Liu N. et al., 2023; Liu et al., 2016; Zhou et al., 2020). The intensive dissolution of feldspars, rock fragments, and calcites forms a large number of dissolution pores (Figure 5J–L), and even some feldspar and calcite almost completely dissolved to form mold pores (Figures 5J, K). As a result, the dissolution pores occupy the absolute dominant position in the pore system of the study area (Table 1). Therefore, the dissolution of feldspar and early calcite by  $CO_2$  in the study area greatly increases the storage space for oil and gas, and simultaneously, the seepage channels are also improved (Figure 4). Mantle-derived  $CO_2$  plays a crucial role in improving the physical properties of reservoirs in the study area.

## 5.2.2 The impacts of cementation by mantle-derived $CO_2$ on reservoirs

Cementation is the most destructive diagenesis in oil and gas reservoirs, and carbonate minerals are the most common types of cement and can negatively impact reservoir properties (Ketzer et al., 2003; Ehrenberg and Baek, 2019). In mantlederived CO<sub>2</sub> gas reservoirs, after a prolonged CO<sub>2</sub>-waterrock interaction, CO2 gas reservoirs will also precipitate carbonate cement such as dawsonite, calcite, siderite, ankerite, and dolomite (Figures 5A-I, e.g., Ahmat et al., 2022; Worden, 2006; Liu et al., 2009). These minerals will occupy the pore spaces (Figures 5A-I), leading to the deterioration of reservoir quality. There are fair negative correlations between the content of carbonate minerals, porosity, and permeability of mantlederived CO<sub>2</sub> reservoirs (Figures 7A, B), indicating that the cementation of carbonate minerals affected by CO<sub>2</sub> is the primary factor leading to the deterioration of CO2 reservoir quality (e.g., Ahmat et al., 2022).

In addition, dawsonite cements are commonly accompanied by authigenic quartz (Figure 5A) (Equations 2–4), and feldspar dissolution can generate authigenic quartz and diverse clay minerals, like kaolinite, I/S (Li et al., 2021). These authigenic quartz fill the pores and reduce the porosity of the reservoirs, while the clay minerals may block the pores and throats and decrease the permeability (Pearce et al., 2019; Wu et al., 2019; Aminu et al., 2018), which can to some extent decrease the quality of the reservoirs.

## 5.3 The impacts of mantle-derived CO<sub>2</sub> on reservoirs in different fluid environments

## 5.3.1 The impacts of mantle-derived $CO_2$ on sandstone reservoirs in open fluid environments

In oil and gas fields with faults, reservoirs near faults typically exhibit relatively good reservoir properties and relatively open fluid environments compared to reservoirs far from faults (Gudmundsson, 2022; Wei et al., 2017; Ward et al., 2016). The good reservoir properties are largely attributed to the dissolution effects of organic acids and the timely migration of dissolution by-products facilitated by formation fluids (Wuelstefeld et al., 2017; Zhou et al., 2020; Li et al., 2016). Similarly, the reservoirs near the faults in the study area all have good porosity and permeability (Figure 4; e.g., Hu, 2016; Wei et al., 2017). For example, the well A, located



near the fault, has an average porosity of 23.6% and an average permeability of 84.0 mD. According to the curves of average porosity and permeability, the vertical variation between porosity and permeability is relatively small (Figure 8). The dissolution of feldspar and early calcite in the reservoir is extremely intensive (Figure 8). The by-products of dissolution, clay minerals, are few and mainly composed of kaolinite (Figure 3). The carbonate cementation in Well A is weak, with only a small amount of other carbonate cement, except dawsonite that can stably exist in high CO<sub>2</sub> concentrations environment (Table 2).

Accordingly, In the open fluid environment near the fault in the study area, the by-products of feldspar dissolution (clay minerals and siliceous cement) are carried out from the reservoir by the flow of formation fluid (e.g., Wuelstefeld et al., 2017; Hu, 2016; Wei et al., 2017). In addition, the high-temperature environment provided by the influx of deep fluids also effectively intensifies the dissolution of feldspars and carbonate minerals (e.g., Wuelstefeld et al., 2017). All of these will lead to a significant improvement in reservoir quality. The abundant presence of kaolinite (Table 2) indicates that the fluid environment is acidic, which can inhibit the carbonate cementation (Boyer, 1983). Therefore, there is only a small amount of carbonate cement in the CO2 reservoirs with open fluid environments (Table 2). Although there may be some dawsonite in high-concentration CO<sub>2</sub> environments, it cannot prevent the overall trend of improving reservoir quality (Figure 8).

In summary, an open fluid environment facilitates the dissolution of reservoirs by  $CO_2$ , facilitating the removal of dissolution products from the reservoirs, and high  $CO_2$  concentration inhibits carbonate mineral cementation, greatly increasing reservoir porosity and permeability.

## 5.3.2 The impacts of mantle-derived CO<sub>2</sub> on sandstone reservoirs in closed fluid environments

In reservoirs located far from faults, the fluid environment is generally more closed, leading to limited fluid flow and complex diagenesis (Macaulay et al., 2001; Wang et al., 2017; Ahmat, et al.,

2022; Yuan et al., 2013b). Clay minerals assemblages were used to identify closed fluid environments (Azzam et al., 2023; Lv et al., 2022). In the study area, the mantle-derived CO<sub>2</sub> reservoirs far away from faults have rare plagioclase, massive carbonate minerals, and I/S (Table 2; e.g., Azzam et al., 2023; Lv et al., 2022), which may imply the relatively closed fluid environment and mineral formation process. Initially, CO2 entered the reservoirs to form an acidic environment, causing significant dissolution of plagioclase, generating dissolution pores and massive I/S. However, due to the difficulty of fluid flow in reservoirs far from faults, the dissolution by-products, such as Na<sup>+</sup>, Ca<sup>2+</sup>, and Al<sup>3+</sup>, were not discharged timely, forming a local alkaline environment. Because I/S cannot be converted into kaolinite in alkaline environments (Yuan et al., 2013b), a large amount of I/S remains in the reservoirs. Meanwhile, the alkaline environments also lead to the precipitation of carbonate minerals in pre-existing dissolution pores (Figures 5C-E; e.g., Macaulay et al., 2001). In addition, reservoirs far from faults generally keep lower CO2 content. For example, the well C is in a low  $CO_2$  content area (Figure 1B), which also proves that the fluid environment of CO<sub>2</sub> reservoirs in well C is relatively closed.

After mantle-derived CO2 enters the trap, CO2 gradually migrates towards the top of the reservoir by the action of buoyancy (Wei et al., 2023; Gudmundsson, 2022), eventually leading to a decrease in  $CO_2$  concentration with increasing depth (Figure 9). The higher the concentration of CO<sub>2</sub>, the stronger the acidity of the fluid and the more intense the dissolution of reservoirs. Therefore, as the depth increases, the dissolution of the reservoir gradually weakens (Figure 9). The acidity of the fluid can also inhibit the cementation of carbonate minerals (Sun et al., 2020), so the carbonate cementation at the top of the reservoir is relatively weak (Figure 9). The prolonged contact between high concentrations of mantle-derived CO2, formation water, and debris particles leads to strong dissolution of feldspar and early carbonate minerals (Figure 5J-L, e.g., Azzam et al., 2023; Lv et al., 2022), resulting in increased salinity of formation water (Giles and Boer, 1990; Yuan et al., 2013b). Due to the vertical difference of CO<sub>2</sub> concentration in the reservoir, the salinity of the formation water



at the top of the reservoir would be higher than that at the bottom after a long-term  $CO_2$ -water-rock interaction. Naturally, high salinity formation water and dissolution by-products migrate and diffuse downwards under the influence of the difference in salinity. Therefore, the dissolution products at the top of the reservoir will also migrate downwards and recrystallize. In addition, as the depth increases, the  $CO_2$  concentration decreases, the fluid alkalinity increases, and  $CO_2$  gradually participates in carbonate minerals cementation (Figure 9).

In summary, within closed fluid environment reservoirs, as depth increases, dissolution gradually weakens while carbonate cementation strengthens (Figure 9). The constructive effects of dissolution and the destructive effects of carbonate cementation work synergistically, resulting in a gradual decrease in porosity and permeability (Figure 7). This trend is observed across each sand layer (Figure 9).

## 5.3.3 Impact pattern of mantle-derived CO<sub>2</sub> on reservoirs in open and closed fluid environments

Based on the argument above, a comprehensive pattern of the impact of  $CO_2$  on reservoirs within open and closed fluid

environments in the study area has been established (Figure 10). The reservoirs near the faults keep an open fluid environment, while the further away from the faults, the more closed the fluid environment is (e.g., Gudmundsson, 2022; Ward et al., 2016; Macaulay et al., 2001; Wang et al., 2017; Ahmat, et al., 2022). As the distance from the fault increases, the dissolution weakens, carbonate cementation strengthens, contents of clay minerals and the relative content of I/S increase, while the relative content of kaolinite decreases (e.g., Azzam et al., 2023; Lv et al., 2022). In an open fluid environment, feldspar strongly dissolves and generates a large number of dissolution pores (e.g., Hu, 2016; Wei and Sun, 2017), and the dissolution by-products (clay minerals) are timely removed from the reservoirs and migrate northward along the faults with the fluids. Moreover, the acidic environment formed by CO<sub>2</sub> inhibits the precipitation of carbonate minerals (Boyer, 1983), so there are only a few carbonate cements. All of these effectively improve the physical properties of reservoirs. In a relatively closed fluid environment, as the depth increases, the CO<sub>2</sub> concentration decreases, the acidity of the formation water weakens, and the dissolution weakens, but the cementation of carbonate minerals strengthens. Due to the higher CO2 content at the top of the reservoir

| Str:<br>eries | tum<br>Member   | Depth<br>(m)  | Lithology   | 10 <u>SP</u> 130 | 5 <u><b>Porosity</b></u> 35 | 0.1 Permeability<br>(mD) 1000 | Photographs                  | Phenomenon<br>description   |
|---------------|---|---|---|------------------|-----------------------------|-------------------------------|------------------------------|---|
|               |   | 1490-<br>   | 9     9     9       9     9     9       9     9     9       9     9     9       9     9     9       9     9     9       •     •     •       •     •     •       •     •     •       •     •     •       •     •     •       •     •     •       •     •     •       •     •     •       •     •     •       •     •     •       •     •     •       •     •     •       •     •     • | and the second   | N=121                       | N=121                         | © 1503.00m                   | CO, content: 65.6%<br>Intensive dissolution<br>of feldspars and<br>slight carbonate<br>cementation,<br>with surface porosity<br>of 22.4%. |
| (T)           | jie Formation (Es <sup>4</sup> )                                    | (2)-<br>1530-<br>1540-<br>(3)-<br>1550-<br>1550-<br>1560- |   | han when         |                             |                               | © 1527.90m                   | CO <sub>2</sub> content: 61.1%<br>Strong dissolution<br>and carbonate<br>cementation, with<br>surface porosity<br>of 16.4%                |
| Eocene (E)    | The 4 <sup>th</sup> Member of Shahejie Formation (Es <sup>4</sup> ) | (4)<br>1570<br>1580<br>1590                               |   | M                | Are with                    | in the second                 | 3 1543.40m                   | CO <sub>2</sub> content: 54.0%<br>Obvious dissolution<br>and carbonate<br>cementation,<br>with surface porosity<br>of 12.6%               |
|               | The   | 1600-<br>1610-<br>1620-<br>(5-<br>1630-                   |   | Mull             | 68<br>6                     |                               | (a) 1569.96m                 | CO <sub>2</sub> content: 48.8%<br>Slight dissolution,<br>strong carbonate<br>cementation,<br>with surface porosity<br>of 8.3%             |
|               |   | 1640-<br>   |   | M                | 2                           | 1                             | <ul> <li>1626.80m</li> </ul> | CO <sub>2</sub> content: 14.1%<br>Weak dissolution and<br>intensive carbonate<br>cementation,<br>with surface porosity<br>of 4.2%         |

compared to the bottom, the  $CO_2$ -water-rock interactions are more intense, leading to differences in the concentration of dissolution by-products between the upper and lower parts of the reservoirs. Although dissolution products cannot be effectively discharged from the reservoir or transported over long distances (Yuan et al., 2013b), they would migrate downwards and recrystallize due to the concentration gradient of the by-products. Ultimately, the porosity and permeability of reservoirs gradually decrease with depth.

Previous experiments have demonstrated that  $CO_2$  primarily participates in mineral dissolution, often overlooking the dual effects of  $CO_2$  on reservoirs and the roles of fluid environments in this context under geological conditions. This is why it's important to select geological cases to illustrate the impact of  $CO_2$  on reservoirs. This work confirms the influence of mantle-derived  $CO_2$  on reservoirs through mineralogy (dawsonite and ankerite) and geochemistry (stable C- and O-isotopes), and further demonstrates the dual effects (dissolution and cementation) of mantle-derived  $CO_2$  on reservoirs integrated with reservoir physical properties, with a particular focus on how  $CO_2$  influences reservoirs in different fluid environments. However, due to the limitation of  $CO_2$  content range (about 15%–70%) in the study area, we are unable to investigate the effects of low-concentration  $CO_2$  on sandstone reservoirs, which may affect the generalizability of this study. Besides, the formation temperature and pressure, and salinity of formation water should be considered when dealing with other cases. Overall, this work can be applied to the studies of reservoir homogeneity and sweet spots in regions with hydrothermal and mantle-derived  $CO_2$  activities.



## 6 Conclusion

In this paper, we take a  $CO_2$  gas reservoir in the Dongying Sag, Bohai Bay Basin, China, as an example, using a multi-technique approach in mineralogy and geochemistry to investigate evidence of the impact of mantle-derived  $CO_2$  on the reservoir. Combining with the charateristics of the reservoir's physical properties, we elucidate the mechanisms through which mantle-derived  $CO_2$  influences the sandstone reservoirs and discuss the patterns of its impact on reservoir properties in open and closed fluid environments. The conclusions can be drawn as follows:

(1) Dawsonite (a special tracer mineral of mantle-derived  $CO_2$ ) and ankerite (its crystallization temperature is much higher than the normal geothermal gradient of the study area) are widely distributed in the  $CO_2$  reservoir of the Dongying Sag. Moreover, the  $\delta^{13}C$  (-9.0‰--1.6‰) and  $\delta^{18}O$  (-21.7--12.7‰) of carbonate cements in the mantle-derived  $CO_2$  gas reservoir basically fall within the range suggested for mantle-derived  $CO_2$  and hydrothermal fluids.

These mineralogical and geochemical evidence indicate that  $CO_2$ -rich hydrothermal fluids have actively participated in water-rock reactions within the reservoir, thereby impacting its diagenesis.

- (2) The CO<sub>2</sub> reservoirs near faults in the study area are an open fluid environment, while the CO<sub>2</sub> reservoirs far from faults keep a relatively closed fluid environment. There are significant differences in the debris particles, mineral composition, and physical properties between reservoirs in open fluid environments and closed fluid environments. The former has better porosity and permeability, more quartz, as well as fewer feldspar, carbonate, and clay mineral cement. Furthermore, the clay minerals in open fluid environments are predominantly kaolinite, whereas these in closed fluid environments primarily consist of I/S.
- (3) The mantle-derived CO<sub>2</sub> has dual effects on sandstone reservoirs. On the one hand, the dissolution by CO<sub>2</sub> greatly increases the reservoir's storage and seepage capacity. On the other hand, the carbonate cement formed by the CO<sub>2</sub>water-rock reaction can also lead to serious deterioration of

reservoir quality. In open fluid environments,  $CO_2$  strongly dissolves feldspar and carbonate minerals in reservoirs, and the dissolution by-products (clay minerals) are carried out from the reservoirs timely, and the acidic environment formed by  $CO_2$  inhibits carbonate cementation, which synergistically improves the physical properties of reservoirs. In closed fluid environments, decreasing  $CO_2$  concentrations with depth leads to diminishing dissolution effects and increased carbonate cementation, resulting in reduced reservoir porosity and permeability.

However, the  $CO_2$  content, the formation temperature and pressure, and salinity of formation water should be considered when dealing with other cases. Overall, this study provides a good understanding for the impacts of mantle-derived  $CO_2$  on sandstone diagenesis and reservoir properties in open and closed fluid environments, which may contribute to the studies of reservoir homogeneity and sweet spots in regions with hydrothermal and mantle-derived  $CO_2$  activities.

### Data availability statement

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

### Author contributions

MW: Conceptualization, Investigation, Methodology, Writing-original draft, Writing-review and editing. JZ: Project administration, Resources, Supervision, Writing-review and editing. CL: Writing-review and editing, Investigation. JQ: Supervision, Validation, Writing-review and editing. WW: Investigation, Writing-original draft. HZ: Investigation, Writing-review and editing. HC: Investigation, Writing-review and editing.

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## Funding

The author(s) declare that financial support was received for the research, authorship, and/or publication of this article. This work is supported by the National Natural Science Foundation of China, Grant number 42302144.

## Acknowledgments

We would like to express our sincere gratitude to the Petroleum Exploration and Development Research Institute of Shengli Oilfield, China Petroleum and Chemical Corporation, for providing us with the core samples and data used in this study. Additionally, we would like to extend our special thanks to the reviewers and editors for their insightful comments and suggestions, which have greatly helped to improve the quality of this article.

### **Conflict of interest**

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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