Response to reviewer 3 (Dr. Giacomo Medici)

General comments

Very good research on geological carbon storage. Please, take into account my comments to improve the manuscript.

R: We appreciate your recognition of the quality of our research and your valuable suggestions for improving the manuscript. Below, we provide a detailed response to each of your comments and explain how we have addressed them in the revised manuscript. The following revisions will be incorporated into the subsequent version.

Specific comments

(1). *Lines 15-87. Please, more emphasis on the fact that your research has a double focus: anisotropy and heterogeneity. My suggestions aim to increase the impact of your research in different sub-fields of applied geoscience.*

R: Thank you. This is a good suggestion. We have revised the introductory section to explicitly state that the objective of this work is twofold. (i) To quantitatively analyze the effect of permeability heterogeneity and anisotropy on the GDC-driven dissolution rate in a wide range of (isotropic and anisotropic) heterogeneous fields with varying degrees of heterogeneity and anisotropy. (ii) To investigate whether the dissolution rate can be predicted based on the finger-tip velocity. Here, we compact the anisotropy and heterogeneity as one focus. We also emphasize how our research can be employed in different sub-fields of applied geoscience by introducing its potential application in other gravity-driven convection processes. We have modified the introduction as follows:

"Therefore, the objective of this work is twofold. (i) To quantitatively analyze the effect of permeability heterogeneity and anisotropy on the GDC-driven dissolution rate in a wide range of (isotropic and anisotropic) heterogeneous fields with varying degrees of heterogeneity and anisotropy. (ii) To investigate whether the dissolution rate can be predicted based on the finger-tip velocity. We do this in two steps. First, performing numerical simulations over a large number of heterogeneous fields of different permeability distributions. Numerical simulations are carried out by a finite-difference numerical program developed by Wang (2022). Permeability fields are generated with the sequential Gaussian simulation method implemented in the SGSIM code (Journel and Huijbregts, 1976). Second, the results of the simulations are analyzed to find relations among the GDC-driven dissolution rate, permeability heterogeneity, anisotropy and finger-tip velocity, and we compare our results against those given in literature. In this step, ordinary-least-squares linear regressions are used. The conclusions from this work may hold significant relevance for other gravity-driven convection processes, where density differences play a crucial role. These processes include contaminant migration, geothermal exploitation, saltwater intrusion, and mineral precipitation/dissolution (Berhanu et al., 2021; Sanz et al., 2022; Guevara Morel and Graf, 2023; Fang et al., 2024; Liyanage et al., 2024)." The revisions will be incorporated into the subsequent version.

We also give a short summary of the shortage of current research before we introduce our objectives:

"Overall, we have a solid understanding of the GDC-driven dissolution process in isotropic homogeneous media, but the GDC-driven dissolution in heterogeneous media needs further study. Especially, we need to quantitatively clarify the impact of the anisotropy ratio on the effective dissolution rate. Moreover, the current predictors are all based on the (equivalent) permeability, and it remains unclear whether we can predict the dissolution rate based on other formation properties or field observations, such as the finger-tip velocity."

(2). *Lines 21-22. "By being sealed under the low permeability caprock". Consider inserting recent papers on low permeability sedimentary layers that incorporate a discussion on applications for CO2 storage.*

- *English, K.L., English, J.M., Moscardini, R., Haughton, P.D., Raine, R.J. and Cooper, M., 2024. Review of Triassic Sherwood Sandstone Group reservoirs of Ireland and Great Britain and their future role in geoenergy applications. Geoenergy, 2(1), pp.geoenergy2023-042.*

- *Medici, G., Munn, J.D., Parker, B.L., 2024. Delineating aquitard characteristics within a Silurian dolostone aquifer using high-density hydraulic head and fracture datasets. Hydrogeology Journal, 32(6), 1663-1691.*

- *Newell, A.J. and Shariatipour, S.M., 2016. Linking outcrop analogue with flow simulation to reduce uncertainty in sub-surface carbon capture and storage: an example from the Sherwood Sandstone Group of the Wessex Basin, UK. Geological Society, London, Special Publications, 436(1), 231-246.*

R: Thank you. We have read these interesting papers and added them in the reference list. You will see them into the subsequent version.

(3). *Line 87. Please, disclose the specific objectives of your research by using numbers (e.g., i, ii, and iii).*

R: Thank you. Inspired by your suggestions we disclose the specific objectives of our research by using numbers (i) and (ii), to clarify the focus of the article, as shown in the following:

"Therefore, the objective of this work is twofold. (i) To quantitatively analyze the effect of permeability heterogeneity and anisotropy on the GDC-driven dissolution rate in a wide range of (isotropic and anisotropic) heterogeneous fields with varying degrees of heterogeneity and anisotropy. (ii) To investigate whether the dissolution rate can be predicted based on the finger-tip velocity." The revisions will be incorporated into the subsequent version.

(4). *Line 133. Be more specific when you mention anisotropy. "Flow anisotropy"? We are not talking about a mechanical anisotropy here due to the presence of rock discontinuities.*

R: This is a good suggestion. Your suggestions help ensure clarity and precision in the manuscript. Here, anisotropy refers to "permeability anisotropy", which specifically describes the directional dependence of permeability (or hydraulic conductivity). It is often used when discussing how permeability varies with direction in a porous medium. For example: "The aquifer exhibits strong permeability anisotropy, with horizontal permeability significantly higher than vertical permeability." It is a key concept in hydrogeology because it directly influences fluid flow patterns can may lead to *flow anisotropy*.

According to your suggestion. We have specified it in this sentence and other sentences using "anisotropy" in the manuscript when necessary. Please check in the upcoming revised version.

(5) Line 295. Too soon for introducing references in this paragraph.

R: Thank you for this suggestion. We have reorganized this paragraph and introduce it later in the paragraph. The changed version is as follows: "For illustrative purposes, we chose a representative permeability realization for each case. These realizations are shown in panel (a) of Figure 4, from which we can see that CO_2 fingering is strongly affected by heterogeneity. In particular, the presence of vertical well-connected high permeability zones (preferential channels) facilitates the initiation and growth of the instability fingers (see for instance the second column of Figure 4). Actually, the white randomness of the top CO_2 mass fraction (needed in homogeneous media to create instabilities) is redundant in heterogeneous porous media as instabilities are controlled by these vertical preferential channels. In all cases, results show that instability make CO_2 fingers grow, merge and re-initiate as also observed in laboratory experiments (Rasmusson et al., 2017; Liyanage, 2018; Tsinober et al., 2022) and numerical simulations (Elenius et al., 2015)." The revisions will be incorporated into the subsequent version.

(6) *Line 367. "Geological Carbon Sequestration (GCS)". You have already defined the acronym.*

R: Thank you. We have removed the redundant definition of GCS to avoid repetition. However, we have retained the definition of Gravity-Driven Convection (GDC) in this paragraph, despite its prior definition. By redefining GDC, we aim to immediately connect readers with its meaning, facilitating smoother comprehension. While acronyms streamline writing, they can occasionally hinder readability, especially in lengthy articles, as readers may need to backtrack to recall their definitions. To mitigate this inconvenience, we have chosen to reintroduce the meaning of GDC at its first appearance in each section. We trust you understand the rationale behind this approach.

(7) *Line 424. Integrate the two relevant papers on the effect of low permeability layers on deep aquifers/reservoirs.*

R: We have carefully read and added English et al. (2024), Medici et al. (2024) and Newell & Shariatipour (2016) in the reference lists. Thank you for your recommendation. These additions provide a more comprehensive perspective on the topic.

Figures and tables

(8) Figure 1a. You made bigger part of the reservoir with a rectangle. The spatial scale is unclear.

R: This is a very good question. The size of instability fingers is a function of permeability, as described by $l_c = 70 \cdot \frac{\mu\phi D_m}{\Delta\rho g \kappa_g}$. This relationship underscores that the size of the instability fingers and the simulation domain are contingent upon the permeability. Given the substantial variability in permeability across different reservoirs, the dimensions of instability fingers can differ markedly. Consequently, it is challenging to assign a precise spatial scale to the general illustration of instability fingers.

Ultimately, Figures 1a and 1b are designed to convey general insights into the development of instability and vertical convection, thereby introducing the objectives of this work. As such, specific temporal and spatial scales are not provided. We trust that you understand this approach.

(9) *Figure 2. Spatial scale unclear also here.*

R: Thank you. We add the following descriptions in the caption instead of putting them inside the figure to make the sketch concise: "The size of the simulation domain is B × L=7.5×7.5 [m^2], and other hydrogeology properties are summarized in Table 2." The revisions will be incorporated into the subsequent version.

(10). *Figure 4. Make the letters larger.*

R: Thank you. In Figure 4, the font size of letters and numbers has been increased to enhance readability. A figure with larger letters and numbers will be found in the subsequent version.

(11). *Figure 4. Make the letters and numbers larger.*

R: Thank you. Probably you mean Figure 5, because you have mentioned Figure 4 in the previous comment. We notice that the letters and numbers are not large enough in Figure 5, and we increase the font size of all labels, letters, and numbers in Figure 5. A figure with larger letters and numbers will be found in the subsequent version.