Response to reviewer 1 (Dr. Yang)

General Comments

It is a pleasure procedure to review this interesting manuscript, which deals with the dissolution trapping of CO_2 *during geological carbon sequestration, an important means to reduce carbon emissions. In the process of sequestering* CO_2 *in the deep saline aquifer, a concern is that the* CO_2 *is less dense than the saline water, which gives rise to the possibility of upward* CO_2 *leak. It is noticed that dissolution of* CO_2 *in the brine is a crucial mechanism that reduces the risk of leak by transforming the supercritical* CO_2 *into aqueous* CO_2*. The significant challenge lies in the fact that the dissolution process is susceptible to instabilities driven by gravity and permeability heterogeneity, both of which are ubiquitous and involve big uncertainties. Therefore, it is important to characterize the dissolution rate of* CO_2 *in the saline water, which is the objective of this manuscript.*

Through reading the manuscript, I can feel that the authors have spent a big effort in preparing this work. They carefully selected the parameters for their numerical model based on the data from a deep literature review. They have conducted a large quantity of numerical simulations to build a data pool for systematic analysis. Finally, they give fruitful analysis and discussions of the results. Based on my own reading of the manuscript, I give my support of publication of this work on HESS. Several revisions are needed before it is accepted for publication.

R: We are grateful for your recognition of the importance of our work and your supportive comments regarding the effort we have invested in this study, including the careful selection of parameters based on a deep literature review, the extensive numerical simulations conducted, and the detailed analysis and discussion of the results. We sincerely thank you for your time and constructive feedback, which has significantly improved the quality of our manuscript.

Below, we provide detailed responses to each comment, along with a clear explanation of the revisions made to the manuscript. All changes will be incorporated into the subsequent revised version.

Major Comments

(1) *This manuscript improves the current predictor for enhanced* CO_2 *dissolution due to gravity driven convection. Most of current predictors for enhanced* CO_2 *dissolution address the homogeneous cases, which may have limitations for the real heterogeneous problem. Two representative works, as shown in Table 5 and Figure 6, have offered the preliminary predictor for the* CO_2 *dissolution in heterogeneous fields. However, while one of them*

simplifies the predictor by neglecting the anisotropic effect, the other may not properly incorporate the ansotropic effect. The authors provided a new explanation of the anisotropic effect on the density driven dissolution. Their numerical results show that for a fixed vertical equivalent permeability increasing the horizontal equivalent permeability can reduce the dissolution efficiency, because increased horizontal permeability can increase horizontal mass exchange, make the horizontal mass distribution more uniform, and thus reduce the instability. My question is: I realize that the gamma_1=0.08 obtained by the data regression is quite similar to that for the 0.09 listed in Table 1. Do they have any relations? This is based on my observation that the alpha_1=1.1 is very close to 1.0 for the homogeneous.

R: Your observation regarding the similarity between γ_1 =0.08 and the value $\gamma = 0.09$ in Table 1 is insightful and highlights an important aspect of our work. The numerical proximity of γ_1 =0.08 to $\gamma = 0.09$ arises because both parameters are rooted in the same underlying physics of gravity-driven convection. However, γ_1 =0.08 is not a direct extension of the homogeneous value but rather a new parameter that incorporates the effects of heterogeneity. The similarity in γ_1 =0.08 to the homogeneous value $\gamma = 0.09$ reflects the consistency between the homogeneous model and the heterogenous model. Interestingly, as mentioned by you, the data regression result shows that $\alpha_1 = 1.1$, which is very close to 1.0, again indicating the same underlying physics governing the vertical mass exchange driven by gravity-driven convection, i.e., the vertical mass flux due to density instability can be approximated by Darcian flux with permeability being represented by equivalent vertical permeability.

To clarify the relationship between these two values, we provide the following detailed explanation:

(i) $\gamma = 0.09$ in Table 1 is the value for the homogeneous field obtained by fitting the mean dissolution rate of 15 homogeneous cases using Equation (1) in the manuscript

$$F_\infty = \gamma X_0^C \rho_0 \frac{\Delta\rho g \kappa}{\bar{\mu}}. \quad (1)$$

(ii) γ_1 =0.08 is obtained by fitting the dissolution rate for the heterogeneous fields using Equation (17) in the manuscript, which accounts for the effects of permeability anisotropy.

$$F_\infty^* = \gamma_1 \left(\frac{\kappa_z^e}{\kappa_g}\right)^{\alpha_1} \left(\frac{\kappa_x^e}{\kappa_z^e}\right)^{\beta_1}. \quad (17)$$

Considering $F_\infty^* = \frac{F_\infty}{X_0^C \rho_0 \frac{\Delta\rho g \kappa}{\bar{\mu}}}$, Equation (17) is equivalent to

$$F_\infty = \gamma_1 X_0^C \rho_0 \frac{\Delta\rho g \kappa}{\bar{\mu}} \left(\frac{\kappa_z^e}{\kappa_g}\right)^{\alpha_1} \left(\frac{\kappa_x^e}{\kappa_z^e}\right)^{\beta_1}. \quad \text{(R1)}$$

If the field is isotropically homogenous, i.e., $\kappa_z^e = \kappa_x^e = \kappa_g$, Equation (R1) should be equivalent to Equation (1), which means γ_1 should be equal to γ in theory. Our value γ_1 =0.08 is very close to $\gamma = 0.09$, indicating the consistency of our theory and good predictivity of the formula.

We note that γ value in our work is similar to those obtained in literature using open top boundaries (e.g., 0.075 in Elenius et al. (2012) using CTZ boundary, 0.065 in Martinez and Hesse (2016) using CTZ boundary, and 0.06 in Rasmusson et al. (2017) using permeable boundary). This similarity to some extent validates the reasonability of our numerical modeling. We also note the γ value in our work is quite different from those research results obtained based on diffuse-only boundary. In our case, the convection of CO_2-saturated brine is allowed to pass the top boundary, which seems more realistic.

(2) *Furthermore, the authors introduced a new predictor using finger velocity, which is particularly intriguing because finger velocity can be measured using optical fiber technology. Given the rapid advancements and expanding applications of optical fibers in the field of geosciences, this novel formula could serve as a valuable tool for monitoring the trapping of CO_2 through dissolution. My question is: The regression value for gamma_2=0.34 is quite different from 0.08 or 0.09, could you please explain why the gamma_2 is so different from gamma_1? I am wondering if it is simply a result of the data regression or maybe it have physical explanations.*

R: The reviewer raises an excellent question regarding the significant difference between γ_1 =0.08 and γ_2 =0.34. This difference arises because γ_1 and γ_2 scale different physical quantities, as explained below:

γ_1 is obtained based on equivalent vertical permeability κ_z^e/κ_g, while γ_2 is based on dimensionless fingertip velocity (v_∞/v_c). When the vertical permeability $\kappa_z^e/\kappa_g = 1$, the value of $v^* = v_\infty/v_c$ is around 0.3, as shown in bottom panel of Figure 3. This means there should be a factor of around 0.3 between γ_1 and γ_2. Interestingly, the value $\gamma_1/\gamma_2 = 0.24$ is very close to the dimensionless fingertip velocity. This consistency makes the predictor physically sound.

(3) *In the study of density driven instability, the fingers are usually irregular, as shown in the Figure 4 of your manuscript. Could you please explain why the fingers in Figure 3 are quite uniform?*

R: Your observation about the uniformity of fingers in Figure 3 versus the irregularity in Figure 4 is meticulous. It is very common to see the irregular instability fingers in literature (e.g., Elenius & Gasda [2021] Farajzadeh et al. [2013]), which is also shown in our work. However, in the very early stage when the instability is just fully developed ($t^* = 1500$ in Figure 3), the finger is quite uniform. These uniform fingers become irregular with time, as shown in Figure R1.

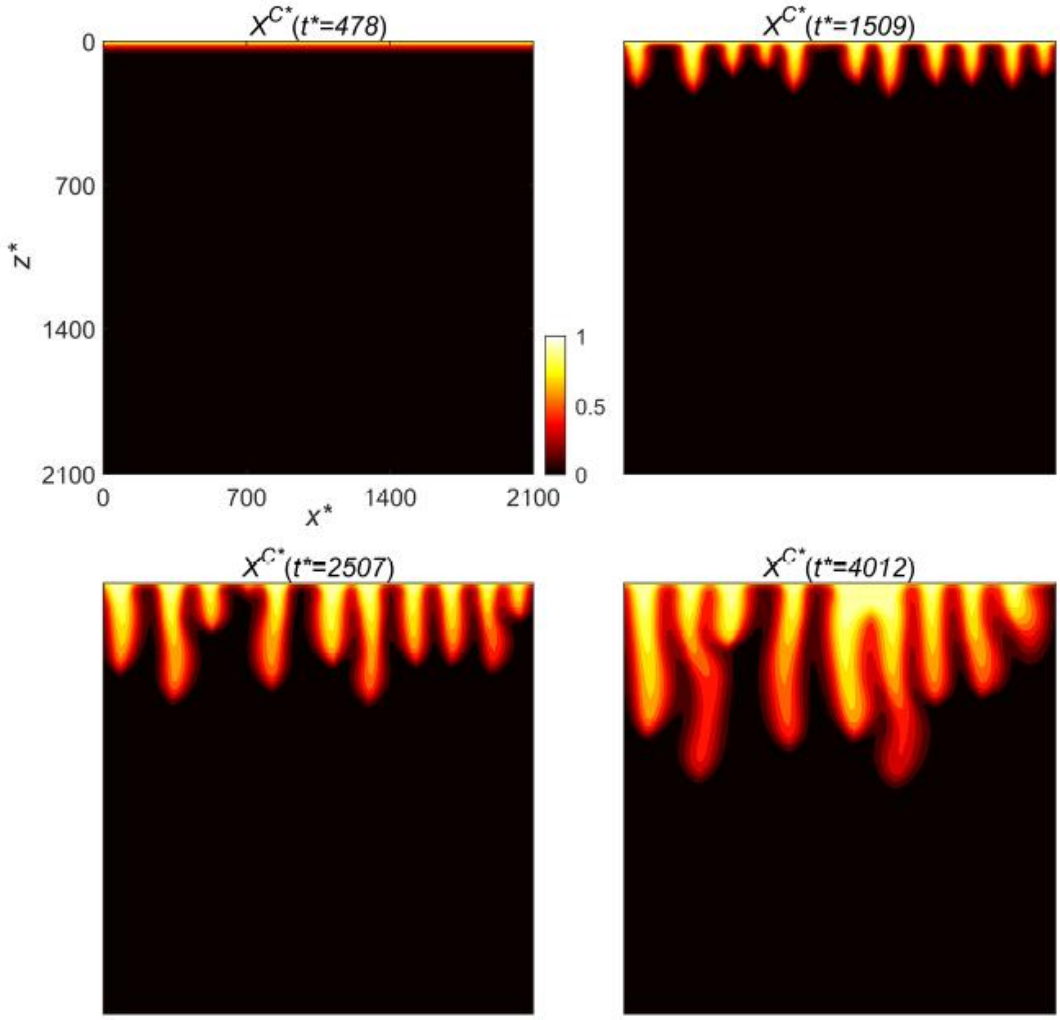

Figure R1: development of instability fingers (c.f., page 117 in Wang [2022], https://upcommons.upc.edu/handle/2117/376077).

(4) *I can understand that the authors and many other researchers use two-dimensional model in both laboratory and numerical simulation, because of the high cost of three-dimensional model. I do agree that the two-dimensional study is very useful, but it would be nice if the authors can provide some discussions on the three-dimensional effect on GDC. I know it is hard to design a new simulation of new parameters, so I would appreciate it if the authors could find some related three-dimensional studies and give a short comparison of the difference of three dimensional simulations and two dimensional simulations. This may give more confidence for readers using the results from this work.*

R: We agree with the reviewer that 3D effects are critical for field-scale applications of gravity-driven convection (GDC). While our study focuses on 2D simulations for computational efficiency, we recognize the importance of discussing 3D effects to provide a more comprehensive understanding of the problem. We study recent researches that have successfully applied 2D-to-3D scaling relationships to bridge the gap between simplified models and real-world applications. The dissolution rate in a real 3D case will be higher than in the 2D cases employed in this work. As pointed out by Pau et al. [2010], the stabilized mass flux in the 3D scenario is observed to be 25% higher than that in a comparable 2D simulation. Although this difference is statistically significant, it is relatively minor considering the substantial variability in permeability often seen in geologic media. Therefore, the impact of additional spatial dimensions on both the onset time and the stabilized mass flux appears to be limited. In the revised manuscript, we will add a concise discussion on the differences between 2D and 3D simulations of GDC.

The following sentences will be added to the conclusion Section 4.1 . "We note that in the real 3D scenario, the dissolution rate may be approximately 25% higher than that observed in 2D cases. However, this difference is relatively minor when compared to the significant variability in permeability commonly encountered in geologic media Wang et al. (2022)." The revisions will be incorporated into the subsequent version.

Minor Comments

(1). *The supplementary materials provided important and comprehensive information about the numerical modeling, but it is not fully referred in the paper. Please, give more clear reference of the supplementary materials in the paper.*

R: We have added explicit references to the Supplementary Materials in Sections 2 and 4.3 to ensure readers are aware of the additional information available. The revisions will be incorporated into the subsequent version.

(2). *Line 195 and 205, the appendix is missing.*

R: Thank you. The appendix has been restored in the end of the manuscript. The revisions will be incorporated into the subsequent version.

(3). *-Line 294, it would be better if we say in panel (a) of Figure 4 rather than in the first panel of Figure 4.*

R: Thank you. We have revised to "panel (a) of Figure 4" for clarity. The revisions will be incorporated into the subsequent version.

(4). -Line 299, I am wondering if you added white randomness ('white noise' maybe) in the heterogeneous fields?

R: Yes. We have added a very small white noise in all simulations to trigger instability. However, we find this white noise has negligible effect in the instability development in the heterogeneous media, where the finger development is strongly influenced by the permeability distribution.

(5). -Line 306, please remove the comma in ‘dissolution process, shows’.

R: Thank you: We have removed the comma in “dissolution process shows”. The revisions will be incorporated into the subsequent version.

(6). -Line 388, please remove the period before the references.

R: Thank you. We have corrected this formatting issue. Now we have moved the related sentence to section 4.7 to make the structure better. The revisions will be incorporated into the subsequent version.

(7). -*The overall structure of this manuscript is very nice, dividing the whole article into logical sections of proper titles. However, it may be better if the authors can reorganize the section ‘6 Conclusions’. We can see that in the section ‘5 Results and Discussion’. The authors first describe the general impact of heterogeneity on the development of instability, and then perform log-linear regressions of the simulation results. However, in the conclusion section the authors do not organize these results in the same order. Moreover, it would be better if the first paragraph is split so that we have a short summary of this work before writing the conclusions. This does not affect the comprehending of this work, but it would be nicer if the authors can reorganize the conclusions.*

R: Thank you. We have split the first paragraph into a brief summary and restructured the conclusions to mirror the Results section’s logic. The revised conclusion now begins with a concise summary of the study’s objectives and key findings, followed by detailed conclusions organized in the same order as the “Results” section. The revisions will be incorporated into the subsequent version.

Finally, we hope that these responses and the corresponding revisions have effectively addressed your concerns. We have also incorporated additional clarifications and refined the formatting throughout the manuscript. Once again, we extend our sincere thanks for your valuable feedback, which has played a crucial role in enhancing the quality of our work.

Reference:

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Pau, G. S., Bell, J. B., Pruess, K., Almgren, A. S., Lijewski, M. J., and Zhang, K.: High-resolution simulation and characterization of density-driven flow in CO2 storage in saline aquifers, Advances in Water Resources, 33, 443 – 455, https://doi.org/https://doi.org/10.1016/j.advwatres.2010.01.009, 2010

Wang, Y.: Numerical Modeling of Geological Carbon Sequestration: Enhanced Dissolution in Randomly Heterogeneous Media, https://doi.org/10.5281/zenodo.6769788 , 2022 .