Dear Anonymous Referee #1,

We sincerely thank the reviewer for the careful reading of our manuscript and for the constructive and insightful comments. We greatly appreciate the positive evaluation of our research. These comments have been very helpful in improving the clarity, rigor, and overall presentation of the manuscript. We have carefully revised the manuscript accordingly. Our detailed responses are provided below.

Comment 1:

In the assumptions section the authors state that the reservoir is homogeneous and isotropic, whereas a later section explicitly investigates heterogeneity through permeability anisotropy. Indeed, heterogeneity plays an important role in CO_2 migration and trapping (see for example, Gas migration and residual trapping in bimodal heterogeneous media during geological storage of CO_2, Advances in Water Resources 142, 103608). The effects of heterogeneity should be investigated. But the manuscript should clearly distinguish the base-case assumptions from the sensitivity-analysis settings. In addition, setting an anisotropy ratio alone cannot fully represent the effects of heterogeneity.

Response: Thank you for this valuable comment. We fully agree that heterogeneity is a key control factor on CO_2 migration and trapping, and that permeability anisotropy alone cannot fully represent the complexity of heterogeneous geological media. In the revised manuscript, we have clarified the distinction between the **base-case model assumptions** and the **sensitivity-analysis scenarios**. Specifically, the assumption of a homogeneous and isotropic reservoir applies only to the reference case used to establish the base behavior of the system, whereas the later section introducing permeability anisotropy is intended as a sensitivity analysis. We have revised the title of Section 4.3 to "Effect of permeability anisotropy". We have added the description regarding the

non-homogeneity assumption in lines 318-321.

"The distribution of the plume in the anisotropic formation at the time of the cessation of injection is shown in fig.11. A permeability anisotropy index γ was used to represent the ratio of vertical permeability to horizontal permeability. It is assumed that the horizontal permeability remains constant at k_0, while the vertical permeability changes according to the permeability anisotropy index."

We have cited the paper recommended by the reviewers, which has enhanced our understanding of how heterogeneity affects the migration of plumes. The influence of this overall heterogeneity has been investigated in our subsequent studies. See lines 41-42:

"The heterogeneity of the reservoir also has a significant impact on the long-term distribution of the plume (Yang et al., 2020)."

Comment 2:

More explicit discussions are needed for several assumptions limiting the model applicability, including the isothermal condition, neglect of precipitation process and the simplified treatment of reactive mineralogy.

Response: We thank the reviewer for this important comment. We agree that these assumptions directly affect the applicability of the proposed model and should be more explicitly discussed. In the revised manuscript, we have expanded the discussion of model assumptions and limitations.

(1) Generally, the geothermal gradient does not exceed 5 ℃ per 100 m (Vilarrasa V, Rutqvist J. Thermal effects on geologic carbon storage. Earth-Science Rev 165:245–256. https://doi.org/10.1016/j.earscirev.2016.12.011). The size of the domain is 50 meters in height, so the temperature variation within the simulation area is relatively small. Here, we assume that the simulated reservoir is isothermal. The isothermal

assumption was adopted to reduce the complexity of the coupled multiphase flow and geochemical transport problem and to focus on the long-term evolution of CO_2 migration, dissolution, and reaction under a prescribed reservoir temperature. In subsequent studies, we will incorporate a coupled analysis of the temperature and mechanical fields. We have added an explanation in lines 216–217:" (7) Based on the geothermal gradient variation data in the reservoir, the temperature variation within the simulation domain is approximately 1–3 °C. We therefore assume the simulation domain to be an isothermal saline aquifer."

(2) Regarding mineral precipitation, we acknowledge that precipitation is a key process for long-term mineral trapping. Salt precipitation could result in pore clogging, which in turn raises wellhead injection pressure. Carbonate precipitation was not included in the present formulation in order to focus on the coupling between multiphase flow, dissolution-driven convection, and calcite reaction over the simulated timescale. We have added a clarification in lines 211–212 (see revised manuscript)" We neglect the salt precipitation process and are therefore unable to capture phenomena such as pore plugging and pressure buildup during plume migration."

(3) The reactive mineralogy is simplified by considering calcite as the only reactive mineral initially present in the formation. The reactive mineralogy was simplified by considering calcite as the only reactive mineral initially present in the formation. This simplification is reasonable for the present timescale because calcite reacts much faster than most silicate minerals, whose dissolution and precipitation typically occur over much longer timescales. Therefore, quartz, feldspar, and other slowly reacting minerals were treated as effectively inert in the present model. We have explained this simplifying assumption in lines 111–116" According to previous studies, calcite have a relatively fast reaction rate among all contents in sandstone (Xu et al., 2019b). The full-cycle model focuses on a relatively short time scale and concentrates on calcium

minerals (calcite, dolomite), considering quartz, feldspar, and other minerals as insoluble substances because their dissolution/precipitation process takes thousands of years (De Silva et al., 2015). The formation of carbonate is considered to be the main way to sequester CO_2 and an important process for achieving permanent trapping in GCS (Bachu et al., 1994). To simplify the model, it is assumed that the reservoir minerals involved in the chemical reaction only contain 5% calcite (Sainz-Garcia et al., 2017b).”

Comment 3:

Some wording choices are misleading or overly strong relative to the presented evidence. For example, “for the first time” in line 64 is a strong priority claim particularly for a manuscript that does not yet demonstrate sufficient rigor in validation or presentation.

Response: We appreciate this comment and acknowledge that the previous wording could cause misunderstanding. These misleading statements have been removed. Accordingly, the final paragraph of the Introduction has been revised to provide a brief, sequential overview of the study, as shown in lines 65–70.” Accordingly, the objectives of this study are to: (i) develop an integrated numerical framework for geological carbon storage that couples multiphase flow, dissolution-driven convection, geochemical reaction, and gravity-induced ripening; (ii) elucidate the spatiotemporal evolution and interactions of major trapping mechanisms governing the long-term migration and phase transition of injected CO_2 in saline aquifers; (iii) quantify how reactive transport and associated porosity–permeability changes influence plume migration and dissolution behaviour; and (iv) evaluate the effects of calcite content and reservoir heterogeneity on the long-term fate of CO_2. ”

Comment 4:

There are some minor grammatical errors. For example, in line 78, “represents” should be “represent…, respectively”. The authors are requested to carefully check the manuscript to avoid similar issues.

Response: We appreciate the reviewer's careful examination and apologize for these grammar errors. The authors have carefully examined the entire manuscript text to avoid similar errors from occurring.

Comment 5:

The wording of some figure titles is inappropriate, such as Figure 4, “The distribution characteristics of the CO_2 saturations”.

Response: Thank you for your valuable suggestion. The wording here is indeed not accurate enough. We have changed the title of Fig. 4 and also made modifications to other figures. The title of Figure 4 has been revised to:” Figure 4: The CO_2 saturation distributions are presented at four timesteps: Stopping injection, 1 year after injection, 10 years after injection, and 50 years after injection. After 50 years of dissolution and chemical reactions, the thickness of the CO_2 plume has decreased from over 10 m to 5 m.”

Comment 6:

The treatment of Ostwald ripening as a post-processing 1D vertical simulation with no horizontal mass transfer needs to clarified. The consequences of this simplification and the reason for it should be discussed.

Response: Thank you for this suggestion. We agree that the assumption of negligible horizontal mass transfer requires further justification to improve the rigor of the model. The ripening process involves mass transfer at a given depth (dz), which tends to

homogenize the CO_2 saturation within a local representative elementary volume (REV). The ripening calculation was simplified as a one-dimensional vertical redistribution process because gravity-induced Ostwald ripening is primarily driven by the depth-dependent solubility contrast caused by hydrostatic pressure differences. Horizontal variations at the same depth are assumed to be secondary after the plume has reached a residual-trapping state. This simplification reduces computational complexity but also means that lateral redistribution of disconnected CO_2 ganglia cannot be explicitly resolved. This behavior is consistent with the fundamental concept underlying the finite element method, where properties within each element are treated as uniform. Consequently, we neglect the ripening-driven mass transfer within individual grid cells. We have added a clarifying statement in lines 149–151 of the revised manuscript: "Horizontal mass transfer would cause the residually trapped CO_2 at the same depth to become more uniform in size and would not lead to appreciable saturation changes at the macroscopic scale."

Comment 7:

The authors have treated the ripening mass transfer process as a post-processing step in the coupled simulation. The readers might be interested in knowing why a fully coupled approach could not be implemented.

Response: We thank the reviewer for this constructive comment. This assumption is based on the fact that the timescale of gravity-induced Ostwald ripening is far longer than the timescales of dissolution and geochemical reactions in our model. The simulation time for convective mixing and geochemical reactions is on the order of 100–1000 years, whereas previous theoretical studies indicate that the time required for ripening-induced CO_2 redistribution in a 50m-thick formation exceeds this by 2–3 orders of magnitude. Incorporating this process directly into the finite element

simulation would significantly reduce computational efficiency, and the amount of mass transferred vertically by ripening is extremely small. Given these considerations, we have implemented a data interface for this long-timescale mass transfer process, which performs a secondary calculation using the finite element simulation results as input.

We have added an explanation of the reasons for adopting this post-processing approach in Section 2.4 of the revised manuscript (lines 172-175): "Since the timescale of gravity-induced Ostwald ripening is 2–3 orders of magnitude larger than that of convective mixing and mineralization, the mass transfer term associated with ripening is very limited. To balance computational efficiency and convergence, we used the final results of the finite element computation as the initial condition for the ripening post-processing calculation."

Comment 8:

Line 317, the case referred to $\gamma = 1$ does not correspond with that in the figure.

Response: Thank you for your careful review. This should be the condition of $\gamma = 0.5$, and we have made the modification in line 321.

Dear Anonymous Referee #2,

We sincerely thank the reviewer for the positive assessment of our work and for recognizing the relevance of the proposed numerical framework to long-term CO_2 sequestration in saline aquifers. We also appreciate the constructive comments, which have helped us clarify several important assumptions and improve the technical rigor of the manuscript. Our point-by-point responses are provided below.

Comment 1:

(Section 2.1) How is the phase partitioning of CO_2 between the liquid and gas phase determined? While Equation 4 is provided, it is not clear to me if it is derived based on the thermodynamic principles.

Response: We thank the reviewer for this expert comment. Here, we indeed adopted a simplified mass transfer model based on Reference (Martinez, M. J., & Hesse, M. A. (2016). Two-phase convective CO_2 dissolution in saline aquifers. Water Resources Research, 52(1). https://doi.org/10.1002/2015WR017085). We did not provide a detailed explanation of this point. We assumed that dissolution occurs only in the two-phase region; therefore, the source term in the brine-saturated region is zero. In this simplified model, CO_2 dissolution is incorporated as a source term in the mass conservation equation. Mass transfer of CO_2 from the CO_2 phase to the brine phase can only take place when $S_g > 0$ and $c < c_{eq}$, i.e., when the brine phase is undersaturated with respect to CO_2. The mass transfer coefficient κ is the inverse time constant for dissolution and diffusion of gaseous CO_2 into fresh brine. This formulation allows us to simulate compositional two-phase flow problems without resorting to the computationally expensive compositional reservoir formulation.

We have added an explanation for adopting this simplified treatment in lines 87–89:" The dissolution of the CO_2 phase into the brine phase is assumed to occur only in the

two-phase region. Treating the dissolution mass as a source term in this way simplifies the simulation of multi-component two-phase flow problems and improves computational efficiency."

Comment 2:

(Section 2.2) The authors mentioned that the precipitation is neglected, yet this is a primary mechanism for mineral trapping. How is mineral trapping quantified in the following sections (e.g., the results shown in Figure 7) if precipitation is excluded?

Response: We thank the reviewer for the comment. We agree that precipitation significantly affects mineral trapping in the reservoir. However, the current model is not yet capable of accounting for mineral precipitation, and this remains a direction for our future research. In this study, we adopted a simplification in which only calcite is reactive in the reservoir, and the total amount of mineral trapping is quantified by the mass change of calcite. Since the timescale of multi-component chemical reactions is 1–2 orders of magnitude greater than that of convective mixing, we selected calcite—which has a relatively fast reaction rate—to focus on the coupling between reaction and convection. This approach is common in similar studies (10.1016/j.ijggc.2016.12.005, 10.1016/j.ijggc.2021.103365 and 10.1016/j.jclepro.2025.145948).

Comment 3:

- (Section 2.3)

(Line 141) Please justify why 500 years is considered reasonable for gas distribution to stabilize.

Response: Thank you for the comment. Our simulation results indicate that the average CO_2 saturation in the reservoir is nearly identical to the residual saturation, and in the region where the CO_2 phase is present, the brine has already reached the maximum concentration at 500 years. As shown by Eq. (4), dissolution will not proceed.

The coexistence of CO_2-saturated brine and residually trapped CO_2 is in fact an important assumption underlying gravity-induced Ostwald ripening. We therefore consider that the gas plume has stabilized after 500 years. We have supplemented this assumption with a clarifying statement in lines 143–146:" The gas distribution stabilizes and attains the initial ripening state under the assumption that the maximum concentration has been achieved throughout the domain. Our simulation results show that after 500 years, the average CO_2 saturation in the reservoir approaches the residual saturation, and according to Eq. (4), dissolution no longer occurs. Therefore, the simulation results at 500 years were selected as the initial condition for the gravity-induced Ostwald ripening process."

Comment 4:

(Line 143) Residual saturation appears to be a critical parameter for the ripening process. Have the authors considered incorporating hysteresis effect to give a more accurate evaluation of residual saturation?

Response: We fully agree with your perspective. We agree that hysteresis can improve the estimation of residual CO_2 saturation, particularly during drainage–imbibition transitions. In the present model, residual saturation was prescribed as an effective parameter to keep the full-cycle simulation tractable. We have now clarified this limitation and noted that incorporating hysteretic relative permeability and capillary pressure functions would be an important extension for more accurately distinguishing mobile, residual, and dissolved CO_2. The work by Wang et al. (2022) provides a detailed description of how to address the issue you raised, and we will consider adopting a similar approach in future work to distinguish residually trapped CO_2. This would greatly facilitate the characterization of transitions among different CO_2 trapping states.

Wang, Y., Vuik, C., & Hajibeygi, H. (2022). Analysis of hydrodynamic trapping interactions during full-cycle injection and migration of CO_2 in deep saline aquifers. *Advances in Water Resources*, *159*(July 2021), 104073. https://doi.org/10.1016/j.advwatres.2021.104073

Comment 5:

(Section 4.1) Ostwald ripening analysis is decoupled from the flow and transport simulation. Given that it takes quite a long time to reach equilibrium, is it physically consistent to incorporate it into the model, as CO_2 may have dissolved completely during this equilibrium stage?

Response: We thank the reviewer for this constructive comment. This assumption stems from the fact that the timescale of gravity-induced Ostwald ripening is far longer than those of dissolution and geochemical reactions in our model. The timescales of convective mixing and geochemical reactions considered here are on the order of 100–1000 years, whereas previous theoretical studies indicate that ripening-induced CO_2 redistribution in a 50-m-thick formation requires timescales 2–3 orders of magnitude longer. Directly incorporating this process into the finite element simulation would significantly reduce computational efficiency; moreover, the vertical mass transfer resulting from ripening is extremely small. Given these considerations, we have implemented a post-processing interface for this long-timescale process: the finite element simulation results are used as input for a subsequent secondary calculation of ripening-induced mass transfer.

We have added an explanation of the reasons for adopting this approach in Section 2.4 of the revised manuscript (lines 172-174): "Since the timescale of gravity-induced Ostwald ripening is 2–3 orders of magnitude larger than that of convective mixing and mineralization, the mass transfer term associated with ripening is very limited. To

balance computational efficiency and convergence, we used the final results of the finite element computation as the initial condition for the ripening post-processing calculation."

Comment 6:

(Section 4.2) Based on the results presented in Figure 9, changes in porosity may not be significant and the reaction has limited impact on the density-driven convective transport. Is this case representative of typical CO_2 storage in saline aquifers? Can the authors comment on this?

Response: We agree with the reviewer on this point. In the present model, the effect of geochemical reactions on the convective process is indeed not significant. The reservoir parameters we selected are primarily representative of typical high-permeability sandstone saline aquifers, and we simplified the system by considering calcite as the only reactive mineral component. Because formation properties and mineral assemblages vary across different reservoirs, the contribution of geochemical reactions to convective flow might not be necessarily as limited as in the case presented here (https://doi.org/10.1016/j.jclepro.2025.145948). In our previous study (https://doi.org/10.1007/s12665-025-12367-1), we have non-dimensionalized this coupled process by introducing the Rayleigh number (Ra) to characterize the intensity of convection and the Damköhler number (Da) to characterize the intensity of reaction, allowing us to systematically evaluate the impact of density-driven reactive flow under a wide range of geological conditions.

Dear Editor,

We sincerely thank you for the careful re-evaluation of our manuscript. We appreciate the editor's recognition that the manuscript has been substantially improved. We have carefully addressed the remaining concerns and revised the manuscript accordingly. Our point-by-point responses are provided below.

Comment 1:

At Line 30, the statement that the four trapping mechanisms "occur sequentially" is overly restrictive, because these mechanisms may overlap and operate concurrently over different timescales. Please revise this statement accordingly.

Response: Thank you for this valuable comment. We agree with the reviewer that describing the four trapping mechanisms as occurring sequentially is overly restrictive. We have revised the sentence at Lines 30–31 as follows:

"structural, residual, dissolution, and mineral trapping may operate concurrently and evolve over different timescales"

Comment 2:

Please carefully check the consistency between the in-text citations and the reference list. For example, Saaltink et al. are cited as both 2013a and 2013b, although only one corresponding reference is listed. Similarly, Wang et al. (2022b) is cited without a corresponding Wang et al. (2022a). These citations should be corrected and standardized throughout the manuscript.

Response: We thank the reviewer for identifying these inconsistencies. We have carefully cross-checked all in-text citations against the reference list and standardized the citation format throughout the manuscript.

Specifically, the citations to "Saaltink et al. (2013a)" and "Saaltink et al. (2013b)"

have been corrected to “Saaltink et al. (2013)”. Similarly, “Wang et al. (2022b)” has been corrected to “Wang et al. (2022).” Other citations and reference entries were also reviewed to ensure that every in-text citation has a corresponding entry in the reference list and that the year suffixes are used only when multiple publications by the same authors in the same year are cited. We also reformatted the complete reference list according to the HESS reference style and verified that every in-text citation has a corresponding reference-list entry and that every listed reference is cited in the manuscript.

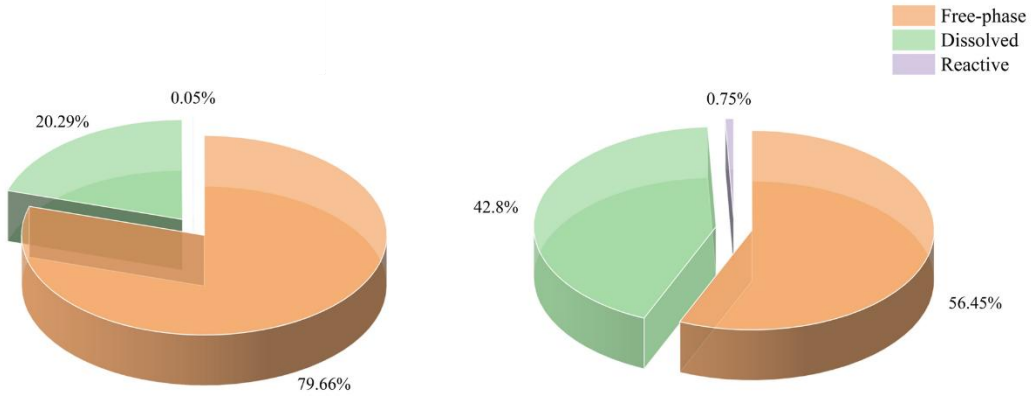
Comment 3:

Figure 7 and the associated interpretation require further revision. The use of a stacked-area plot with a logarithmic y-axis makes it difficult to interpret the stated relative contributions of approximately 60% physical trapping, 40% dissolution trapping, and less than 1% mineral trapping. It is unclear how these percentages can be directly derived from the current figure. Please revise the figure or its presentation so that the relative contributions of the different trapping mechanisms can be clearly and quantitatively identified.

More importantly, the treatment of calcite dissolution as “mineral trapping” requires reconsideration. Mineral trapping conventionally refers to the net incorporation of injected CO_2 into solid carbonate minerals through precipitation. Calcite dissolution may influence aqueous carbon storage and provide cations for subsequent mineral precipitation, but it does not by itself represent mineral trapping. The authors should therefore re-examine the definition and calculation of the mineral-trapping contribution reported in Figure 7.

Response: We sincerely thank you for this important comment. We agree that the original stacked-area plot with a logarithmic y-axis did not allow the relative

contributions of the different CO_2 states to be identified directly and quantitatively. To improve the clarity of Figure 7, we have replaced the original time-dependent stacked-area plot with two pie charts showing the relative proportions of CO_2 in the different modeled states at 1 year and 500 years, respectively. The percentage contribution of each state is now explicitly labeled in the figure. This revised presentation provides a direct comparison between the early and late stages of the simulation and more clearly illustrates the progressive transformation of CO_2 from the free-phase to the dissolved phase over time.



(a) 1year (b)500 years

Figure 7: Relative proportions of CO_2 in the free-phase state, dissolved state, and reactive state at simulation times of 1 and 500 years. The two pie charts illustrate the redistribution of injected CO_2.

We also agree with the reviewer that calcite dissolution should not be referred to as "mineral trapping." The present model considers calcite dissolution and associated geochemical reactions but does not include secondary carbonate precipitation. Therefore, the previously used term "mineral trapping" was inaccurate. Accordingly, we have replaced "mineral trapping" with "reactive CO_2" throughout the revised manuscript and Figure 7. See lines L273-279:

"Figure 7 illustrates the temporal evolution of the contribution of each trapping mechanism in the study case. Given the practical difficulty in distinguishing between

structural and residual trapping, these are collectively categorized as free-phase CO_2. The results indicate that free-phase CO_2 remains the dominant mechanism throughout the simulated period, accounting for 79.66% of total trapped CO_2 during the early injection period (within 1year). As dissolution and geochemical reactions progress, the proportion of free-phase CO_2 gradually decreases to 56.45% (500 years). After 500 years, dissolved CO_2 accounted for 42.80% of the total sequestered fraction. Reactive CO_2 constitutes less than 1% of the total. As dissolved CO_2 concentration declines in later stages, the reactive fraction stabilizes at 0.75%."