

1 Dear Anonymous Referee #2,

2 We sincerely thank the reviewer for the positive assessment of our work and for
3 recognizing the relevance of the proposed numerical framework to long-term CO₂
4 sequestration in saline aquifers. We also appreciate the constructive comments, which
5 have helped us clarify several important assumptions and improve the technical rigor
6 of the manuscript. Our point-by-point responses are provided below.

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8 **Comment 1:**

9 (Section 2.1) How is the phase partitioning of CO₂ between the liquid and gas phase
10 determined? While Equation 4 is provided, it is not clear to me if it is derived based on
11 the thermodynamic principles.

12 **Response:** We thank the reviewer for this expert comment. Here, we indeed adopted
13 a simplified mass transfer model based on Reference (Martinez, M. J., & Hesse, M. A.
14 (2016). Two-phase convective CO₂ dissolution in saline aquifers. *Water Resources*
15 *Research*, 52(1). <https://doi.org/10.1002/2015WR017085>). We did not provide a
16 detailed explanation of this point. We assumed that dissolution occurs only in the two-
17 phase region; therefore, the source term in the brine-saturated region is zero. In this
18 simplified model, CO₂ dissolution is incorporated as a source term in the mass
19 conservation equation. Mass transfer of CO₂ from the CO₂ phase to the brine phase can
20 only take place when $S_g > 0$ and $c < c_{eq}$, i.e., when the brine phase is undersaturated
21 with respect to CO₂. The mass transfer coefficient κ is the inverse time constant for
22 dissolution and diffusion of gaseous CO₂ into fresh brine. This formulation allows us
23 to simulate compositional two-phase flow problems without resorting to the
24 computationally expensive compositional reservoir formulation.

25 We have added an explanation for adopting this simplified treatment in lines 87–89:”

26 **The dissolution of the CO₂ phase into the brine phase is assumed to occur only in the**

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

30 **Comment 2:**

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

34 **Response:** We thank the reviewer for the comment. We agree that precipitation
35 significantly affects mineral trapping in the reservoir. However, the current model is not
36 yet capable of accounting for mineral precipitation, and this remains a direction for our
37 future research. In this study, we adopted a simplification in which only calcite is
38 reactive in the reservoir, and the total amount of mineral trapping is quantified by the
39 mass change of calcite. Since the timescale of multi-component chemical reactions is
40 1–2 orders of magnitude greater than that of convective mixing, we selected calcite—
41 which has a relatively fast reaction rate—to focus on the coupling between reaction and
42 convection. This approach is common in similar studies ([10.1016/j.ijggc.2016.12.005](https://doi.org/10.1016/j.ijggc.2016.12.005),
43 [10.1016/j.ijggc.2021.103365](https://doi.org/10.1016/j.ijggc.2021.103365) and [10.1016/j.jclepro.2025.145948](https://doi.org/10.1016/j.jclepro.2025.145948)).

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45 **Comment 3:**

46 - (Section 2.3)

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

49 **Response:** Thank you for the comment. Our simulation results indicate that the
50 average CO₂ saturation in the reservoir is nearly identical to the residual saturation, and
51 in the region where the CO₂ phase is present, the brine has already reached the
52 maximum concentration at 500 years. As shown by Eq. (4), dissolution will not proceed.

53 The coexistence of CO₂-saturated brine and residually trapped CO₂ is in fact an
54 important assumption underlying gravity-induced Ostwald ripening. We therefore
55 consider that the gas plume has stabilized after 500 years. We have supplemented this
56 assumption with a clarifying statement in lines 143–146:” The gas distribution
57 stabilizes and attains the initial ripening state under the assumption that the maximum
58 concentration has been achieved throughout the domain. Our simulation results show
59 that after 500 years, the average CO₂ saturation in the reservoir approaches the residual
60 saturation, and according to Eq. (4), dissolution no longer occurs. Therefore, the
61 simulation results at 500 years were selected as the initial condition for the gravity-
62 induced Ostwald ripening process.”

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64 **Comment 4:**

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

68 **Response:** We fully agree with your perspective. We agree that hysteresis can
69 improve the estimation of residual CO₂ saturation, particularly during drainage–
70 imbibition transitions. In the present model, residual saturation was prescribed as an
71 effective parameter to keep the full-cycle simulation tractable. We have now clarified
72 this limitation and noted that incorporating hysteretic relative permeability and
73 capillary pressure functions would be an important extension for more accurately
74 distinguishing mobile, residual, and dissolved CO₂. The work by Wang et al. (2022)
75 provides a detailed description of how to address the issue you raised, and we will
76 consider adopting a similar approach in future work to distinguish residually trapped
77 CO₂. This would greatly facilitate the characterization of transitions among different
78 CO₂ trapping states.

79 Wang, Y., Vuik, C., & Hajibeygi, H. (2022). Analysis of hydrodynamic trapping
80 interactions during full-cycle injection and migration of CO₂ in deep saline aquifers.
81 *Advances in Water Resources*, 159(July 2021), 104073.
82 <https://doi.org/10.1016/j.advwatres.2021.104073>

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84 **Comment 5:**

85 (Section 4.1) Ostwald ripening analysis is decoupled from the flow and transport
86 simulation. Given that it takes quite a long time to reach equilibrium, is it physically
87 consistent to incorporate it into the model, as CO₂ may have dissolved completely
88 during this equilibrium stage?

89 **Response:** We thank the reviewer for this constructive comment. This assumption
90 stems from the fact that the timescale of gravity-induced Ostwald ripening is far longer
91 than those of dissolution and geochemical reactions in our model. The timescales of
92 convective mixing and geochemical reactions considered here are on the order of 100–
93 1000 years, whereas previous theoretical studies indicate that ripening-induced CO₂
94 redistribution in a 50-m-thick formation requires timescales 2–3 orders of magnitude
95 longer. Directly incorporating this process into the finite element simulation would
96 significantly reduce computational efficiency; moreover, the vertical mass transfer
97 resulting from ripening is extremely small. Given these considerations, we have
98 implemented a post-processing interface for this long-timescale process: the finite
99 element simulation results are used as input for a subsequent secondary calculation of
100 ripening-induced mass transfer.

101 We have added an explanation of the reasons for adopting this approach in Section
102 2.4 of the revised manuscript (lines 172-174): "**Since the timescale of gravity-induced**
103 **Ostwald ripening is 2–3 orders of magnitude larger than that of convective mixing and**
104 **mineralization, the mass transfer term associated with ripening is very limited. To**

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

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109 **Comment 6:**

110 (Section 4.2) Based on the results presented in Figure 9, changes in porosity may not
111 be significant and the reaction has limited impact on the density-driven convective
112 transport. Is this case representative of typical CO₂ storage in saline aquifers? Can the
113 authors comment on this?

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

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