

1 Dear Anonymous Referee #1,

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

7

8 **Comment 1:**

9 In the assumptions section the authors state that the reservoir is homogeneous and
10 isotropic, whereas a later section explicitly investigates heterogeneity through
11 permeability anisotropy. Indeed, heterogeneity plays an important role in CO₂ migration
12 and trapping (see for example, Gas migration and residual trapping in bimodal
13 heterogeneous media during geological storage of CO₂, Advances in Water Resources
14 142, 103608). The effects of heterogeneity should be investigated. But the manuscript
15 should clearly distinguish the base-case assumptions from the sensitivity-analysis
16 settings. In addition, setting an anisotropy ratio alone cannot fully represent the effects
17 of heterogeneity.

18 **Response:** Thank you for this valuable comment. We fully agree that heterogeneity
19 is a key control factor on CO₂ migration and trapping, and that permeability anisotropy
20 alone cannot fully represent the complexity of heterogeneous geological media. In the
21 revised manuscript, we have clarified the distinction between the **base-case model**
22 **assumptions** and the **sensitivity-analysis scenarios**. Specifically, the assumption of a
23 homogeneous and isotropic reservoir applies only to the reference case used to establish
24 the base behavior of the system, whereas the later section introducing permeability
25 anisotropy is intended as a sensitivity analysis. We have revised the title of Section 4.3
26 to “**Effect of permeability anisotropy**”. We have added the description regarding the

27 non-homogeneity assumption in lines 318-321.

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

33 We have cited the paper recommended by the reviewers, which has enhanced our
34 understanding of how heterogeneity affects the migration of plumes. The influence of
35 this overall heterogeneity has been investigated in our subsequent studies. See lines 41-
36 42:

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

39

40 **Comment 2:**

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

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

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

53 assumption was adopted to reduce the complexity of the coupled multiphase flow and
54 geochemical transport problem and to focus on the long-term evolution of CO₂
55 migration, dissolution, and reaction under a prescribed reservoir temperature. In
56 subsequent studies, we will incorporate a coupled analysis of the temperature and
57 mechanical fields. We have added an explanation in lines 216–217:” (7) Based on the
58 geothermal gradient variation data in the reservoir, the temperature variation within the
59 simulation domain is approximately 1–3 °C. We therefore assume the simulation
60 domain to be an isothermal saline aquifer.”

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

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

79 minerals (calcite, dolomite), considering quartz, feldspar, and other minerals as
80 insoluble substances because their dissolution/precipitation process takes thousands of
81 years (De Silva et al., 2015). The formation of carbonate is considered to be the main
82 way to sequester CO₂ and an important process for achieving permanent trapping in
83 GCS (Bachu et al., 1994). To simplify the model, it is assumed that the reservoir
84 minerals involved in the chemical reaction only contain 5% calcite (Sainz-Garcia et al.,
85 2017b).”

86

87 **Comment 3:**

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

92 **Response:** We appreciate this comment and acknowledge that the previous wording
93 could cause misunderstanding. These misleading statements have been removed.
94 Accordingly, the final paragraph of the Introduction has been revised to provide a brief,
95 sequential overview of the study, as shown in lines 65–70.” **Accordingly, the objectives**
96 **of this study are to: (i) develop an integrated numerical framework for geological**
97 **carbon storage that couples multiphase flow, dissolution-driven convection,**
98 **geochemical reaction, and gravity-induced ripening; (ii) elucidate the spatiotemporal**
99 **evolution and interactions of major trapping mechanisms governing the long-term**
100 **migration and phase transition of injected CO₂ in saline aquifers; (iii) quantify how**
101 **reactive transport and associated porosity–permeability changes influence plume**
102 **migration and dissolution behaviour; and (iv) evaluate the effects of calcite content and**
103 **reservoir heterogeneity on the long-term fate of CO₂. ”**

104

105 **Comment 4:**

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

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

112

113 **Comment 5:**

114 The wording of some figure titles is inappropriate, such as Figure 4, “The distribution
115 characteristics of the CO₂ saturations”.

116 **Response:** Thank you for your valuable suggestion. The wording here is indeed not
117 accurate enough. We have changed the title of Fig. 4 and also made modifications to
118 other figures. The title of Figure 4 has been revised to:” **Figure 4: The CO₂ saturation
119 distributions are presented at four timesteps: Stopping injection, 1 year after injection,
120 10 years after injection, and 50 years after injection. After 50 years of dissolution and
121 chemical reactions, the thickness of the CO₂ plume has decreased from over 10 m to 5
122 m.”**

123

124 **Comment 6:**

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

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

131 homogenize the CO₂ saturation within a local representative elementary volume (REV).
132 The ripening calculation was simplified as a one-dimensional vertical redistribution
133 process because gravity-induced Ostwald ripening is primarily driven by the depth-
134 dependent solubility contrast caused by hydrostatic pressure differences. Horizontal
135 variations at the same depth are assumed to be secondary after the plume has reached a
136 residual-trapping state. This simplification reduces computational complexity but also
137 means that lateral redistribution of disconnected CO₂ ganglia cannot be explicitly
138 resolved. This behavior is consistent with the fundamental concept underlying the finite
139 element method, where properties within each element are treated as uniform.
140 Consequently, we neglect the ripening-driven mass transfer within individual grid cells.
141 We have added a clarifying statement in lines 149–151 of the revised manuscript:
142 “Horizontal mass transfer would cause the residually trapped CO₂ at the same depth to
143 become more uniform in size and would not lead to appreciable saturation changes at
144 the macroscopic scale.”

145

146 **Comment 7:**

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

150 **Response:** We thank the reviewer for this constructive comment. This assumption is
151 based on the fact that the timescale of gravity-induced Ostwald ripening is far longer
152 than the timescales of dissolution and geochemical reactions in our model. The
153 simulation time for convective mixing and geochemical reactions is on the order of
154 100–1000 years, whereas previous theoretical studies indicate that the time required for
155 ripening-induced CO₂ redistribution in a 50m-thick formation exceeds this by 2–3
156 orders of magnitude. Incorporating this process directly into the finite element

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

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

169

170 **Comment 8:**

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

172 **Response:** Thank you for your careful review. This should be the condition of $\gamma =$
173 **0.5**, and we have made the modification in line 321.