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# The impact of local-scale processes on large-scale CO<sub>2</sub> migration and immobilization

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#### Abstract

Storage security of injected carbon dioxide (CO2) is an essential component of risk management for geological carbon sequestration operations. During the injection and early post-injection periods, CO2 leakage may occur along faults and leaky wells, but this risk may be partly managed by proper site selection and sensible deployment of monitoring and remediation technologies. On the other hand, long-term storage security is an entirely different risk management problem—one that is dominated by a mobile CO<sub>2</sub> plume that may travel over very large spatial distances, over long time periods, before it is trapped by a variety of different physical and chemical processes. In the post-injection phase, the mobile CO<sub>2</sub> plume migrates in large part due to buoyancy forces, following the natural topography of the geological formation. The primary trapping mechanisms are capillary and solubility trapping, which evolve over thousands to tens of thousands of years and can immobilize a significant portion of the mobile, free-phase CO<sub>2</sub> plume. However, both the migration and trapping processes are inherently complex, involving a combination of small and large spatial scales and acting over a range of time scales. Solubility trapping is a prime example of this complexity, where small-scale density instabilities in the dissolved CO<sub>2</sub> region leads to convective mixing that has that has a significant effect on the large-scale dissolution process over very long time scales. Another example is the effect of capillary forces on the evolution of mobile CO<sub>2</sub>, an often-neglected process except with regard to residual trapping. As the plume migrates due to buoyancy and viscous forces, local capillary effects acting at the CO<sub>2</sub>-brine interface lead to a transition zone where both fluids are present in the mobile state. This small-scale effect may have a significant impact on large-scale plume migration as well as long-term residual and dissolution trapping. Using appropriate models that can capture both large and small-scale effects is essential for understanding the role of these processes on the long-term storage security of CO<sub>2</sub> sequestration operations.

There are several approaches to modeling long-term CO<sub>2</sub> trapping mechanisms. One modeling option is the use of traditional numerical methods, which are often highly sophisticated models that can handle multiple complex phenomena with high levels of accuracy. However, these complex models quickly become prohibitively expensive for the type of large-scale, long-term modeling that is necessary for risk assessment applications such as the late post-injection period. We present an alternative modeling option that combines vertically-averaged governing equations with an upscaled representation of the dissolution-convective mixing process and the local capillary transition zone at the CO<sub>2</sub>-brine interface. CO<sub>2</sub> injection is solved numerically on a coarse grid, capturing the large-scale injection problem and the post-injection capillary trapping, while the upscaled dissolution and capillary fringe models capture these subscale effects and eliminate the need for expensive grid refinement to capture the subscale instabilities associated with convective mixing or the details of the capillary transition zone. With this

\* Corresponding author. Tel.: +1-919-966-5430. *E-mail address*: sgasda@unc.edu. modeling approach, we demonstrate the effect of different modeling choices associated with dissolution and capillary processes for typical large-scale geological systems.

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# 1. Introduction

Geological carbon sequestration is a promising strategy for reducing anthropogenic carbon dioxide (CO<sub>2</sub>) emissions from large stationary sources and, as a result, can stabilize atmospheric levels of CO<sub>2</sub> and lessen the impact of climate change [1]. Storage security of injected CO<sub>2</sub> during the injection and post-injection phases is an essential component of the ultimate success of this technology. Short-term storage security involves mitigation of vertical CO<sub>2</sub> migration through the overlying caprock, which may in part be minimized by proper site selection and sensible deployment of monitoring and remediation strategies. However, long-term post-injection storage security involves ensuring the eventual immobilization of free-phase CO<sub>2</sub> in a more permanent residual or dissolved state, a process that may take thousands to tens of thousands of years to occur [1]. During this time, buoyancy forces and topographical features may allow the free-phase, mobile CO<sub>2</sub> to migrate laterally over very large distances.

An important aspect of long-term solubility trapping of CO<sub>2</sub> is the onset of convective mixing at small length scales, which can greatly enhance and accelerate the dissolution process during the post-injection period [2]. In addition, local capillary forces can significantly effect the migration of the CO<sub>2</sub> plume over large distances [3]. Understanding the effect of these small-scale processes over large spatial and temporal scales is a critical component of long-term risk assessment. However, traditional numerical methods [4] are unable to capture both small and large-scale processes in an efficient manner, and analytical methods [5,6] are limited in their scope. Ultimately, these open issues in long-term storage security can only be addressed through the development of upscaled numerical models that use subscale approximations for critical physical processes. In this paper, we employ approximations to extend our previous work on vertically-averaged numerical models of CO<sub>2</sub> injection and migration to include dissolution-convection processes and local-scale capillary effects. We use this modeling approach to explore the sensitivity of long-term storage security on these subscale processes.

# 2. Model Approach

This work is based upon a set of vertically-averaged models that are presented in greater detail elsewhere [7,8]. The model employs a vertical-equilibrium assumption, which allows for derivation of a set of governing equations for the CO<sub>2</sub>-brine two-phase system, while also accounting for the dissolved mass fraction of CO<sub>2</sub> in brine. As depicted in Figure 1, the system is composed of four macroscopic interfaces, where  $\zeta_T = \zeta_T(x_I, x_2)$  and  $\zeta_B = \zeta_B(x_I, x_2)$  are the top and bottom surfaces of the formation, respectively, and  $\zeta_M = \zeta_M(x_I, x_2, t)$  and  $\zeta_R = \zeta_R(x_I, x_2, t)$  are the bottom interfaces of the Mobile and Residual CO<sub>2</sub> macroscopic regions, respectively.

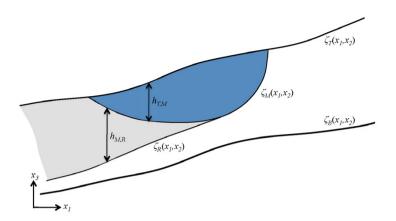


Figure 1 Schematic of CO<sub>2</sub>-brine system. The relevant macroscopic interfaces are the top and bottom surfaces of the formation,  $\zeta_T$  and  $\zeta_B$ , and the bottom of the mobile and residual CO<sub>2</sub> regions,  $\zeta_M$  and  $\zeta_R$ .

The conservation of mass equations for each component  $M^{\alpha}$  are

$$\frac{\partial M^{\alpha}}{\partial t} + \nabla \cdot \mathbf{F}^{\alpha} = 0, \quad \alpha = c, b$$
 (1)

The conservation of total dissolved  $CO_2$  in the brine phase,  $M_b$ , is modeled with the following transport equation,

$$\frac{\partial M_b^c}{\partial t} + \nabla \cdot \int_{\varsigma_B}^{\varsigma_T} \boldsymbol{u}_b \rho_b m_b^c dx_3 = C_{diss}(\omega, \varsigma_R)$$
(2)

where the second term on the left hand side represents the total flux of dissolved  $CO_2$  in the brine phase, and the right hand side represents kinetic mass exchange for dissolved  $CO_2$  that is governed by the vertically-averaged concentration dissolved  $CO_2$  in brine,  $\omega$ . Equation (2) holds subject to the constraint that  $0 \le \omega \le 1$ .

Equation (2) captures two dissolution mechanisms: 1) equilibrium between the mobile CO<sub>2</sub> phase and region of pure residual brine during drainage and residual CO<sub>2</sub> and mobile brine during imbibition; and 2) mass transfer of CO<sub>2</sub> into the brine region through dissolution combined with convective mixing. For the latter kinetic process, a number of detailed numerical and experimental studies have found that the overall mass transfer rate due to the dissolution-convection process is roughly constant in time [9,10], which is adopted in this model.

The final equation accounts for hysteresis and evolution of the residual  $CO_2$  interface  $\zeta_R$ . This is done by considering the conservation equation for mobile  $CO_2$ ,

$$-\phi(1-s_{c,r}-s_{b,r})\frac{\partial \zeta_M}{\partial t} + \nabla \cdot \int_{\varsigma_B}^{\varsigma_T} \boldsymbol{u}_c \rho_c m_c^c dx_3 = 0$$
(3)

where  $\phi$  is the porosity,  $s_{\alpha,r}$  are the residual saturations for each phase, and the second term represents the vertically-integrated CO<sub>2</sub> phase velocity  $u_c$ .

In previous work [8], the model derivation presented above employed an additional assumption regarding complete gravity segregation and a sharp interface between the two fluids. In this study, the sharp-interface assumption has been relaxed to include the presence of local capillary effects that creates a transition zone of  $CO_2$  saturation, referred to as the capillary fringe. The inclusion of the capillary fringe in vertically-integrated models is described in detail elsewhere [11,3]. In regard to Figure 1, the interface  $\zeta_M$  indicates the bottom of the capillary transition zone.

The above system of equations, (1)-(3), is solved in an IMPES-type strategy whereby a pressure is solved implicitly under the assumption of an incompressible system. For the transport equations, we consider a splitting-type approach, evolving  $\zeta_M$  first, then  $\zeta_R$ , and finally the concentration  $\omega$ . The system is solved numerically using a standard finite-difference approximation.

# 3. Model Application

The above-described model is applied to the Johansen formation [12], which is a large saline aquifer approximately 3,000 below the North Sea and is a prospective site for injection of onshore Norwegian emissions. The Johansen formation is characterized by large areal extent, heterogeneity, a significant dip, and a major fault line splitting the formation [13]. For this problem,  $CO_2$  is injected through a vertical well (x = 32.25 km, y = 29.75 km) at a constant rate of 30 kg/s for 50 years and then allowed to migrate due to buoyancy forces until the plume is immobilized by residual and/or solubility trapping mechanisms.

We investigate the effect of four different modeling choices on the evolution of mobile and trapped  $CO_2$  over tens of thousands of years. The first case considers only structural  $CO_2$  trapping and no solubility or residual trapping. The second case only considers trapping of  $CO_2$  in the residual phase and employs the sharp-interface assumption. The third case includes capillary fringe effects with residual trapping but does not model dissolution. The local capillary-saturation model employed for this case is  $Pc = \alpha(S_{w,n})^{-1/2}$  with the parameter  $\alpha = 2 \times 10^4$  Pa. Finally, the last case extends the second case by modeling the additional mechanism of dissolution through convective mixing but does not include capillary fringe effects. The upscaled dissolution rate is modeled as  $C_{diss} = 1 \text{ kg/m}^2/\text{yr}$  unless otherwise noted.

For the above cases, we consider an incompressible system and assign the fluid and porous media properties defined in the original benchmark definition [12,8]. The post-injection period is modelled for 5,000 years or until the mobile plume reaches the edge of the domain, a minimum distance of 25 km from the injection point for this problem.

### 4. Results and Discussion

When the sharp-interface model is applied to the Johansen formation (Figure 2) without considering trapping by either residual or solubility processes, the CO<sub>2</sub> travels as far as 10 km from the injection well in the upslope direction within the first 50 years post-injection. After 1,000 years, the maximum travel distance is approximately 30 km, and the far right edge of the plume reaches the outer boundary, which is 25 km away, within that time. The plume thickness varies significantly over its areal extent, which is due to a combination of structural and heterogeneity effects. Eventually, most of the CO<sub>2</sub> will reach the outer boundary when residual and solubility trapping mechanisms are not modeled.

In comparison, Figure 3 shows the effect of residual trapping, where in this case the residual saturation of CO<sub>2</sub> is assigned to be 50%. The maximum travel distance is nearly the same after the first 100 years. However, by 1,000 years, the mobile CO<sub>2</sub> travels as far as the far right boundary, but by this time, approximately 90% immobilized by residual trapping mechanisms.

The model results with capillary effects are shown in Figure 4. At the end of the 50-year injection period, the maximum extent CO<sub>2</sub> plume is approximately 5 km in the updip direction.

After 100 years, the mobile CO<sub>2</sub> only migrates a few more kilometers upslope while trapping approximately 40% of the CO<sub>2</sub> mass in residual phase. After 1,000 years, some mobile CO<sub>2</sub> remains but is essentially immobilized by a combination of capillary effects and topographical features. The CO<sub>2</sub> mass trapped residually is approximately 70% of the total mass.

The dissolution model results in Figure 5 show that plume migrates approximately 5 km upslope from the injection well by the end of injection, which increases to 7 km within 100 years. By 1,000 years, the CO<sub>2</sub> has been almost completely immobilized by either residual or solubility trapping processes. The extent of the dissolved CO<sub>2</sub> shows that mobile CO<sub>2</sub> traveled as far as 10 km from the injection well, but since the outer plume edge is very thin under the sharp interface assumption, the mobile CO<sub>2</sub> in those regions is dissolved relatively quickly. At 100 years, approximately 50% of the total mass is trapped residually and 15% is trapped in the dissolved state. After 1,000 years, 90% of the total mass is immobilized, with 40% in the residual state and 50% trapped in the dissolved state. Over time, the residual CO<sub>2</sub> dissolves until the thickness of the dissolved region reaches the bottom of the aquifer.

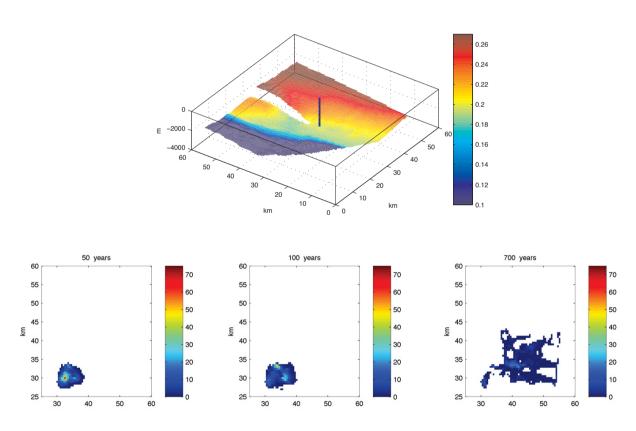


Figure 2 Sharp-interface model results of post-injection CO<sub>2</sub> migration without trapping in the Johansen formation. The geometry and porosity distribution of the formation and the location of the injection well are shown at top. At bottom is the thickness in meters of the mobile CO<sub>2</sub> over 1,000 yrs.

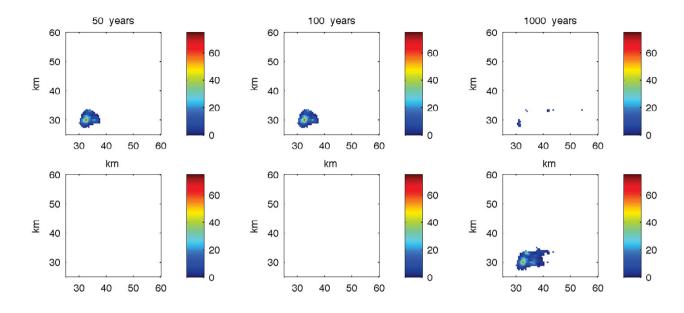


Figure 3 Sharp-interface model results of post-injection CO<sub>2</sub> migration with residual trapping in the Johansen formation. Shown is the thickness in meters of the mobile CO<sub>2</sub> in the top panels and of the residual CO<sub>2</sub> in the bottom panels over 1,000 yrs.

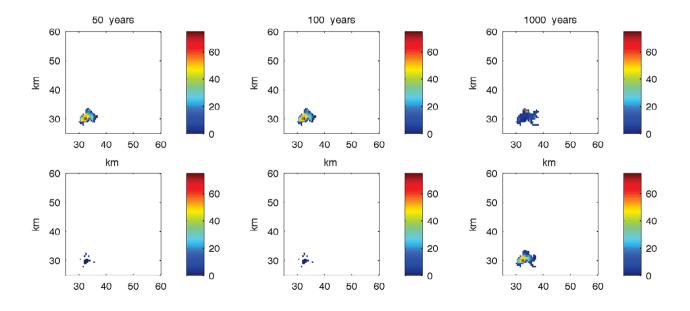


Figure 4 Capillary fringe model results of post-injection CO<sub>2</sub> migration and trapping in the Johansen formation. Shown is the thickness in meters of the mobile CO<sub>2</sub> in the top panels and of the residual CO<sub>2</sub> in the bottom panels over 1,000 yrs

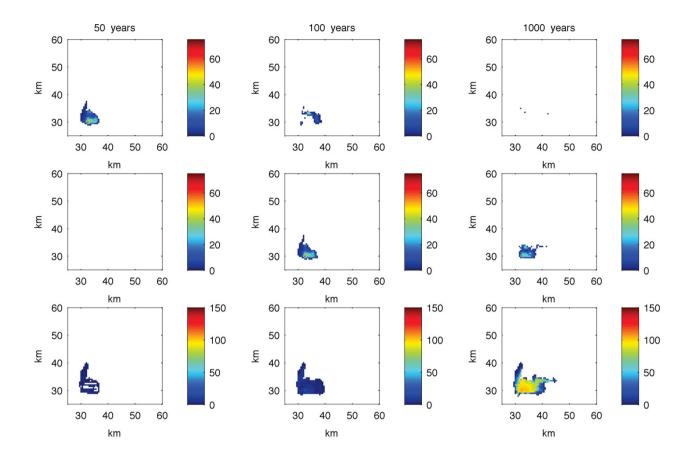


Figure 5 Dissolution model results of post-injection CO<sub>2</sub> migration and trapping in the Johansen formation. Shown is the thickness in meters of the mobile CO<sub>2</sub> in the top panels, of the residual CO<sub>2</sub> in the middle panels, and of the dissolved CO<sub>2</sub> in the bottom panels over 1,000 yrs.

## 5. Conclusions

- Vertically-integrated approaches to modeling CO<sub>2</sub> injection can be used to investigate CO<sub>2</sub> migration and trapping over length scales on the order of 100 km and time scales greater than 10,000 years.
- Subscale models for local effects such as a capillary fringe and convective-mixing enhanced dissolution can be included in vertically-integrated models and applied to realistic geologic systems.
- Capillary fringe effects result in slower rate of upslope migration and decreased maximum travel distances by several kilometers for systems with large capillary transition zones.
- CO<sub>2</sub> dissolution enhanced by convective mixing can immobilize injected CO<sub>2</sub> within a few hundred years.
- Vertically-integrated models can be used to assess the impact of different processes on CO<sub>2</sub> immobilization and are important tools for managing long-term risk of CO<sub>2</sub> injection operations.

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