Spreading and convective dissolution of carbon dioxide in vertically confined, horizontal aquifers

Christopher W. MacMinn, Jerome A. Neufeld, Marc A. Hesse, and Herbert E. Huppert

1. Introduction

Injection of carbon dioxide (CO₂) into saline aquifers is a promising tool for reducing anthropogenic CO₂ emissions. At reservoir conditions, the injected CO₂ is buoyant relative to the ambient groundwater. The buoyant plume of CO₂ rises toward the top of the aquifer and spreads laterally as a gravity current, presenting the risk of leakage into shallower formations via a fracture or fault. In contrast, the mixture that forms as the CO₂ dissolves into the ambient water is denser than the water and sinks, driving a convective process that enhances CO₂ dissolution and promotes stable long-term storage. Motivated by this problem, we study convective dissolution from a buoyant gravity current as it spreads along the top of a vertically confined, horizontal aquifer. We conduct laboratory experiments with analog fluids (water and a mixture of methanol and ethylene glycol) and compare the experimental results with simple theoretical models. Since the aquifer has a finite thickness, dissolved buoyant fluid accumulates along the bottom of the aquifer, and this mixture spreads laterally as a dense gravity current. When dissolved buoyant fluid accumulates slowly, our experiments show that the spreading of the buoyant current is characterized by a parabola-like advance and retreat of its leading edge. When dissolved buoyant fluid accumulates quickly, the retreat of the leading edge slows as further dissolution is controlled by the slumping of the dense gravity current. We show that simple theoretical models predict this behavior in both limits, where the accumulation of dissolved buoyant fluid is either negligible or dominant. Finally, we apply one of these models to a plume of CO₂ in a saline aquifer. We show that the accumulation of dissolved CO₂ in the water can increase the maximum extent of the CO₂ plume by several fold and the lifetime of the CO₂ plume by several orders of magnitude.

understanding of both the onset [Ennis-King et al., 2005; Riaz et al., 2006; Hidalgo and Carrera, 2009; Slim and Ramakrishnan, 2010; Backhaus et al., 2011] and the subsequent rate of the convective dissolution of a stationary layer of CO₂ overlying a reservoir of water [Kneafsey and Pruess, 2010; Pau et al., 2010; Neufeld et al., 2010; Backhaus et al., 2011]. These results have been used to incorporate upscaled models for convective dissolution into models for the spreading and migration of buoyant plumes of CO₂ after injection [Gasda et al., 2011; MacMinn et al., 2011]. However, convective dissolution has not been studied experimentally in the context of a gravity current that spreads as it dissolves, and the interaction between these two processes is not understood. In addition, in an aquifer of finite thickness the accumulation of dissolved CO₂ in the water beneath the spreading plume may strongly influence the rate of convective dissolution.

[5] Here, we consider the simple model problem of convective dissolution from a buoyant gravity current as it spreads along the top boundary of a vertically confined, horizontal aquifer (Figure 1). We first study this system experimentally using analog fluids. Motivated by our experimental observations, we then study theoretical models for this system based on the well-known theory of gravity currents [Bear, 1972; Huppert and Woods, 1995], which has recently been used to develop physical insight into CO₂ injection [Lyle et al., 2005; Nordbotten and Celia, 2006] and postinjection spreading and migration [Hesse et al., 2007, 2008; Juanes et al., 2010; MacMinn et al., 2011; de Loubens and Ramakrishnan, 2011; Golding et al., 2011]. We show that the interaction between buoyant spreading, convective dissolution, and the finite thickness of the aquifer has a strong influence on the maximum extent and the lifetime of the buoyant current.

\[ h(z, t) \]

**Figure 1.** A sketch of the simple model problem considered here: a buoyant gravity current (dark blue) spreads beneath a horizontal caprock in a vertically confined aquifer, shrinking as the buoyant fluid dissolves into the ambient fluid via convective dissolution (light blue).
152 influence on the propagation of the gravity currents or the 
153 rate of convective dissolution. After each experiment, we 
154 drain the flow cell, remove the beads, and wash and dry 
155 both the beads and the cell.

156 We use the resulting time-lapse images from each 
157 experiment to quantify the rate at which the buoyant cur- 
158 rent spreads by measuring the position $x_N$ of its leading 
159 edge or ‘nose’ as a function of time [e.g., Huppert, 1982; 

160 Huppert and Woods, 1995]. We measure the nose position 
161 in the images either by eye or programmatically based on a 
162 light intensity threshold; the two methods give similar 
163 results because the interface between the two fluids is sharp.

164 We measure the void fraction or porosity $\phi$ of the 
165 bead pack for each experiment directly by measuring the 
166 volume of fluid added to the cell during filling. Porosity 
167 measurements for different packings of the same bead size 
168 were repeatable to within a few percent.

169 [13] Rather than measuring the permeability of each 
170 individual bead pack directly, we instead infer the effective 
171 permeability $k$ of these packings from a series of bench- 
172 mark experiments where a buoyant gravity current of fresh 
173 water spreads over ambient saltwater. We review the well-
174 known theoretical model for this problem [Bear, 1972; 
175 Huppert and Woods, 1995] in section 4.1. We prepare and 
176 initiate these benchmark experiments following the proce-
177 dure described at the beginning of this section. Since the 
178 rate of spreading is directly proportional to the permeability 
179 of the bead pack, and all other parameters are known, we 
180 infer the value of the permeability as that which gives the 
181 best agreement between the experimental measurements 
182 and the predictions of the model (Figure 4). We repeat 
183 these measurements several times to ensure reproducibility, 
184 and we take the result to be representative of the permeabil-
185 ity of all such packings of beads of the same size, prepared 
186 following the same protocol.

187 [14] We work with three different bead sizes, with nomi-
188 nal diameters of 1, 2, and 3 mm. We report the measured 
189 porosity and effective permeability of packings of these 
190 beads in Table 2. The effective permeability values are in 
191 all cases within a factor of about 2 of the values estimated 
192 from the Kozeny-Carman relation. The measured values 
193 are consistently smaller than the Kozeny-Carman values, 
194 which may be due to the fact that the beads were not 
195 monodisperse.

3. Experimental Results

We first study the spreading and convective dissolu-
198 tion of buoyant currents of the 59.1 wt % MEG. We 
199 observe that the nose of the current gradually decelerates 
200 until it reaches a maximum position, and subsequently

Table 1. Properties of the Three Pairs of Buoyant and Ambient Fluids Used in the Experiments$^a$

<table>
<thead>
<tr>
<th>Buoyant Fluid</th>
<th>Ambient Fluid</th>
<th>$\rho_b$ (g cm$^{-3}$)</th>
<th>$\mu_b$ (P)</th>
<th>$\rho_a$ (g cm$^{-3}$)</th>
<th>$\mu_a$ (P)</th>
<th>$\Delta \rho$ (g cm$^{-3}$)</th>
<th>$\Delta \mu$ (g cm$^{-3}$)</th>
<th>$M$</th>
</tr>
</thead>
<tbody>
<tr>
<td>59.1 wt % MEG</td>
<td>water</td>
<td>0.971</td>
<td>0.031</td>
<td>0.999</td>
<td>0.095</td>
<td>0.028</td>
<td>0.0026</td>
<td>0.31</td>
</tr>
<tr>
<td>65.4 wt % MEG</td>
<td>water</td>
<td>0.991</td>
<td>0.039</td>
<td>0.999</td>
<td>0.095</td>
<td>0.008</td>
<td>0.0109</td>
<td>0.24</td>
</tr>
<tr>
<td>Water</td>
<td>saltwater</td>
<td>0.999</td>
<td>0.095</td>
<td>1.007</td>
<td>0.095</td>
<td>0.008</td>
<td>1</td>
<td>1</td>
</tr>
</tbody>
</table>

$^a$The saltwater is water with 1.075 wt % NaCl.
retracts at an accelerating rate until the current dissolves completely. We find that the current typically breaks into a series of discrete patches as it nears complete dissolution (Figure 5).

[16] For buoyant currents of 65.4 wt % MEG, we see different behavior. As with the 59.1 wt % MEG, the nose of the current decelerates, reaches a maximum position, and then retreats to the left. Rather than accelerating on the retreat, however, the nose retreats at a rate that is approximately constant (Figure 6).

[17] The buoyant current of 59.1 wt % MEG advances quickly (driven by \( \Delta \rho \)) relative to the rate at which water with dissolved MEG falls downward (driven by \( \Delta \rho_w \)). The buoyant current of 65.4 wt % MEG advances more slowly and dissolves more quickly (\( \Delta \rho \) is smaller and \( \Delta \rho_w \) is larger), and we observe that water with dissolved MEG reaches the bottom of the flow cell and accumulates there, building and spreading as a dense gravity current. This accumulation increases the density of the ambient fluid beneath the buoyant current, slowing the process of convective dissolution and retarding the retreat of the nose.

### 4. Theoretical Models

[18] We now consider theoretical models for this problem. We assume that the buoyant fluid has constant density \( \rho_b \) and viscosity \( \mu_b \), and that the ambient fluid has constant density \( \rho_a \) and viscosity \( \mu_a \). We assume that the aquifer is vertically confined, and has uniform porosity \( \phi \), permeability \( k \), and thickness \( H \) (Figure 1). For simplicity, we consider a planar, two-dimensional system and assume no fluid flow or variations in fluid velocities along the third (y) dimension (into the page). The shape of the buoyant current is characterized by its initial length \( L \) in the x direction, height \( H \) in the z direction, and width \( W \) in the y direction, so that the buoyant current initially occupies a volume \( V = LWH \) of the aquifer and contains a corresponding volume \( \phi V \) of buoyant fluid.

[19] We assume vertical flow equilibrium (i.e., the Dupuit approximation), neglecting the vertical component of the fluid velocity relative to the horizontal one and taking the pressure field to be hydrostatic [Coats et al., 1971; Yortsos, 1995]. We further neglect the capillary pressure relative to typical gravitational and viscous pressure changes, and also assume that the interface between the two fluids remains sharp. Although both capillary pressure and saturation gradients can be included in such models [Nordbotten and Dahle, 2011; Golding et al., 2011; Zhao et al., 2012], the interaction of these effects with convective dissolution is not clear, and they are not present in our experimental analog system because the fluids are perfectly miscible. Lastly, we require for mass conservation that there be no net flux of fluid through any cross section of the aquifer, because it is vertically confined.

#### 4.1. Buoyant Spreading Without Convective Dissolution

[20] In the absence of convective dissolution, the model for the spreading of a buoyant current in a confined porous layer can be written [e.g., Bear, 1972, p. 535, equation 9.5.64]

\[
\frac{\partial h}{\partial t} - U \frac{\partial}{\partial x} \left[ (1-f)h \frac{\partial h}{\partial x} \right] = 0, \tag{1}
\]

where \( h(x,t) \) is the unknown thickness of the current (Figure 1), \( U = \Delta \rho g k / \phi \mu_a \) is the characteristic buoyancy velocity of the current. The function \( f(h) \) is given by

\[
f = \frac{Mh}{(M-1)h + H}, \tag{2}
\]
with mobility ratio $M = \mu_\rho / \mu_s$. The presence of $f(h)$ reflects the fact that the aquifer is vertically confined, so that the buoyant fluid must displace the relatively viscous ambient fluid in order to spread. Flow of the ambient fluid becomes unimportant at late times as the current becomes thin relative to the aquifer thickness ($f(h) \ll 1$ when $h \ll H$). The spreading behavior becomes independent of $M$ in this unconfined limit [Barenblatt, 1992; Hesse et al., 2007].

To rewrite equation (1) in dimensionless form, we define scaled variables

$$\tilde{h} = \frac{h}{H}, \quad \tilde{t} = \frac{UH}{L^2}t, \quad \tilde{x} = \frac{x}{L}. \quad (3)$$

and obtain

$$\frac{\partial \tilde{h}}{\partial \tilde{t}} = \frac{\partial}{\partial \tilde{x}} \left[ (1-f) \tilde{h} \frac{\partial \tilde{h}}{\partial \tilde{x}} \right] = 0. \quad (4)$$

In the unconfined limit, equation (4) has a similarity solution from Barenblatt [1992],

$$h(\tilde{x}, \tilde{t}) = \frac{1}{6} \tilde{t}^{-1/3} \left( 9^{2/3} - \frac{\tilde{x}^2}{\tilde{t}^{1/3}} \right), \quad (5)$$

valid for $|\tilde{x}| \leq \tilde{x}_N(\tilde{t})$ and $\tilde{t} > 0$. Equation (5) indicates that the position $\tilde{x}_N$ of the nose of an unconfined current will increase monotonically in time according to the power law

$$\tilde{x}_N(\tilde{t}) = (9\tilde{t})^{1/3}. \quad (6)$$

The solution to equation (4) will converge to equation (5) as the current becomes thin for any initial shape with compact support. The convergence time is small for $M \ll 1$ and increases strongly with $M$ [Hesse et al., 2007; MacMinn and Juanes, 2009].

We compare the predictions of equation (4) with benchmark experiments in which buoyant water spreads over water with experimental measurements (black circles) and the predictions of the model (solid black curves and gray shaded regions) for three different bead sizes. (b–e) Snapshots of the experiment in 2 mm beads, along with the predictions of the model (blue curves). Nose positions and times in Figure 5a are dimensionless, scaled as discussed in section 4.1. We discuss the model in section 4.2.

Figure 5. (a) The scaled position of the nose of a buoyant current of 59.1 wt % MEG spreading over water with experimental measurements (black circles) and the predictions of the model (solid black curves and gray shaded regions) for three different bead sizes. (b–e) Snapshots of the experiment in 2 mm beads, along with the predictions of the model (blue curves). Nose positions and times in Figure 5a are dimensionless, scaled as discussed in section 4.1. We discuss the model in section 4.2.

Figure 6. (a) The scaled position of the nose of a buoyant current of 65.4 wt % MEG spreading over water with experimental measurements (black circles) and the predictions of the model (solid black curves and gray shaded regions) for two different bead sizes. (b–e) Snapshots of the experiment in 2 mm beads, along with the predictions of the model (blue and cyan curves). Nose position and time are scaled as discussed in section 4.1. We discuss the model in section 4.2.
over ambient saltwater (Figure 4). The ambient saltwater contains 1.075% NaCl by weight, so the driving density difference is $\Delta \rho = 7.52 \text{ kg m}^{-3}$ and the mobility ratio is $M = 1$ (Table 1). As discussed in section 2.2, we use these experiments to infer the effective permeability of the flow cell.

### 4.2. Buoyant Spreading With Convective Dissolution

Previous studies of convective dissolution have shown that a stationary layer of CO$_2$ will dissolve into a semi-infinite layer of water at a rate that is roughly constant in time [Hidalgo and Carrera, 2009; Kneafsey and Prew, 2010; Pau et al., 2010]. When the water layer has a finite thickness, recent results suggest that the dissolution rate is small relative to the thickness of the CO$_2$ layer that dissolves per unit bulk fluid-fluid interfacial area per unit time. Rewriting equation (7) in dimensionless form using (3), we obtain

\[
\frac{\partial h}{\partial \tau} - U \frac{\partial}{\partial x} \left[ (1-f)h \frac{\partial h}{\partial \xi} \right] = -\frac{q_d}{\phi},
\]  

(7)

where the dissolution rate $q_d$ is the volume of buoyant fluid that dissolves per unit bulk fluid-fluid interfacial area per unit time. Rewriting equation (7) in dimensionless form using (3), we obtain

\[
\frac{\partial \tilde{h}}{\partial \tilde{\tau}} - \frac{\partial}{\partial \tilde{x}} \left[ (1-f)^{\tilde{h}} \frac{\partial \tilde{h}}{\partial \tilde{\xi}} \right] = -\epsilon,
\]  

(8)

where

\[
\epsilon = \left( \frac{L}{H} \right)^2 \frac{q_d}{N \phi},
\]  

(9)

is the dimensionless dissolution rate. Pritchard et al. [2001] studied equation (8) in the unconfined limit ($f = 0$) in a different context, developing the explicit solution

\[
\tilde{h}(\tilde{x}, \tilde{\tau}) = \frac{1}{6} \tilde{\tau}^{-1/3} \left( g^{2/3} - \frac{\tilde{x}^2}{\tilde{\tau}^{2/3}} \right)^{-3/4} \tilde{\tau},
\]  

(10)

valid for $|\tilde{x}| \leq \tilde{x}_N(\tilde{\tau})$ and $\tilde{\tau} > 0$. As discussed by Pritchard et al. [2001], equation (10) implies that the position of the nose of the current evolves according to

\[
\tilde{x}_N(\tilde{\tau}) = (9 \tilde{\tau})^{1/3} \sqrt{1 - \frac{1}{18} \epsilon (9 \tilde{\tau})^{1/3}}.
\]  

(11)

Equations (10) and (11) reduce to equations (5) and (6), respectively, when $\epsilon = 0$.

Equation (11) predicts that convective dissolution will have a strong impact on the spreading of the current in the unconfined limit. Without convective dissolution ($\epsilon = 0$), the nose of the current advances for all time following the power law $x_N \propto t^{1/3}$ (equation 6). With convective dissolution, the nose reaches a maximum position $\tilde{x}_N^{\text{max}} = (8/3)^{1/4} \epsilon^{-1/4}$ and then retreats to the origin as the volume of the current decreases to zero at time $\tilde{t}^{\text{end}} = (8/9)^{1/4} \epsilon^{-3/4}$ (Figure 7a).

We refer to this time $\tilde{t}^{\text{end}}$ as the ‘lifetime’ of the current.

[23] The spreading behavior for nonnegligible $M$ is qualitatively similar to the unconfined behavior predicted by equations (6) and (11), but the current spreads more slowly as $M$ increases (Figure 7b). Accordingly, the maximum extent of the current decreases with $M$ while the lifetime of the current increases with $M$. However, the scalings of these quantities with $\epsilon$ show only minor deviations from the unconfined predictions of $\tilde{x}_N^{\text{max}} \sim \epsilon^{-1/4}$ and $\tilde{t}^{\text{end}} \sim \epsilon^{-3/4}$ (Figure 8).

[27] Equation (10) is not strictly an asymptotic solution of equation (8) because convective dissolution causes some memory of the initial shape to be retained throughout the evolution, as with residual trapping [Kochina et al., 1983; Barenblatt, 1996]. In addition, the concept of asymptotics has limited relevance here because the current dissolves completely in finite time.

[28] The predictions of equation (8) are in qualitative agreement with our experimental observations for currents of the faster-spooling, slower-dissolving MEG (59.1 wt %). Quantitative comparison requires an estimate of the dissolution rate $q_d$. Expressions for $q_d$ for a stationary layer have been presented by Pau et al. [2010] and Neufeld et al. [2010]. The latter, in particular, performed experiments with the same pair of analog fluids used here. Based on those experiments, we determine the dissolution rate for our system to be

\[
q_d = 10 \text{ mmol m}^{-2} \text{ s}^{-1}.
\]
results in conjunction with high-resolution numerical simulations, Neufeld et al. [2010] suggested that

\[ q_d = b \left( \frac{\phi \chi^* D}{H} \right) \left( \frac{\Delta \rho \phi D}{\Delta \rho \mu D} \right), \]

where \( \chi^* = \rho_d \chi / \rho_b \) measures the volume of buoyant fluid dissolved in one unit volume of ambient fluid containing the maximum (saturated) mass fraction \( \chi_m^* \) of dissolved buoyant fluid, \( D \approx 1 \times 10^{-5} \text{ cm}^2 \) is the molecular diffusivity of aqueous buoyant fluid in a porous medium, and \( b \approx 0.12 \) and \( n \approx 0.84 \) are dimensionless constants. Although the characteristic vertical scale here should be the depth of the layer of ambient fluid below the buoyant current, we use the total depth of the fluid layer \( H \approx 14 \text{ cm} \) for simplicity since the buoyant current is thin for most of its evolution (\( h \ll H \)). Dissolution due to diffusion and dispersion are not included in this estimate of \( q_d \) since these are negligible compared to convective dissolution [Ennis-King et al., 2005].

[28] We begin by estimating the dissolution rate from this expression for 59.1 wt % MEG dissolving into water. We then treat the dissolution rate as a fitting parameter, calibrating its value around this estimate by comparing the predictions of the model with experimental measurements. We present the estimated and calibrated dissolution rates.

### Table 3. Parameters for the 59.1 wt % MEG

<table>
<thead>
<tr>
<th>Bead Diameter (cm)</th>
<th>( U (\text{cm s}^{-1}) )</th>
<th>( q_d^* (\text{cm s}^{-1}) )</th>
<th>( q_d (\text{cm s}^{-1}) )</th>
<th>( \epsilon )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1</td>
<td>0.017</td>
<td>6.61 \times 10^{-4}</td>
<td>4.55 \times 10^{-4}</td>
<td>4.6 \times 10^{-5}</td>
</tr>
<tr>
<td>0.2</td>
<td>0.071</td>
<td>2.07 \times 10^{-4}</td>
<td>1.21 \times 10^{-4}</td>
<td>2.0 \times 10^{-5}</td>
</tr>
<tr>
<td>0.3</td>
<td>0.120</td>
<td>3.29 \times 10^{-4}</td>
<td>1.45 \times 10^{-4}</td>
<td>1.2 \times 10^{-5}</td>
</tr>
</tbody>
</table>

\( q_d^* \) and \( q_d \) respectively, in Table 3. The calibrated values are within about a factor of two of the estimated values. That they do not agree exactly is not surprising, given that the correlation of Neufeld et al. [2010] was developed in the context of a stationary layer of MEG dissolving into water. Diffusion and flow-induced dispersion in the present context, where the interface has both advancing and receding portions, may enhance or inhibit convective dissolution relative to the case of a stationary layer.

[30] We compare these experiments with the predictions of equation (8) in Figure 5. We compare the evolution of the nose position for all three bead sizes, as well as the evolution of the shape of the current for the 2 mm beads. We include an envelope around the nose position corresponding to \( \pm 10 \% \) around the calibrated dissolution rate \( q_d \) to illustrate the sensitivity of the model to this parameter.

[31] These results suggest that the assumption of a constant rate of convective dissolution can capture the qualitative and quantitative features of the impact of convective dissolution on a buoyant current in this system, provided that dissolved buoyant fluid accumulates slowly beneath the buoyant current.

### 5. Two-Current Model for Spreading With Convective Dissolution

[32] Experimental and numerical studies of convective dissolution have thus far focused on dissolution from a stationary layer of CO2 overlying a deep or semi-infinite layer of water. In a confined aquifer, we expect that the accumulation of dissolved CO2 in the water beneath the buoyant current will limit the rate at which the CO2 can dissolve. Here, we extend the model discussed above (equation (8)) to include this accumulation effect in a simple way.

[33] In our experiments with buoyant currents of slower-spreading, faster-dissolving MEG (65.4 wt %), we observed that the accumulation of dissolved MEG in the water played a strong role in the dynamics of the buoyant current. In particular, water with dissolved MEG accumulated on the bottom of the aquifer and slapped downward relative to the ambient fluid because of its larger density. Although the details of convective dissolution and subsequent mass transport are complex, we develop a simple model for this process by assuming that it primarily transports dissolved buoyant fluid vertically from the buoyant current down to a dense current of ambient fluid with dissolved buoyant fluid (Figure 9). We assume that this dense current consists of ambient fluid with a constant and uniform mass fraction \( \chi_m \) of dissolved buoyant fluid, with corresponding density \( \rho_d \) and viscosity \( \mu_d \), and we denote its unknown local thickness by \( h_d(x,t) \). Note that \( \chi_m \) may be less than the maximum (saturated) mass fraction \( \chi_m^* \). We...
Finally, we redefine the dimensionless convective dissolution rate \( \epsilon \) to be conditional,

\[
\epsilon = \begin{cases} 
(L/H)^2 (q_d/\phi U) & \text{if } h + h_d < 1, \\
0 & \text{if } h + h_d = 1,
\end{cases}
\]
...when the two currents touch, the fact that although we have assumed that convective dissolution study the accumulation of dissolved buoyant fluid. Also, experiments and high-resolution numerical simulations to this model is that we do not have an a priori estimate of the 

\[ \text{[42]} \] These results suggest that this model captures the and also the properties of the dense current via the parameters \( \delta \) and \( M_d \).

\[ \text{[43]} \] Motivated by the Sleipner site in the North Sea, we consider an aquifer for which \( \mu_b \approx 500 \text{ kg m}^{-1} \text{s}^{-1} \), \( \mu_w \approx 4 \times 10^{-2} \text{ Pa s} \), \( \rho_b \approx 1000 \text{ kg m}^{-3} \), and \( \rho_w \approx 6 \times 10^{-4} \text{ Pa s} \) [Bickle et al., 2007], giving a mobility ratio of \( \mathcal{M} \approx 15 \).

This value of \( \mathcal{M} \) is much larger than in the MEG-water system (\( \mathcal{M} \approx 0.31 \) for the 59.11 wt % MEG and \( \approx 0.24 \) for the 65.4 wt % MEG). As a result, the CO2 plume will be much more strongly tongued than the MEG plume, presenting much more interfacial area for convective dissolution and increasing its effectiveness [MacMinn et al., 2011].

\[ \text{[44]} \] For an aquifer of thickness \( H \approx 20 \text{ m} \), porosity \( \phi \approx 0.375 \), and permeability \( k \approx 2.5 \times 10^{-12} \text{ m}^2 \), the characteristic spreading rate of the CO2 plume is \( U \approx 5.8 \times 10^{-5} \text{ m s}^{-1} \) and Neufeld et al. [2010] estimated the dissolution rate to be \( q_d \approx 1 \times 10^{-9} \text{ m}^3 \text{s}^{-1} \). The estimate of Pau et al. [2010] for the dissolution rate in the Carrizo-Wilcox aquifer in Texas is of the same order. For a relatively large sequestration project in such an aquifer, where \( M \approx 10 \text{ Mt of CO2} \) is injected along a linear array of wells [Nicot, 2008] of length \( W \approx 10 \text{ km} \), the characteristic length is \( L = M/2 \rho_b \phi H W \approx 130 \text{ m} \) and the corresponding dimensionless dissolution rate is \( \epsilon = q_d L^2/\phi U H L^2 \approx 0.002 \).

Although quite sensitive to the specific injection scenario, this value is comparable to the values from the experiments.

### 6. Application to Carbon Sequestration

...when the two currents touch since it remains possible to generate mixtures at the interface that are denser than the surrounding fluid. The comparison between this model and our experimental results implies that this model captures the fundamental behavior of the MEG-water system, but further study will be necessary to assess the limits of this model in practice.

![Figure 10](image.png)

Figure 10. (a) The evolution of the nose of the buoyant current from numerical solutions to the two-current model for several values of \( \varepsilon \) without (dashed black lines) and with (solid gray lines) accumulation. The presence of the dense current retards the nose of the buoyant current weakly due to hydrodynamic interactions before the two currents touch and then strongly by inhibiting convective dissolution after the two currents touch (\( M = 0.25, M_d = 1, \delta = 1.1, x_r = 0.2 \)). (b) Solutions for \( \varepsilon = 0.01 \) and several values of \( x_r \) (other parameters unchanged). The two currents touch earlier as \( x_r \) decreases, and the nose retreats more slowly thereafter.

Table 4. Parameters for the 65.4 wt % MEG

<table>
<thead>
<tr>
<th>Bead Diameter (cm)</th>
<th>( U (\text{cm s}^{-1}) )</th>
<th>( u_d^g (\text{cm s}^{-1}) )</th>
<th>( q_d (\text{cm}^3 \text{s}^{-1}) )</th>
<th>( \epsilon )</th>
<th>( x_r )</th>
<th>( M_d )</th>
<th>( \delta )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1</td>
<td>3.6 x 10^{-3}</td>
<td>2.0 x 10^{-4}</td>
<td>1.8 x 10^{-4}</td>
<td>0.055</td>
<td>0.30</td>
<td>0.51</td>
<td>2.14</td>
</tr>
<tr>
<td>0.2</td>
<td>1.5 x 10^{-2}</td>
<td>6.8 x 10^{-4}</td>
<td>6.8 x 10^{-4}</td>
<td>0.040</td>
<td>0.30</td>
<td>0.51</td>
<td>2.14</td>
</tr>
<tr>
<td>0.3</td>
<td>2.5 x 10^{-2}</td>
<td>1.1 x 10^{-3}</td>
<td>8.2 x 10^{-4}</td>
<td>0.028</td>
<td>0.28</td>
<td>0.53</td>
<td>2.06</td>
</tr>
</tbody>
</table>

*We report the properties of the pure MEG in Table 1. We use \( L = 9 \text{ cm} \) and \( H \approx 14 \text{ cm} \) in these and all other experiments. Here \( x_m \) and \( x_r \) are approximately equal.*
XXX

(e \approx 0.001-0.005 for the 59.11 wt % MEG and \approx 0.03-0.06 for the 65.4 wt % MEG).

[46] Unlike in our experiments, the solubility of CO₂ in groundwater is only a few percent by weight at typical aquifer conditions [Duan and Sun, 2003]. For χ_{aq} \approx 0.01, the corresponding increase in the density of the water is \Delta \rho_{w} \approx 10 \text{ kg m}^{-3} \ [García, 2001] and the change in its viscosity is negligible, so we expect \chi_{w} \approx 0.02, \delta \approx 0.02, and \mathcal{M}_0 \approx 1. These values of \chi_{w} and \delta are about an order of magnitude smaller than those in our experiments.

[47] Based on these values of \epsilon, \delta, and \chi_{aq}, we expect dissolved CO₂ to accumulate very quickly and slump downward very slowly relative to the rate at which the buoyant current spreads. As a result, we expect the rate at which CO₂ is trapped to be controlled not by the rate of convective dissolution, but by the amount of dissolved CO₂ the water can hold (i.e., \chi_{aq}) and by the rate at which this water slumps downward. In Figure 11, we show that this is indeed the case: both the maximum extent and the lifetime of a plume of CO₂ decrease as the dissolution rate increases, but both quantities approach limiting values that are independent of the dissolution rate if this rate is sufficiently large. The dissolution rate of \epsilon \approx 0.002 estimated above is about 2 orders of magnitude above this threshold value. As a result, the spreading and convective dissolution of the plume is completely controlled by the accumulation of dissolved CO₂ in this setting, and the plume spreads several times further and persists for several orders of magnitude longer than it would without this accumulation.

7. Discussion and Conclusions

[48] We have shown via experiments with analog fluids that simple models are able to capture the impact of convective dissolution on the spreading of a buoyant gravity current in a vertically confined, horizontal layer.

[49] When dissolved buoyant fluid accumulates slowly beneath the buoyant current, our experiments have confirmed that the complex dynamics of convective dissolution can be upscaled to a constant mass flux [Pau et al., 2010; Kneafsey and Pruess, 2010; Neufeld et al., 2010] and incorporated into a simple model [Gasda et al., 2011; MacMinn et al., 2011] (Figure 5).

[50] When dissolved buoyant fluid accumulates quickly beneath the buoyant current, our experiments have shown that this accumulation can have an important limiting effect on the dissolution process. To capture the accumulation of dissolved buoyant fluid, we have developed a two-current model where a dense gravity current of ambient fluid with dissolved buoyant fluid grows and spreads along the bottom of the aquifer. We have used this model to show that the accumulation of dissolved buoyant fluid beneath the buoyant current can slow convective dissolution, and we have confirmed this prediction experimentally (Figure 6).

[51] Using this two-current model, we have shown that we expect CO₂ spreading and dissolution in a horizontal aquifer to be controlled primarily by the mass fraction at which CO₂ accumulates in the water, and to be nearly independent of the dissolution rate (Figure 11). This can be the case even in the presence of aquifer slope or background groundwater flow, both of which drive net CO₂ migration and expose the plume to fresh water, when slope- or flow-driven migration is sufficiently slow [MacMinn et al., 2011].

[52] The planar models used here rely on the fact that the transverse width of the buoyant current is much larger than its length in the x direction, \( W \gg L \), which is typically the case when large amounts of CO₂ are injected via a line drive configuration [Nicot, 2008; Szułczewski et al., 2012]. The models presented here can be readily adapted to a radial geometry for injection from a single well where appropriate. Where neither geometric approximation is appropriate, use of a more complicated, two-dimensional model will be necessary.

[53] We have also assumed here an idealized rectangular initial shape for the plume of CO₂. In practice, the specific details of the injection scenario will have some quantitative impact on the maximum extent and lifetime of the CO₂, but should have little qualitative impact on the interaction between plume spreading, convective dissolution, and the accumulation of dissolved CO₂.

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References


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