COUPLED SUBSURFACE–SURFACE LAYER GAS TRANSPORT AND DISPERSION FOR GEOLOGIC CARBON SEQUESTRATION SEEPAGE SIMULATION

Curtis M. Oldenburg and André A. J. Unger

Earth Sciences Division
Lawrence Berkeley National Laboratory
Berkeley, CA, 94720, USA

e-mail: cmoldenburg@lbl.gov, ajunger@lbl.gov

ABSTRACT

The accumulation of vast quantities of injected carbon dioxide (CO₂) in geologic sequestration sites may entail health and environmental risks from potential seepage of CO₂ into the near-surface environment. We are developing and applying a coupled subsurface and atmospheric surface layer modeling capability based on the TOUGH2 framework to predict CO₂ concentration distributions under a variety of seepage scenarios and geologic, topographic, and atmospheric conditions. These concentration distributions will provide the basis for determining above-ground and near-surface instrumentation needs for carbon sequestration monitoring and verification, as well and for assessing health, safety, and environmental risks. A key feature of CO₂ is its large density (ρ = 1.8 kg m⁻³) relative to air (ρ = 1.2 kg m⁻³), a property that may allow small leaks to cause concentrations above the occupational exposure limit of 4% in low-lying and enclosed areas such as valleys and basements where dilution rates are low. The approach we take to coupled modeling involves development of T2CA, a TOUGH2 module for modeling transport of water, brine, CO₂, gas tracer, and air in the subsurface. For the atmospheric surface layer advection and dispersion, we use a logarithmic vertical velocity profile to specify time-averaged ambient winds, and well established atmospheric dispersion approaches to model mixing due to eddies and turbulence. Initial simulations with the coupled model show that atmospheric dispersion quickly dilutes diffuse CO₂ seepage fluxes to negligible concentrations, and that rainfall infiltration causes CO₂ to return to the subsurface.

INTRODUCTION

From the point of view of human and environmental risk associated with exposure to carbon dioxide (CO₂) from leaking geologic carbon sequestration sites, it is advection and dispersion above the ground surface in the biosphere that is most significant since this is where the key receptors are located. Yet the advection and dispersion processes occurring in the atmospheric surface layer will be coupled to subsurface processes since (1) the subsurface is the source of the seeping CO₂, (2) ambient air can flow into and out of the subsurface in response to atmospheric pressure changes, and (3) CO₂ is a dense gas that will tend to migrate downwards and hug the ground relative to ambient air. Therefore simulation models for atmospheric dispersion of CO₂ that neglect processes involving the subsurface may not be appropriate except in certain limited situations. A schematic of potential CO₂ leakage and seepage from a geologic sequestration site is shown in Figure 1 along with associated processes and features.

**Figure 1. Schematic showing unexpected leakage and seepage of CO₂ from a geologic carbon sequestration site.**

Motivated by the need to predict CO₂ concentrations in the unlikely event that a geologic sequestration site would leak leading to significant upward migration and eventual CO₂ seepage at the ground surface, we have developed a fully coupled subsurface–surface layer simulation capability called T2CA, for TOUGH2 CO₂ and Air. T2CA can be used for risk assessment and for designing monitoring and verification instrumentation and strategies for geologic carbon sequestration. This new simulation capability can be used to answer questions about what the expected concentrations will be in the surface layer and shallow subsurface resulting from assumed leakage fluxes. This information can then be used to (1) assess the potential exposure to CO₂ for humans and other environmental receptors, and (2) develop specifications and designs of monitoring...
equipment and strategies for sequestration verification.

In the case of catastrophic failures involving large seepage fluxes, for example due to a well blowout, the health risks are obvious and could have potentially lethal effects, thus subordinating the verification issue in favor of safety assurance. We expect the challenging issue to be health, safety, and environmental risk assessment, as well as monitoring and verification, associated with diffuse or very slow seepage phenomena that are hard to detect. For this reason, our simulation capability is designed for cases of diffuse seepage as opposed to catastrophic failures. The purpose of this paper is to present our approach to modeling subsurface and surface-layer CO₂ migration and dispersion of leakage and seepage from geological carbon sequestration sites, and to show some initial results.

**BACKGROUND**

Carbon dioxide is a dense gas (ρ = 1.8 kg m⁻³) relative to air (ρ = 1.2 kg m⁻³) as shown in Figure 2, where we have plotted gas density and viscosity for mixtures of CO₂ and air calculated from the NIST14 Database (NIST, 1992; Magee et al., 1994). Although CO₂ is ubiquitous and essential to life as part of the natural carbon cycle, it is dangerous at high concentrations. The current ambient CO₂ concentration in the atmosphere is approximately 375 ppmv (0.0375 %); concentrations of 4% can cause immediate danger to humans (NIOSH, 1981). As such, CO₂ can be considered a dense hazardous gas, a class of substances that has received considerable attention over the years for leak and spill risk assessment of industrial gases (e.g., Britter and Griffiths, 1982; Hanna and Steinberg, 2001). For example, liquefied propane gas (LPG), liquefied natural gas (LNG), and many others are dense hazardous gases upon release to the atmosphere. Motivated by the need to assess risks associated with the mass production and transport of dense gases, a great deal of experimental, analytical, and modeling work has been focused on the problem of dense gas dispersion in the surface layer. This work is summarized in the review article by Britter (1989).

The result of many field experiments of dense gas dispersion processes has been the development of correlations involving the most important parameters controlling atmospheric dispersion such as wind speed, density of released gas, and release flux (Britter and McQuaid, 1988).

These correlations were developed based on simple scale and dimensional analysis. One of these correlations relates the seepage flux and average wind speed at an elevation of 10 m to the form of the dispersion process, i.e., whether it is active (density-dependent) or passive like a gas tracer. In Figure 3, we have plotted this correlation with values appropriate for CO₂-air mixtures for various source area length scales along with the typical amount of CO₂ emitted and taken up by plants, soil, and roots known as the net ecosystem exchange (NEE) (e.g., Baldocchi and Wilson, 2001). As shown in Figure 3, seepage fluxes have to be quite high (note logarithmic scale) for windy situations for the resulting dispersive mixing process to be active. Note that wind conditions are averages over a period of 10 minutes.

In prior work (Oldenburg and Unger, 2003), we have simulated subsurface migration of leaking CO₂ through the unsaturated zone with rainwater infiltration for various leakage rates specified at the water table. These leakage rates were given as annual mass leakage percentages of the total stored CO₂. Typical seepage fluxes for the 0.1% yr⁻¹ leakage rate were on the order of 10⁻⁵–10⁻⁶ kg m⁻² s⁻¹. As shown in Figure 3, seepage fluxes of this magnitude lead to passive dispersion for all but the calmest wind conditions. Therefore, our approach is for the case of passive mixing associated with diffuse CO₂ emissions rather than catastrophic failures.
Figure 3. Correlation for active (i.e., density-dependent) and passive dispersion in the surface layer as a function of seepage flux and wind velocity.

**COUPLED MODELING APPROACH**

**Introduction**

In order to simulate the coupled subsurface–surface layer advection and dispersion of CO₂, we have developed T2CA, an extension of the EOS7R module (Oldenburg and Pruess, 1995; Pruess et al., 1999). T2CA handles five components (H₂O, brine, CO₂, a gas tracer, air) and heat. Real gas mixture properties are calculated so the full range of high-pressure sequestration-site conditions to low-pressure ambient surface layer conditions can be modeled. T2CA adds atmospheric surface layer advection and dispersion capabilities to create a fully coupled subsurface–surface layer simulator.

The purpose of this section is to present the methods implemented in T2CA. These methods are derived from the current practice in atmospheric dispersion modeling. Because subsurface transport in T2CA is unchanged from the standard approach used in TOUGH2, we focus our discussion on the methods we apply in the surface layer to model atmospheric dispersion. While the discussion below focuses on CO₂ transport, all of the gas-phase components are treated identically, and an analogous treatment can be developed for heat.

**Transport of Dilute CO₂ as a Passive Gas**

Transport of CO₂ as a passive gas implies that it advects and disperses in the atmosphere without influencing the flow field. In order for this assumption to hold, CO₂ must be at sufficiently low concentrations that it does not affect the density or viscosity of the ambient atmosphere. Under this assumption, we discuss below the underpinnings of the use of an ambient wind profile as well as advection and dispersion in the lower layers of the atmosphere as developed in the atmospheric transport literature (e.g., Slade, 1968; Pasquill, 1974; Stull, 1988; Arya, 1999).

**Logarithmic Velocity Profile**

The ambient time-averaged wind profile has been shown theoretically to follow a logarithmic profile. An excellent review of the assumptions and calculations involved in the logarithmic profile, as well as experimentally derived parameters obtained from calibration to field data is provided in Slade (1968). The logarithmic wind profile as shown on Figure 4 is given as:

\[ u_x(z) = \frac{u_z}{k^*} \ln \left( \frac{z}{z_0} \right) \]  

where \( u(z) \) is the ambient wind velocity as a function of height, \( u_* \) is the friction velocity (a parameter that governs the shape of the wind profile near the ground surface for various surface types (Slade, 1968)), \( k^* \) is von Karman’s constant \((k^* = 0.4)\), \( z \) is the elevation, and \( z_0 \) is a roughness height such that \( u_x(z) = 0 \) at \( z = z_0 \) and is also a function of various surface types.

**Advevtive-Dispersive Transport**

Transport of CO₂ as a passive gas in the atmospheric surface layer follows the linear advection-dispersion equation. A general background including all assumptions in the formulation of this equation can be found in Slade (1968) and Arya (1999). In general, this process can be simplified to the following partial differential equation for the three-dimensional \((x, y, z)\) transport of a component (such as CO₂) at concentration \( c \):

\[
\frac{\partial c}{\partial t} + u_x \frac{\partial c}{\partial x} + u_y \frac{\partial c}{\partial y} + u_z \frac{\partial c}{\partial z} - D_{xx} \frac{\partial^2 c}{\partial x^2} - D_{yy} \frac{\partial^2 c}{\partial y^2} - D_{zz} \frac{\partial^2 c}{\partial z^2} = 0
\]  

(2)
For atmospheric transport modeling, the coordinate system is arranged so that $x$ is aligned in the downwind direction. Advection is generated by the velocity terms so that with the coordinate system transformation, $u_y = u_z = 0$, and $u_x$ is the ambient wind. Dispersion is governed by $D_{xx}$, $D_{yy}$, and $D_{zz}$ which are the diagonal elements of a dispersion tensor and are related to the standard deviation ($\sigma$) in the concentration distribution by

$$\begin{align*}
\sigma_x^2 &= 2D_{xx}t \\
\sigma_y^2 &= 2D_{yy}t \\
\sigma_z^2 &= 2D_{zz}t
\end{align*}$$

(3)

where $t$ is time (e.g., Arya, 1999).

**Pasquill-Gifford Dispersion**

The empirically derived Pasquill-Gifford (P-G) dispersion curves provide a commonly accepted and practical means of determining atmospheric dispersion, and are discussed in detail in Slade (1968) and Arya (1999). Essentially, large-scale eddies in the convective motion of the lower atmospheric layers are assumed to result in dispersion of passive constituents that can be mathematically represented as a Fickian diffusion process. The P-G scheme was developed from experiments conducted over a wide variety of terrain (e.g., project Prairie Grass and British diffusion experiments (Pasquill, 1961; Gifford, 1961)) and atmospheric conditions (ranging from class A-extremely unstable, class B-moderately unstable, class C-slightly unstable, class D-neutral, class E-slightly stable, to class F-moderately stable). The P-G curves are shown on Figure 5 and provide a value of $\sigma_y$ and $\sigma_z$ as a function of downwind distance under a specific atmospheric condition (classes A–F), with $\sigma_x$ set equal to zero. The empirically derived P-G dispersion scheme is based on large-scale experiments and is valid for large-scale eddies in the lowest 100 m of the atmosphere evolving over length scales ranging from 100 m to 10,000 m downwind from the source. Figure 5 includes values of $\sigma_y$ and $\sigma_z$ extrapolated from 100 m down to 1 m, the purpose of which will be discussed below.

**Smagorinski Model Dispersion**

In the context of CO$_2$ transport, the source zone typically represents a surface seep. Health and environmental risks resulting from the transport of CO$_2$ need to be resolved at distances from the source that may be smaller than strictly applicable for the P-G dispersion curves discussed above. On the scale of 100 m or less, small-scale eddies in the convective motion of the near surface atmospheric layers are assumed to result in dispersion of passive constituents within these layers (Arya, 1999). These eddies are assumed to arise from the shear stress resulting from the viscosity of the air in contact with the ground surface. Note that this identical mechanism is responsible for the logarithmic wind profile discussed earlier, in which wind velocities approach zero near the ground surface.

The Smagorinski Model is the simplest and most widely used small-scale eddy dispersion model and is described in detail by Arya (1999). In general, the Smagorinski Model provides a methodology to develop a dispersion tensor for use in the conservation of momentum equation when using the Navier-Stokes equations to model air flow near the ground surface. Briefly, the dispersion of momentum ($D$) is assumed to be proportional to the vertical, $z$, and horizontal, $x$, gradient in air velocity as given by:

$$D = \frac{l^2}{\sqrt{2}} \left( \frac{\partial u_x}{\partial z} + \frac{\partial u_z}{\partial x} \right) = \frac{l^2}{\sqrt{2}} \frac{u_x}{k^*} \frac{1}{z}$$

(4)

where $l$ is a grid-related length scale, and $u_x$ is obtained from the logarithmic wind profile given by Eq. 1, while $u_z$ is zero in the logarithmic wind profile. Scalars such as a passive tracer or heat (energy) are assumed to undergo Fickian diffusion where the diagonal elements of the dispersion tensor are given by the momentum diffusivity in Eq. 4 multiplied by a constant ranging from 2.0 to 3.0 (Arya, 1999).
Summary
The most commonly accepted and practical means of modeling large-scale atmospheric transport of CO₂ as a passive gas involves solving the 3-D advection-dispersion equation for various flux or concentration source conditions with advection obtained from the logarithmic wind profile and dispersion obtained using the P-G dispersion curves. Within the immediate vicinity of the source zone, the same approach can be used with the dispersion coefficient estimated by the Smagorinski Model. The extrapolation of the P-G curves to smaller length scales is done so the transition from small-scale (i.e., Smagorinski Model) to large-scale (i.e., P-G) dispersion will be smooth. Because Fickian diffusion is a linear flux operator in the context of the advection-dispersion equation, the P-G curves and the Smagorinski Model can be applied simultaneously along with pure molecular diffusion with the largest term controlling dispersion.

IMPLEMENTATION IN TOUGH2
Specification of the Logarithmic Wind Profile
The simulation of atmospheric advection and dispersion by the above methods begins by creating a logarithmic wind profile within the TOUGH2 framework. This step involves generating a grid with sufficient layers (i.e., parallel to the ground surface) to discretize the wind profile to the desired accuracy. Next, a static gas-phase pressure profile in the $z$-direction is used along with a constant pressure difference between the upstream and downstream boundaries of the surface layer

$$\Delta P = P_2 - P_1, \quad R_1 > R_2$$  \hspace{1cm} (5)

where $P_1$ and $P_2$ are the upstream and downstream pressures, respectively, within a layer. TOUGH2 computes the phase velocity using Darcy’s equation

$$u = \frac{k_D}{\phi \mu} \nabla (P - \rho g z)$$  \hspace{1cm} (6)

where $k_\alpha$ is the intrinsic (Darcy) permeability, $\phi$ is the porosity, $\mu$ is the gas viscosity, $\rho$ is the mass density of the gas phase, $g$ is the gravitational acceleration and $z$ is height. Setting the porosity of the surface layer materials to unity, the velocity of the atmospheric air will be proportional to the permeability of the layer and pressure difference, $\Delta P$, for horizontal layers. Given that $\Delta P$ is a constant for all layers, the individual permeability variations of the layers will combine to produce the logarithmic wind profile. Note that the thickness of each layer must be constant to ensure a constant air velocity within the layer across the length of the domain. Note further that the permeability is a pseudo-permeability with no physical significance; its purpose is simply to create the desired velocity profile.

Calculating Atmospheric Dispersion
Within the TOUGH2 framework, transport of CO₂ as a passive gas will follow the linear advective-dispersive transport equations already used to calculate the transport of species within the gas phase. Ambient atmospheric dispersion of CO₂ using either P-G dispersion curves or the Smagorinski Model modified for the dispersion of a scalar is implemented by using a spatially dependent effective molecular diffusivity in the surface layer region.
With this approach, the diagonal of the tensor representing Fickian diffusion of CO₂ is modified to be the sum of molecular diffusion, as well as \( D_{xx} \) and \( D_{zz} \) from the P-G curves, as well as a contribution from the Smagorinsky Model. This enables T2CA to include the dispersion of the CO₂ plume due to molecular diffusion and both small- and large-scale eddies in the convective motion of the atmosphere.

**Computing Dispersion Coefficients**

In order to compute values of \( D_{xx} \) and \( D_{zz} \) from the P-G curves at the interface between two nodes within the mesh, the distance from the source must be computed internally within TOUGH2. This distance is then used to interpolate values \( \sigma_x \) and \( \sigma_z \) from the curves given in Figure 5. Note that values of \( \sigma_x \) and \( \sigma_z \) are extrapolated from a downwind distance of 100 m to 1 m in order to prevent a jump in the value of the dispersion tensor as the plume moves a downwind distance of 100 m. Next, the time \( t \) required for the CO₂ plume to travel to the interface is estimated as the travel distance divided by the wind velocity at a specified height. This assumes that the centroid of the plume advects at a constant elevation above the ground surface, i.e., under neutral stability conditions. Finally, values of \( \sigma_x \) and \( \sigma_z \) and \( t \) in Eq. 3 to compute values of \( D_{xx} \) and \( D_{zz} \). These parameters are specified in the TOUGH2 input file by means of the SELEC data block.

**Active vs. Passive Dispersive Transport**

Transport of CO₂ can occur both as a dense or as a passive gas, depending on the local CO₂ concentration. Although we have restricted the treatment to passive gas transport, note in Eq. 6 that the body force term remains. Therefore, if significant density effects ever arise, the surface layer velocity will be affected and will deviate from the logarithmic velocity profile. If this occurs, it is an indication that the atmospheric dispersion process is active as opposed to passive, and the user should proceed carefully to assess whether other methods should be applied to model active dense gas dispersion.

**Summary**

Implementing the coupled subsurface–surface layer CO₂ flow and transport model in TOUGH2 involves the assumption of an average logarithmic wind velocity profile and the use of dispersion coefficients to model mixing due to small- and large-scale eddies. This approach is standard in atmospheric dispersion modeling. What is new in our approach is the coupling of the atmospheric surface layer to the subsurface region. This coupling is important because CO₂ is a dense gas that may seep out of—but also possibly back into—the subsurface. While our approach in the surface layer is strictly correct only for passive mixing, it may also prove to be acceptable in calm conditions for active (density-dependent) gas dispersion, pending additional investigation.

**PRELIMINARY RESULTS**

We present in this section preliminary results to demonstrate the capabilities of T2CA. The properties of an idealized two-dimensional unsaturated zone and atmospheric surface layer are shown in Table 1. In the model system, CO₂ is being injected at the water table to model the arrival of leaking CO₂ from a deep geologic sequestration site. The CO₂ migrates upwards through the unsaturated zone and seeps out of the subsurface into the surface layer. We inject pure water at a constant rate of 10 cm yr⁻¹ at the ground surface to model rainfall infiltration. The subsurface part of this system is a cartesian version of the radial system we have studied earlier (Oldenburg and Unger, 2003). We use the same leakage rate of 0.1% yr⁻¹ of an assumed 4 x 10⁷ kg CO₂ sequestration site giving rise to a leakage rate of 4 x 10⁷ kg yr⁻¹. If we assume this leakage occurs over 10⁷ m², the seepage flux is approximately 1.3 x 10⁻⁴ kg m⁻² s⁻¹. The surface layer part of the system has porosity equal to unity and a logarithmic velocity profile that we specify by using variable permeabilities in the layers above the ground surface. We define a reference velocity at an elevation of 10 m above the ground to be 1 m s⁻¹ and 5 m s⁻¹ to test two different wind conditions. The simulation is run for six months allowing time for the CO₂ to migrate upward through the unsaturated zone, and seep out of the ground where it is advected and dispersed by wind in the atmospheric surface layer. The simulation is isothermal at 15 °C.

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Subsurface</strong></td>
<td></td>
</tr>
<tr>
<td>Permeability ((k = k_s))</td>
<td>1 x 10⁻¹² m²</td>
</tr>
<tr>
<td>Porosity ((\phi))</td>
<td>0.2</td>
</tr>
<tr>
<td>Infiltration rate ((i))</td>
<td>10. cm yr⁻¹</td>
</tr>
<tr>
<td>Residual water sat. ((S_w))</td>
<td>0.1</td>
</tr>
<tr>
<td>Residual gas sat. ((S_f))</td>
<td>0.01</td>
</tr>
<tr>
<td>van Genuchten (1980) (\alpha)</td>
<td>1 x 10⁻⁴ Pa⁻¹</td>
</tr>
<tr>
<td>van Genuchten (1980) (m)</td>
<td>0.2</td>
</tr>
<tr>
<td><strong>Surface Layer</strong></td>
<td></td>
</tr>
<tr>
<td>Pasquill-Gifford stability class</td>
<td>F</td>
</tr>
<tr>
<td>Friction velocity for (u = 1 \text{ m s}^{-1})</td>
<td>0.0869 m s⁻¹</td>
</tr>
<tr>
<td>Friction velocity for (u = 5 \text{ m s}^{-1})</td>
<td>0.434 m s⁻¹</td>
</tr>
<tr>
<td>Reference height ((z_0))</td>
<td>0.10 m</td>
</tr>
<tr>
<td>Reference velocity at (z = 10 \text{ m})</td>
<td>1 or 5 m s⁻¹</td>
</tr>
</tbody>
</table>

Results are shown in Figures 6 and 7. As shown in the figures, concentrations of CO₂ are quite high in the unsaturated zone because the CO₂ sweeps through the pores and displaces existing soil gas. A sharp gradient in concentration is maintained at the ground...
surface because of the large amount of dilution afforded by the wind which advects air into the seeping CO$_2$ and carries it downwind. Note that we have assumed zero background CO$_2$ concentration in the system to examine the CO$_2$ added by the leakage and seepage processes. As shown in Figures 6 and 7, the CO$_2$ concentrations rise strongly in the subsurface, but the CO$_2$ concentrations in the surface layer due to this seepage flux and wind condition are practically negligible. Indeed, Figures 6 and 7 show that the concentrations increase by approximately 0.0001 (100 ppmv) just above the source area and far less above and downwind from it. Such concentration increases would be easily detectable relative to a background CO$_2$ concentration of 0.000375 (375 ppmv), but would not be a health hazard (NIOSH, 1981). Dispersion is higher in the 5 m s$^{-1}$ case than in the 1 m s$^{-1}$ case as expected.

Note the interesting result that CO$_2$ re-enters the subsurface through dissolution in infiltrating rainwater. This is an example of the need for coupled modeling approaches, that include interactions between the surface layer and subsurface that may be significant in some situations. Although the results presented here are two-dimensional, T2CA is a fully three-dimensional model as long as the $x$-coordinate is aligned with the wind direction.

![Image](image1.jpg)

**Figure 6.** Gas-phase mass fraction of CO$_2$ and velocity in the fully coupled subsurface–surface layer model domain six months after CO$_2$ seepage begins for a reference velocity of 1 m s$^{-1}$.

![Image](image2.jpg)

**Figure 7.** Gas-phase mass fraction of CO$_2$ and velocity in the fully coupled subsurface–surface layer model domain six months after CO$_2$ seepage begins for a reference velocity of 5 m s$^{-1}$.

**CONCLUSIONS**

We have developed simulation capabilities for coupled subsurface–surface layer advection and dispersion of CO$_2$ that may potentially seep from the ground after leaking from geologic carbon sequestration sites. The purpose of this research is to examine potential health, safety, and environmental risks, as well as to make specifications for instrumentation and design monitoring strategies that can be used to verify carbon sequestration and ensure minimal health and environmental risk. The approach we have taken for the dense gas CO$_2$ is to focus on the difficult-to-detect cases of diffuse gas seepage where fluxes are small and surface layer concentrations are low. In these scenarios, dispersion in the atmospheric surface layer is passive, and the steady logarithmic velocity profile can be used to model time-average winds. Dispersion in the atmosphere is estimated based on distance- and time-dependent empirical dispersion coefficients that are added to the effective diffusion coefficient in T2CA.

Application of the method to a CO$_2$ leakage and seepage scenario shows that while high concentrations of CO$_2$ can develop in the subsurface, in the surface layer dilution strongly attenuates the seepage plume. Our preliminary simulation shows that while such seepage would be readily detectable by conventional instrumentation which can detect in the ppmv range, the additional CO$_2$ would not constitute a significant health or environmental hazard. Furthermore, as testimony to the need for coupled models, we observed that rainfall infiltration is capable of bringing CO$_2$ back into the subsurface through dissolution into rainwater infiltrating into the subsurface.

**ACKNOWLEDGMENT**

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