Modeling of Near-Surface Leakage and Seepage of CO_2 for Risk Characterization

Chapter Authors

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Abstract

The injection of carbon dioxide (CO_2) into deep geologic carbon sequestration sites entails risk that CO_2 will leak away from the primary storage formation and migrate upwards to the unsaturated zone from which it can seep out of the ground. We have developed a coupled modeling framework called T2CA for simulating CO_2 leakage and seepage in the subsurface and in the atmospheric surface layer. The results of model simulations can be used to calculate the two key health, safety, and environmental (HSE) risk drivers, namely CO_2 seepage flux and near-surface CO_2 concentrations. Sensitivity studies for a subsurface system with a thick unsaturated zone show limited leakage attenuation resulting in correspondingly large CO_2 concentrations in the shallow subsurface. Large CO_2 concentrations in the shallow subsurface present a risk to plant and tree roots, and to humans and other animals in subsurface structures such as basements or utility vaults. Whereas CO_2 concentrations in the subsurface can be high, surface-layer winds reduce CO_2 concentrations to low levels for the fluxes investigated. We recommend more verification and case studies be carried out with T2CA, along with the development of extensions to handle additional scenarios such as calm conditions, topographic effects, and catastrophic surface-layer discharge events.

Introduction

The injection of carbon dioxide (CO_2) into deep geologic formations for carbon sequestration

involves the risk that CO_2 will unexpectedly leak away from the target formation and migrate generally upward eventually reaching the shallow subsurface where CO_2 could seep out of the ground. In the near-surface environment, defined here roughly as within 10 m of the ground surface either above or below ground, high concentrations of CO_2 can pose significant health, safety, and environmental (HSE) risks. The assessment of HSE risks is an essential part of public acceptance, planning, and permitting of geologic carbon sequestration projects. Risk assessment in general can be divided into three parts: (1) definition of scenarios of what can go wrong; (2) assessment of the likelihood of those scenarios; and (3) assignment of a measure of severity to the consequences arising from a given scenario. When applying this approach to substances that pose a hazard to human health and ecosystems, the risk assessment process includes hazard identification and risk characterization. For geologic carbon sequestration, a recognized HSE hazard is CO_2 leakage and seepage from the storage site leading potentially to exposure by humans, plants, and animals to elevated CO_2 concentrations in air and water. Risk characterization requires the estimation or calculation of elevated CO_2 concentrations to which humans, plants, and animals may be exposed in the given failure scenarios. The research described here focuses on calculating CO_2 concentrations and fluxes using a coupled subsurface and atmospheric-surface-layer numerical simulator.

A formal and consistent terminology is needed to describe the different modes of CO_2 migration. We define *leakage* as migration away from the primary sequestration target formation, whereas *seepage* is CO_2 migration through an interface such as the ground surface, a basement floor or wall, or the bottom of a body of surface water. In Figure 1, we present a schematic of some of the important features that may affect HSE risk characterization for CO_2 leakage and seepage in the near-surface environment, a region that we define as within approximately 10 m of the ground surface, either below (i.e., in the subsurface) or above (i.e., in the atmospheric surface layer). These features include a house with a basement and cracked floor through which CO_2 can seep, and a water well through which water with high dissolved CO_2 content could be produced if CO_2 leaked up through the aquifer. Also shown are plants, a tree, and roots that may be sensitive to

elevated CO_2 concentrations in the shallow subsurface. We also show animals that live in the ground and therefore may be susceptible to elevated CO_2 concentrations in soil, along with their burrows that may provide fast-flow paths for CO_2 that enhance mixing by barometric pumping of soil gas and ambient air. Snow cover or ice (not shown) can also affect CO_2 flow and transport. In addition, we show in Figure 1 the saturated zone, unsaturated zone, surface water, and wind in the atmospheric surface layer all of which may be capable of diluting and attenuating leaking and seeping CO_2.

In this chapter, we summarize our research into the development and demonstration of the coupled modeling framework T2CA applicable to the leakage and seepage of CO_2 from geologic carbon sequestration sites. The purpose of the coupled model is to calculate CO_2 fluxes and concentrations in the near-surface environment where risk to humans, plants, and animals is highest. The underlying premise of our approach is that the fundamental drivers of the HSE risk are the CO_2 flux and near-surface CO_2 concentrations, and that a capability to calculate these quantities is essential for a defensible HSE risk assessment. A new coupled model is required because to our knowledge there is no existing model that handles both subsurface and atmospheric surface-layer transport and dispersion along with the coupling at the subsurface–surface-layer interface at length scales of order 10^2–10^3 m. The focus of our approach is on diffuse and low level leakage that could occur through the natural barriers in the subsurface as opposed to catastrophic leakage such as may occur through abandoned wells or well blowouts.

Methodology

Key Concepts

The methodology and structure of the coupled modeling framework are based on the following key concepts: (1) the human, plant, and animal receptors span the interface between the subsurface and surface layer; (2) the flow processes involved in leakage and seepage are

coupled; and (3) the main risk drivers are CO_2 flux and concentration. Before describing the methods and structure, we elaborate on these three key concepts and discuss the time and length scales appropriate to our approach.

First, HSE risk assessment applies to humans, plants, and animals. These environmental receptors live generally near the ground surface but may be entirely below, entirely above, or in both regions at different times. As examples of the importance of the subsurface, surface-layer, and in-between environments, consider the house and basement and the burrowis of animals shown in Figure 1. Clearly the house and the burrow are open to gas flow from both the subsurface and surface layer and therefore CO_2 in either the subsurface or surface layer has the potential to affect the environment in which people or animals live. The plants and trees and their roots similarly will be affected by CO_2 leakage and seepage in both the subsurface and surface-layer environments. Because exposure to CO_2 in the near-surface environment is the main risk associated with CO_2 leakage and seepage, we have developed a coupled modeling framework that focuses on this region.

Second, CO_2 leakage and seepage are coupled transport processes. Specifically, CO_2 gas in the near-surface environment will flow by advection and diffusion as controlled by pressure, density, and concentration gradients. For example, seeping CO_2 will be strongly advected by surface winds above the ground surface, while atmospheric pressure variations (i.e., barometric pumping) will cause CO_2 to move in and out of the subsurface. However, the low permeability of soils will tend to dampen subsurface advective transport driven by pressure variations and wind in the surface layer. Rainfall infiltration containing dissolved CO_2 can be another mechanism for CO_2 to return from the surface layer to the subsurface. Because these apparent coupled processes occur between the surface layer and subsurface, a coupled modeling framework capable of modeling these interactions is required.

Third, if high CO_2 concentrations are the fundamental adverse condition for HSE risk, then CO_2

seepage flux and near-surface CO_2 concentration are the main risk drivers. Seepage flux in terms of mass has units of kg CO_2 m^{-2} s^{-1} and is a measure of the rate at which CO_2 is passing out of the ground per unit area. If CO_2 is the only component of the gas stream seeping out of the ground, then flux and concentration are directly correlated. However, if the CO_2 is contained within a stream of another component (e.g., with steam in a geothermal vent), then there can be a high CO_2 flux with low CO_2 concentrations. In this sense, flux and CO_2 concentration must be considered independently. In the case where the only component in the seeping gas is CO_2, the seepage flux is a good indicator of whether the given surface-layer winds, surface-water flows, or plant uptake rates are capable of reducing CO_2 concentrations to near-ambient levels. Annual leakage rates given as percentages per year of given CO_2 sequestration projects should not be used for characterizing risk since they do not provide information on the form or nature of the leakage process. As for CO_2 concentrations, the location of the occurrence of high concentration and nature of the receptor control the attendant risk. For example, high CO_2 concentrations at a depth of 1 m in the ground may cause negligible risk to humans because people live mostly above the ground surface, while such concentrations would pose a serious risk to burrowing animals or to plants through exposure to their roots.

Given these key concepts, it is apparent that a quantitative coupled modeling capability is required to make defensible estimates of CO_2 flux and concentration for various expected leakage and seepage scenarios. Overly simplified models of the subsurface or surface layer alone may not stand up to public and scientific scrutiny. We have used a methodology and structure that is based on multiphase and multicomponent reservoir simulation. The fluxes and concentrations calculated by the coupled framework can be used as inputs to exposure models to calculate defensible HSE risks. The direct output from the present coupled modeling framework is also useful by itself since CO_2 flux and concentration are primary risk drivers. The approach we have taken can be used to model the whole leakage pathway from deep sequestration site to the surface, but here we focus the model description on the region where the main HSE hazards occur, namely the near-surface environment containing the unsaturated zone and surface layer.

Length and Time Scales

With CO_2 storage and sequestration operations potentially occurring on a large and widespread industrial scale, the length and time scales of interest to CO_2 risk characterization are quite large. Because broad and diffuse CO_2 seepage may occur over large areas for long periods of time, such leakage and seepage may be hard to detect and difficult to mitigate. As such, diffuse seepage is an important focus for risk assessment and risk management. Catastrophic events such as well failures are also relevant, but such events are obviously serious HSE risks and everything possible will be done to stop such events. We have focused on the 10 m to 10^3 m length scale, and the 1 month to 10 year time scale consistent with the diffuse seepage scenario that is our focus. Over these length and time scales, averaging is defensible. For example, constant wind speed, pressure, rainfall infiltration, and other weather-related processes can be used along with appropriate parameterizations since the time scale is relatively long. While the coupled model is capable of nonisothermal simulations, we consider here only isothermal situations and we parameterize turbulence using variable-K theory to model atmospheric dispersion.

Subsurface Flow and Transport

The coupled modeling framework we are using is built on the TOUGH2 code [1], a multiphase and multicomponent integral finite difference reservoir simulator. Briefly, TOUGH2 uses a multiphase version of Darcy's law for fluid flow and the advective-dispersive model for component transport. Readers interested in greater detail and information on the theory or practical implementation of TOUGH2 should consult the users guide [1] and the website (http://www-esd.lbl.gov/TOUGH2). The coupled model handles five components (H_2O, brine, CO_2, a gas tracer, air) and heat. Air is a pseudocomponent that is approximated as a mixture of 21% oxygen

and 79% nitrogen by volume. Real gas mixture properties are calculated so the full range from high-pressure sequestration-site conditions to low-pressure ambient surface-layer conditions can be modeled. We refer to the coupled model as T2CA, for TOUGH2 for CO_2 and Air. While the discussion below focuses on the CO_2 transport, all of the gas-phase components are modeled in the TOUGH2 multicomponent framework, and an analogous treatment can be developed for heat.

Atmospheric Dispersion

The approach we use for atmospheric surface-layer transport is based on gradient transport and variable-K theory (e.g., [2]). In this approach, the advection and dispersion of CO_2 are modeled with an advective-dispersive transport equation in which advection is unidirectional in the *x*-direction, velocity varies with height according to the logarithmic velocity profile applicable for neutral stability conditions, and the dispersivities K_y and K_z model eddy diffusion. The advective-dispersive transport equation with *x*-axis aligned with the unidirectional flow field can be written for CO_2 concentration (*c*) as

$$\frac{\partial c}{\partial t} + u\frac{\partial c}{\partial x} - \frac{\partial}{\partial y}\left(K_y \frac{\partial c}{\partial y}\right) - \frac{\partial}{\partial z}\left(K_z \frac{\partial c}{\partial z}\right) = 0 \qquad (1)$$

where we assume advection dominates transport in the *x*-direction. The logarithmic velocity profile for neutral stability conditions is given by the equation

$$u(z) = \frac{u_*}{k}\ln\left(\frac{z}{z_0}\right) \qquad (2)$$

[2, 3] where u^* is the friction velocity, k is von Karman's constant (k = 0.4), z_o is the roughness length, and z is height above the ground surface. Turbulent eddies act to disperse gaseous components, and these eddies become larger with elevation above the ground surface. Arya [2] recommends use of an increasing K_z with elevation for neutral stability conditions according to

$$K_z = k\ u_*\ z \qquad (3)$$

Lateral dispersion K_y in variable-K theory is less well understood, and we avoid consideration of how to parameterize K_y by adopting a 2-D model problem that neglects lateral dispersion of CO_2 and will therefore be conservative in that CO_2 concentrations will be overestimated relative to a case with lateral dispersion.

In summary, for atmospheric dispersion in the surface layer we use variable-K theory and we assume neutral stability and a logarithmic velocity profile The logarithmic velocity profile represents time-averaged surface winds to model advection in the surface layer, with turbulent mixing parameterized by a variable K_z. The velocity field in the surface layer is prescribed as an initial condition and stays constant throughout the simulation. The surface layer is defined simply by setting porosity to unity and layer permeabilities to a range of values orders of magnitude larger than the subsurface parts of the domain and that specify the desired logarithmic profile for the given boundary conditions. The entire coupled subsurface–surface-layer calculation is carried out using a single grid. Hence, the model regions are implicitly coupled. Full multiphase and multicomponent flow and transport are used throughout the domain.

Field experiments of dense gas dispersion have been used to develop correlations involving the most important parameters controlling atmospheric dispersion such as wind speed, density of released gas, and release flux [4, 5]. These correlations were developed based on simple scale and dimensional analyses. 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 density-dependent or passive (not density-dependent) as appropriate for a gas tracer. In density-dependent dispersion of a dense gas like CO_2, the gas can flow in response to its own density gradient relative to air, and it can resist mixing if contained in a low-lying area such as a valley or other topographic depression. In Figure 2, we have plotted this correlation with values appropriate for CO_2-air mixtures for various source-area length scales along with the typical ecological flux of CO_2 emitted and taken up by plants, soil, and roots known as the net ecosystem exchange (NEE) [6]. As shown in Figure 2, seepage fluxes have to be quite high (note

logarithmic scale) for windy situations for the resulting dispersive mixing process to be density-dependent. Note that wind conditions are averages over a period of 10 minutes.

In prior work [7], we have simulated subsurface migration of leaking CO_2 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 CO_2 on the order of 10^9 kg through a circular region with radius 100 m. 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 2, seepage fluxes of this magnitude lead to passive dispersion for all but the calmest wind conditions. It must be emphasized that deriving a leakage rate from annual percent leakage is case-specific in that doing so produces a leakage rate that is dependent on the mass of stored CO_2, i.e., the size of the sequestration project. For example in this case, if the project were 100 times larger (stored CO_2 on the order of 10^{11} kg), seepage fluxes on the order of 10^{-5}–10^{-6} kg m^{-2} s^{-1} would result from leakage rates of 0.001% yr^{-1} for the same leak geometry. Similarly, 0.1% yr^{-1} leakage from a project 100 times larger would produce fluxes of order 10^{-3}–10^{-4} kg m^{-2} s^{-1} for the same geometry, which could produce density-dependent dispersion at higher wind speeds as shown in Figure 2. In general, the CO_2 leakage and seepage flux are the important quantities governing flow behavior, while percent leakage per year provides information only about mass loss and requires definition of the project size and leakage or seepage area.

Results and Discussion

Verification

The subsurface flow and transport methods in T2CA are well established by virtue of the long history of TOUGH2, the novel part being the equation of state module for the mass components water, brine, CO_2, gas tracer, and air. We have compared physical properties of the gas mixtures in T2CA against independent predictions and observed good agreement [7]. Real gas mixture

properties are calculated because they are needed at depth, e.g., below approximately 800 m where CO_2 becomes supercritical, although our focus here is on the unsaturated zone and surface layer where pressures are approximately 1 bar (0.1 MPa).

Here we present verification of the surface-layer methods in T2CA for the special case of uniform velocity and constant eddy diffusivity, in which the approach reduces to the well-known Gaussian plume dispersion model for which there are simple analytical solutions. We present in Figure 3 results of a verification study in which we compared the T2CA result of a 3-D Gaussian plume dispersion problem against the analytical solution. In this problem, u = 1 m s^{-1}, $D_{xx} = D_{yy} = D_{zz}$ = 5 m s^{-2}. The point-source strength $Q_{1/4}$ = 0.0785 kg s^{-1}, where $Q_{1/4}$ is the source strength for the one-quarter domain used in the T2CA simulation that takes adavantage of the symmetry planes in the horizontal and vertical directions parallel to the flow direction. The main part of Figure 3 shows the 3-D plume, while the upper inset shows the *y-x* plane with comparison of the T2CA result to the analytical solution given by Arya [2]. The agreement is very good and confirms our implementation of surface layer atmospheric dispersion processes in T2CA.

Unsaturated Zone Attenuation

The purpose of this application is to examine the extent to which the unsaturated zone can attenuate CO_2 leakage, full details of which can be found in [7]. We consider a radial system with a thick (30 m) unsaturated zone into which a CO_2 leakage flux enters from below. The leakage fluxes are arbitrarily set at 4.04 x 10^{-6}, 4.04 x 10^{-7}, and 4.04 x 10^{-8} kg m^{-2} s^{-1}. For reference, a leakage flux of 4.04 x 10^{-6} kg m^{-2} s^{-1} would correspond to an annual loss through a 100 m radius region of 0.1% yr^{-1} of a 4 x 10^{9} kg CO_2 sequestration project, or 0.001% yr^{-1} of a project 100 times bigger (4 x 10^{11} kg CO_2). We point this out to emphasize again that leakage flux rather than annual percentage loss controls leakage and seepage processes. In some cases leakage rate and leakage flux will be loosely correlated because leakage area may scale with size of project, but in general these quantities represent distinct measures of sequestration integrity. The leakage area was one of the many properties of the system that was varied as part of the

sensitivity analysis discussed below. Rainfall infiltration flows downward through the section and acts to dissolve CO_2 and transport it downward. Additional properties of the system for the base case are provided in Table 1.

Figure 4 shows the steady-state simulation results for the base case at the three different arbitrary leakage rates (0.1% yr^{-1} (4 x 10^6 kg yr^{-1}), 0.01% yr^{-1} (4 x 10^5 kg yr^{-1}) and 0.001% yr^{-1} (4 x 10^4 kg yr^{-1})). Steady state is reached after approximately 0.3, 5, and 30 yrs for the three cases, respectively. Carbon dioxide concentrations in the shallow subsurface increase with increasing leakage rate, as diffusion and the specified rainfall infiltration are overwhelmed by larger leakage fluxes. Note further the limited degree to which the CO_2 spreads outward in the unsaturated zone despite the density contrast. Pressure gradients induced by the active leakage flux dominate over gravity effects here and thus lead to predominantly vertical CO_2 flow through the vadose zone to the ground surface [7].

Figure 5 shows seepage flux and near-surface CO_2 concentration (mole fraction) for a large number of simulations carried out as part of a sensitivity analysis [7]. For reference, we have plotted the typical ecological flux or net ecosystem exchange (NEE) 4.4 x 10^{-7} kg m^{-2} s^{-1} [6] and the soil-gas CO_2 mole fraction (x_{gas}^{CO2} = 0.3) that appears to have caused tree mortality at Mammoth Mountain, California [8]. As shown, the leakage flux exerts the strongest control on flux and concentration at the ground surface. Permeability and permeability anisotropy are also very important in controlling CO_2 seepage flux and near-surface concentrations. Simulations of barometric pumping presented in prior work [7] show that pressure variations produce local temporal changes in flux and concentration but have little effect on long-term average values for this leakage scenario. The fundamental observation of the simulation results presented here is that subsurface CO_2 concentrations from leakage and seepage can be high in the near-surface environment, even when the fluxes are of the same order of magnitude as the NEE [6].

Subsurface–Surface-Layer Coupling

We have also applied the new simulation capability to a coupled subsurface–surface-layer cartesian system, properties of which are listed in Table 2. The domain discretization and boundary conditions are shown in Figure 6. The bottom boundary is held at constant pressure, while the top boundary is closed. The side boundaries are closed in the unsaturated zone, and held at constant pressure in the surface layer to prescribe the logarithmic velocity profile. Further details of our modeling approach and of this application can be found in [9]. We present in Figure 7 simulation results after 6 months of leakage showing subsurface and surface-layer CO_2 concentrations (mass fraction) and gas-phase velocity vectors for the cases of winds of 1, 3, and 5 m s^{-1} at a height of 10 m from the ground surface and neutral atmospheric conditions (e.g., [2, 3]). In Figure 7d we show a summary of the temporal evolution of the CO_2 gas mass fraction at x = 645 m (approximately 100 m downstream from the edge of the source). Figure 7a, b, c shows the strong effects of wind and atmospheric dispersion on seeping CO_2. Concentrations downwind from the source are strongly attenuated by turbulent mixing. Note further in Figure 7a, b, c the downward migration of CO_2 into the subsurface downwind of the source. This process is due to CO_2 dissolution in rainwater that is infiltrating at 10 cm yr^{-1}. It is important to note that in all of the simulations we have assumed a zero background CO_2 concentration to emphasize the additional CO_2 that seeps from the ground in the various scenarios. Note that the mass fraction scale in Figure 7 shows that CO_2 concentrations in the surface layer are very low, barely above the background concentration of 370 ppmv which would be 0.00056 by mass fraction. The fundamental conclusion is that surface winds and atmospheric dispersion appear to be very effective at diluting diffuse CO_2 seepage fluxes over flat ground. We note that calm conditions, topographic depressions, and higher CO_2 seepage fluxes not yet analyzed can cause larger CO_2 concentrations to develop.

Given that HSE risks will be calculated based on exposures at certain locations in the flow field, we present in Figure 7d downwind-CO_2 concentrations as a function of time for the test problem.

Note that concentrations are conservative because of the assumption of a 2-D system, and the use of a closed boundary at the top of the surface layer. For this case of diffuse CO_2 seepage, concentrations would be elevated above background by approximately 23 ppmv (3.5×10^{-5} mass fraction) for the 1 m s^{-1} case, and concentrations decrease approximately linearly with reference wind speed. Although this test problem is 2-D, the coupled modeling framework is a fully 3-D capability.

Conclusions

Main Points

We have demonstrated the use of a coupled modeling framework for modeling CO_2 fluxes and concentrations for risk characterization. This work is relevant and important to the development of geologic carbon sequestration because it provides a modeling capability for simulating CO_2 flow and transport from the deep CO_2 storage site all the way to the atmosphere. The approach is built on the assumption that the near-surface environment is the main region in which HSE risks will arise. In this region, CO_2 flux and concentration are the main risk drivers. The coupled model handles subsurface and atmospheric surface-layer flow and transport assuming that dispersion in the surface-layer is passive and that the wind is described by a logarithmic velocity profile. Model results show limited unsaturated zone attenuation of leakage flux, with correspondingly large CO_2 concentrations possible in the shallow subsurface. These results suggest that if leakage leads to CO_2 migrating as far as the vadose zone, high CO_2 concentrations can occur in the root zone of the shallow subsurface with potentially harmful effects on plants, as well as on humans or other animals in poorly ventilated subsurface structures such as basements or burrows. Coupled subsurface–surface-layer demonstration simulations show the large degree of dilution that occurs in the surface layer, and the possible reflux of CO_2 to the subsurface that occurs when CO_2 dissolves in infiltrating rainwater.

Recommendations

We recommend development of additional capabilities for risk characterization related to leakage and seepage, along with further verification and testing of model approaches. Although the coupled modeling framework T2CA is applicable to many important leakage and seepage scenarios, it is not applicable to absolute calm conditions where dense-gas dispersion occurs, nor is it applicable to very high fluxes such as might occur from an open well or catastrophic tank or pipeline release into the open atmosphere. In addition, buildings are neglected even though it is well established that exposures to people by soil-gas contaminants (e.g., radon) are most likely to occur indoors. We recommend that future research funding be directed toward model development for simulation of the foregoing processes. Finally, the surface-layer methods in T2CA should be compared against other atmospheric dispersion models for verification, and the methods should be refined if necessary.

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List of Acronyms and Abbreviations

2–D	Two–dimensional
3–D	Three–dimensional
CO_2	Carbon dioxide
HSE	Health, safety and environmental (risks)
NEE	Net ecosystem exchange (for CO_2)
TOUGH2	Reservoir simulator, Transport Of Unsaturated Groundwater and Heat 2.
T2CA	for TOUGH2 for CO_2 and Air.
x	mole fraction.
X	mass fraction.

Table 1. Hydrogeological properties of the unsaturated zone for the base case.

Property	Value
Permeability ($k_r = k_Z$)	1×10^{-12} m^2 (1 Darcy)
Porosity (ϕ)	0.2
Infiltration rate (i)	10.0 cm yr^{-1}
Temperature (T)	15 ˚C
Residual water saturation (S_{lr})	0.1
Residual gas saturation (S_{gr})	0.01
van Genuchten [10] α	1×10^{-4} Pa^{-1}
van Genuchten [10] m	0.2

Table 2. Properties of the coupled subsurface–surface-layer model system.

Property	Value
Subsurface	
Subsurface region extent (x x y x z)	1 km x 1 m, 0 m < z < 35 m
Discretization (Nx x Ny x Nz)	100 x 1 x 35
Permeability ($k_X = k_Z$)	1×10^{-12} m^2
Porosity (ϕ)	0.2
Infiltration rate (i)	10.0 cm yr^{-1}
CO_2 flux region	450 m < x < 550 m
CO_2 mass flux (0.1, 0.01, and 0.001% yr^{-1})	4.04×10^{-6},10^{-7},10^{-8} kg m^{-2} s^{-1}
Residual water sat. (S_{lr})	0.1
Residual gas sat. (S_{gr})	0.01
van Genuchten [14] α	1×10^{-4} Pa^{-1}
van Genuchten [14] m	0.2
Surface Layer	
Surface-layer region extent (x x y x z)	1 km x 1 m, 35 m < z < 45 m
Discretization (Nx x Ny x Nz)	100 x 1 x 20
Pressure in surface layer	1 bar (0.1 MPa)
Temperature (isothermal)	15 ^oC
Atmospheric stability	neutral
Velocity profile	logarithmic
Reference velocity at z = 10 m	1, 3, or 5 m s^{-1}
Friction velocity for u_x = 1, 3, 5 m s^{-1}	0.0868, 0.261, 0.434 m s^{-1}
Roughness length (z_0)	0.10 m

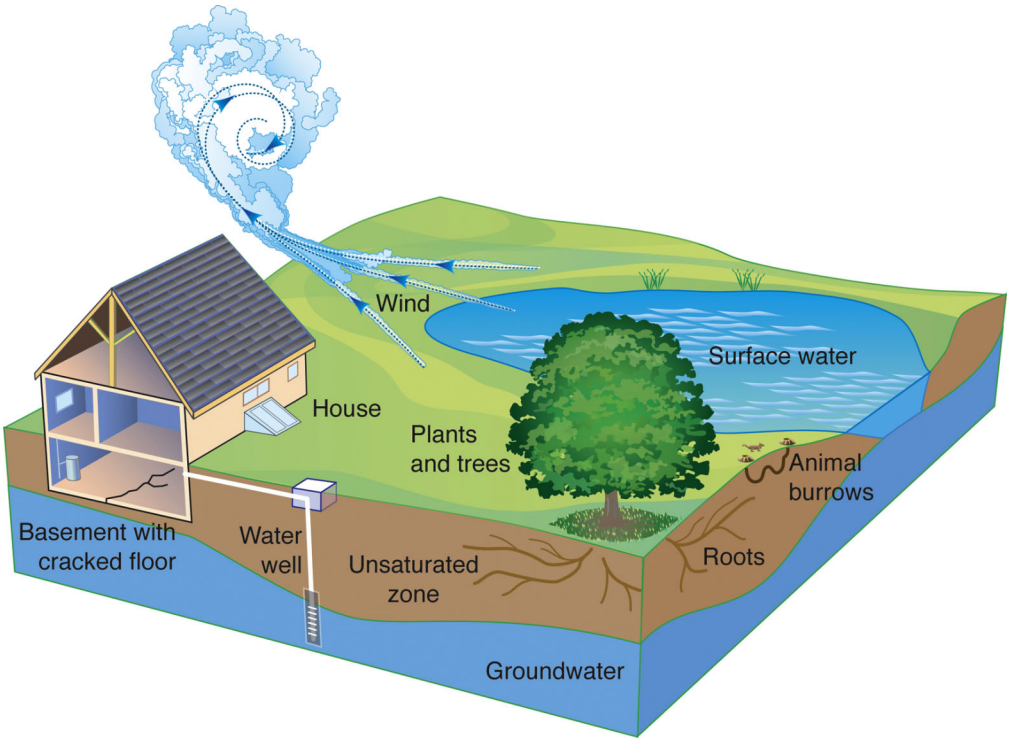

Figure 1. Sketch of near-surface environment with accompanying features relevant to HSE risk associated with CO_2 leakage and seepage.

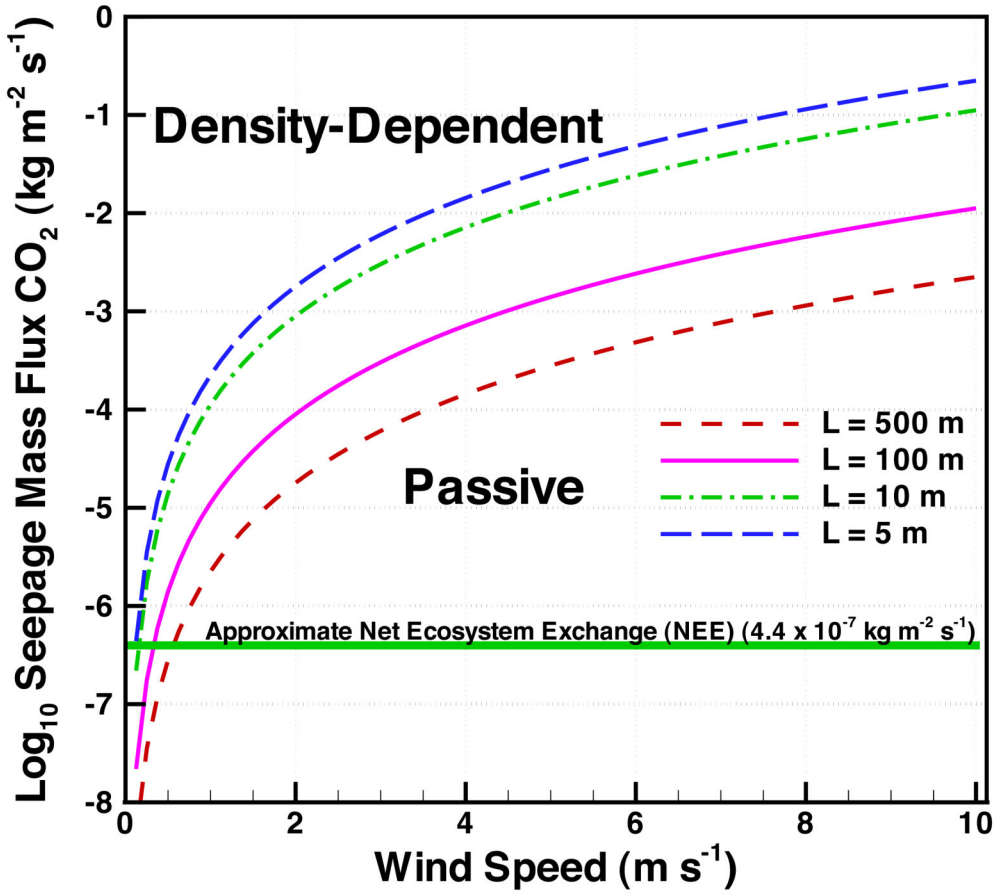

Figure 2. Correlation for density-dependent and passive dispersion in the surface layer as a function of seepage flux and wind speed for four different source length scales.

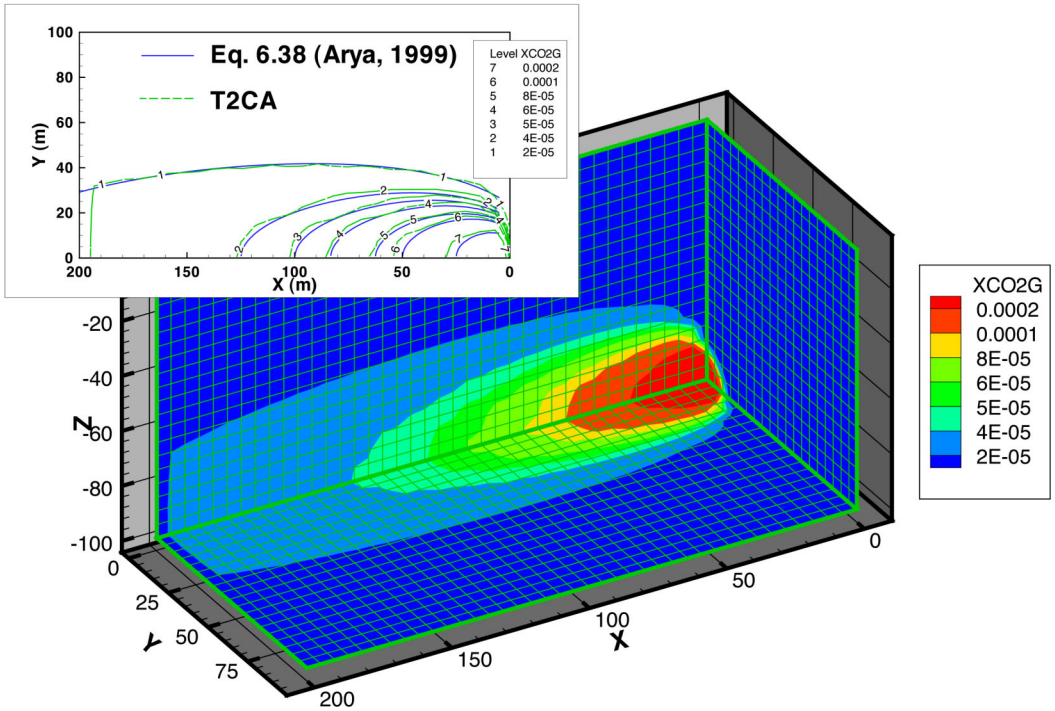

Figure 3. Contours of kg CO_2 m^{-3} gas from T2CA for the Gaussian plume dispersion verification problem in 3-D, and comparison to analytical solution in the *x-y* plane (inset).

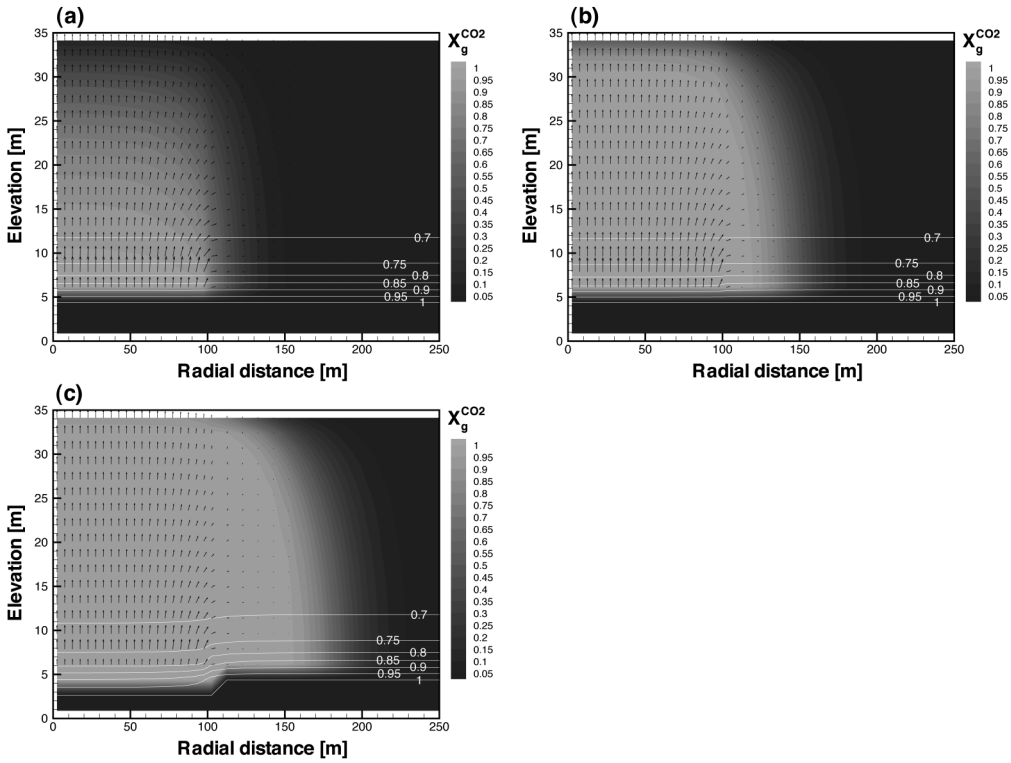

Figure 4. Simulation results for leakage in a thick unsaturated zone where shading indicates mass fraction of CO_2 in the gas phase, and labeled contour lines indicate water saturation, and vectors show gas phase pore velocity for steady-state leakage rates of 4 x 10^4, 4 x 10^5, and 4 x 10^6 kg yr^{-1}. The maximum vector size represents values of approximately (a) 0.057, (b) 0.53, and (c) 3.6 m d^{-1}.

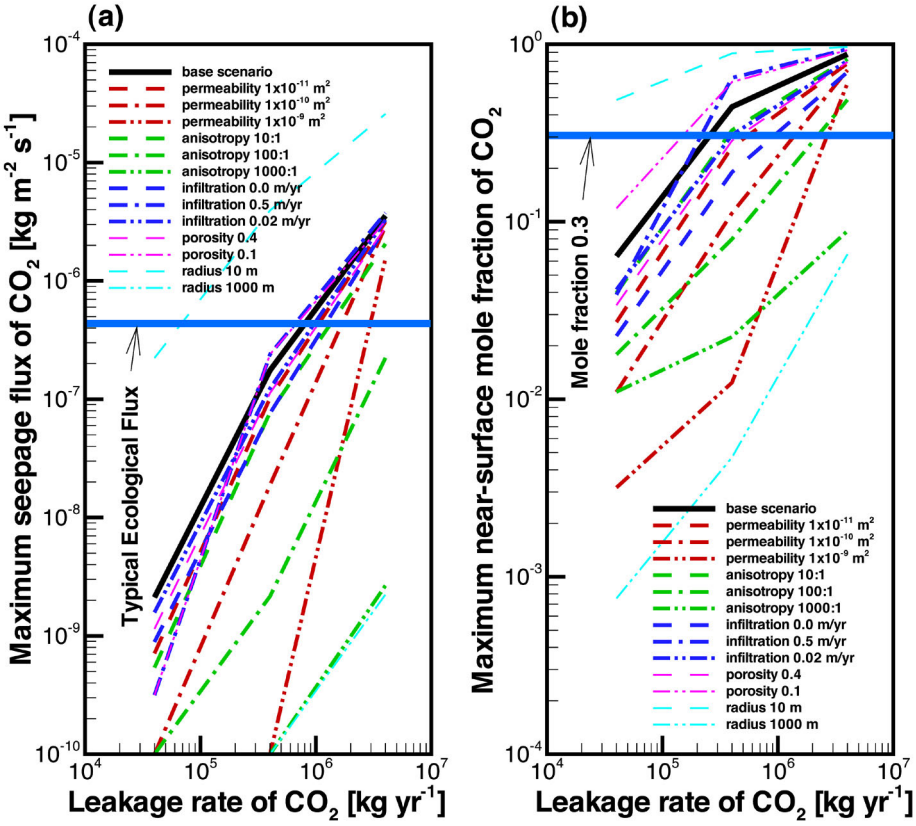

Figure 5. Maximum seepage flux of CO_2 and maximum near-surface gas mole fraction CO_2 as a function of leakage rate at steady-state seepage conditions.

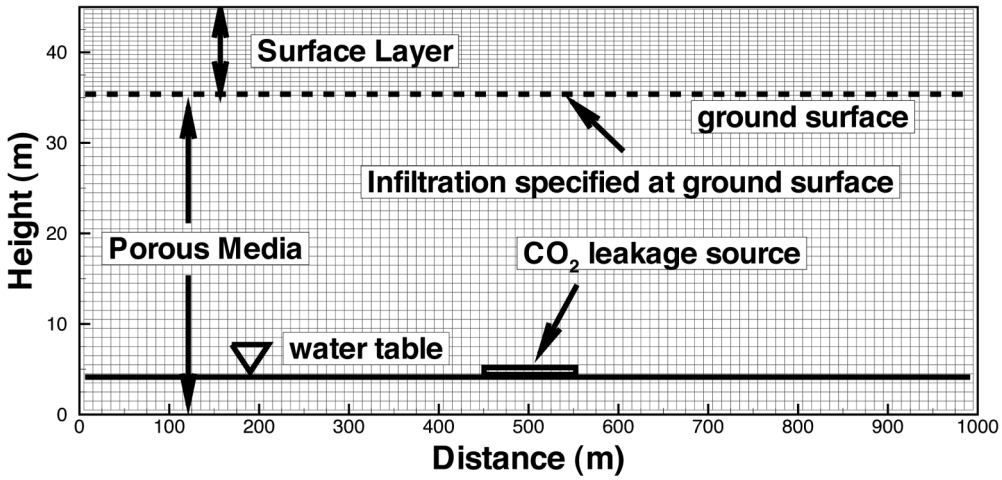

Figure 6. Domain and discretization used in the coupled subsurface–surface-layer test problem.

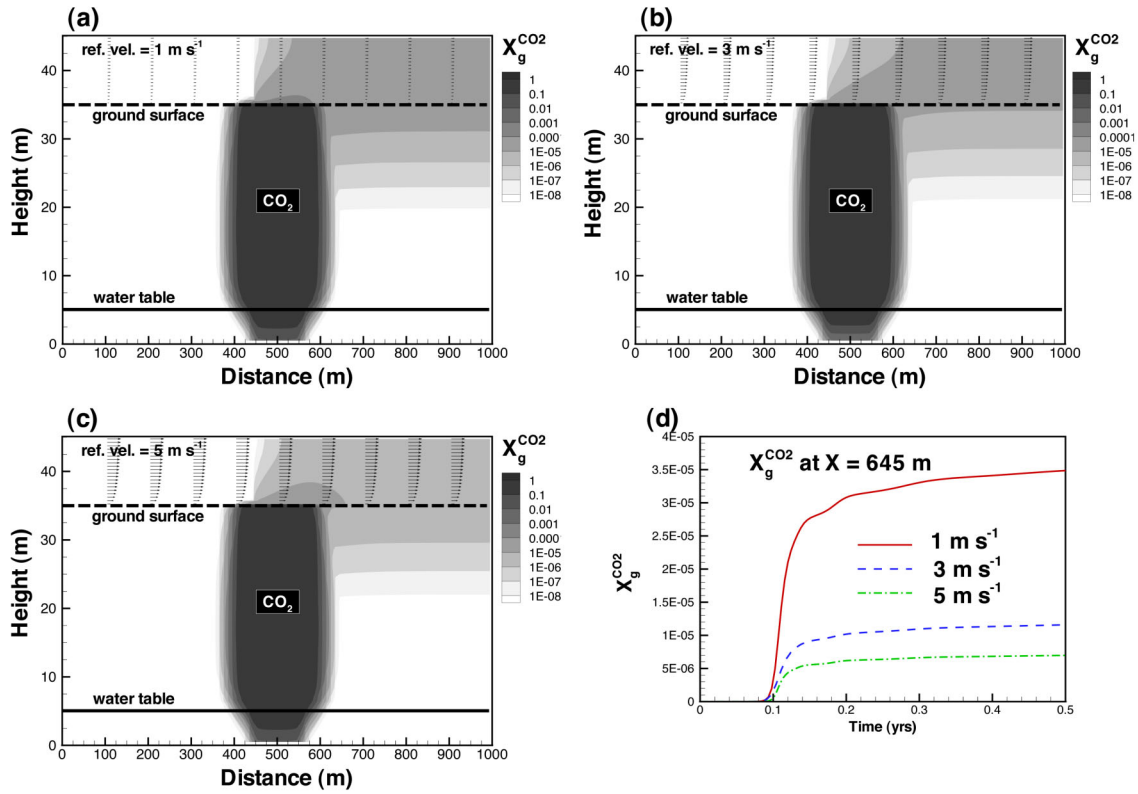

Figure 7. Simulation results for the coupled subsurface–surface-layer problem showing mass fraction of CO_2 in the gas phase and gas velocity vectors. (a) wind speed 1 m s^{-1}; (b) wind speed 3 m s^{-1}; (c) wind speed 5 m s^{-1}; (d) mass fraction CO_2 in the gas vs. time at *x* = 645 m.