

CO₂ Sequestration in Deep Aquifers

Brian J. O. L. McPherson (brian@nmt.edu; 505-835-5259)
Hydrology Program, Earth and Environmental Science
New Mexico Institute of Mining and Technology
Socorro, NM 87801

Peter C. Lichtner (lichtner@lanl.gov; 505-667-3420)
Los Alamos National Laboratory
Earth & Environmental Sciences Division
(EES-6) MS F-649
Los Alamos, NM 87545

Abstract

Disposal and long-term sequestration of anthropogenic "greenhouse gases" such as CO₂ is a proposed approach to reducing global warming. Deep, regional-scale aquifers in sedimentary basins are possible sites for sequestration, given their ubiquitous nature. We used a mathematical sedimentary basin model, including multiphase flow of CO₂, groundwater, and brine, to evaluate residence times in possible aquifer storage sites and migration patterns and rates away from such sites in the Powder River Basin of Wyoming. We also used the model to simulate CO₂ flow through fractures, to evaluate partitioning between fracture and rock matrix. These simulations provide insight regarding the ultimate propensity of permeability reductions versus permeability increases in the fracture zone associated with carbonate reactions.

Regional-scale hydrologic properties, including the presence of fracture zones, were calibrated using surface heat flow data. Our initial results suggest that, in general, long-term (~1000 years or more) sequestration in deep aquifers is possible, if subsurface structure and permeability are well characterized. However, additional risks are involved. In addition to CO₂ escape from sequestration aquifers into other aquifers or to the land surface, another environmental threat posed by subsurface sequestration is contamination by brines. We evaluated the potential for such unintended aquifer contamination by displacement of brines out of adjacent sealing layers such as marine shales. Results suggest that sustained injection of CO₂ may incur wide-scale brine displacement out of adjacent sealing layers, depending on the injection history, initial brine composition, and hydrologic properties of both aquifers and seals.

Introduction

Anthropogenic greenhouse gases, such as carbon dioxide, are considered a major contributor to global warming. Sequestration of power plant generated CO₂ by injection into groundwater aquifers and (petroleum and gas) reservoirs has been proposed as a possible alternative for the reduction of excessive greenhouse gases in the atmosphere. Ideally, injected CO₂ will migrate through an aquifer from injection wells to remote storage sites, and remain isolated from the atmosphere for a considerable period of time.

CO₂ has been used for enhanced oil recovery (EOR) purposes since the 1950s. For example, the first carbonated water floods were tested as early as 1951, and slugs of pure CO₂ for oil displacement were tested as early as 1963. Research on use of CO₂ for EOR continues in earnest today.

However, research concerning terrestrial sequestration of CO₂ for environmental purposes is relatively recent. Fundamental topics of interest in sequestration research have concerned not just scientific and technical aspects, but practical concerns such as the economic feasibility of storage, safety, and the maximum possible amount of CO₂ storage globally and for specified regions.

An economically sound approach to CO₂ sequestration is to identify storage sites close to CO₂ sources such as fossil-fuel power plants. Subsurface groundwater aquifers are suggested, and we further refine this idea to groundwater aquifer systems and petroleum reservoir aquifers such as those found in sedimentary basins. These aquifer systems are large in size, possibly permitting storage of large quantities of CO₂. More importantly, however, these aquifers exist throughout large portions of the continents, improving the chance that CO₂ sources are near storage sites and minimize engineering and transport costs.

Approach

Our hypothesis is that sedimentary basin aquifers and petroleum reservoir systems are good candidates for sequestering CO₂, and further contend that the capability of an aquifer to transmit and store CO₂ is controlled by the fundamental geology: depositional environment, structure, stratigraphy, and pressure/temperature conditions. To test this hypothesis, we examined specific aquifers in a typical intracontinental sedimentary basin, using conceptual and numerical modeling.

Fundamental research questions include: 1. Can typical regional scale aquifers provide the residence time needed to achieve stated global temperature reduction goals? 2. What geologic conditions, e.g., rock properties, depositional environments, structure, etc., provide optimal CO₂ flow and transport, and optimal storage in a sedimentary basin? We approached these questions in a preliminary numerical analysis of a sedimentary basin, the Powder River Basin of Wyoming. The numerical model includes multiphase flow of CO₂, groundwater, and brine, allowing evaluation of residence times in possible aquifer storage sites, and migration patterns and rates away from such sites in the Powder River Basin of Wyoming. We also used the model to simulate CO₂ flow through fractures, to evaluate partitioning of CO₂ between the fracture network and rock matrix. These simulations provide insight regarding the ultimate propensity of permeability reduction versus permeability enhancement in the fracture zone associated with carbonate reactions.

Project Description: Mathematical Model

A two-dimensional finite volume model of the Powder River Basin cross-section SW-NE (Figure 1) is used for the simulations analyses. The model domain is 230 km horizontal by 5 km vertical, consisting of 5000 grid-blocks (100 horizontal by 50 vertical), each 2300 m by 100 m. Eight model units are delineated in the cross-section of Figure 1(a). The computer code TOUGH2 (Pruess, 1991), which includes coupled flow of heat, groundwater, and brine, was used to carry out the simulations.

TOUGH2 was adapted to include an extended equation-of-state for CO₂ (McPherson and Cole, 2000). The approach of Weir, White, and Kissling (1996) was used. These authors developed an extended Redlich-Kwong equation based on the work of Kerrick and

Jacobs (1981) to describe the transport and thermodynamic properties of CO₂ mixtures as a function of pressure, temperature, and CO₂ concentration. Using this extended Redlich-Kwong equation in the TOUGH2 model provides appropriate thermodynamic and transport properties of supercritical CO₂ for the range of pressures and temperatures observed in a typical sedimentary basin.

Results and Application

Calibration of Regional Scale Permeability

The first step for investigating the suitability of the Powder River Basin for sequestering CO₂, is to calibrate the model to present-day conditions. The thermal regime of the Powder River Basin was evaluated by McPherson and Chapman (1996), who analyzed temperature and geologic data from over 3000 oil and gas wells within a 180 km by 30 km transect. The cross-section of Figure 1(a) is the center of that transect. The cause of anomalously high heat flow (>200 mW m⁻²)

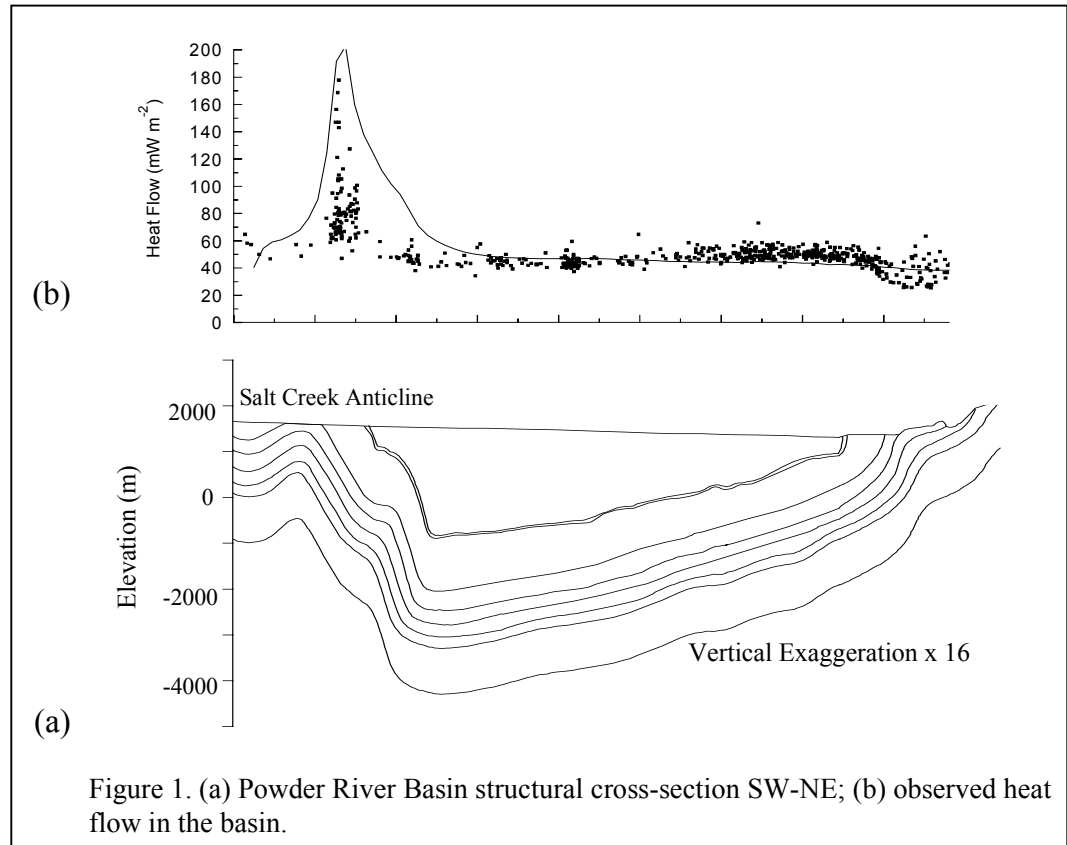


Figure 1. (a) Powder River Basin structural cross-section SW-NE; (b) observed heat flow in the basin.

observed over the Salt Creek anticline on the southwestern side of the transect (Figure 1b) was not definitively resolved. The anomalous thermal regime observed in the southern half of the basin cannot be explained by variation in conductivity alone. It is likely caused by advection of heat in a basin-scale groundwater flow system, including groundwater driven by the topography of the Black Hills in South Dakota, sweeping heat from deeper portions of the basin and discharging in the area of the Salt Creek Anticline. We analyzed this possible flow system using our extended model of coupled groundwater-CO₂-heat flow that accounts for thermal advection, conduction, and heat generation. By matching modeled surface heat flow to the observed distribution, the model provided estimates of effective basin-scale permeabilities of Powder River Basin aquifers, including candidate aquifers for CO₂ sequestration.

The best match to the surface heat flow distribution (Figure 1b) was obtained by assuming a simple but heterogeneous permeability structure: aquifers were assigned effective basin-scale aquifer permeability ranges of $K_{PB} = 5.0 \times 10^{-14} \text{ m}^2$ to $2.0 \times 10^{-13} \text{ m}^2$ and $K_{NB} = 1.0 \times 10^{-17} \text{ m}^2$ to $1.0 \times 10^{-16} \text{ m}^2$ (where K_{PB} = permeability parallel to the bedding plane and K_{NB} = permeability normal to the bedding plane), and shaley units were assigned regional permeability values of $< 10^{-17} \text{ m}^2$. These permeability distributions were used in our simulations of CO₂ transport and storage.

Probably the most interesting result of the permeability calibration analysis was the necessity to simulate fractures in the apex of the Salt Creek Anticline with higher, isotropic ($K_{PB} = K_{NB}$) permeability in order to match model surface heat flow to observed heat flow at the anticline (Figure 1b). Thus, for the regional groundwater flow system to be the primary cause of the anomalously high heat flow observed over the anticline, the anticline must be fractured enough to provide a high permeability conduit.

CO₂ Residence Time and Migration Rates

In our numerical model simulations of the Powder River Basin, we injected supercritical CO₂ into the Fox Hills Sandstone, an Upper Cretaceous formation, at ~1800 meters depth (location shown on Figure 2). The unit above the Fox Hills is the Lance Formation, a Paleocene sandy shale unit. Groundwater flow in this area of the Fox Hills formation is relatively slow, inasmuch as the hydraulic head gradient is weak. Migration of supercritical CO₂ is towards the area of lowest surface elevation adjacent to the Black Hills (left to right on Figure 2). Because the density of supercritical CO₂ is lower than that of normal groundwater, buoyancy as well as hydrodynamic gradients drive the flow of CO₂. Dissolved CO₂ in the aqueous solution tracks the movement of supercritical CO₂. Lower relative and saturated permeability mitigate the vertical movement of CO₂ through the Lance Formation above the Fox Hills.

Figure 3 illustrates lateral migration rates of supercritical CO₂ as a function of matrix permeability. Higher permeability induces higher migration rates, on the order of 100 meters per year. At permeability less than 10^{-15} m^2 (~1 mD), migration rates become negligible, suggesting that intact, low permeability shales should be effective sealing layers. Overpressures associated with high injection rates can induce local migration rates approaching 1 km per year, at higher permeability values (e.g., $\sim 10^{-12} \text{ m}^2$). A sensitivity analysis suggested that the other primary factors affecting migration rate include relative permeability and capillarity, both of which are

poorly characterized in the literature.

Recall that fracture permeability in the Salt Creek area was necessary to mimic observed heat flow patterns. Often such fracture zones in the subsurface are not discovered until drilling, begging the question about effective CO₂ migration

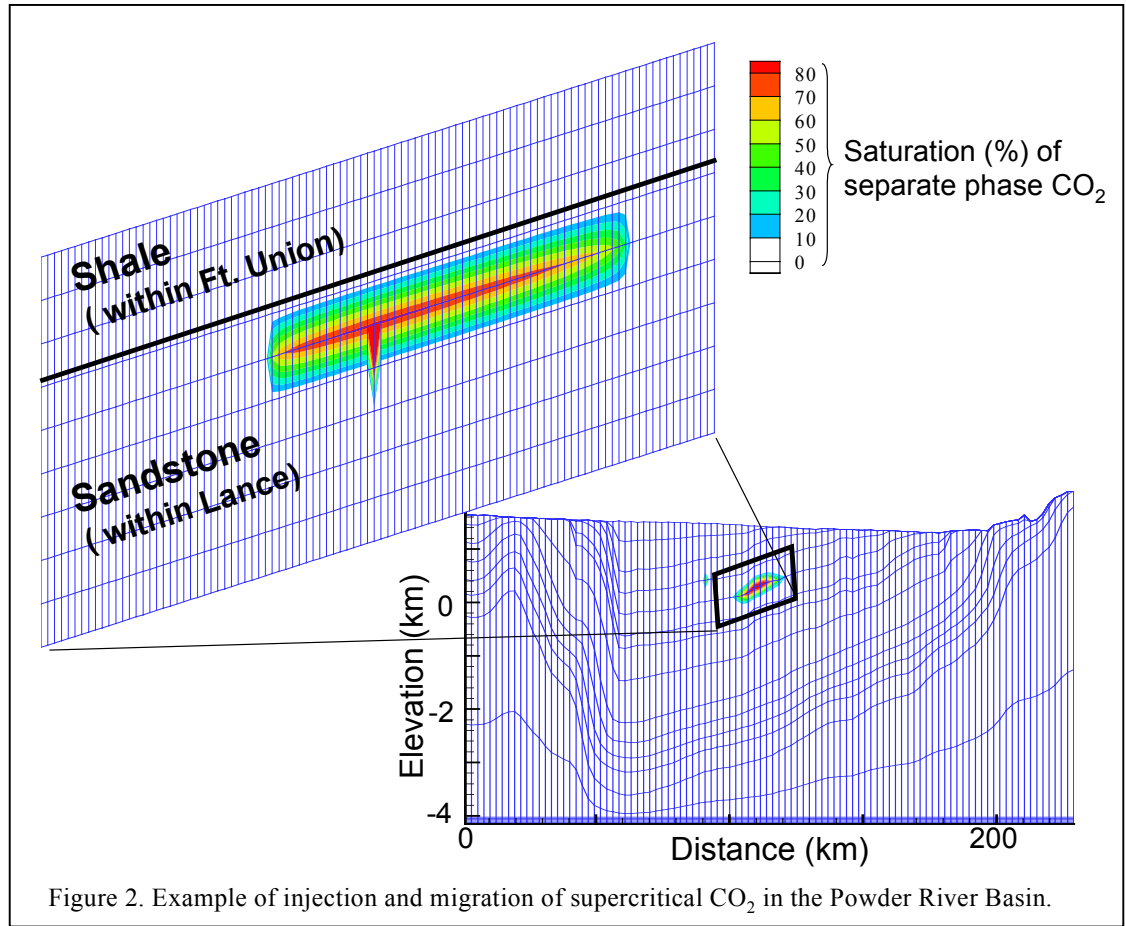


Figure 2. Example of injection and migration of supercritical CO₂ in the Powder River Basin.

rates through fractures, and possible partitioning of CO₂ between fracture and matrix. We investigated this issue using the dual permeability approach incorporated in the TOUGH2 code (Pruess, 1991).

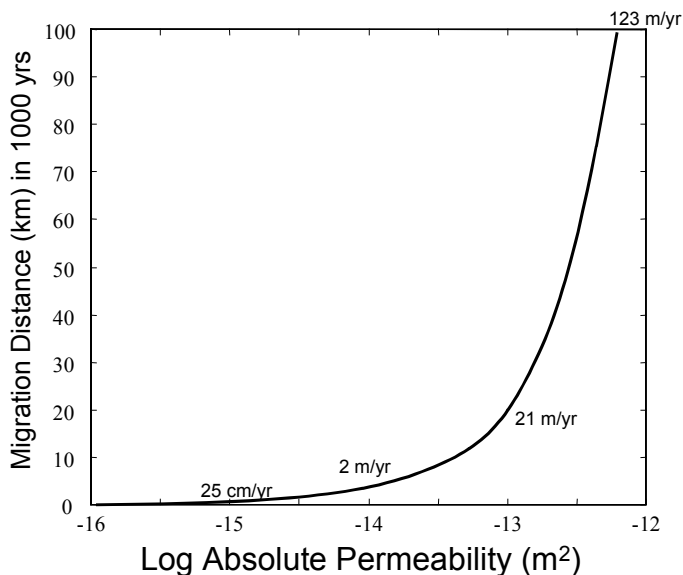


Figure 3. Migration rate of supercritical CO₂ as a function of permeability.

To simulate a fracture through a sealing layer, we assigned a vertical column of dual permeability cells within the sealing layer of the basin model, local to the injection area. Thus, high fluid pressures associated with injection (approaching 30 MPa, whereas ambient hydrostatic pressure is ~17 MPa) are the principal driving force of CO₂ through the vertical fracture. In this generic fracture model, the fracture volume is assigned to be 2% of the cell, fracture porosity is 1%, and permeability is 10⁻¹⁵ m² (~1 mD). The surrounding medium was assigned an order of magnitude lower permeability (10⁻¹⁶ m²). Results of this

simulation indicate that, in the absence of chemical reactions, most CO₂ will move through the fracture network per se (Figure 4). A migration rate of ~30 m/year through the fracture resulted for both dissolved and supercritical phases of CO₂. Plotting the ratio of mass of dissolved phase CO₂ to total mass CO₂ illustrates that the leading edge of the migrating CO₂ is primarily dissolved phase CO₂. No supercritical phase migrates into the rock matrix bordering the fracture walls, and the only significant penetration of dissolved phase CO₂ occurs very close to the lower fracture opening, where fluid pressure gradients are highest.

Unintended Impact: Brine Displacement

Some risks are associated with geological sequestration of CO₂. In addition to escape of CO₂ from sequestration aquifers into other aquifers or to the land surface, another environmental threat posed by subsurface sequestration is

contamination of sweet water aquifers by brines. We evaluated the potential for such unintended aquifer contamination by displacement of brines out of adjacent sealing layers such as marine shales. In the model simulation, we assigned a salinity of 300 g/l, or ~25% by weight brine saturation, (which is high compared to typical values of 10% or less), in the Lance shale unit above the Fox Hills sandstone (the CO₂ sequestration aquifer). We examined how migration of CO₂ into the Lance unit (not fractured in this simulation) from below displaced brine out of the Lance unit at its top.

Figure 5 illustrates that after 40 years of injection into the Fox Hills, high fluid pressures cause significant displacement of brine out of the top of the Lance unit. The movement of the brine front is summarized in Figure 6, illustrating that permeability of the sealing layer is the primary factor of concern, in addition to injection rates and associated high fluid pressures. A sensitivity analysis indicated that capillarity had a nominal effect on brine displacement.

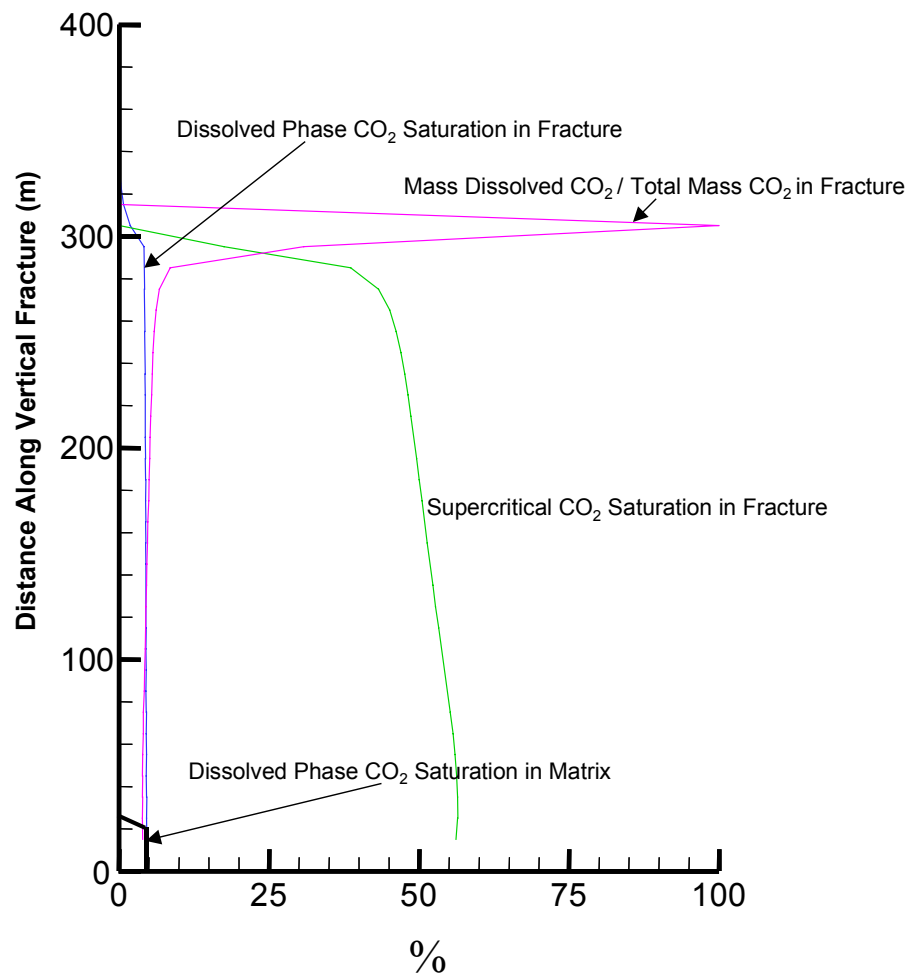


Figure 4. Distribution of CO₂ within fracture simulated using a dual permeability approach.

In sum, results suggest that sustained injection of CO₂ may incur wide-scale brine displacement out of adjacent sealing layers, depending on the injection history, initial brine composition, and hydrologic properties of both aquifers and seals.

While the results presented here are unique to the simplified model of the Powder River Basin used in this analysis, they may provide general implications for other basins of interest for CO₂ sequestration.

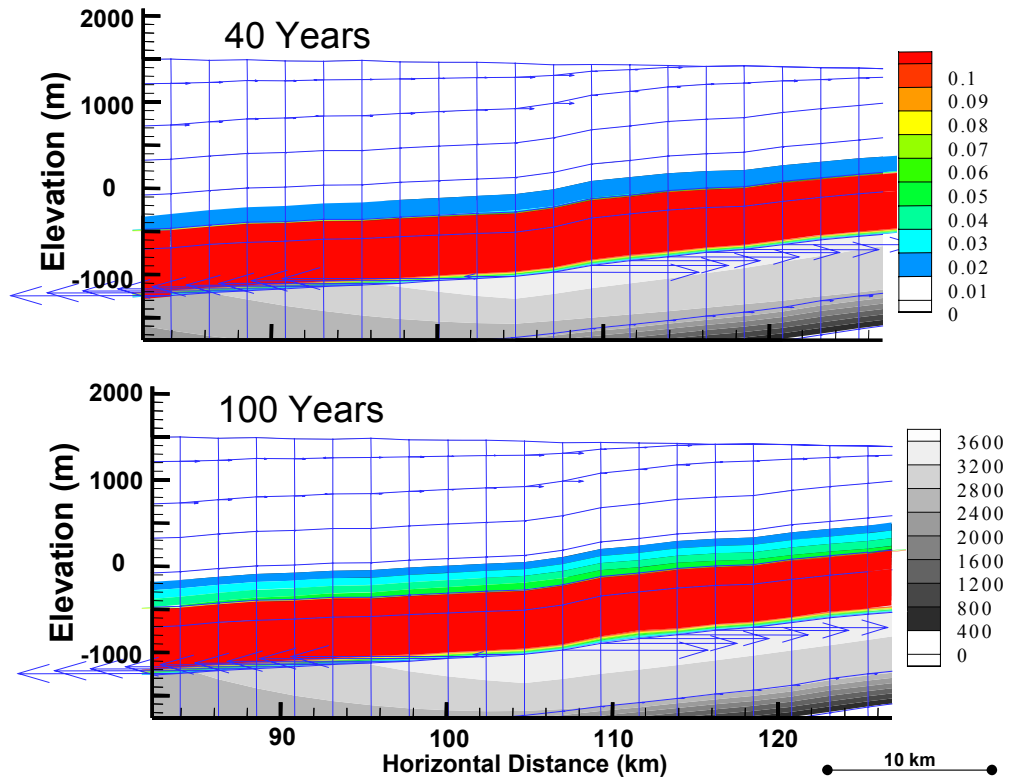
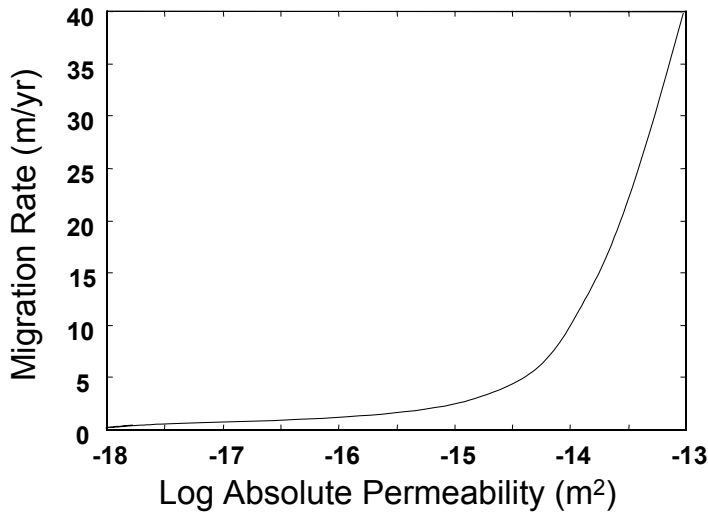


Figure 5. Simulation results illustrating displacement of brine out of a shale sealing layer as a consequence of CO₂ injection in aquifer below. Color contours are brine saturation in the sealing layer and aquifer above; grayscale contours are equivalent hydraulic head in the aquifer below. Arrows are flow vectors illustrating flow away from the injection well.

Future Activities

Current and future activities include laboratory experiments to characterize relevant chemical reactions associated with geological sequestration of CO₂. We are examining a limited number of reactions among saturated brines, supercritical CO₂ solutions, and the rock matrix at elevated pressures and temperatures. Simultaneously, we are analyzing brines and reaction products for a broad suite of minor and trace elements to survey for possible changes. We are particularly interested in these experiments in the stability of hydrated silicates and aluminosilicates that comprise reactive components of reservoir injection zones. These flow-through experiments will be used to characterize pore structure changes and general behavior of brine and CO₂ during alteration. Precipitation rates for magnesite will also be investigated.

Results of these experiments will be used to parameterize reactive transport model aspects using the computer code FLOTRAN (Lichtner, 2000), which we are coupling to our extended



TOUGH2 CO₂ flow and transport simulation model. Similar model simulations as described here, but with reactive transport aspects, will be conducted to evaluate possible effects on in situ permeability evolution associated with geological sequestration of CO₂, including mineral trapping

Figure 6. Rate of brine front movement as a function of matrix permeability.

References

- Kerrick, D. M., and G. K. Jacobs, A modified Redlich–Kwong equation for H₂O, CO₂, and H₂O-CO₂ mixtures, *Amer. J. Sci.*, v. 281, p. 735–767, 1981.
- Lichtner, P.C., FLOTRAN: User's Manual, Los Alamos National Laboratory Report, 2000.
- McPherson, B.J.O.L. and Chapman, D.S., 1996, Thermal Analysis of the Southern Powder River Basin, Wyoming, *Geophysics*, v. 61, p. 1689-1701, 1996.
- McPherson, B. J. O. L., and Cole, B. S., Multiphase CO₂ Flow, Transport and Sequestration in the Powder River Basin, Wyoming, U.S.A., *Journal of Geochemical Exploration*, v. 69-70, p. 65-69, 2000.
- Pruess, K., TOUGH2-A general-purpose numerical simulator for multiphase fluid and heat flow, *Rep. LBL - 29400*, Lawrence Berkeley Lab., September 1991.
- Weir, G. J., S. P. White, and W. M. Kissling, Reservoir storage and containment of greenhouse gases, *Transport in Porous Media*, v. 23, p. 37–60, 1996.