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Buoyancy effects on upward brine displacement caused by CO_2 injection

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Abstract

Upward displacement of brine from deep reservoirs driven by pressure increases resulting from CO_2 injection for geologic carbon sequestration may occur through improperly sealed abandoned wells, through permeable faults, or through permeable channels between pinch-outs of shale formations. The concern about upward brine flow is that, upon intrusion into aquifers containing groundwater resources, the brine may degrade groundwater. Because both salinity and temperature increase with depth in sedimentary basins, upward displacement of brine involves lifting fluid that is saline but also warm into shallower regions that contain fresher, cooler water. We have carried out dynamic simulations using TOUGH2/EOS7 of upward displacement of warm, salty water into cooler, fresher aquifers in a highly idealized two-dimensional model consisting of a vertical conduit (representing a well or permeable fault) connecting a deep and a shallow reservoir. Our simulations show that for small pressure increases and/or high-salinity-gradient cases, brine is pushed up the conduit to a new static steady-state equilibrium. On the other hand, if the pressure rise is large enough that brine is pushed up the conduit and into the overlying upper aquifer, flow may be sustained if the dense brine is allowed to spread laterally. In this scenario, dense brine only contacts the lower-most region of the upper aquifer. In a hypothetical case in which strong cooling of the dense brine occurs in the upper reservoir, the brine becomes sufficiently dense that it flows back down into the deeper

reservoir from where it came. The brine then heats again in the lower aquifer and moves back up the conduit to repeat the cycle. Parameter studies delineate steady-state (static) and oscillatory solutions and reveal the character and period of oscillatory solutions. Such oscillatory solutions are mostly a curiosity rather than an expected natural phenomenon because in nature the geothermal gradient prevents the cooling in the upper aquifer that occurs in the model. The expected effect of upward brine displacement is either establishment of a new hydrostatic equilibrium or sustained upward flux into the bottom-most region of the upper aquifer.

Introduction

The current global dependence on fossil fuels for over 80% of mankind's primary energy supply (IEA, 2010) is causing atmospheric CO_2 concentrations to rise resulting in climate change (IPCC, 2007a). To avoid the most serious effects of climate change, measures need to be taken now to reduce net CO_2 emissions by at least one-half (IPCC, 2007b). A combination of approaches has the best chance of reducing CO_2 emissions at the scale and rate needed to avoid the most serious effects of climate change (Pacala and Socolow, 2004). Of the six most feasible options proposed by Pacala and Socolow (2004), capture of CO_2 from fossil-fuel power plants and other stationary industrial sources with Geologic Carbon Sequestration (GCS) is the only option that permits a bridging from current carbon-based energy sources to an energy-supply future that uses only low-carbon energy sources. The current concept for large-scale GCS is the direct injection of supercritical CO_2 into deep geologic formations which typically contain brine (IPCC, 2005).

In order to inject CO_2 into deep brine-filled aquifers, over-pressure must be applied to drive the CO_2 into the formation and displace the brine outward to accommodate the injected CO_2. By this process, injection of CO_2 causes pressure increases in the brine formations (e.g., Nicot, 2008; Zhou et al., 2010; Birkholzer and Zhou, 2009, Birkholzer et al., 2009). Because brine and the porous matrix are not very compressible, pressure will propagate rapidly to large distances away from the injection well (e.g., Zhou et al., 2008). The resulting pressure gradients provide a driving force for brine flow, which may be upwards if there are vertical conduits. Potential conduits for this upward flow could be (1) an open well, (2) poorly cemented well annulus, (3) permeable faults or fracture zones, etc. Although such conduits are not expected to be present, this paper addresses the question of the dynamics of upward brine flows assuming a conduit exists. We note that the conduit does not

3

convey CO_2 upward as the injected CO_2 may be many kilometers away, but rather it conveys over-pressured brine that results from CO_2 injection some distance away.

The issue of brine upflow into groundwater resources is critically important for environmental risk assessment of GCS. The context for upward brine flow in GCS systems involves several key features that make the process interesting:

1) Pressures in deep systems are nearly hydrostatic, so small over- or under-pressures can cause brine flow provided there is sufficient permeability.
2) Brine flow can be continuous and steady, or it can be short-lived and end with establishment of a new hydrostatic equilibrium, or it can be transient or oscillatory.
3) Brine flow is affected not only by pressurization or depressurization, but also by buoyancy of the brine relative to surrounding groundwater.
4) Buoyancy of the brine is controlled by both compositional (salinity) and thermal effects.
5) Thermal and salinity effects operate very differently in porous media due to thermal conduction into solid grains of the matrix, resulting in the process known as thermal retardation.
6) Diffusion of heat is much faster than diffusion of dissolved salt in brine.
7) Salinity generally increases with depth as does temperature, tending to compensate each other with respect to fluid density.

In this paper, we illustrate some of the features above through numerical modeling of the dynamics of upward brine flow. In so doing, we illuminate key parameters that control upward brine flow allowing an estimate of behavior of various systems based on their site-specific parameter values. The model system is highly idealized to focus attention on the thermal and solutal effects.

The conceptual model for the system considered here is shown in Figure 1, which shows a conduit assumed to be a leaky well. In fact, the system we model is more generic and models upward flow through a 5 m-wide conduit with the same permeability as the reservoir rocks. As such, the model is not site-specific nor is it representative of any actual GCS site. Instead the purpose of the modeling presented here is to demonstrate concepts of the establishment of a new hydrostatic equilibrium upon reservoir pressurization, sustained flow, and oscillatory dynamic solutions in the brine up flow

system. The use of an idealized model is made purely as an end-member case to examine the

potentially interesting dynamics that could arise in some special circumstances. Although it is

necessary to understand what might happen if brine upflow occurs, in general, highly conductive

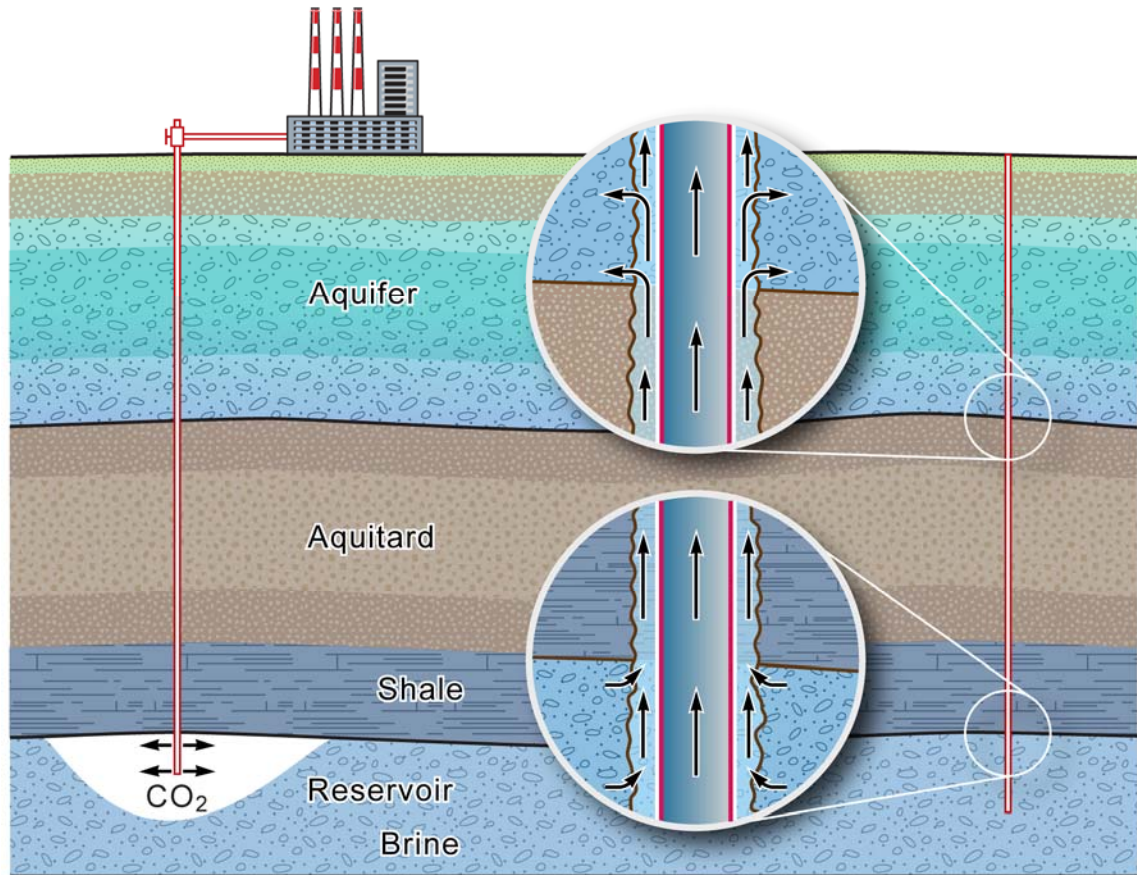features such as those assumed here will not be present in any GCS system.

Figure 1. Conceptualization of CO_2 injection causing pressure rise that leads to brine up flow in an abandoned well.

Prior Work

The structure of deep sedimentary basins and associated flow of groundwater has been studied in the

context of understanding deep hydrologic systems (e.g., Bethke, 1985; Gupta and Bair, 1997),

geothermal resources (Bachu and Burwash, 1991), ore deposits (Garven and Freeze, 1984; Bethke,

1986), oil migration (e.g., Hindle, 1997), and GCS (Gunter et al., 2004). In the case of GCS, upward migration of CO_2 has been studied extensively through various combinations of overburden and conductive and non-conductive features by analytical (e.g., Silin et al., 2009; Hayek et al., 2009) and numerical approaches (e.g., Pruess, 2008; Doughty and Myer, 2009). The large potential for upward vertical flow (i.e., leakage) through fast paths such as abandoned wells was pointed out by Nordbotten et al. (2004) who also provided semi-analytical approaches to estimating the magnitude of such leakage. While much of the early focus of potential impacts of GCS was on upward CO_2 leakage into groundwater resources, more recently concern is being focused on upward brine displacement and associated degradation of groundwater. One reason for this interest is that pressure increases due to injection can extend tens of km or more, making it difficult to characterize the area thoroughly enough to guarantee absence of conduits capable of conveying brine upward. The hazard is that up-flowing deep brines will increase Total Dissolved Solids (TDS) and potentially other components such as heavy metals (e.g., lead, arsenic, etc.) in shallower aquifers potentially leading to degradation of groundwater quality (Zheng et al., 2009; Apps et al., 2010).

The overall topic of well-bore integrity relative to upward leakage is an area of ongoing research critical to GCS safety and effectiveness (e.g., Gasda et al., 2004; Carey et al., 2007). The potential for brine flowing upward in wells was analyzed by Nicot et al. (2009a; 2009b) who considered density effects and drilling mud in the wellbore in a hydrostatic context as possible mitigating features for upward brine flow. While the properties of faults remain even more enigmatic than that of well bores, recent efforts have used percolation theory and fuzzy rules to estimate connectivity of fault networks that could give rise to a connected flow path if the individual faults are conductive (Zhang et al, 2010).

When deep brine flows upward into cooler regions along the geothermal gradient, both thermal and solutal effects of buoyancy come into play. The advection of heat and salt by flowing brine in viscous liquid systems (e.g., Turner, 1973) is very different from the flow of heat and salt in porous media (e.g., Nield, 1968). Specifically, solute is transported at the pore velocity in porous media whereas heat is transported approximately at the Darcy velocity due to thermal conduction into the solid grains of the matrix (Phillips, 1991). This interesting behavior gives rise to plume separation (Oldenburg and Pruess, 1999), that is, the solute plume tends to advance ahead the thermal plume when a parcel of hot, salty water moves through cooler, fresher porous media, a phenomenon that will be observed in some of the results presented below.

Finally by analogy to viscous liquid systems (no porous media) such as the oceans, we note that researchers have identified the different kinds of behavior that can be expected depending on the stratification of heat and salt. Shown in Figure 2 are two conceptual depictions of configurations borrowed from early oceanographic work in this area (e.g., see reviews in Turner, 1973) that lead to very different forms of natural convection. Specifically, as a thought experiment consider the situation when cold, fresh water is displaced upward in a hypothetical flexible pipe that exchanges heat but not salt with its surroundings. In this case, the flow is unstable and accelerates upwards as seen in Figure 2a. In contrast, warm, salty water displaced upward in the hypothetical pipe tends to return to its original location as it cools but remains salty at higher levels in the system. These behaviors were observed by Stommel and Fedorov (1967) and related topics were covered in depth by Turner (1973). One interesting feature of these thought experiments is that because thermal diffusivity is much greater than solutal diffusivity (giving rise to so-called double-diffusive convection), the pipe does not have to be present for these fingering or oscillatory convective patterns to develop. We mention these previously recognized aspects of double-diffusive convection

in viscous liquid systems simply to point out the importance of multiple sources of buoyancy and the

interesting convective phenomena that they can cause in viscous liquid and in porous media systems

(e.g., layering, as described in Oldenburg and Pruess, 1998). In the geologic carbon sequestration

case, warm, salty water can potentially be pushed upwards by CO_2 injection into cooler, fresher
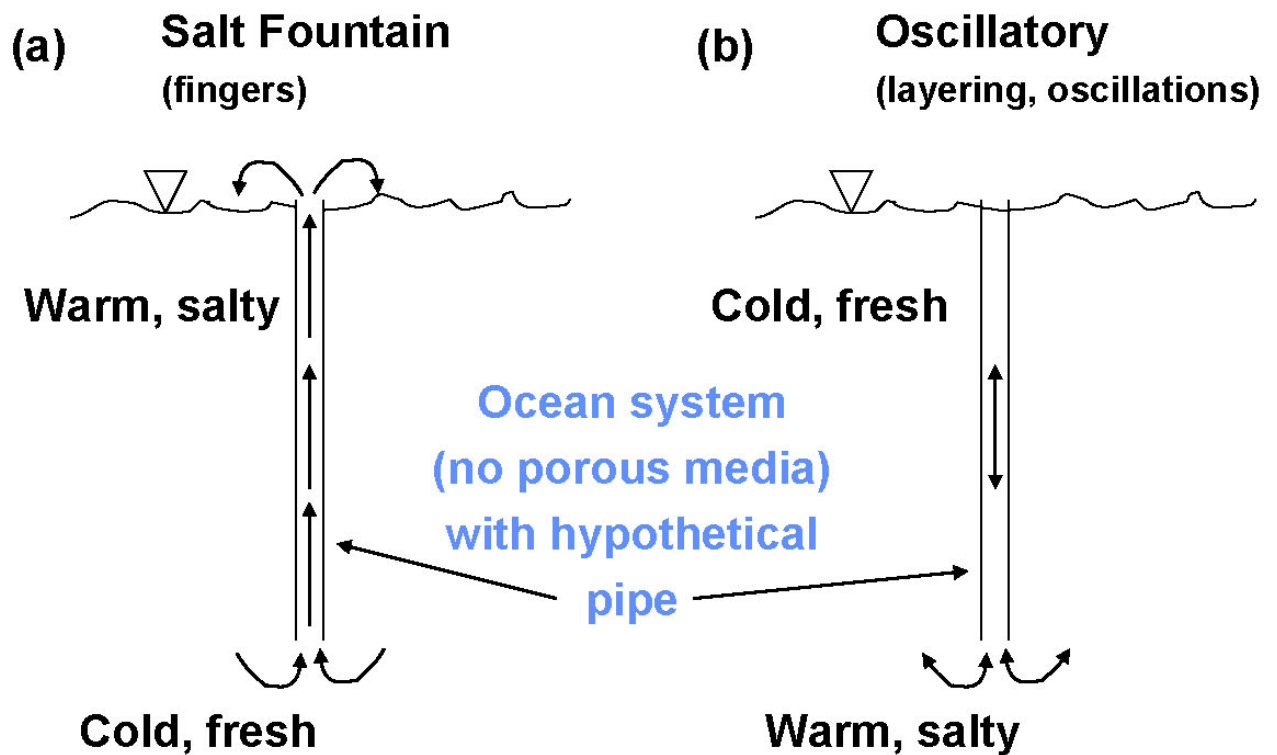
aquifers as will be discussed in detail below.

Figure 2. Conceptual depictions of double-diffusive convection in the finger regime (a) and the oscillatory regime (b). Note that differences in diffusivity between heat and salt mean that the pipe is not necessary to develop fingers (a) and layered or oscillatory convection (b).

Methods

We have simulated the upward single-phase flow of deep brine into a shallower aquifer using the

non-isothermal reservoir simulation model TOUGH2 with the EOS7 equation-of-state module

(Pruess et al., 1999). The equations solved in TOUGH2/EOS7 for single-phase conditions and three

components (water, brine, and air) are shown in Table 1 (symbols are defined in Nomenclature).

Briefly, we solve the advection-diffusion equation using implicit time-stepping with a fully-coupled

residual-based convergence criterion that can handle strong density contrasts (e.g., Oldenburg and

Pruess, 1995).

Table 1. Governing equations solved in TOUGH2/EOS7 for single-phase non-isothermal cases.

Description	Equation
Conservation of mass and energy	$\dfrac{d}{dt}\displaystyle\int_{V_n} M^K dV = \int_{\Gamma_n} \mathbf{F}^K \cdot \mathbf{n}\, d\Gamma + \int_{V_n} q_v{}^K dV$
Mass accumulation	$M^K = \phi\, \rho_l X_l{}^K,\; K = 1, NK$
Thermal energy accumulation	$M^{NK+1} = (1-\phi)\rho_R C_R T + \phi \rho_l u_l$
Phase flux	$\mathbf{F}_l = -k\dfrac{\rho_l}{\mu_l}(\nabla P_l - \rho_l \mathbf{g})$
Component flux	$\mathbf{F}^K = X_l{}^K \mathbf{F}_l - \phi\tau_o \rho_l d_l{}^K \nabla X_l{}^K,\; K = 1, NK$
Molecular diffusion	$f_l{}^K = -\phi\tau_o \rho_l d_l{}^K \nabla X_l{}^K$
Thermal energy flux	$\mathbf{F}^{NK+1} = -\lambda\nabla T + h_l \mathbf{F}_l$

Table 2. Properties used in the brine upflow model.

	Lower and Upper Aquifers and Conduit
Temperature (T)	Variable (geothermal gradient)
Porosity (ϕ)	0.30
Permeability (k)	Variable (1 x 10^{-11}, 1 x 10^{-12}, 1 x 10^{-13} m^2)
Thermal conductivity (λ)	2.51 W m^{-1} K^{-1}
Molec. diffusivity coefficients ($d_\beta{}^K$)	Liquid: 10^{-10} m^2 s^{-1}
Tortuosity (τ_0)	1.0

The brine component in EOS7 is defined as a concentrated NaCl brine, a more convenient choice

than solid NaCl and water components because the brine and water volumes are approximately

linearly additive whereas NaCl and water volumes are not linearly additive. The density of pure

water as a function of temperature is calculated using the results of the IFC (1967). Under natural

conditions, temperature and salinity both tend to increase with depth, with opposite impacts on

density. The net effect is that density of brine increases with depth. Given that ρ_{H2O}(1 bar, 15 ^{o}C) =

999 kg m^{-3}, and the densest brine considered is 1150 kg m^{-3} which is assumed to correspond to a

brine with 25% salinity, we approximate salinity for any reference brine density by the formula

$$S = \frac{(\rho_{ref\,brine} - 999)}{(1150 - 999)} 0.25 \qquad (1)$$

Which gives salinities of 0.25, 0.17, and 0.08 for the reference brines of density 1150, 1100, and

1050 kg m^{-3}, respectively. The density as calculated in EOS7 of brine-water mixtures with

maximum density of 1150 kg m^{-3} are shown in Figure 3 as a function of temperature and salinity.
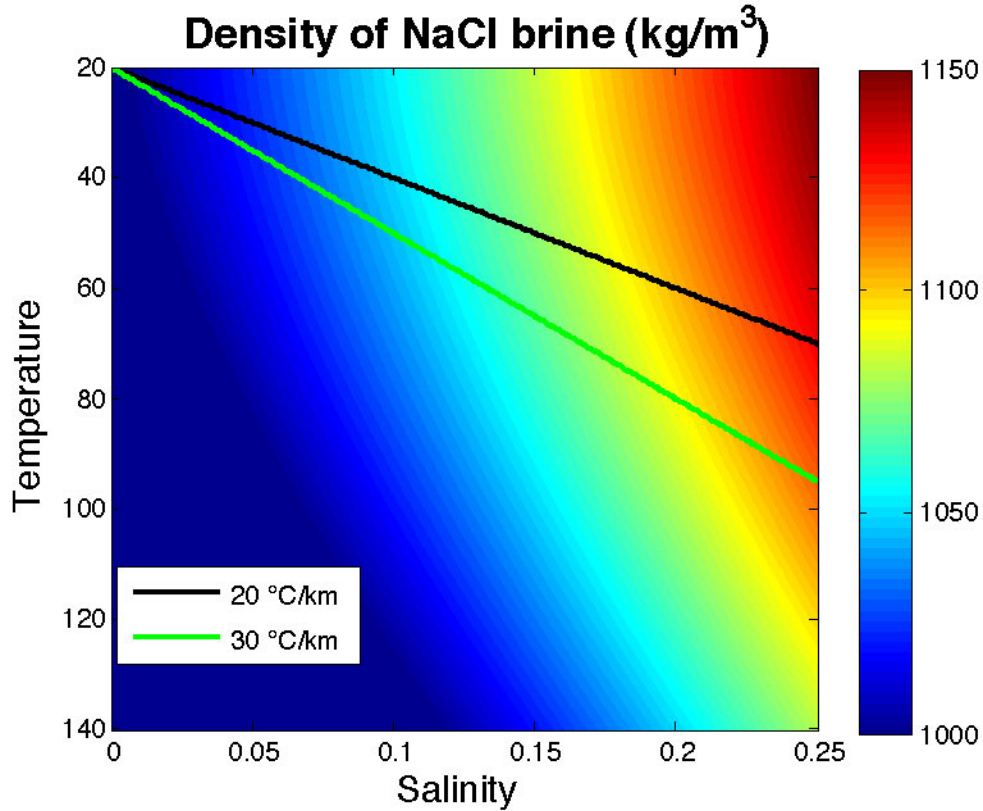
Figure 3. Variation of density of NaCl brine as a function of temperature and salinity as calculated in TOUGH2/EOS7 with some typical geothermal gradients shown assuming zero salinity at the ground surface and salinity of 0.25 at a depth of 2.5 km.

The conceptual model of the multilayered hydrologic system with GCS underway was shown in

Figure 1. The sub-system that we consider is taken to be far away (laterally displaced) from the CO_2

injection well and addresses only the upward flow of brine (no CO_2 present) through a conduit such

as a well that cuts through a thick aquitard into an upper shallow aquifer. The model system consists

of a lower reservoir (brine formation) with closed boundaries and a grid block at the lower left-hand

corner at which the pressure is raised impulsively to represent CO_2 injection at a large distance away

from the conduit. The boundary conditions at the top of the system are open (constant $T = 20\ °C$, 1

bar) and the sides of the system are closed (no flow). The system was discretized as shown in Figure

4 with a 5 m-wide conduit. The choice of closed boundaries on the lower reservoir was made to

ensure a significant pressure rise. As for the upper aquifer, the closed side boundaries represent a system with lateral compartmentalization. We also ran some cases with open side boundaries in the upper reservoir as discussed below.
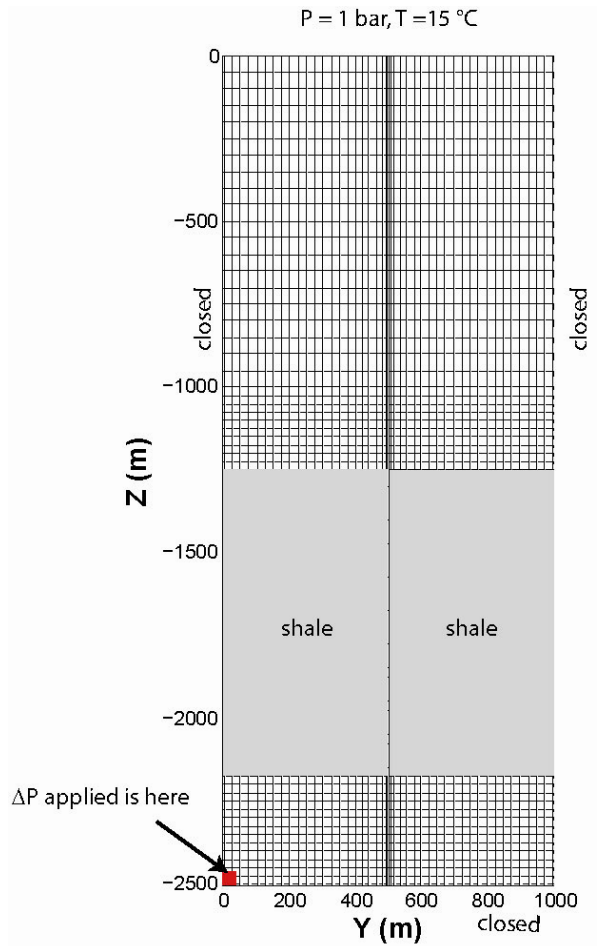

Figure 4. Idealized model system showing boundary conditions and discretization.

Results

Overview

Before showing the details of the results, we summarize the overall behavior of the system. First, the initial condition consists of hydrostatic brine linearly stratified in salinity and temperature from the top to the bottom. The overpressure applied at the lower left-hand corner of the system

12

propagates rapidly through the lower reservoir and serves to drive warm, salty brine up the conduit. If the brine density is high enough or the overpressure low enough, the warm brine moves only part way up the conduit and finds a new hydrostatic equilibrium.

If the brine density is low or the overpressure is high, the warm brine flows all the way up the conduit and into the upper aquifer. With closed boundaries 500 m away on each side of the well in the model, the brine cannot flow laterally for very long, and ponds in the upper aquifer increasing hydrostatic pressure and eventually finding a new hydrostatic equilibrium. If the lateral boundaries are open or the upper aquifer lateral extent were very large, brine exiting the conduit into the upper aquifer would flow laterally along the lower-most regions of the upper aquifer as controlled by buoyancy; this flow can be sustained for as long as the lateral flow occurs. In the hypothetical case that brine ponds in the upper aquifer and cools, the brine can become dense enough to overcome the original overpressure of the lower reservoir that drove it up the conduit. The result is that the brine is sufficiently dense to flow back down the conduit. Once in the lower reservoir, it heats up again and the cycle continues. This is the essential mechanism of the oscillatory solutions to be discussed below.

Base Case

We present first the details of the simulation result for the case of the highest salinity brine we considered, approximately 25% salinity (sea water has a salinity of approximately 3.5%) and a geothermal gradient of 30 °C/km. Shown in the Figure 5 are temperature, salinity, and brine density in contour form and profiles along the conduit after 500 yr of a pressure perturbation of 0.2 MPa and permeability (k) equal to 1×10^{-12} m^2. As shown, the resulting system is nearly static with the dense brine from the lower aquifer positioned part way up the conduit. Note the temperature field remains

equal to the initial geothermal gradient while the salinity becomes higher in the conduit. This is due to the well-known effect of thermal retardation during the initial upward displacement whereby the solute front moves out ahead of the thermal front in the porous medium (e.g., Oldenburg and Pruess, 1999) and then the solute anomaly persists for a very long time as controlled by the slow diffusion of salinity. This effect is reflected in the density profile which shows fluid density in the conduit reflecting anomalous brine concentration.
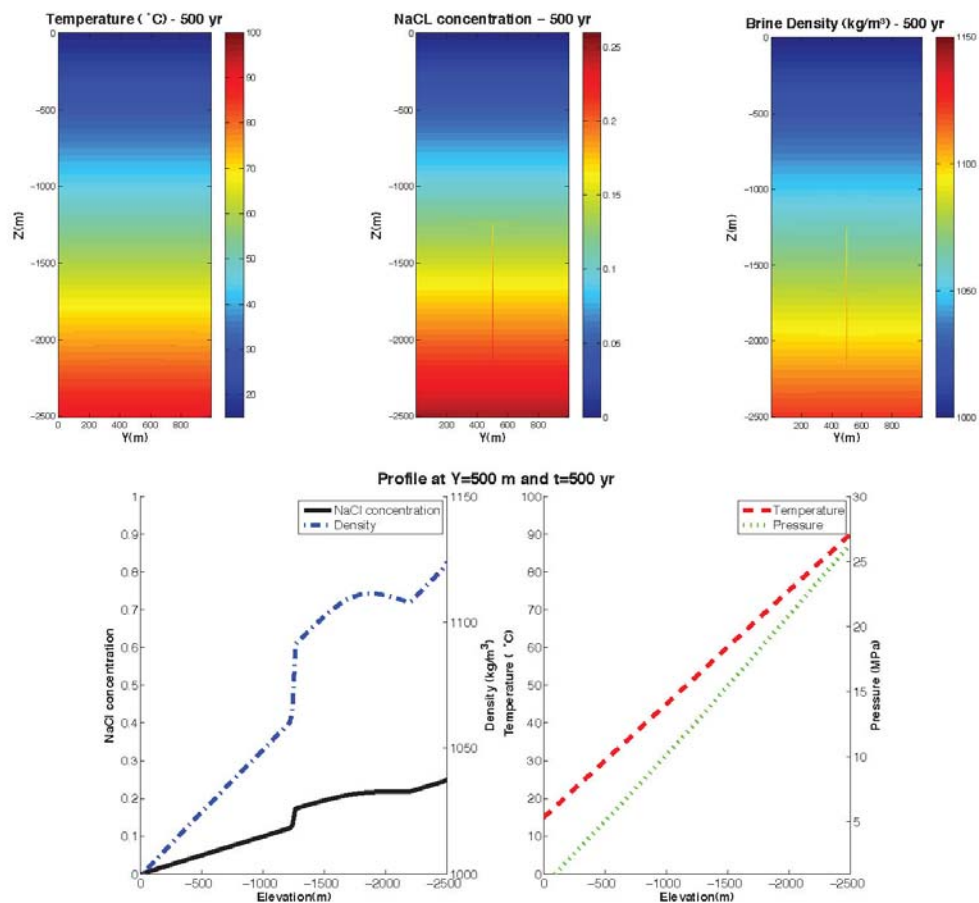
Figure 5. High-salinity brine is displaced up into the conduit where it is held in a static equilibrium due to pressure rise of 0.2 MPa.

In Figure 6 we show details of results for the case of brine salinity equal to 17% and pressure perturbation 0.2 MPa for $k = 1 \times 10^{-12}$ m². As shown, in this case the 0.2 MPa pressure rise is able to

14

push the warm brine up the conduit and into the lower part of the upper aquifer where it spreads out at $t = 500$ yrs. This spreading will occur in this system until the brine ponds to a depth sufficient to match the pressure perturbation supporting the column of brine in the conduit. If the model domain were laterally much larger in the upper aquifer, the upward flow could be sustained for a long time with dense brine underplating the upper aquifer as controlled by buoyancy. Note the vertical profile of density shows an inversion in the conduit resulting from nearly uniform salinity during upflow with geothermal-gradient-controlled temperature. This profile shows the system is still evolving at $t = 500$ yrs.
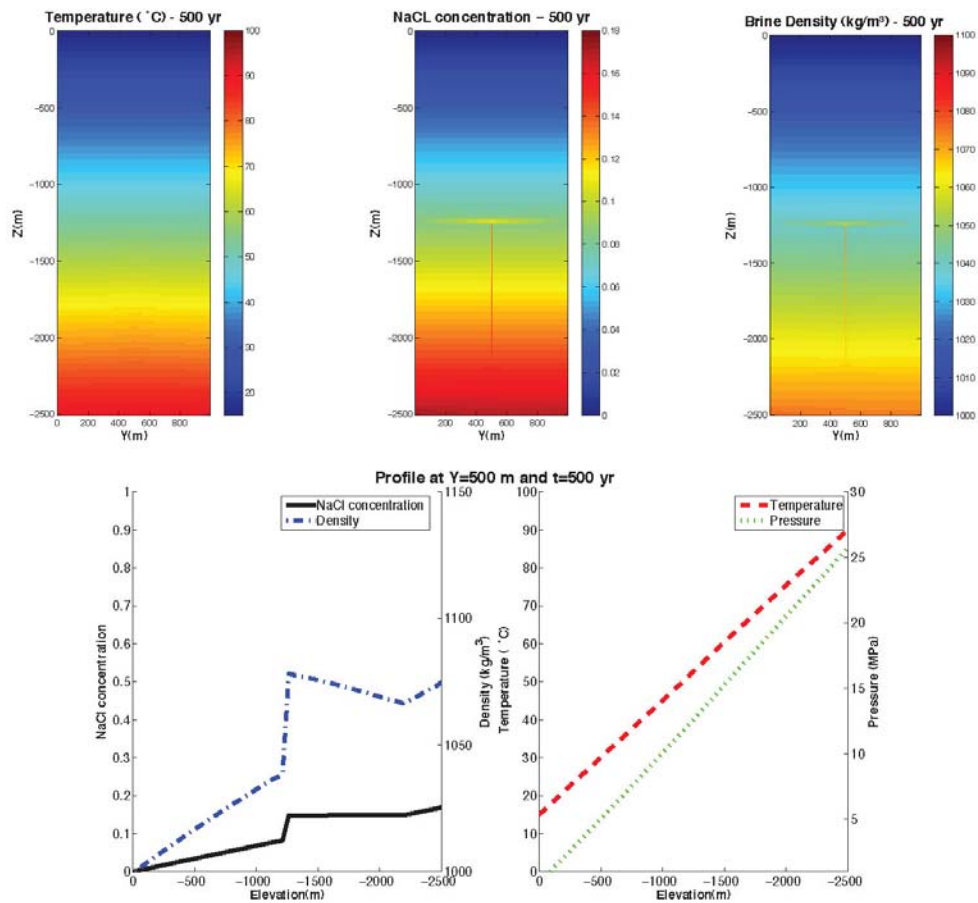
Figure 6. Medium-salinity brine flowing up the conduit due to pressure rise of 0.2 MPa.

The case with all properties the same as in Figures 5 and 6 except the maximum salinity is 8% is shown in Figure 7. Here the brine up flow into the upper aquifer is correspondingly stronger, and the system is dynamic at $t = 500$ yrs as brine underplates the upper aquifer. As in Figure 6, the salinity in the conduit as shown in Figure 7 is relatively uniform resulting in a density inversion.
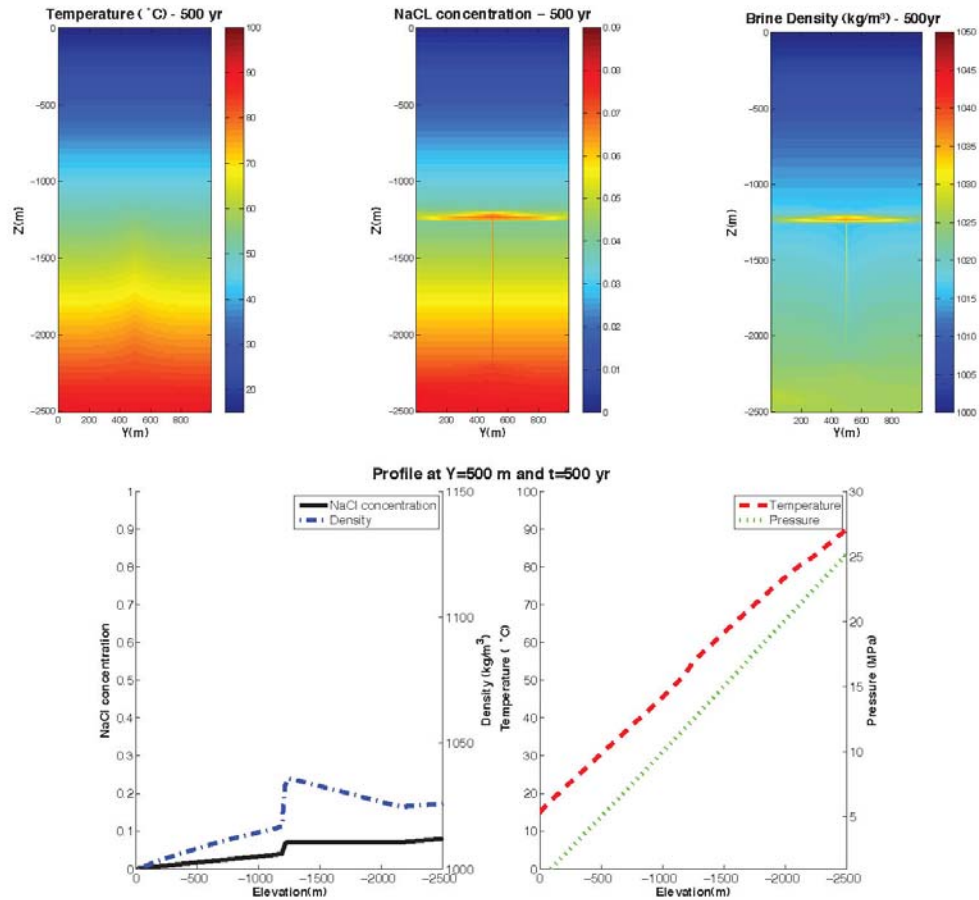

Figure 7. Low-salinity brine flowing up the conduit due to pressure rise of 0.2 MPa.

Oscillatory Solutions

In the interest of examining all possible situations, we study next the consequences of a hypothetical situation not generally expected in sedimentary basin systems but potentially relevant in systems with varying geothermal gradients with depth. The situation is illustrated in Figure 8 which shows

16

details of results for a system in which the conduit is thermally insulated from the shale, an effect achieved by removing the shale layer from the simulation, causing warm brine to arrive at the upper aquifer. In this case, the simulation shows a very strong density inversion in the vertical profile along the conduit. This arises from the hot brine entering the upper aquifer and undergoing significant cooling. During this cooling process, the density increases strongly, and can become so large that the fluid overcomes the over-pressure that drove it upward to the upper aquifer. The result is that the brine begins to move back down the conduit and into the lower aquifer as it cools. Once in the lower aquifer, the brine heats up again and rises up the conduit. This oscillatory behavior appears to go on indefinitely with time scales of oscillation on the order of thousands of years but dependent on system properties.

We show in Figure 9 some oscillatory behavior by plotting the height of the brine layer in the upper aquifer versus time for various combinations of parameters and $k = 1 \times 10^{-12}$ m^2. Note the long time scales for this oscillation. Note also that this is a hypothetical situation in which no heat transfer occurs from the fluid to the walls of the conduit, and in which the geothermal gradient in the upper aquifer is not maintained but rather is controlled by fluid up- and down-flow and the top boundary condition. Nevertheless, we believe this behavior is worth noting and may be relevant in some situations with anomalous geothermal gradients.
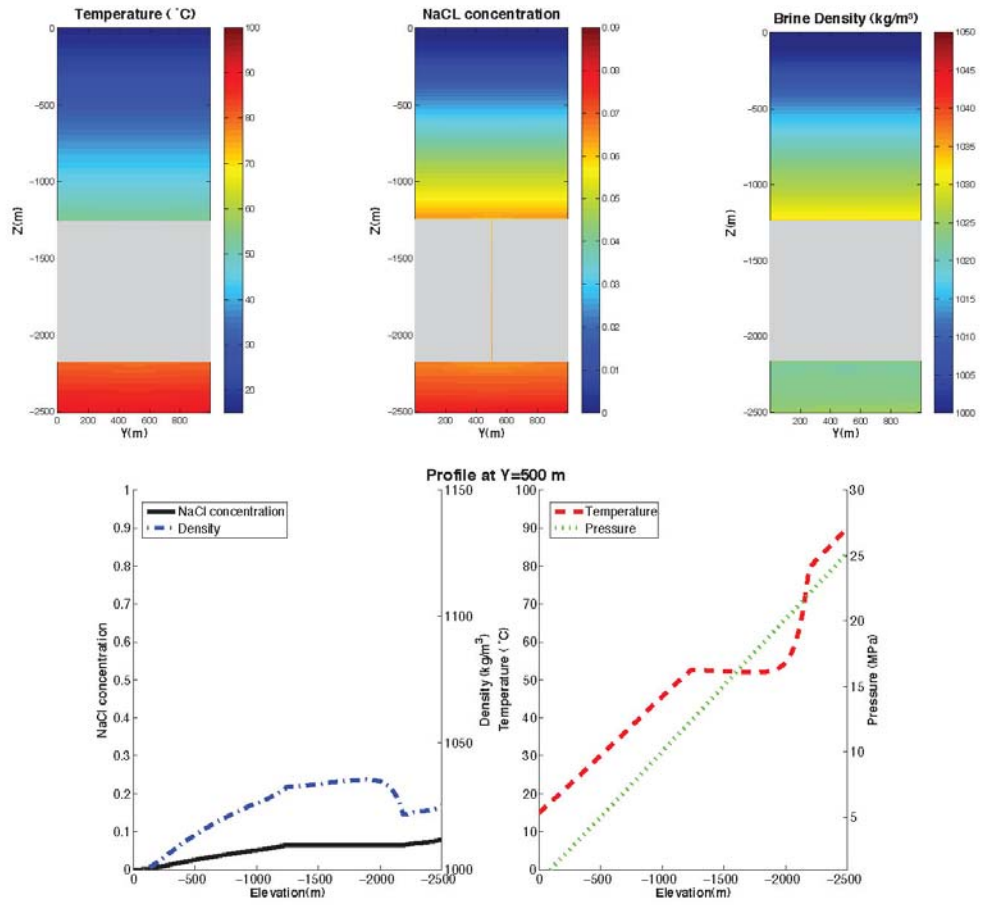
Figure 8. Dynamic change of height of brine intrusion with time showing oscillatory behavior.
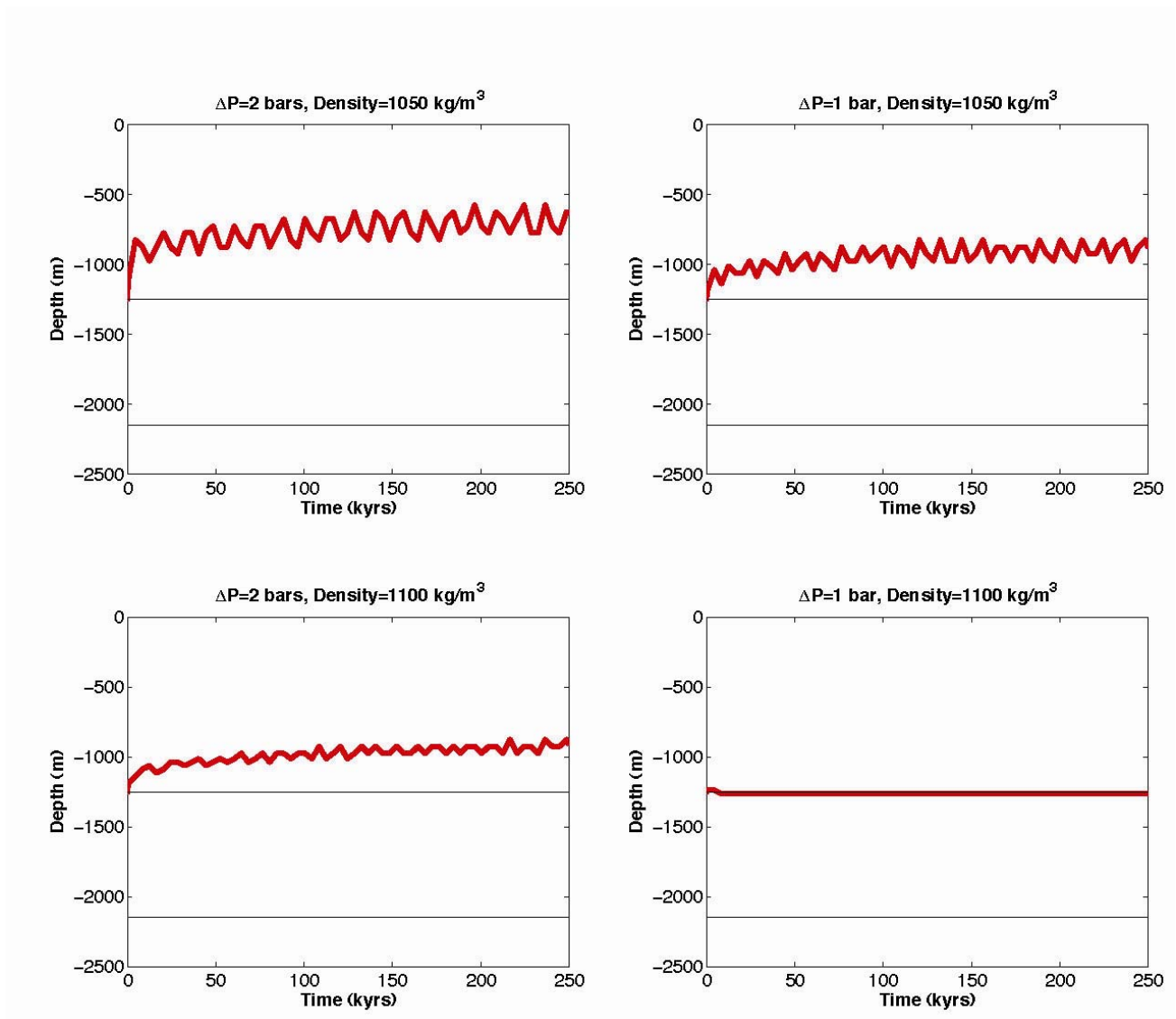
Figure 9. Oscillatory and static solutions shown by the height of the brine layer in the upper aquifer as a function of time.

Parameter Study of Oscillatory and Static Solutions

For the case in which no heat transfer occurs between fluid and conduit, and the geothermal gradient in the upper aquifer is controlled by fluid flow, we mapped out oscillatory and steady static solutions for 27 combinations of the parameters including (1) pressure perturbation (ΔP), (2) salinity as given by brine density at standard conditions, and (3) permeability. Values used were $\Delta P = 0.03, 0.1,$ and

0.2 MPa, ρ_{brine} = 1050, 1100, and 1150 kg m^{-3}, and k = 10^{-13}, 10^{-12}, and 10^{-11} m^2. We present in

Figure 10 results showing that oscillatory solutions tend to occur for large ΔP, small salinity, and

large permeability. Again we emphasize that this mapping of parameter space corresponds to

hypothetical conditions of heat transfer that are not likely to occur, but that are consistent with the

trend that static steady states are the expected result of weak upflow of dense brine in low

permeability conduits.
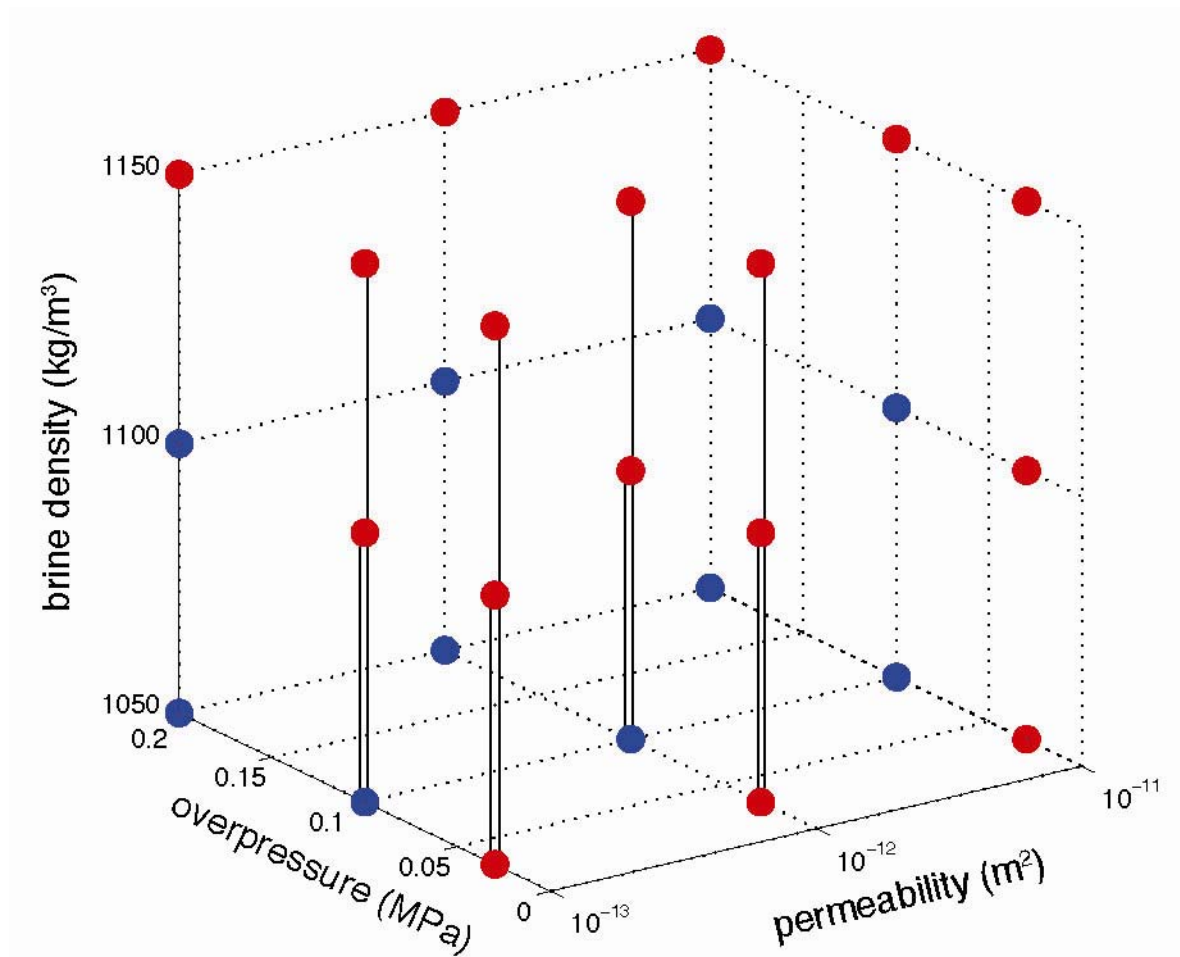
Figure 10. Regions of steady-state static (red dots) and oscillatory (blue dots) solutions for the
upward brine displacement as a function of pressure rise, salinity, and permeability.

Conclusions

The injection of large quantities of CO_2 for geologic carbon sequestration will increase pressure in brine formations and tend to displace brine upwards if high-permeability conduits (e.g., permeable fracture zones or faults) are present. The extent of upward flow of warm brine in the conduit will be affected by overpressure and the density of the upward-displaced fluid. Fluid density in saline systems is controlled by both temperature and salinity. Because of the large difference in thermal diffusivity relative to salt diffusivity, porous media double-diffusive convective effects may arise. In addition, the porous medium alone tends to cause plume separation due to thermal retardation. For high salinity or low over-pressure, brine tends to move up a vertical conduit until it finds a new hydrostatic equilibrium. For low salinity or high overpressure, brine tends to move up the conduit and out into the upper aquifer. In the hypothetical case that the conduit has very low thermal conductivity, warm brine may move up the conduit into an upper aquifer where it will cool. Once the brine cools, its density may be sufficient to overcome the over-pressure and cause brine to flow back down the conduit. Once in the warm lower aquifer, heating will occur again and the cycle may repeat itself. Our simulations show the existence of oscillatory solutions in which this cycle repeats itself at least for hundreds of thousands of years.

Acknowledgement

Nomenclature

C_R	heat capacity of the formation	J kg^{-1} K^{-1}
d	molecular diffusivity	m^2 s^{-1}

g	acceleration of gravity vector	m s^{-2}
F	Darcy flux vector	kg m^2 s^{-1}
h	enthalpy	J kg^{-1}
k	permeability	m^2
M	mass accumulation term	kg m^{-3}
n	outward unit normal vector	
NK	number of components	
P	total pressure	Pa
q_v	volumetric source term	kg m^{-3} s^{-1}
S	salinity	fraction
t	time	s
T	temperature	$^{\mathrm{o}}$C
u	internal energy	J kg^{-1}
V	volume	m^3
X	mass fraction	
Y	Y-coordinate	
Z	Z-coordinate (positive upward)	

Greek symbols

Γ	surface area	m^2
K	mass components (superscript)	
λ	thermal conductivity	J s^{-1} m^{-1} K^{-1}
μ	dynamic viscosity	kg m^{-1} s^{-1}
ρ	density	kg m^{-3}
τ_o	reference tortuosity	
ϕ	porosity	

Subscripts
l liquid
R rock (formation)

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