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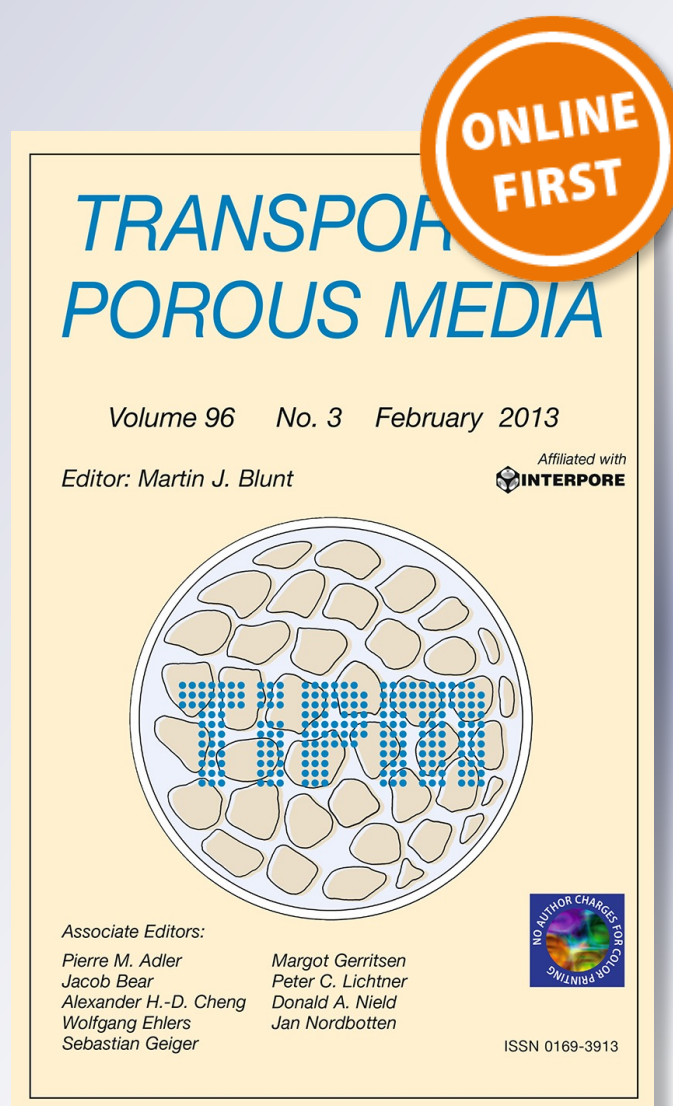
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Semianalytical Solution for CO₂ Plume Shape and Pressure Evolution During CO₂ Injection in Deep Saline Formations

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Abstract The injection of supercritical carbon dioxide (CO₂) in deep saline aquifers leads to the formation of a CO₂ rich phase plume that tends to float over the resident brine. As pressure builds up, CO₂ density will increase because of its high compressibility. Current analytical solutions do not account for CO₂ compressibility and consider a volumetric injection rate that is uniformly distributed along the whole thickness of the aquifer, which is unrealistic. Furthermore, the slope of the CO₂ pressure with respect to the logarithm of distance obtained from these solutions differs from that of numerical solutions. We develop a semianalytical solution for the CO₂ plume geometry and fluid pressure evolution, accounting for CO₂ compressibility and buoyancy effects in the injection well, so CO₂ is not uniformly injected along the aquifer thickness. We formulate the problem in terms of a CO₂ potential that facilitates solution in horizontal layers, with which we discretize the aquifer. Capillary pressure is considered at the interface between the CO₂ rich phase and the aqueous phase. When a prescribed CO₂ mass flow rate is injected, CO₂ advances initially through the top portion of the aquifer. As CO₂ is being injected, the CO₂ plume advances not only laterally, but also vertically downwards. However, the CO₂ plume does not necessarily occupy the whole thickness of the aquifer. We found that even in the cases in which the CO₂ plume reaches the bottom of the aquifer, most of the injected CO₂ enters the aquifer through the layers at the top. Both CO₂ plume position and fluid pressure compare well with numerical simulations. This solution permits quick evaluations of the CO₂ plume position and fluid pressure distribution when injecting supercritical CO₂ in a deep saline aquifer.

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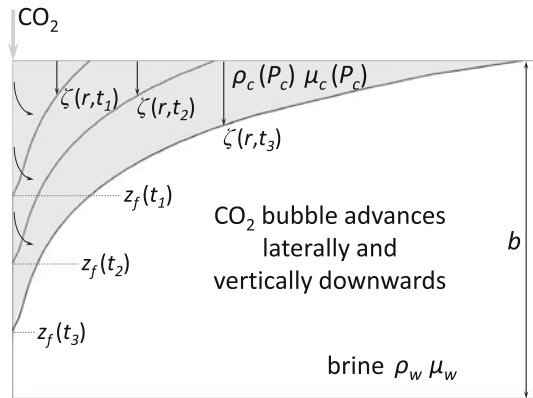
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List of symbols

b	Thickness of the aquifer
b_c	Thickness of the CO ₂ plume at the injection well
d	Thickness of a layer
g	Gravity
h_α	Head of α -phase ($\alpha = c, w$)
J_α	Mass flow rate per unit thickness of α -phase ($\alpha = c, w$)
k	Intrinsic permeability
$k_{r\alpha}$	Relative permeability of α -phase ($\alpha = c, w$)
n	Number of layers
N_g	Gravity number
P_0	Reference fluid pressure corresponding to the hydrostatic pressure at depth z_0
P_{cc}	Capillary entry pressure
P_α	Fluid pressure of α -phase ($\alpha = c, w$)
Q_0	CO ₂ Volumetric flow rate
Q_m	Prescribed CO ₂ mass flow rate
Q_w	Brine volumetric flow rate
q_α	Volumetric flux of α -phase ($\alpha = c, w$)
R	Radius of influence
r	Radial coordinate
r_i	Radial position of the interface at the depth z
r_{if}	Interface position at the bottom of the CO ₂ plume
r_p	Radius of the injection well
S_α	Degree of saturation of α -phase ($\alpha = c, w$)
S_{rw}	Residual degree of saturation of brine
S_s	Specific storage coefficient
t	Time
z	Vertical coordinate
z_0	Reference depth
Δz_j	Thickness of layer j
z_f	Depth of the bottom of the CO ₂ plume
α	Phase index, c for the CO ₂ rich phase and w for the aqueous phase
β	CO ₂ compressibility
ζ	Vertical position of the CO ₂ plume with respect to z_0
Φ_α	Potential of α -phase ($\alpha = c, w$)
Φ_i	Potential at the interface
Φ_R	Potential at the radius of influence
φ	Porosity
μ_α	Viscosity of α -phase ($\alpha = c, w$)
ρ_0	A reference CO ₂ density corresponding to the reference pressure P_0
ρ_α	Fluid density of α -phase ($\alpha = c, w$)

Fig. 1 CO₂ injection in a deep saline aquifer. The CO₂ plume thickness at the injection well progressively increases with time as CO₂ is injected. CO₂ density and viscosity are dependent on pressure. Note that CO₂ remains in the upper part of the aquifer because of buoyancy and it is not necessarily injected through the whole thickness of the aquifer



1 Introduction

Greenhouse gas emissions to the atmosphere can be reduced through the injection of supercritical carbon dioxide (CO₂) in deep saline aquifers. Under injection conditions, the density (450–900 kg/m³) and viscosity (0.03–0.1 mPa s) of CO₂ are highly dependent on pressure and temperature (Garcia 2003). This density is sufficiently high for storage purposes, but it is much lower than that of typical resident waters (1020–1200 kg/m³). Thus, the CO₂ plume tends to float above the resident brine and its thickness progressively increases as CO₂ pressure builds (Fig. 1).

Existing analytical and most numerical solutions to this problem assume that the injection takes place uniformly along the whole thickness of the aquifer (Saripalli and McGrail 2002; Nordbotten et al. 2005; Nordbotten and Celia 2006; Dentz and Tartakovsky 2009a; Vilarrasa et al. 2010a; Manceau and Rohmer 2011; Houseworth 2012). This assumption is unrealistic because of buoyancy. Instead, one should expect that CO₂ flows preferentially through the top portion of the aquifer, where the difference between CO₂ and resident water pressures are largest. In fact, the CO₂ plume may never reach the bottom of the aquifer (Fig. 1). Even analytical solutions that predict CO₂ plume evolution in the post-injection period consider that the CO₂ plume occupies the whole thickness of the aquifer at the end of the injection period (Hesse et al. 2007, 2008; Juanes et al. 2010). This may underestimate the CO₂ volume in free-phase at late times because the shape of the CO₂ plume at the end of injection affects its post-injection behaviour when capillary trapping is considered (MacMinn and Juanes 2009).

Supercritical CO₂ is highly compressible relative to water and determining its density in the reservoir is complicated because of highly nonlinear and coupled relationships. On the one hand, CO₂ density depends on fluid pressure. On the other hand, fluid pressure buildup during injection depends on CO₂ density, because it determines the volume of displaced brine. Pressure buildup is also controlled by other factors, like caprock permeability (Birkholzer et al. 2009) or the nature of aquifer boundaries (Zhou et al. 2008). Resident brine can easily migrate out laterally in open aquifers. This limits pressure buildup, but may salinize adjacent freshwater bodies. By contrast, fluid and rock compressibility may limit storage capacity in the presence of low-permeability boundaries (Zhou et al. 2008; Mathias et al. 2011). Evidence that CO₂ density is difficult to estimate can be found both in situ (Nooner et al. 2007) and analytically (Vilarrasa et al. 2010a). While 3D seismic data gave an average in situ CO₂ density of 530 ± 65 kg/m³ at the Utsira formation (Sleipner, Norway), the CO₂ density estimates prior to actual measurements ranged from 650 to 700 kg/m³ (Nooner et al. 2007).

CO₂ density, which is a priori unknown, has to be chosen when using an analytical solution to determine the CO₂ plume position. Neglecting CO₂ compressibility in these solutions can lead to errors greater than 50 % in the CO₂ plume position at the top of the aquifer (Vilarrasa et al. 2010a).

In addition to the evolution of the CO₂ plume, it is important to understand the evolution of pressure. Pressure affects the required compression energy, the CO₂ density and the mechanical stability of the caprock (Rutqvist et al. 2007; Ferronato et al. 2010; Vilarrasa et al. 2012). Mathias et al. (2009) assumed the Nordbotten et al. (2005) solution for the CO₂ plume position to calculate a vertical average of the fluid pressure in the aquifer. The same result was obtained by Vilarrasa et al. (2010a), who extended the computation to the solution of Dentz and Tartakovsky (2009a) and to calculate fluid pressure at every point of the aquifer. However, the slope of CO₂ pressure as a function of the logarithm of distance obtained from these analytical solutions differs significantly from that of numerical solutions (Vilarrasa et al. 2010a).

This paper aims to develop a semianalytical solution to the CO₂ plume position and fluid pressure evolution accounting for CO₂ compressibility and buoyancy effects in the injection well, while acknowledging that CO₂ flux into the aquifer is not uniform along the aquifer thickness. Thus, the extent and the thickness of the CO₂ plume as well as the overpressure can be quickly assessed. We formulate the problem and present the methodology for solving it when the CO₂ mass flow rate or the CO₂ pressure are prescribed at the injection well. Finally, we present an application of this methodology and compare the results with full numerical simulations.

2 Problem Formulation

Consider the injection of compressible CO₂ through a vertical well in a deep homogeneous horizontal confined brine aquifer. Mass conservation of these two fluids can be expressed as (Bear 1972)

$$\frac{\partial (\rho_\alpha S_\alpha \varphi)}{\partial t} = -\nabla \cdot (\rho_\alpha \mathbf{q}_\alpha), \quad \alpha = c, w, \quad (1)$$

where ρ_α is fluid density of the α -phase, S_α is the saturation of the α -phase, φ is porosity, t is time and $\mathbf{q}_\alpha$ is the volumetric flux of the α -phase, which can be either c , the CO₂ rich phase, or w , the aqueous phase.

Momentum conservation is expressed using Darcy's law

$$\mathbf{q}_\alpha = -\frac{k k_{r\alpha}}{\mu_\alpha} (\nabla P_\alpha + \rho_\alpha g \nabla z), \quad \alpha = c, w, \quad (2)$$

where k is intrinsic permeability, $k_{r\alpha}$ is relative permeability of the α -phase, μ_α is viscosity of the α -phase, P_α is fluid pressure of α -phase, g is gravity, and z is the vertical coordinate (positive upwards).

This two-phase flow is affected by buoyancy effects because CO₂ is lighter than brine. To quantify the relative influence of buoyancy we define a gravity number, N_g , as the ratio of gravity to viscous forces. Gravity forces can be represented by the buoyancy force ($\Delta \rho g$, where $\Delta \rho$ is the difference between fluids density) when Darcy's law is expressed in terms of equivalent head. Viscous dissipation forces correspond to the horizontal pressure gradient, which can be approximated as ($Q_0 \mu / (2\pi r b k k_{r\alpha})$), where Q_0 is the volumetric flow rate, r is radial distance and b is aquifer thickness). Still, since CO₂ is compressible, we prefer to

express the volumetric flow rate in terms of the mass flow rate, Q_m , as $Q_0 = Q_m/\rho$. Thus, N_g becomes

$$N_g = \frac{2\pi r_{ch} b k k_{rc} \Delta \rho g \rho_{ch}}{\mu_c Q_m}, \quad (3)$$

where ρ_{ch} is a characteristic density and r_{ch} is a characteristic distance. Large values of the gravity number ($N_g \gg 1$) indicate that buoyancy forces dominate. On the other hand, small gravity numbers ($N_g < 1$) indicate that viscous forces dominate. Note that buoyancy forces will always dominate far from the injection well, where r_{ch} is sufficiently large, whereas the opposite is generally true near the well, except for small mass flow rates and aquifers with a high permeability, in which case buoyancy forces will dominate also in the vicinity of the well.

Assuming that fluid density depends only on fluid pressure, the head of the α -phase is defined as (Bear 1972)

$$h_\alpha = z - z_0 + \frac{1}{g} \int_{P_0}^{P_\alpha} \frac{dP'_\alpha}{\rho_\alpha(P'_\alpha)}, \quad (4)$$

where h_α is the head of the α -phase, z_0 is a reference depth and P_0 is the hydrostatic fluid pressure corresponding to depth z_0 .

Darcy's law can be expressed in terms of head provided that density is not affected by other variables (i.e., under isothermal conditions) by combining Eqs. (2) and (4)

$$\mathbf{q}_\alpha = -\frac{k k_{r\alpha}}{\mu_\alpha} \rho_\alpha g \nabla h_\alpha. \quad (5)$$

When flow rate is prescribed at the injection well, CO₂ will penetrate initially along the top portion of the aquifer because its pressure is not sufficient to displace brine along the entire aquifer thickness. As injection continues and CO₂ pressure builds up, the portion of the well occupied by CO₂ grows up to the depth where CO₂ pressure equilibrates with that of brine. As a result, the plume advances not only laterally, but also vertically downwards. Its thickness also increases with time (Fig. 1). Brine and CO₂ pressures are related at the porous medium interface via the capillary entry pressure

$$P_c(r_i, z) = P_w(r_i, z) + P_{cc}, \quad (6)$$

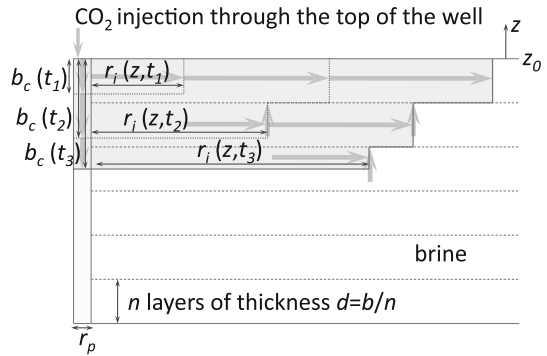
where P_{cc} is the capillary entry pressure and r_i is the radial position of the interface at depth z .

Neglecting mass transfer across the interface, the problem is defined by the two differential equations in (1), one for each phase, which are coupled by the equilibrium equation in (6) and by the continuity of flux at the interface

$$\mathbf{q}_c(r_i, z) = \mathbf{q}_w(r_i, z). \quad (7)$$

The boundary condition at the injection well is applied at the top of the aquifer, where it is possible to prescribe either the total CO₂ mass flow rate or the CO₂ pressure. This formulation is realistic and leads naturally to non-uniform CO₂ flux into the aquifer along the well depth. As for the outer boundary, we consider an infinite aquifer.

Fig. 2 Schematic representation of CO₂ injection evolution taking into account that CO₂ first enters through the top of the aquifer because of buoyancy. The aquifer is divided into n layers through which CO₂ advances laterally and vertically downwards. The CO₂ plume at the well reaches, at every time step, the depth at which CO₂ pressure equals brine pressure



3 Semianalytical Solution

3.1 Radial Injection of Compressible CO₂

To address the problem of CO₂ injection defined in the previous section we assume that the CO₂ rich phase and the formation brine are separated by a sharp interface. The validity of this assumption has been discussed extensively in previous studies on analytical solutions (Nordbotten et al. 2005; Dentz and Tartakovsky 2009a,b; Lu et al. 2009). Capillary pressure is considered at the interface between the CO₂ rich phase and the formation brine (Eq. 6). Thus, there is a jump in fluid pressure at the interface equal to the entry pressure. CO₂ dissolution into the brine, which may induce density-driven convective cells (Lindeberg and Wessel-Berg 1997; Riaz et al. 2006; Hidalgo and Carrera 2009; Pau et al. 2010), is not considered here.

We solve the problem by vertically discretizing the aquifer into n layers of equal thickness $d = b/n$ (Fig. 2). The time evolution of the problem is solved using discrete time steps to overcome nonlinearities and coupling difficulties. The interface advances laterally in the layers that contain CO₂, but also moves vertically downwards as fluid pressure builds up. CO₂ at the bottom of the CO₂ plume may fill the thickness of a layer only partially. Once the CO₂ plume reaches a thickness equal to md , where m is the number of layers filled with CO₂, the following layer $m + 1$, previously devoid of CO₂, begins to fill with CO₂. The part occupied by CO₂ in this new layer has a thickness equal to $b_c - md$, where b_c is the CO₂ plume thickness at the well (Fig. 2).

To calculate the CO₂ plume thickness in the injection well, we assume hydrostatic conditions in it,

$$\frac{dP_\alpha}{dz} = -g\rho_\alpha(P_\alpha), \quad \alpha = c, w. \quad (8)$$

This assumption is motivated by observations from high-resolution numerical simulations. Therefore, CO₂ enters the aquifer only through the part of the injection well occupied by CO₂; the mass flux will be higher at the top of the aquifer because of the larger difference between CO₂ and brine pressure.

To calculate the lateral advance of CO₂ in each layer, we assume that the hydraulic response within the CO₂ plume is much shorter than transport of the front. Therefore, we consider a quasi-steady (sequence of steady-states) description of the moving fronts in Eq. (1), i.e., the left-hand side of Eq. (1) cancels. In addition, we make the Dupuit approximation of horizontal flow. Furthermore, the density of the CO₂ phase will vary in space due to changes in the CO₂ pressure and due to the high compressibility of CO₂. Therefore, conservation should not be

expressed in terms of volumetric fluxes, but mass fluxes. The total mass flow rate, per unit aquifer thickness, in a radial injection varies with depth but is constant at a given z within each phase

$$J_{\alpha} = 2\pi r \rho_{\alpha} q_{r\alpha}, \quad (9)$$

where J_{α} is the mass flow rate per unit thickness of α -phase and $q_{r\alpha}$ is the horizontal component of the volumetric flux of α -phase. This mass flow rate per unit thickness will vary from layer to layer. At the interface in each layer, we impose continuity of flux in the radial direction

$$q_{rc}(r_j, z) = q_{rw}(r_j, z), \quad (10)$$

where r_j is the radius of the interface in layer j .

Since we adopt a sharp interface approximation, the saturation in the CO₂ rich phase is taken as constant. Thus, the interface position advances as

$$\frac{dr_j}{dt} = \frac{q_{r\alpha}(r_j)}{\varphi(1 - S_{rw})}, \quad (11)$$

where S_{rw} is the residual degree of saturation of brine.

To eliminate complexities associated with nonlinearity, we define the following potential to formulate the problem assuming that relative permeability and fluid density are solely a function of fluid pressure

$$\Phi_{\alpha}(h_{\alpha}) = \int_0^{h_{\alpha}} 2\pi g k k_{r\alpha}(h'_{\alpha}) \frac{(\rho_{\alpha}^*(h'_{\alpha}))^2}{\mu_{\alpha}} dh'_{\alpha}, \quad (12)$$

where $\rho_{\alpha}^*(h'_{\alpha}) = \rho_{\alpha}^*(P_{\alpha}(h'_{\alpha}))$.

Combining Eqs. (5), (9), and (12), flow rate in a layer becomes

$$r \frac{d\Phi_{\alpha}}{dr} = -J_{\alpha}. \quad (13)$$

Though we make the Dupuit approximation of horizontal flow within a layer, we acknowledge vertical CO₂ leakage between layers. Acknowledging that flow towards the top of the aquifer is largely buoyancy driven, we impose that the vertical mass flow rates between layers occur punctually, for simplicity. The distance at which the vertical flow rates are injected to the adjacent layer depends on the gravity number computed in the vicinity of the injection well (Eq. 3). This is because vertical flow rates are expected to occur when gravity forces dominate, i.e., large gravity numbers. Then, vertical flow rates will occur close to the injection well for large gravity numbers. On the other hand, vertical flow rates will occur far from the injection well for small gravity numbers computed close to the injection well. Therefore, the distance at which the vertical flow rates occur is inversely proportional to the gravity number. The vertical mass flow rate of a layer to its adjacent one is given by

$$J_{cz,j} = \int_{r_p}^{\bar{r}_j} 2\pi \rho_c q_{zc} dr, \quad (14)$$

where r_p is the radius of the well and q_{zc} is the vertical component of the volumetric CO₂ flux. We will inject this flow rate at a radius $\bar{r}_j = \min(r_{j+1}, b/(2N_g))$. The term $b/(2N_g)$ reflects the fact that the injection distance is inversely proportional to the gravity number as

mentioned above. For simplicity, we have adopted r_{j+1} as $\bar{r}_j$. Introducing Eq. (5) into Eq. (14) and assuming that the CO₂ head varies linearly with the logarithm of distance to the well (we assume this based on previous experience and observations from high resolution numerical simulations, which will be presented in further detail in Sect. 5), after some algebra, yields

$$J_{cz,j} = \frac{2\pi k k_{rc} \rho_c^2 g}{\mu_c \Delta z_j} (u_j - u_{j+1}) \left(r_{j+1} - r_p - r_{j+1} \ln \frac{r_{j+1}}{r_p} \right), \quad (15)$$

where u_j is the logarithmic slope of the CO₂ head in layer j , Δz_j is the thickness of layer j and CO₂ density is evaluated at the point of the vertical flow rate injection. Note that the subscript of the layers increases with depth, i.e., layer j is placed above layer $j + 1$.

The integration of Eq. (13), accounting for the fact that now J_c is a function of the radial distance due to the vertical flow rates, yields the solution of the problem

$$\Phi_{w,j} = \Phi_R + J_{w,j} \ln \left(\frac{R}{r} \right), \quad r \geq r_j \quad (16a)$$

$$\Phi_{c,j} = \Phi_j + (J_{c,j} + J_{cz,j+1}) \ln \left(\frac{r_j}{r} \right), \quad r_{j+1} < r < r_j \quad (16b)$$

$$\Phi_{c,j} = \Phi_j + J_{c,j} \ln \left(\frac{r_j}{r} \right) + J_{cz,j+1} \ln \left(\frac{r_j}{r_{j+1}} \right), \quad r < r_{j+1} \quad (16c)$$

where Φ_R is the potential at the radius of influence, R is the radius of influence, the subscript after the comma indicates the layer (e.g., j indicates layer j) and Φ_j is the potential at the interface in layer j . Φ_R is known and constant because it refers to the initial fluid pressure in the aquifer. Φ_j can be determined by evaluating Eq. (16a) at the interface. The radius of influence corresponds to the distance affected by the pressure buildup cone caused by injection and grows with the square root of time as

$$R = \sqrt{\frac{2.25 k \rho_w g t}{\mu_w S_s}}, \quad (17)$$

where S_s is the specific storage coefficient. This result comes from combining the classical Theis and Thiem solution and is further discussed in Vilarrasa et al. (2010a).

The CO₂ mass flow rate at the injection well for layer j can be determined from Eq. (16c) as

$$J_{c,j} = \frac{\Phi_{c,j}(r_p) - \Phi_j - J_{cz,j+1} \ln(r_j/r_{j+1})}{\ln(r_j/r_p)}. \quad (18)$$

This CO₂ mass flow rate is different in every layer and will change with time as the CO₂ plume grows. The evolution of the CO₂ plume is calculated using a time stepping algorithm. Integrating Eq. (11) and using Eq. (9) yields the interface position for a given time step

$$r_j^{l+1}(z) = \sqrt{\left(r_j^l\right)^2 + \frac{J_{c,j}^l(r_j) \Delta t^{l+1}}{\pi \varphi (1 - S_{rw}) \rho_{ci,j}^l}}. \quad (19)$$

where superscript l denotes the time step, Δt is increment of time between step l and step $l + 1$ and $\rho_{ci,j}$ is the CO₂ density at the interface in layer j .

The volumetric flow rate of brine, Q_w , at a radial distance r from the well can be calculated once the interface position is known. Due to the continuity of fluxes at the interface, we obtain

$$Q_w(r) = \int_{z_f}^{z_0 - \zeta(r)} \frac{J_c(r)}{\rho_c(r)} dz, \quad (20)$$

where z_f is the depth of the bottom of the CO₂ plume and ζ is the vertical position of the CO₂ plume from the top of the aquifer.

The volume of displaced brine at radius r is equal to the volume of injected CO₂. The flow rate of brine is driven by the overpressure produced by the injected CO₂, which is assumed to be distributed through the portion of the aquifer thickness occupied by the formation brine

$$Q_w(r) = - \int_{z_0 - b}^{z_0 - \zeta(r)} 2\pi r \frac{k}{\mu_w} \frac{\partial P_w}{\partial r} dz. \quad (21)$$

Integrating Eq. (21) yields the following expression for brine pressure

$$P_w - P_0 = \frac{\mu_w Q_w(r)}{2\pi k (b - \zeta(r))} \ln \frac{R}{r}. \quad (22)$$

As mentioned above, two types of boundary conditions can be adopted at the injection well. Either a prescribed CO₂ mass flow rate or a prescribed CO₂ pressure can be imposed. In either case, the condition is imposed at the depth that coincides with the top of the aquifer. Both are discussed below.

3.2 Prescribed CO₂ Mass Flow Rate

When injecting a prescribed CO₂ mass flow rate, the CO₂ plume advances both laterally and vertically downwards as CO₂ is injected. Since this problem presents two unknowns at every time step, i.e., the CO₂ head at the well and the thickness of the CO₂ plume at the well, two conditions are needed. First, hydrostatic conditions are assumed in the well (Eq. 8). And second, mass balance must be satisfied. The mass inlet at a given time step corresponds to the mass flow rate multiplied by the time increment. This mass is distributed through the layers containing CO₂ proportionally to the mass flow rate per unit thickness and the thickness of each layer. Furthermore, the mass that occupies the volume corresponding to the increment of the plume thickness in each time step has to be accounted for, resulting in

$$Q_m = \sum_{j=1}^m J_{c,j} \Delta z_j + \bar{\rho}_c \pi r_{if}^2 \varphi (1 - S_{rw}) \frac{\Delta z_f}{\Delta t}, \quad (23)$$

where Q_m is the prescribed CO₂ mass flow rate, m is the total number of layers in which CO₂ is present, $\bar{\rho}_c$ is the mean CO₂ density in the layer that coincides with the bottom of the CO₂ plume, r_{if} is the interface position at the bottom of the CO₂ plume and Δz_f is the increment of the CO₂ plume thickness at the well at a given time step.

3.3 Prescribed CO₂ Pressure

Since the head at the well is known when imposing the CO₂ pressure, there is only one unknown: the thickness of the CO₂ plume at the well. Hence, imposing hydrostatic conditions

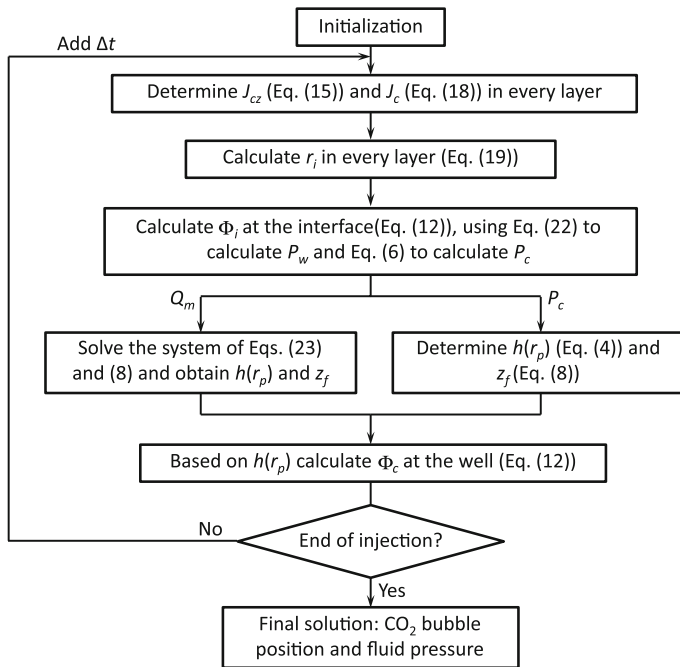


Fig. 3 Time stepping algorithm to calculate the position of the CO₂ plume and the fluid pressure

in the well (Eq. 8) and knowing that the bottom of the CO₂ plume in the well coincides with where the CO₂ pressure equals the brine pressure allows the problem to be solved.

4 Algorithm

The evolution of the position of the CO₂ plume is calculated using a time stepping algorithm. The process is very similar for the two possible injection boundary conditions and is repeated for each time step (Fig. 3). The procedure has to be initialized, using a small time increment, as follows

- The CO₂ plume is assumed to grow slightly in the top layer, i.e., the interface position advances laterally a fraction of the well radius and the thickness of the CO₂ plume is a fraction of d .
- The volumetric flow rate is assumed equal to the mass flow rate divided by density at the reference CO₂ pressure.

These two assumptions allow for initialization of the overpressure (using Eq. (22) and the volumetric flow rate), the potential at the interface (using Eq. 12), the depth that the CO₂ plume reaches, the head at the injection well and the potential at the well (using Eq. 12). No vertical flow rates exist in this initialization. After this, the time stepping algorithm can then be used. It is as follows

1. Determine the vertical CO₂ mass flow rate (Eq. 15) and the horizontal CO₂ mass flow rate in each layer evaluating Eq. (18) at the well. We use the potential at the interface and at the well and the interface position evaluated at the previous time step.

2. Calculate the new interface position in every layer using Eq. (19). The CO₂ mass flow rate is the one calculated in step 1 and the CO₂ density at the interface is the one evaluated at the previous time step.
3. Calculate the potential at the interface (Eq. 12), using Eq. (20) to calculate the volumetric flow rate and Eq. (22) to calculate the brine pressure, and Eq. (6) to calculate the CO₂ pressure at the interface.
- 4a. Impose a prescribed CO₂ mass flow rate: solve the system of two equations (Eqs. 23 and 8) and determine the CO₂ head at the well and the thickness of the CO₂ plume at the well.
- 4b. Impose a prescribed CO₂ pressure: the head at the well can be determined by using Eq. (4) and the thickness of the CO₂ plume at the well can be calculated by imposing Eq. (8).
5. Based on the head at the well calculated in step 4, compute the potential at the well using Eq. (12).

These five steps are repeated, applying a time increment after every loop, until the desired time of CO₂ injection is completed. The time increment should be small (some seconds) at the beginning of injection and can progressively increase (up to some hours). We found a condition that the thickness by which the CO₂ plume increases should be less than a small percentage of the thickness of a layer (typically less than a 3 %) in order to guarantee convergence.

5 Application

5.1 Spreadsheet Programming

In order to evaluate this methodology, we programmed it in a spreadsheet that can be downloaded from [GHS \(2012\)](#). We programmed it this way to highlight the ease of implementation and use by non-expert programmers. This implementation considers 25 layers. We chose 25 layers, because we found this sufficient to resolve all features and that the result barely changed as this number increases. In fact the solution already changes very little from 10 to 25 layers as illustrated in Fig. 4. Note that the solutions presented in Fig. 4 do not incorporate vertical flows. The implementation requires a prescribed mass flow rate, constant properties of the brine (density and viscosity), constant CO₂ viscosity and the CO₂ density is defined to vary linearly with CO₂ pressure

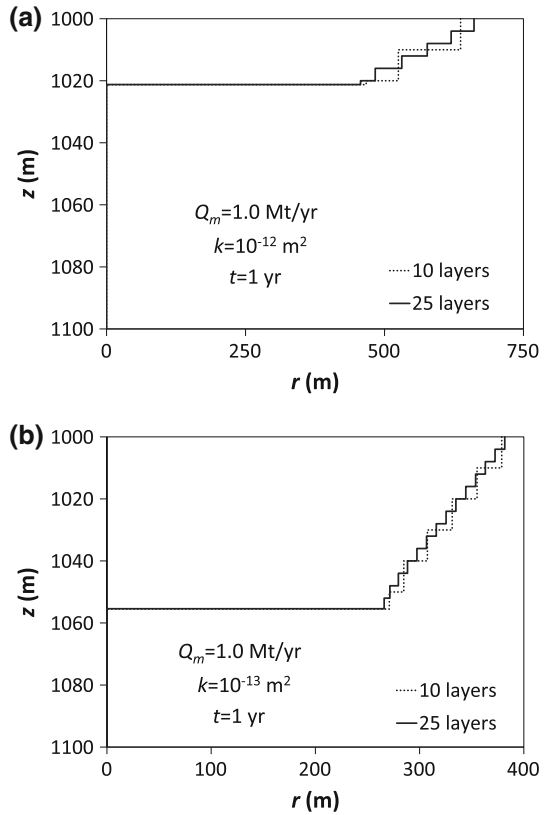
$$\rho_c = \rho_0 + \beta (P_c - P_0), \quad (24)$$

where ρ_0 is the reference density for the reference pressure P_0 and β is CO₂ compressibility. For the range of pressures in this study this linear approximation appears to provide good results. The parameter values in (24) are taken from the data tables given by [Span and Wagner \(1996\)](#).

With this linear approximation of the CO₂ density, the potential for the CO₂ can be obtained by integrating Eq. (12), which yields (see Appendix 1)

$$\Phi_c = \frac{\pi k \rho_0^2}{\mu_c \beta} e^{-2g\beta(z-z_0)} \left(e^{2g\beta h_c} - 1 \right). \quad (25)$$

Fig. 4 Effect of the number of layers (10 or 25 layers) contained in the semianalytical solution, without incorporating vertical flows, on the CO₂ plume position, after 1 year of injection of 1 Mt/year, for permeabilities, k , **a** $k = 10^{-12} \text{ m}^2$ and **b** $k = 10^{-13} \text{ m}^2$. Note that the differences are small and that become negligible as the number of layers increases further



Furthermore, CO₂ pressure can be expressed as a function of the potential (Appendix 1)

$$P_c - P_0 = \sqrt{\frac{\mu_c}{\pi k \beta}} \Phi_c + \frac{\rho_0^2}{\beta^2} e^{-2g\beta(z-z_0)} - \frac{\rho_0}{\beta}. \quad (26)$$

Note that CO₂ overpressure varies with the square root of the logarithm of the distance to the injection well [see the form of the potential Φ_c in Eq. (16b,16c)].

The solution of the system of two equations (Eqs. 8 and 23) for finding the CO₂ head and the thickness of the CO₂ plume at the well is shown in Appendix 2. CO₂ pressure is calculated by solving the system of two equations with two unknowns presented in Appendix 2 and brine pressure is given by the overpressure that CO₂ generates when brine is displaced. The mean density appearing in Eq. (23) is calculated in Appendix 3.

5.2 Model Setup

We represent a 100-m thick saline aquifer whose top is located at a depth of 1,000 m. The aquifer is assumed to be infinite-acting, homogeneous, and isotropic. The permeability of the aquifer is either 10^{-12} or 10^{-13} m^2 , its porosity is 0.1 and the rock compressibility is $1.2 \times 10^{-10} \text{ Pa}^{-1}$. The temperature is assumed to be constant and equal to 320 K. The density of brine is 1087.5 kg/m^3 , its viscosity is 0.6 mPa s and its compressibility is $4.5 \times 10^{-10} \text{ Pa}^{-1}$. Thus, the specific storage coefficient yields a value of $1.76 \times 10^{-6} \text{ m}^{-1}$. The reference CO₂

density ρ_0 , corresponding to the reference pressure $P_0 = 10$ MPa (hydrostatic pressure at the top of the aquifer), is 448.28 kg/m^3 and its compressibility β is $5.56 \times 10^{-5} \text{ kg/m}^3 \text{ Pa}^{-1}$ (Span and Wagner 1996). β is the product of the actual CO₂ compressibility and a density. The actual CO₂ compressibility at the pressure and temperature of the aquifer is $1.48 \times 10^{-7} \text{ Pa}^{-1}$. Note that CO₂ compressibility, for the range of pressure and temperature of this study, is three orders of magnitude higher than that of brine. The CO₂ viscosity is calculated using the expression proposed by Altunin and Sakhabetdinov (1972). Though constant, CO₂ viscosity is case specific and depends on the overpressure and aquifer temperature, so that a representative value can be adopted according to the mean CO₂ density. CO₂ viscosity is set, according to pressure and temperature, to 0.03 and 0.04 mPas for the aquifer with a permeability of 10^{-12} and 10^{-13} m^2 , respectively. The entry pressure equals 0.02 MPa and the residual degree of saturation of brine is 0.025. The injected mass flow rate is 1.0 Mt/year. An injection ramp is used to progressively increase the mass flow rate from zero to the desired mass flow rate. Doing so, the increments in the CO₂ plume thickness are small. This injection ramp lasts less than 50 s, so its effect can be considered as negligible over the course of the full injection period for practical purposes.

The finite element numerical code CODE_BRIGHT (Olivella et al. 1994, 1996), extended for CO₂ sequestration (Villarrasa et al. 2010b), has been used to validate the results of this semianalytical solution with those of numerical results. The aquifer is represented by an axisymmetric model, with a radius of 100 km, in which CO₂ is injected at the top of the injection well. The radius of the injection well is 0.5 m. The well is represented by a zone of unit porosity and high permeability (four orders of magnitude higher than that of the aquifer). The inclusion of the injection well in the model allows us to reproduce a realistic CO₂ injection, with non-uniform CO₂ flux along the whole thickness of the aquifer. The grid is structured and has 25 elements in the vertical coordinate, which matches the 25 layers adopted in the application of the semianalytical solution. We used a van Genuchten retention curve (van Genuchten 1980), with an entry pressure of 0.02 MPa and a shape parameter of 0.8. The relative permeability functions, for both the CO₂ and the brine, are linear with the degree of saturation of each phase. These retention curve and relative permeability functions produce a CO₂ plume with an almost constant CO₂ saturation and a narrow capillary fringe. Thus, the numerical solutions are close to the assumption of the abrupt interface approximation assumed for the semianalytical solution. The CO₂ saturation 90 % isoline has been chosen to represent the position of the CO₂-brine interface. The effect of changing the retention curve would be a wider capillary fringe and a non constant CO₂ saturation within the CO₂ plume. However, the shape of the plume would be maintained. Thus, the geometry of the CO₂ plume can be calculated precisely by introducing into the semianalytical solution an appropriate value of the residual degree of saturation of brine. CO₂ pressure would change slightly due to capillarity, but its profile with respect to the logarithm of distance would be maintained. However, the curvature of the relative permeability curves affects the thickness of the CO₂ plume tip at the top of the aquifer (Gasda et al. 2008). A more concave relative permeability curve, such as the cube of the degree of saturation of each phase compared to a linear relationship with the degree of saturation, yields a wider CO₂ plume tip.

5.3 Validation of the Semianalytical Solution

We compare the results of the semianalytical solution with those of the numerical solution. In addition, the analytical solutions of Nordbotten et al. (2005) and Dentz and Tartakovsky (2009a), in which the method proposed by Villarrasa et al. (2010a) to incorporate CO₂

Fig. 5 CO₂ mass flow rate per unit thickness at the injection well as a function of depth after 1 year of injecting 1 Mt/year for two permeabilities. Note that in both cases the vast majority of CO₂ is injected through the top portion of the aquifer, rather than being uniformly injected along the whole thickness of the aquifer

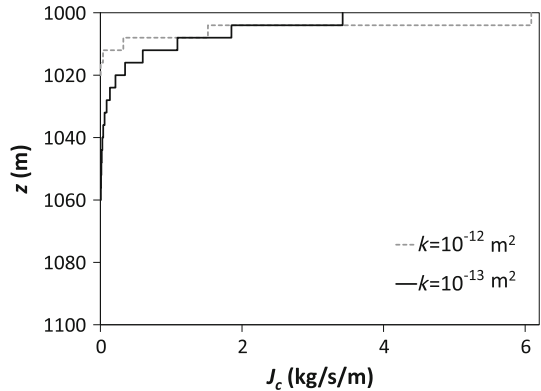
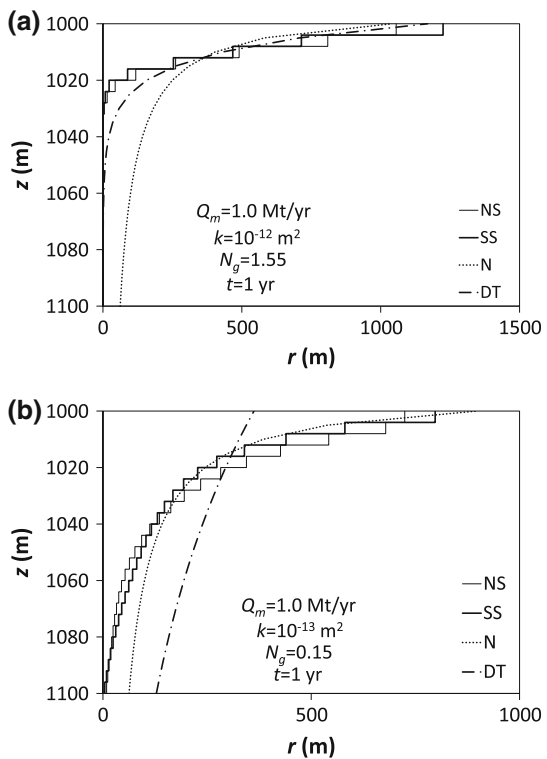


Fig. 6 Comparison of the CO₂ plume position between semianalytical (SS) and numerical solution (NS) after 1 year of injection of 1 Mt/year, and permeabilities, k , **a** $k = 10^{-12} \text{ m}^2$ and **b** $k = 10^{-13} \text{ m}^2$. In addition, the analytical solutions of Nordbotten et al. (2005) (N) and Dentz and Tartakovsky (2009a) (DT) after using the method of Vilarrasa et al. (2010a) to account for CO₂ compressibility are presented for comparison. Note that, these analytical solutions inject CO₂ uniformly along the whole thickness of the aquifer



compressibility has been applied, are presented for comparative purposes. As mentioned earlier, these analytical solutions inject CO₂ uniformly along the whole thickness of the aquifer. Figure 5 displays the mass flow rate per unit thickness as a function of depth in the injection well, showing that most of the CO₂ enters into the aquifer through its top portion, rather than uniformly along the whole thickness of the aquifer. Whereas this may not be desirable from a storage point of view, where it is desirable to maximize the use of pore space, it will occur whenever the flow rate is small for the aquifer permeability.

Figure 6 displays the CO₂ plume position for the analytical, semianalytical and numerical solutions after 1 year of injecting 1 Mt/year of CO₂ for two aquifer permeabilities.

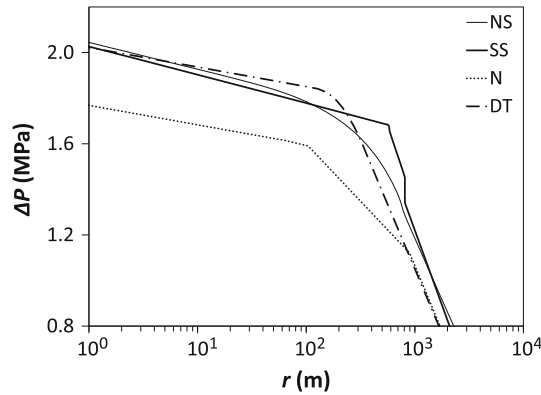


Fig. 7 Comparison of the fluid overpressure at the top of the aquifer between analytical, semianalytical (SS) and numerical solutions (NS) after 1 year injecting 1.0 Mt/year in an aquifer with a permeability of 10^{-13} m^2 . The analytical solutions of Nordbotten et al. (2005) (N) and Dentz and Tartakovsky (2009a) (DT) after using the method of Vilarrrasa et al. (2010a) to account for CO₂ compressibility are presented for comparison. The first change in slope beginning from the right hand side of the figure indicates the CO₂-brine interface position for each solution

We consider a case with high permeability ($k = 10^{-12} \text{ m}^2$) in which gravity forces dominate and another case with low permeability ($k = 10^{-13} \text{ m}^2$) in which viscous forces dominate. The semianalytical solution compares well with the numerical solution in both cases. The CO₂ plume occupies only the top portion of the aquifer when gravity forces dominate (CO₂ is injected into the aquifer through the top 32 m) (Fig. 6a). On the other hand, the CO₂ plume reaches the bottom of the aquifer when viscous forces dominate (Fig. 6b). The Nordbotten et al. (2005) solution gives a better approximation when viscous forces dominate, while the Dentz and Tartakovsky (2009a) solution predicts a better CO₂ plume position when gravity forces dominate. However, both analytical solutions differ from the numerical solution at the bottom of the CO₂ plume due to the fact that they consider a uniform injection along the whole thickness of the aquifer.

Figure 7 compares fluid overpressure at the top of the aquifer as a function of the distance to the injection well resulting from the semianalytical, numerical and analytical solutions when injecting 1.0 Mt/year in an aquifer with a permeability of 10^{-13} m^2 . Fluid overpressure obtained from the semianalytical solution compares well with that of the numerical solution, presenting the same slope in the region occupied by CO₂ close to the injection well. The pressure drop increases sharply at the distance where CO₂ from the second layer is injected in the top layer. This is in contrast with the numerical solution, where the gradient increases smoothly. As a result, our solution is somewhat more abrupt than the numerical solution near the interface. Existing analytical solutions, i.e. those of Nordbotten et al. (2005) and Dentz and Tartakovsky (2009a), fail to give good fluid pressure predictions. First, they predict a lower brine overpressure because they underestimate the volumetric flow rate of brine. This is because CO₂ is injected along the whole thickness of the aquifer and since CO₂ density increases with depth, the mean CO₂ density becomes higher than in the semianalytical and numerical solutions. In addition, the slopes of fluid pressure inside the CO₂ plume are lower than that of numerical simulations. In contrast, the slope of the semianalytical solution is the same as that of the numerical solution close to the injection well in the region occupied by CO₂. Hence, it can be concluded from this semianalytical solution that while brine overpressure is proportional to the logarithm of distance from the injection well, CO₂

Fig. 8 Comparison of the time evolution of the injection pressure at the top of the aquifer between analytical, semianalytical (SS) and numerical solutions (NS) when injecting 1.0 Mt/year in an aquifer with a permeability of 10^{-13} m^2 . The analytical solutions of Nordbotten et al. (2005) (N) and Dentz and Tartakovsky (2009a) (DT) after using the method of Vilarrasa et al. (2010a) to account for CO_2 compressibility are presented for comparison

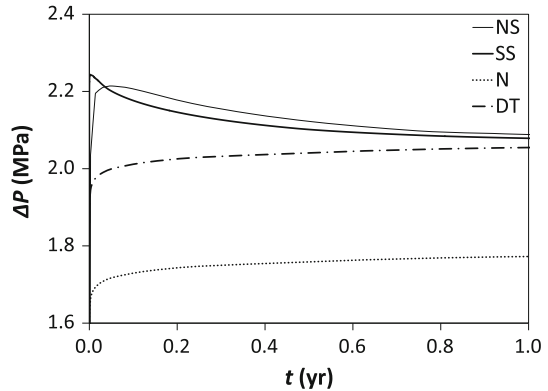
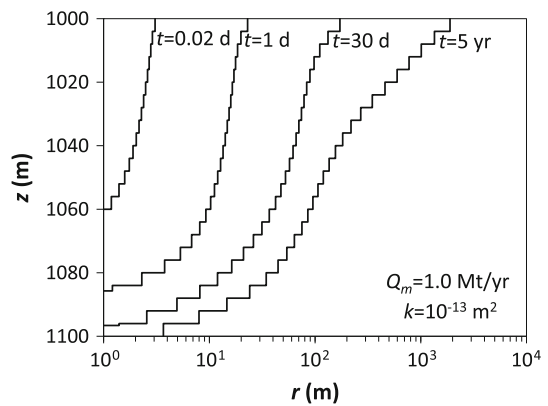


Fig. 9 CO_2 plume evolution given by the semianalytical solution for several injection times when injecting 1.0 Mt/year in a 100-m thick aquifer with a permeability of 10^{-13} m^2 . Note that, the thickness of the CO_2 plume progressively increases with injection time



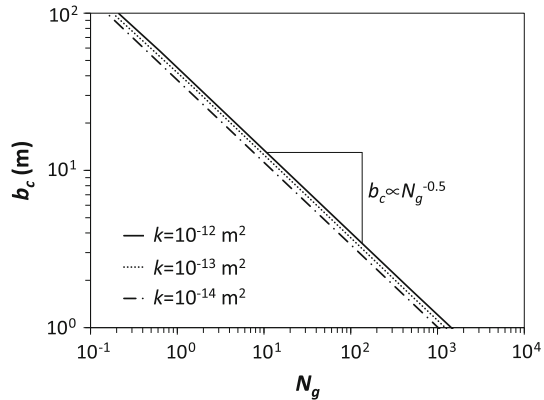
overpressure is proportional to the square root of the logarithm of the distance from the injection well (recall Eq. 26).

Figure 8 displays the evolution of the overpressure at the injection well when injecting 1.0 Mt/year in an aquifer with a permeability of 10^{-13} m^2 . While the overpressure predicted by the analytical solutions of Nordbotten et al. (2005) and Dentz and Tartakovsky (2009a) increases continuously, fluid overpressure decreases after reaching a maximum at the beginning of injection for the semianalytical and numerical solutions. Mukhopadhyay et al. (2012) found that overpressure increases continuously when injecting CO_2 in depleted gas reservoirs, whose fluid pressure is significantly below hydrostatic conditions. However, a pressure drop after the initial pressure buildup was observed in situ in the Ketsin test site, Germany (Henniges et al. 2011) and numerically by Vilarrasa et al. (2010b), who argued that pressure drops because the overpressure that occurs in the capillary fringe due to relative permeability reduction is distributed over a larger area as the CO_2 plume increases and because the viscosity of the CO_2 is much lower than that of the brine.

5.4 CO_2 Plume Thickness

Figure 9 shows the CO_2 plume position evolution for several injection times given by the semi-analytical solution when injecting 1.0 Mt/year in an aquifer with a permeability of 10^{-13} m^2 . CO_2 advances laterally and vertically downwards with injection time. Note that, the CO_2

Fig. 10 CO₂ plume thickness at the injection well after 1 year of injection as a function of the gravity number for several aquifer permeabilities. Note that, the logarithm of the CO₂ plume thickness decreases linearly with the logarithm of the gravity number



plume advances preferentially through the top of the aquifer for increasing injection times. This is because gravity forces become dominant as CO₂ flows away from the injection well. Note that, in this case the CO₂ plume reaches the aquifer bottom for an injection time longer than 30 days (actually, it occurs after 162.6 days of injection). However, the CO₂ plume would not reach the bottom of the aquifer in a more permeable aquifer, like the one presented in Fig. 6a.

Figure 10 displays the CO₂ plume thickness at the well after 1 year of injection as a function of the gravity number computed at 1 m from the injection well (Eq. 3) for several aquifer permeabilities. The curves are obtained by varying the mass flow rate. The logarithm of the CO₂ plume thickness decreases linearly with the logarithm of the gravity number, presenting a slope of $-1/2$. The CO₂ plume is thinner than the aquifer thickness for high gravity numbers (buoyancy forces dominate). In contrast, it reaches the bottom of the aquifer for gravity numbers lower than 0.15 (viscous forces dominate). The effect of permeability is small, but not negligible because permeability affects fluid overpressure and thus CO₂ density. These curves are useful for quickly estimating the CO₂ thickness at the well of a CO₂ injection project.

6 Discussion and Conclusions

We have presented a semianalytical solution for the CO₂ plume geometry and fluid pressure evolution that accounts for CO₂ compressibility and buoyancy in the well. The latter enables us to study the non-uniform injection rate along the aquifer thickness that will occur in industrial injection sites (Fig. 5). CO₂ compressibility is taken into account by assuming that CO₂ density varies linearly with CO₂ pressure. This approximation is reasonable because CO₂ pressure is relatively constant during injection (recall Fig. 8). However, if the range of variation were wider, an exponential approximation would adjust better to the actual CO₂ density variation with pressure. To achieve a non-uniform injection along the aquifer thickness, we assume that fluid pressure is hydrostatic within the well, though its magnitude changes with time. This reflects the low viscosity of CO₂ and the slow velocities that will occur in real sites. Indeed, fluid pressure is observed to be hydrostatic inside the well in the numerical simulations of the detailed problem. While the general methodology presented allows for variable viscosity the specific implementation in this work assumes constant viscosity for simplicity. However, since it is an input parameter, it can be adjusted according to the aquifer

temperature and the overpressure induced by injection in each case. This implementation has 25 horizontal layers, which provides a reasonable resolution in many cases, except for very thick aquifers (of 250–300 m thick or thicker), where more layers may be necessary.

We found that both CO₂ plume position and fluid pressure obtained from the proposed semianalytical solution compare well with those given by numerical simulations. Analytical solutions, which have been corrected to account for CO₂ compressibility to make them comparable with the semianalytical and numerical solutions, give acceptable results of the CO₂ plume position depending on the gravity number (Fig. 6). However, the semianalytical solution gives good estimates regardless of the gravity number. Nonetheless, a weakness of the semianalytical solution is that vertical flows have to be considered explicitly between adjacent layers, otherwise the CO₂ plume shape does not resemble that of numerical simulations (compare Figs. 4, 6). Another limitation is that the semianalytical solution yields good estimates of the CO₂ plume tip only for a relative permeability curve that is linear with the degree of saturation of each phase. For more concave relative permeability curves (e.g., the cube of the degree of saturation of each phase), the CO₂ plume tip becomes thicker and cannot be precisely reproduced by the semianalytical solution. Nevertheless, the approximation of the semianalytical solution has a clear advantage over numerical solutions in terms of the time required for calculation. One should bear in mind that simulating CO₂ injection through the well instead of injecting it uniformly along the whole thickness of the aquifer results in high computational cost. By contrast, the semianalytical solution gives immediate results. Furthermore, analytical and semianalytical solutions can be coupled with numerical models in order to speed up their calculations (Celia and Nordbotten 2009; McDermott et al. 2011).

This solution facilitates quick evaluation of the lateral extension and thickness of the CO₂ plume, which may not reach the bottom of the aquifer, for a given injection time. The calculation of the CO₂ plume thickness accounting for buoyancy is innovative and significant because it points out important differences to calculations that are based on the commonly accepted assumption that the CO₂ plume occupies the whole aquifer thickness. The CO₂ plume thickness is a function of the gravity number (see Fig. 10). This knowledge can be useful to support decision-making concerning the operation of CO₂ injection projects. In addition, this solution can be helpful in designing and interpreting CO₂ injection tests in pilot projects. In this context, it is important to bear in mind that, from a storage point of view, it is desirable to inject over the whole aquifer thickness to maximize the use of pore space. As we have seen, this goal is limited by buoyancy, which dominates far from the injection well. However, even near the well, injecting over a partial thickness may be profitable during early stages because it promotes CO₂ dissolution into the brine, which in turn may cause mineral dissolution and stimulation (i.e., increasing the permeability of the aquifer, so that injectivity increases and the required overpressure to inject a given mass flow rate decreases, which reduces injection costs). This stimulation close to the well can propagate relatively far from the injection well by promoting CO₂ advance through the top of the aquifer. For such goal, our solution, generalized for varying permeability, would be extremely useful.

Finally, the slope of CO₂ pressure as a function of the logarithm of distance from the well calculated with the semianalytical solution is the same as that of the numerical solution. In the semianalytical solution, the CO₂ overpressure varies with the square root of the logarithm of the distance to the injection well (Eq. 26). This is interesting because this variation with distance to the well differs from those of existing analytical solutions. In addition, the semianalytical solution reproduces a CO₂ injection pressure evolution similar to the one observed in numerical solutions and in field-scale injection projects, i.e., fluid pressure drops after an initial abrupt fluid pressure buildup. This behaviour, which appears naturally in this semianalytical solution, is not reflected by other existing analytical solutions.

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Appendix 1

Here we develop the mathematical formulation of the problem for the case in which CO₂ density varies linearly with pressure (Eq. 24), and CO₂ viscosity and brine properties are constant.

First, we integrate Eq. (4) for the CO₂ phase, which yields

$$h_c = z - z_0 + \frac{1}{g\beta} \ln \left(1 + \frac{\beta}{\rho_0} (P_c - P_0) \right). \quad (27)$$

The inverse of Eq. (27) gives the CO₂ pressure as a function of the head as

$$P_c - P_0 = \frac{\rho_0}{\beta} \left(e^{g\beta(h_c - (z - z_0))} - 1 \right). \quad (28)$$

Integrating Eq. (12) and using Eq. (28) gives the following expression for the CO₂ potential

$$\Phi_c = \frac{\pi k \rho_0^2}{\mu_c \beta} e^{-2g\beta(z - z_0)} \left(e^{2g\beta h_c} - 1 \right). \quad (29)$$

Since the exponent $2g\beta h_c$ is small, $(e^{2g\beta h_c} - 1)$ can be approximated as $2g\beta h_c$. Therefore, the CO₂ potential can be expressed as:

$$\Phi_c \approx 2\pi g k \frac{\rho_0^2}{\mu_c} h_c e^{-2g\beta(z - z_0)}, \quad (30)$$

where the potential is composed of a part corresponding to a constant CO₂ density (ρ_0) multiplied by a correction due to CO₂ compressibility [the exponential in the right-hand side of Eq. (30)]. Combining Eqs. (27) and (29) and operating, yields an expression of CO₂ pressure as a function of the potential

$$P_c - P_0 = \sqrt{\frac{\mu_c}{\pi k \beta} \Phi_c + \frac{\rho_0^2}{\beta^2} e^{-2g\beta(z - z_0)}} - \frac{\rho_0}{\beta}. \quad (31)$$

Note that the head (Eq. 27) at the interface can be expressed as a function of the CO₂ density as

$$h_{ci} = z - z_0 + \frac{1}{g\beta} \ln \left(\frac{\rho_0 + \beta (P_{ci} - P_0)}{\rho_0} \right) = z - z_0 + \frac{1}{g\beta} \ln \left(\frac{\rho_{ci}}{\rho_0} \right), \quad (32)$$

where the subscript i indicates interface. Combining Eq. (29) with Eq. (32) yields the following expression for the CO₂ potential at the interface

$$\Phi_i = \frac{\pi k \rho_0^2}{\mu_c \beta} \left(\frac{\rho_{ci}^2}{\rho_0^2} - e^{-2g\beta(z - z_0)} \right). \quad (33)$$

In the brine phase, integration of Eq. (4) yields

$$h_w = z - z_0 + \frac{P_w - P_0}{\rho_w g} \quad (34)$$

which is the expression of the head for an incompressible fluid. Integrating Eq. (12) gives the potential in the brine phase as

$$\Phi_w = 2\pi g k \frac{\rho_w^2}{\mu_w} h_w. \quad (35)$$

Combining Eqs. (34) with (35), gives the following expression for the brine pressure

$$P_w - P_0 = \frac{\mu_w}{2\pi k \rho_w} \Phi_w - (z - z_0) \rho_w g. \quad (36)$$

Note that, the brine pressure varies with the logarithm of the distance to the injection well [see the form of the potential in Eq. (16a)].

Appendix 2

A system of two equations with two unknowns has to be solved in step 4 of the time stepping algorithm when a mass flow rate is prescribed at the injection well. The unknowns are the head at the well and the thickness of the CO₂ plume at the well. The two equations are Eqs. (23) and (8).

Combining Eq. (23) with Eqs. (18), (29), and (33), after some algebra, gives the following expression for the head at the well as a function of the increment of the CO₂ plume thickness

$$e^{2g\beta h_c} = \frac{Q - A_1 \Delta z_f + A_2 - A_3}{A_4}, \quad (37)$$

where

$$Q = \frac{\mu_c \beta}{\pi k \rho_0^2} Q_m, \quad (38a)$$

$$A_1 = \frac{\mu_c \beta}{\pi k \rho_0^2} \frac{\bar{\rho}_c \pi r_{if}^2 \varphi (1 - S_{rw})}{\Delta t}, \quad (38b)$$

$$A_2 = \sum_{j=1}^m \frac{\rho_{ci,j}^2 / \rho_0^2}{\ln(r_j / r_p)} \Delta z_j, \quad (38c)$$

$$A_3 = \frac{\mu_c \beta}{\pi k \rho_0^2} \sum_{j=1}^n \frac{J_{cz,j+1} \ln(r_{j+1} / r_j)}{\ln(r_j / r_p)} \Delta z_j, \quad (38d)$$

$$A_4 = \sum_{j=1}^m \frac{e^{-2g\beta(z_j - z_0)}}{\ln(r_j / r_p)} \Delta z_j. \quad (38e)$$

Next, assuming hydrostatic conditions at the well (Eq. 8) and that the bottom of the CO₂ plume coincides where brine pressure equals CO₂ pressure and using Eqs. (29) and (31) give the second equation of the system of equations

$$e^{g\beta h_c} = p + \frac{\rho_w}{\rho_0} g\beta \Delta z_f, \quad (39)$$

where

$$p = \frac{\beta}{\rho_0} P_w(z_{f-1}) + 1, \quad (40)$$

where $P_w(z_{f-1})$ is the brine pressure evaluated at the depth reached by the CO₂ plume in the previous time step. The combination of Eqs. (37) and (39) gives the following quadratic equation

$$\Delta z_f^2 + B \Delta z_f + C = 0, \quad (41)$$

where

$$B = \frac{2p\rho_0}{g\beta\rho_w} + \frac{\rho_0^2 e^{-2g\beta(z_f-z_0)}}{\rho_w^2 g^2 \beta^2} \frac{A_1}{A_4}, \quad (42a)$$

$$C = \frac{\rho_0^2}{\rho_w^2 g^2 \beta^2} \left(p^2 - \frac{Q + A_2 - A_3}{A_4} e^{-2g\beta(z_f-z_0)} \right). \quad (42b)$$

Once Eq. (41) is solved and the increment of the CO₂ plume thickness at the well in a given time step is known, the head at the well can be calculated from Eq. (37) as

$$h_c(r_p) = \frac{1}{2g\beta} \exp\left(\frac{Q - A_1 \Delta z_f + A_2 - A_3}{A_4}\right). \quad (43)$$

Appendix 3

The mean CO₂ density in a given layer has to be calculated in order to apply Eq. (23). Assuming that CO₂ density varies linearly with pressure (Eq. 24), and using Eqs. (31), (16c), and (33), after some algebra, the following expression for the CO₂ density is obtained

$$\rho_c = \sqrt{\rho_{ci}^2 + \frac{\mu_c \beta}{\pi k} J_c \ln \frac{r_j}{r}}. \quad (44)$$

The mean CO₂ density in a layer is obtained from dividing the CO₂ mass in a given layer by the volume that it occupies

$$\bar{\rho} = \frac{1}{V} \int_{z_{j-1}}^{z_j} \int_{r_p}^{r_j} 2\pi r \varphi (1 - S_{rw}) \rho_c dr dz. \quad (45)$$

Introducing Eq. (44) in Eq. (45) and integrating yields

$$\begin{aligned}\bar{\rho} &= \frac{1}{(r_j^2 - r_p^2)} \left[r^2 \sqrt{a - b \ln r} - \frac{1}{2} \sqrt{\frac{\pi}{2}} \sqrt{b} e^{2a/b} \operatorname{erf} \left(\sqrt{\frac{2}{b}} \sqrt{a - b \ln r} \right) \right]_{r_p}^{r_j} \\ &= \frac{1}{(r_j^2 - r_p^2)} \left[r_j^2 \rho_{ci} - r_p^2 \rho_{cp} + \frac{r_j^2}{2} \sqrt{\frac{b\pi}{2}} \exp \left(\frac{2\rho_{ci}^2}{b} \right) \left(\operatorname{erf} \left(\sqrt{\frac{2}{b}} \rho_{ci} \right) - \operatorname{erf} \left(\sqrt{\frac{2}{b}} \rho_{cp} \right) \right) \right],\end{aligned}\quad (46)$$

where

$$a = \rho_{ci}^2 + \frac{\mu_c \beta}{\pi k} J_c \ln r_j, \quad (47a)$$

$$b = \frac{\mu_c \beta}{\pi k} J_c. \quad (47b)$$

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