Preliminary study on density-dependent flow and transport in the Lake Karachai area (UE RaCoS project): adequacy of CODESA-3D to model a plume of contaminant brine.

CRS4

Environment Group

Erminia Leonardi & Claudio Gallo

February 1999

Contents

List of Tables

List of Figures

1 Introduction

Groundwater flow and transport modeling can be used to determine the direction of dissolved contaminant migration, to define the limits of a capture zone for a contamination recovery well (or well field), or to delineate a water well protection area (or recharge area) for a water supply. A number of computer groundwater modeling programs have been developed allow for rapid and efficient assessment of groundwater flow under conditions that may involve the addition of simulated wells and/or simulated sources of recharge in an existing flow field. The models often generate contour maps that illustrate relevant data that are related to groundwater flow. Thus, the transport of various dissolved organic and inorganic compounds in groundwater can be evaluated in a number of geologic situations by means of numerical models.

Many problems in subsurface hydrology imply the study of flow and transport of solute coupled via the fluid density. Usually, the density of the groundwater is nearly constant or it may vary slightly as a result of small variations in either temperature or pressure or as a result of the presence of trace quantities of dissolved contaminants. However, a number of environmentally important problems requires the analysis of brine dynamics, the movement of very dense contaminants in subsurface systems, caused by disposal of hazardous (toxic or radioactive) waste in crystalline or salt rock formations, infiltration of leachates from landfills, and industrial waste disposals. A typical example of weak coupling between flow and transport is found when dealing with problems of saltwater intrusion in coastal aquifers, caused by overpumping in sensitive portions of water supply aquifers [*Bear*, 1979; *Frind*, 1982].

When the density variations become larger than $> 3-4\%$, flow and transport begin to be strongly coupled, and the problem becomes increasingly nonlinear. A primary coupling arises in the equations through the body force term of the fluid flow equation and the advection term of the solute transport equation; a second coupling enters the equations through velocity-dependent hydrodynamic dispersion. Density variations in excess of 20% occur in salt domes and bedded-salt formations which are currently being considered for radioactive waste repositories [*Oldenburg*, 1995] .

In the past years, computer speed has represented a strong limitation in the development of numerical codes able to describe complex scenarios such as the transport dynamics in a strongly coupled groundwater-brine flow system.

Transport of brine is by advection and hydrodynamic dispersion only (no molecular diffusion), and only since about ten years ago the problem has begun to be extensively examined using a number of numerical codes such as SWIFT, METROPOLE, SUTRA [*Souza*, 1987]. *Oldenburg and Pruess* [1995] presented the results of their simulations for strongly coupled transport/flow systems obtained by implementing a methodology capable of attaining a verifiable steady state for a problem (transport of brines) where other models could not, using their finite difference simulator TOUGH2.

The mathematical model proposed by *Kolditz et al.* [1996] comprises a set of nonlinear, coupled, partial differential equations to be solved for pressure/hydraulic head and mass fraction/concentration of the solute component. Different levels of the approximation of density variations in the mass balance equations are used for convection problems (e.g., the Boussinesq approximation or full density coupling). Henry, Elder and salt dome problems have been used as benchmarks and studied by using different finite element simulators varying in details such as temporal discretization, spatial discretization, finite elements basis functions, iteration schemes, and linear solvers. *Hassanizadeh and Leijnse* [1995] have analyzed the transport problem in porous media under the effect of high-concentration-gradient dispersion, using a theoretical derivation of the Fickian dispersion equation, where in addition to the longitudinal and transverse dispersivity, a new parameter, independent of the fluid properties, is introduced to obtain a non-linear Fickian dispersion equation.

One of the main objectives in this preliminary study was to implement CODESA-3D (COupled variable DEnsity and SAturation) code [*Gambolati* 1998] for the Lake Karachai site and to run preliminary simulations under a variety of conditions, but with density ratio coefficient no larger than 0.07 (density variations of maximum 7%).

Because CODESA-3D is a finite element numerical code for coupled flow and transport that is normally used for problems of seawater intrusion, where density variations are around 3%, part of our work has been devoted to determine the limit of applicability of the code. Elder's problem has been considered to test the accuracy of the model in representing fluid flow driven purely by density differences. To this aim, the test case of *Voss and Souza* , has been reproduced using CODESA-3D to verify the possibility of studing the movement of brines, even with density ratio coefficient larger than 0.07.

Using as test cases the simulations of brine transport in the aquifer located under the Lake Karachai waste disposal site, we have evaluated the optimized set of parameters (e.g., time step size, best temporal integration scheme, mesh size, and so on) necessary to minimize the numerical instability of the solution due to the strong non linearity of the problem.

In this context, a vertical section of the Lake Karachai waste disposal site has been considered. Leakage from Lake Karachai in South Ural (Russia), operated since the 1950s as a storage reservoir for medium level radioactive liquid wastes, has formed a subsurface contamination plume and presents a grave threat to the environment. The plume has been observed to be moving towards local areas of groundwater discharge, potentially contaminating springs and sources of drinking water with radionuclide releases. The Lake Karachai waste disposal site is considered one of the most radioactively contaminated sites in the world, and special water protection and remediation measures need to be urgently implemented.

Due to the complexity of the problem under study, two-dimensional vertical section of the Lake Karachai area was selected, and the initial test problem has been identified on the basis of experimental data provided by the Department of Environmental Geology of Russian Accademy of Science. A series of test cases is run in order to test the capabilities of the numerical code to reproduce the physical behaviour of the plume spreading and to produce accurate results with reasonable computational effort. Part of the work was aimed at evaluating the relationship between CPU time, mesh size, and density ratio (contaminant to water) in the current implementation of the CODESA-3D code.

2 Mathematical Model

The mathematical model of density-dependent flow and transport in groundwater is expressed here in terms of an equivalent freshwater head h, defined as [*Huyakorn et al.*, 1987, *Frind*, 1982, *Gambolati et. al.*, 1993]

$$h = \psi + z$$

where $\psi = p/(\rho_o g)$ is the equivalent freshwater pressure head, p is the pressure, ρ_o is the freshwater density, g is the gravitational constant, and z is the vertical coordinate directed upward. The density ρ of the saltwater solution is written in terms of the reference density ρ_o and the normalized salt concentration c:

$$\rho = \rho_o(1 + \epsilon c) \tag{1}$$

where $\epsilon = (\rho_s - \rho_o)/\rho_o$ is the density ratio, typically $\ll 1$, and ρ_s is the solution density at the maximum normalized concentration $c = 1$. Depending on the application, ρ_s can represent, for instance, the density of seawater, or of the solution nearest a surface salt mound or around an underground saline diapir. The dynamic viscosity μ of the saltwater mixture is also expressed as a function of c and of the reference viscosity μ_o as

$$\mu = \mu_o(1 + \epsilon' c) \tag{2}$$

where $\epsilon' = (\mu_s - \mu_o)/\mu_o$ is the viscosity ratio and μ_s is the viscosity of the solution at $c = 1$. With these definitions, the coupled system of variably saturated flow and miscible salt transport equations is [*Gambolati et. al.*, 1999]

$$\begin{aligned}
\sigma\frac{\partial\psi}{\partial t} &= \nabla\cdot\left[K_s\frac{1+\epsilon c}{1+\epsilon' c}K_r\left(\nabla\psi + (1+\epsilon c)\eta_z\right)\right] \\
&\quad - \phi S_w \epsilon\frac{\partial c}{\partial t} + \frac{\rho}{\rho_o}q \qquad (3)\\
\mathbf{v} &= -K_s\frac{1+\epsilon c}{1+\epsilon' c}K_r\left(\nabla\psi + (1+\epsilon c)\eta_z\right) \qquad (4)\\
\phi\frac{\partial S_w c}{\partial t} &= \nabla\cdot(D\nabla c) - \nabla\cdot(c\mathbf{v}) + qc^* + f \qquad (5)
\end{aligned}$$

where ∇ is the gradient operator, K_s is the saturated hydraulic conductivity tensor at the reference density, $K_r(\psi)$ is the relative conductivity, η_z is a vector equal to zero in its x and y components and 1 in its z component, $\sigma(\psi, c)$ is the general storage term or overall storage coefficient, t is time, ϕ is the porosity, $S_w(\psi)$ is the water saturation, q is the injected (positive)/extracted (negative) volumetric flow rate, $\mathbf{v}$ is the Darcy velocity vector, D is the dispersion tensor, c^* is the normalized concentration of salt in the injected/extracted fluid, and f is the volumetric rate of injected (positive)/extracted (negative) solute that does not affect the velocity field.

Initial conditions and Dirichlet, Neumann, or Cauchy boundary conditions are added to complete the mathematical formulation of the flow and transport problem expressed in

(3)-(5). For the flow equation, these take the form

$$\psi(\mathbf{x}, 0) = \psi_o(\mathbf{x}) \tag{6}$$
$$\psi(\mathbf{x}, t) = \psi_p(\mathbf{x}, t) \qquad \text{on } \Gamma_1 \tag{7}$$
$$\mathbf{v} \cdot \mathbf{n} = -q_n(\mathbf{x}, t) \qquad \text{on } \Gamma_2 \tag{8}$$

where $\mathbf{x} = (x, y, z)^T$ is the Cartesian spatial coordinate vector, superscript T is the transpose operator, ψ_o is the pressure head at time 0, ψ_p is the prescribed pressure head (Dirichlet condition) on boundary Γ_1, $\mathbf{n}$ is the outward normal unit vector, and q_n is the prescribed flux (Neumann condition) across boundary Γ_2. We use the sign convention of q_n positive for an inward flux and negative for an outward flux, consistent with the convention used for q and f in system (5).

For the transport equation, the initial and boundary conditions are [*Galeati and Gambolati*, 1989]

$$c(\mathbf{x}, 0) = c_o(\mathbf{x}) \tag{9}$$
$$c(\mathbf{x}, t) = c_p(\mathbf{x}, t) \qquad \text{on } \Gamma_3 \tag{10}$$
$$D\nabla c \cdot \mathbf{n} = q_d(\mathbf{x}, t) \qquad \text{on } \Gamma_4 \tag{11}$$
$$(\mathbf{v}c - D\nabla c) \cdot \mathbf{n} = -q_c(\mathbf{x}, t) \qquad \text{on } \Gamma_5 \tag{12}$$

where c_o is the initial concentration, c_p is the prescribed concentration (Dirichlet condition) on boundary Γ_3, q_d is the prescribed dispersive flux (Neumann condition) across boundary Γ_4, and q_c is the prescribed total flux of solute (Cauchy condition) across boundary Γ_5. The sign convention for q_d and q_c is the same as for q_n, q, and f.

The numerical discretization of the mathematical model is a standard Galerkin scheme, with tetrahedral elements and linear basis functions, complemented by weighted finite differences for the discretization of the time derivatives [*Gambolati et. al.,* 1999] .

3 Description of the area around Lake Karachai

The area under study is located within Chelyabinsk, one of the South Ural provinces in Russia (Figures 1, 2). There are several surface waste reservoirs between two rivers that have been used for over 45 years for storage of medium-and-low-radioactivity liquid wastes. The largest reservoir of the zone, known as Karachai Lake, contains a total of 120 mln Ci, which makes it one of the most contaminated (by radionuclides) sites in the world. The leakage from the reservoir has resulted in a contamination plume traveling through the aquifer represented by fractured metavolcanic rocks [*Mironenko et. al.*, 1986], .

The high waste density and aquifer heterogeneity with no well-developed aquitard has caused a complex 3D spreading of the plume. Increasing in volume, the plume moves towards the zones of groundwater discharge threatening surface water and groundwater wellfields.

Subsurface radionuclide transport at the Lake Karachai site is accompained by a range of physical and chemical processes and interactions including: advection, mechanical dispersion, chemical diffusion, radioactive decay, chemical reaction, and adsorption. Depending on the process or interaction, the mass transport potential of the contaminant may be either enhanced or deteriorated.

The regional geological cross section is represented by fractured moderately metamorphosed effusive Lower-Silurian rocks including porphyrites, tuffs, and occasional metamorphosed shells covered by a thin layer of weathering products. The body of effusive rocks has an areally and cross-sectionally complex fractured structure formed as a result of tectonic processes and weathering. Three tectonic fracturing systems are present. The largest of them is associated with disjunctive faults striking submeridionally. Regional fracturing of the weathered rock attenuates at a depth on the order of 100 m. However, in linear fault zones, fracturing is found at a depth of 300 m. As a rule, the fractures are filled with the products of weathering to depth of 20-40 m. The interaction of regional and tectonic fracturing forms a body represented by relatively "strong" blocks broken by "weakened" narrow tectonic zones [*Mironenko et. al.*, 1986] .

One of the objectives of this work was to numerically reproduce the monitored hydrogeological data of a water flow which moves into north-eastern directions while the actual plume spreads to the south. To this aim, as mentioned in the Introduction, a simplified vertical section of the complex Karachai site was considered under different kinds of boundary conditions, in order to study the effects of sloping bottoms in contrasting the advective movement of a contaminant plume. The 2D domain under study has the following dimensions: 12 km in length and 85 m in height. The Lake Karachai waste disposal is 400 m long and is almost centered within the domain of interest. According to data acquired by field measurements (obtained by means of a well network), three main zones have been identified on the basis of differences in hydraulic conductivity values (refer to Figure **??**):

$$\begin{array}{ll} \text{zone-1:} & 0.1 < \mathbf{K} < 0.5 \text{ m/day} \\ \text{zone-2:} & 0.5 < \mathbf{K} < 5.0 \text{ m/day} \\ \text{zone-3:} & \quad\;\; \mathbf{K} < 0.1 \text{ m/day} \end{array}$$

Precipitation is the primary recharge mechanism for the underlying aquifer, thus prescribed fluxes (Neumann condition) q have been considered as boundary conditions for the problem. The bottom of the system under study is assumed to be impermeable (given the presence of a clay lens). The left side of the domain (refer to the Figure **??**) is assumed to be a watershed divide and can be assumed to be impermeable as well. The right hand side of the domain is assumed to be governed mainly by a horizontal groundwater flow motion.

The contaminant (a dense salty mixture of nitrates - NO_3^- - and radioactive ions - Cs, Sr) is introduced in the system throughout Lake Karachai at variable concentrations. The density dependent flow arising from the presence of nitrates (in this context, radionuclides can be assumed to be transported as "passive" tracers), whose concentration rises up to 150 g/l, should be be strongly influenced by the presence of aquifer heterogeneities described above.

4 Test case description

The objectives of this work, as described in the Introduction, were to evaluate the ability of the CODESA-3D code [*Gambolati et. al.*, 1998] to simulate the physical behaviour of a plume of dense contaminant moving in a saturated medium, identify the order of magnitude of some of the relevant parameters such as time-step size and mesh-size, and to evaluate the effects of a sloping aquifer bottom on the brine transport.

Since the CODESA-3D simulator is three-dimensional, a section was simulated by using a vertical slab, whose thickness (y) was much smaller than the other two dimensions (x and z). Simulations were performed considering increasingly complex scenarios: no-flow recharge vs flow-recharge, flat vs sloping aquifer bottom, and homogeneous vs heterogeneous conditions.

The vertical section of the Lake Karachai site is very long (12000 m) and narrow (85 m), and this is surely a problem when we discretize the representative domain. In fact, unless we use a very dense grid of points (which results in runs prohibitively time consuming), the tetrahedal elements are strongly deformed along a preferential direction, and this can cause numerical instabilities in finite element numerical procedures. The simplest way of operating in these cases is to act on the longitudinal or transverse dispersivity coefficients, in order to satisfy the Peclet constraints [*Perrochet et. al.*, 1993] . But this implies, sometimes, very large values for these parameters, which could adversely affect the correct description of the physical phenomena of interest, producing, for instance, unrealistically large dispersion. Moreover, it must be pointed out that the use of very distorted elements makes difficult to describe contaminant plume motion through heterogeneous regions, or its spreading at the bottom of a domain even if dispersion is small. Thus, in the test cases described below, we often need to find the best compromise between a good physical description of the phenomena and a satisfactory stability of the numerical solution.

At first, a "small physical size" test case has been considered in order to evaluate the contaminant behavior when accounting for soil heterogeneity, bottom slope, and water recharge.

Part of our work was aimed at studying the fingering effect, that is, the fingered shape of the plume caused by gravitational instabilities (physical effect) or propagation of numerical oscillations of the solution (numerical effects). Fingering is "mesh size" convergent [*Kolditz et. al*, 1996] , that is, strong discretization effects have been observed, and it is possible to consecutively refine the mesh until convergence is achieved and the fingered shape of the plume never changes. Here a series of test cases was aimed to evaluate the relationship between mesh size, time step size, density ratio coefficient and fingering effects.

A series of test cases was also aimed at reproducing the case of a contaminant introduced inside the domain during a period of 20 years, and to follow its fate for the subsequent 80 years when no more contaminant is introduced.

Unless otherwise specified, we have assumed negligible the effect of the molecular diffusion, while the density difference ratio has always been chosen equal to 0.07, and the porosity equal to 0.25.

4.1 Test case A: influence of sloping bottoms vs homogeneous or heterogeneous domains, when different flow boundary conditions are considered.

Because the aim of these simulations is to test the performances of the CODESA-3D code, a small size domain has been chosen, discretized with quite regular tetrahedral elementary cells. Therefore, a domain 500 m long, 1 m wide, and 100 m high is considered. The domain is discretized in 100 5 m subdivisions along the x direction, 20 5 m subdivisions in the z directions, and a single 1-m strip in the y direction, by means of a structured mesh composed by 200 triangular elements in the xy-plane. The discretization is replicated in the z direction for 20 layers to obtain a 4242-node/12000-tetrahedral element mesh.

We have considered four cases with different sloping bottoms, corresponding to slope with $\theta = 0^o, 0.1^o, 2^o$, and 5^o, respectively. Moreover, we note that the aquifer layers, corresponding to the zones of different heterogeneity, are maintained parallel to the aquifer bottom when the 3D-mesh is generated; thus, the heterogeneity mapping is strongly affected by the slope of the domain. Figure **??** illustrates a simplified sketch of the domain.

The boundary conditions considered are the following:

- Flow: in a first series of simulations, all the sides of the domain (DE, EF, FA, and AD in Figure **??**) are impermeable boundary. We indicate this case as the "no flow" case. In a second series of simulations the sides DE and EF are impermeable boundaries, on the sides AB-CD and BC a Neuman boundary condition of dispersive flux $q_D = 5 \times 10^{-10}$m/s/m, and $q_D = 7 \times 10^{-9}$m/s/m, respectively are imposed, and, finally, on the side FA a Dirichlet condition of prescribed potential head $h = 0$ m is assumed. We indicate this case as the "with flow" case.

- Transport: on side BC a Dirichlet boundary condition of concentration C=1 (C is normalized concentration) was fixed.

The domain was assumed to be heterogeneous and three main layers were identified. Soil properties and parameter values are detailed in Table 1, where K_{xx}, K_{yy}, and K_{zz} are the components of the hydraulic conductivity tensor, S_s is the specific storage, α_L and α_T are the longitudinal and transverse dispersivity coefficients, respectively, and n is the porosity.

An initial condition of prescribed pressure head and concentration equal to zero over the entire domain has been assumed.

Integration in time of equations (3) and (5) is performed using a weighted finite difference scheme with weighting parameter equal to 0.5 (Crank-Nicolson scheme) for the flow equation and 1.0 (Euler fully implicit backward difference scheme) for the transport equation. The time step size chosen is $\Delta t = 50$ days, which is sufficiently small to avoid numerical instabilities.

4.2 Test case B: fingering effect as a function of the mesh size, time step, and density coefficient ϵ

Here, some insights regarding the relationship between mesh size, time step size, density ratio coefficient and fingering effects are considered. The domain, 1400 m long, 1 m wide, and 85 m high, was considered homogeneous, and Table 2 gives the soil and solute properties.

A "coarse mesh" domain discretized in 280 5 m subdivisions along the x direction, 15 5.6 m subdivisons in the z directions, and a single 1-m strip in the y direction by means of a structured mesh composed by 560 triangular elements in the xy-plane is considered; then, the 2D domain has been replicated along the z direction for 15 layers in order to obtain a 8992-node/25200-tetrahedral element mesh.

The "fine mesh" domain is discretized in 1400 1 m subdivisions along the x direction, 15 5.6 m subdivisions in the z directions, and a single 1-m strip in the y direction by means of a structured mesh composed by 2800 triangular elements in the xy-plane. By replicating the 2D domain along the z direction for 15 layers a 44832-node/126000-tetrahedral element mesh is obtained.

The boundary conditions are the following:

- Flow: a Neumann condition of dispersive flux equal to zero was imposed on every boundary so that the domain is impermeable.
- Transport: a constant contaminant recharge C=1 (C is the normalized concentration) is assumed at the top of the domain between $500 < x < 900$ m.

The simulations begin with an initial condition of concentration and potential head zero over the entire domain. The flow equation is integrated in time using the Crank-Nicholson scheme, while the fully implicit backward difference scheme has been used to integrate in time the transport equation.

4.3 Test case C: simulations using Cauchy boundary condition.

The objectives of this test case were to verify the capability of the CODESA-3D code to simulate the fate of a contaminant plume when, after a period of 20 years, we stop the introduction of contaminant and follow its development for the subsequent 80 years.

A quasi three-dimensional domain 12000 m long, 10 m wide, and 85 m high has been considered, discretized in 12000 10 m subdivisions along the x direction, 17 5 m subdivisions in the z directions, and a single 5-m strip in the y direction (refer to Figure **??**) by means of a structured mesh composed by 480 triangular elements in the xy-plane and it has been replicated along the z direction to obtain a 6534 node/24480 tetrahedral element mesh.

The boundary conditions are the following (refer to Figure **??**):

- Flow: the sides AB and AH are impermeable boundaries. On side HF is imposed a Dirichlet condition of prescribed potential head h = 85 m; furthermore, a condition of h= 92 m is considered at the top of the domain at point C of the scketch. On the remaining points at the top of the domain a Neumann condition of constant dispersive flux q_D was imposed; in particular, on the nodes between D and E (corresponding to the lake) $q_D = 7 \times 10^{-9}$ m/s/m, and on the nodes between A and D and between E and F $q_D = 5 \times 10^{-10}$ m/s/m.
- Transport: during the first 20 years of the simulation, on side DE, $C = 1$ (C is normalized concentration) was imposed, while for the successive 80 years a Cauchy condition of total flux q_C zero everywhere was imposed at the top of the domain.

The domain was assumed to be heterogeneous and three main layers were identified. Soil properties and parameter values are detailed in Table 8.

An initial condition of prescribed potential head was obtained from a steady state flow simulation while a concentration equal to zero over the entire domain has been assumed.

An Euler back stepping scheme was used for the temporal integration of both flow and transport equations, while the time step size was chosen equal to 1 day.

5 Test case results

5.1 Test case A: influence of sloping bottoms vs homogeneous or heterogeneous domains, when different flow boundary conditions are considered.

Monitoring field data [*Mironenko et. al.*, 1986] from the Lake Karachai area seem to indicate the presence of a sloping bottom in the aquifer; this may explain the movement of the contaminant plume in a direction not consistent with the flow field. Thus we verified the possibility of simulating such a behaviour by means of the CODESA-3D code.

Figures **??**.a, **??**.b, **??**.c, and **??**.d show the results obtained when considering the domain completely impermeable (i.e., no-flow conditions on sides AF, AB, BC, DC of the sketched domain of Figure **??**). Figure **??**.a shows the plume of contaminant after 10,000 days for the case of a flat bottom of the domain under study. The plume was symmetrical and it widened or shrunk when encountering a zone of either smaller or larger permeability, respectively. In Figures **??**.b, **??**.c, and **??**.d a non-flat bottom was considered. In the case of Figure **??**.b, a slope of 0.2^o was imposed, while in the case of Figure **??**.c, it was increased to 1^o, and in the case of Figure **??**.d a slope of 5^o was considered. The simulated plume distribution appears significantly affected by the presence of a non-flat domain, expecially when a very sloping domain is considered.

Figures **??**.a, **??**.b, **??**.c, and **??**.d show the results when the domain is only partially impermeable, that is, impermeable on sides DE and EF of Figure **??**. On side AF it was assumed an equivalent pressure head equal to 0.0, on sides AB ($0 < x < 225$ m) and CD ($260 < x < 500$ m) a Neumann condition of prescribed flux $q_D = 5.0 \times 10^{-10}$ m^2/day, while on side BC ($225 < x < 260$ m) a flux $q_D = 7.0 \times 10^{-9}$ m^2/day. A slope of 0.2^o (Figure **??**.b) does not significantly affect the development of the contaminant plume (compare with Figure **??**.a), when $\theta = 0^o$): the contaminant tends to move in the flow direction. Further increasing the slope begins to affect the dense plume : at 1.0^o (Figure **??**.c) the plume motion downwards along the slope prevails, becoming the driving force at 5.0^o (Figure **??**.d).

The numerical instabilities are negligible, since the Peclet constraint is generally satisfied. The undershoot concentrations are always less than 10^{-5}, even when the domain is strongly deformed (e.g. $\theta = 5^o$). The average number of linear iterations for the flow equation per time step does not notably change if we consider or not flow boundary conditions. The average number of nonlinear iterations for solving the coupled system per time step is generally very small (2-3 iterations) while the number of average linear iterations for the flow equation per nonlinear iteration is about 100. Finally, the mean total CPU time for the simulations is about 2650 s (on IBM560 RISK/6000 workstations).

5.2 Testcase B: fingering effect as a function of the mesh size, time step, and density coefficient ϵ

In order to understand the influence of different temporal or spatial discretizations, or different values of the density coefficient ϵ on fingering patterns, three series of simulations have been considered.

In the first one, fingering is analyzed as a function of the time step size. At first, the "coarse mesh" domain described in subsection 4.2 is considered, and the concentration patterns are plotted after 500, 4000, and 8000 days, using different time steps, e.g.,$\Delta t = 50, 10, 1$ days, respectively (see Figure **??**). The numerical stability, here in terms of undershoots of the computed concentrations, is reported in Table 3, where $Figure$ refers to the plot of the concentration profile, N is the number of negative concentrations greater than 10^{-30}, C^* is the maximum negative concentration, and $\%N$ is the percentage of negative concentration values. A quite unexpected behavior appears from the analysis of the results summarized in Table 3: it appears that the best results are obtained with the largest time step, $\Delta t = 50$ days, rather than $\Delta t = 10$ days, or $\Delta t = 1$ day, although we observe that the percentage of undershoot concentrations can always be considered negligible. The concentration patterns at time $T = 500, 4000, 8000$ days, respectively, are shown in Figures **??**. Note that the Peclet constraint is widely satisfied, being $Pe \approx \frac{\Delta x}{\alpha_L} = 1$. When the contaminant plume reaches the bottom of the domain it begins to spread, as expected.

In a second series of simulations, fingering has been studied as a function of the spatial discretization, and crossed comparisons with different temporal discretizations have contributed to a better comprehension of the phenomena. Figure **??** (cases *a, c, e*) refers to the "coarse mesh" domain at time 500, 4000 and 8000 days, respectively, while Figure **??** (cases *b, d, f*) shows the corresponding results obtained with the "fine mesh" domain, as defined in subsection 4.2 . Here, the time step size is $\Delta t = 10$ days, and the density ratio coefficient ϵ is equal to 0.07. Fingering appears both in the "coarse" and "fine" mesh domain, although in this last case, where the numerical instability is lower (see Table 4), fingering is greater. It may be that fingering in the "coarse mesh" case is mainly due to numerical instabilities, while when the mesh is "fine" it is possible to better describe its physical component due to gravitational instabilities. The use of the smaller time step size, $\Delta t = 1$ day, futher stabilizes the numerical solution when the mesh is "fine", while there are not significant improvements when the mesh is "coarse" (see Table 5). On the other hand, the concentration patterns in Figure **??** show a smaller fingering when the mesh is "coarse", and a bigger one when the mesh is "fine" by comparing these results with those obtained with $\Delta t = 10$ days. Thus, it appears that the improvement of the temporal discretization mainly contributes to the numerical stability when a "fine" spatial discretization is already considered: the shape of the contaminant plume is significantly influenced, being increased the fingering effect in the "fine mesh" case (compare Figures **??**-cases *b, d, f* with Figure **??**-cases *b, d, f*), while, in the "coarse" mesh case, the smaller time step makes fingering less evident (compare Figures **??** (cases *a, c, e*) with Figures **??** (cases *a, c, e*).

In this context, *Bixio et. al.* have run a series of simulations using CODESA-3D to test the numerical solution for the case of the Elder's problem. They have reproduced the test case of *Voss and Souza* obtaining results in excellent agreements with those, and thus demonstrating the convergency of the fingered shape of the plume when the mesh size become sufficiently large. Because in their test case, Voss and Souza consider a density ratio equal to 0.2, Bixio et. al. have also demonstrated the numerical stability of CODESA-3D, even when the density ratio is that of a typical brine.

The effect of density ratio ϵ on fingering has also been analyzed. Thus, in the case of the "coarse mesh", Figure **??** shows the differences in the concentration patterns between the case with $\epsilon = 0.07$ (cases *a, c, e*) and $\epsilon = 0.02$ (cases *b, d, f*), when $\Delta t = 1$ day, and Table 6 shows the results in terms of numerical stability. The finer mesh plays a dominant role over the density ratio coefficient to create fingering of the plume.

Here, it is not not clear if fingering is essentially of numerical or physical nature, and little information is avaible in the literature. For example, *Kolditz et. al.*, (1996). have considered fingering dependecy on different levels of the density approximation but they did not study the influence on fingering of increasingly larger ϵ values. The gravitational instability increases with ϵ; moreover small ϵ values means small coupling between flow and transport processes (see equations (3)-(5)), and thus a more linear and less instable numerical solution. Indeed, what is observed is that, when ϵ is 0.02, the number of undershoot concentrations is greater with respect to ϵ equal to 0.07. Fingering appears to increase with ϵ, but it is not possible to distinguish the contributions of gravitational instabilities and numerical oscillations, this behavior being confirmed from further comparisons shown in Figures **??** and Figures **??**, where a fine mesh domain is considered, and calculations with $\epsilon = 0.07$ and $\epsilon = 0.02$ are done with two different temporal discretizations, $\Delta t = 10$ days (see Figures **??**), and $\Delta t = 1$ day, respectively (see Figures **??** and Table 7).

5.3 Test case C: simulations using Cauchy boundary conditions.

This simulation begins with the calculation of the steady state flow to ensure a stable starting flow field for the coupled flow-transport problem. In order to avoid the presence of y components of the velocity field on the lateral sides of the domain and z component at the bottom which could cause significant loss of mass by advection when the contaminant movement is also considered, the hydraulic conductivity along the y direction is set very small (10^{-6} m). The same artificial correction is taken for the z component at the bottom (see Table 8). Figure **??** shows the high directionality of the velocity field, which is essentially along the xz-plane, and is quite small (order of 10^{-3}m/day along x, 10^{-9}m/day along y, and 10^{-5}m/day along z when it reaches its maximum values) because of the small pressure gradient imposed. Figure **??** shows the steady state potential head and pressure. The pressure decrease at 3300 m is caused by the imposition of the condition $h = 92$ m, which makes the corresponding point C of Figure **??** a sink point.

After 20 years the coupling between flow and transport has significantly perturbed the velocity field (see Figure **??**). The potential head, pressure and contaminant concentration profiles after 20 years are shown in Figure **??**. The effect of dispersion masks the spreading of the contaminant plume when crossing zones of different permeability.

To simulate the spreading of the contaminant plume when its immission is interrupted, a Cauchy boundary condition of total flux equal to zero has been imposed at the top surface of the domain, leaving all the other boundary conditions and parameters unchanged. The contaminant begins to spread, and Figure **??** shows its situation after 40 and 60 years. In order to establish the variations of contaminant mass inside the domain during the simulation, we have used a simple procedure, although with strong limitations on its applicability; that is, the mass content at a prescribed time t is evaluated by a summation of the nodal concentrations (absolute values) times the cell-volume associated to each node. Changes in mass content within a given period of time are easily evaluated, being, of course, sure that in the meantime the contaminant has not escaped from the domain. In this sense, mass conservation is well satisfied, with a decrease of 0.4% from 20 to 40 years, and a further decrease of 2.5% from 40 to 60 years. The concentration map at $t = 100$ years (end of the simulation) is shown in Figures **??** and **??**.

These results (labelled as "case.steady") have been compared with those obtained from an analogous test case (labelled as "case.h85"), where the transient simulation begins with the initial condition of h=85 m over the entire domain, and no artificial constraints are imposed on the hydraulic conductivity to ensure a velocity field strictly directed along the xz-plane.

At t=20 years, the "case.steady" velocity field has a maximum v_y value of the order of 10^{-8} s, v_z at the bottom is of the order of 10^{-7} s, and the there not negative values of concentration greater than -10^{-6} (undershoot concentrations). On the other hand, the "case.h85" velocity field has a maximum v_y value of the order of 10^{-2} s, v_z at the bottom is of the order of 10^{-3} s, and there are 15 undershoot concentrations greater than -10^{-6}, which, although few in number, are a demonstration of a more unstable numerical solution. The not negligible velocity component along the y direction and along z at the bottom of the domain are responsible for the advective movement of the contaminant through the lateral boundaries, and through the bottom, which indeed should be impermeable, thus causing, at t=40 years, a loss of contaminant mass of 32%.

A sloping bottom has also been simulated. To this aim, three different slopes, namely 0.0^o, 0.1^o, and 0.2^o, have been considered. The concentration patterns at $t = 20, 40$, and 100 years are shown in Figure **??**. The contaminant tends to follow the slope, although the velocity pattern is directed in the opposite direction. This tendency becomes more evident as the slope increases. We note the effect of the Dirichlet flow boundary condition ($h = 92$ at point $x = 3300$ m) which results in a sink point for the contaminant. For a slope of 0.2^o, most of the contaminant has already exited the domain from the point $h = 92$ (see Figure **??**, case i) The influence of the condition $h = 92$ m on the brine has been verified by removing that condition leaving all the others untouched. Figure **??** shows the comparison of the concentration maps at 20 and 40 years: in this case the plume moves towards the permeable boundary (side FH of Figure **??**) and exits the domain.

6 Conclusions and future work

The main purpose of this preliminary study was to verify the capability of the CODESA-3D code to simulate the transport of a very dense contaminant plume (density ratio coefficient $\epsilon = 0.07$) in a saturated porous medium under different sets of conditions. Also, we were interested in examining the effects of the sloping bottom of an aquifer on plume distribution.

Three series of test cases, differing in domain size, boundary conditions, time stepping, permeability, dispersivity, and density coefficient have been analyzed.

From the first series of simulations (on a small domain), some general features of the phenomena under study have been identified. The effect of the slope angle of the aquifer bottom was quite evident. The movement of the contaminant was correct described in so far as being affected by both the slope and the flow direction. The longitudinal and transverse dispersivities were set (according to field measurements) to 5 and 1 m, respectively. Since the horizontal and vertical node spacing is lower than 6 m, an approximate evaluation of the Peclet number yielded $Pe < 2$. This should have guaranteed that our simulations were performed within the range of numerical stability.

In a second series of simulations we have found that fingering slightly increases with the time step size, while it is notably affected by the spatial discretization. In this last case, as also documented in the literature, it is possible to reach a convergence; that is, further refinements do not further contribute to create fingering. The influence of the parameter ϵ is ambiguous the gravitational instabilities increase with ϵ; however, although the coupling between flow and transport equations decreases with ϵ, making the system more linear, we have found an increase in numerical instabilities as ϵ decreases.

The last series of tests was focused on the simulation of the fate of a contaminant inside the domain, when, after a prescribed period of time, its immission is interrupted. To simulate this behavior, a Cauchy boundary condition of total flux zero was imposed at the top surface of the domain. A proper parameter setup has been necessary to ensure a high directionality of the velocity field and to avoid contaminant losses from lateral boundaries of the domain, such as the imposition of a thin layer at the bottom of the domain with a very small vertical hydraulic conductivity K_z, and a very small K_y component over the entire domain. The sink point, with the Dirichlet flow boundary condition $h = 92$, plays a foundamental role on the contaminant movement, because of the shape assumed by the velocity field, which tends to split the plume into two "branches". This is also confirmed by further investigation, when, for example, we eliminate such boundary conditions and consider a strong slope ($\theta = 0.2^o$) at the bottom: the contaminant plume migrates toward the boundary FH.

Further CODESA-3D simulations and a number of code upgrades are planned in the next phase of the RaCoS project:

- **Dual-Porosity transport** CODESA-3D will be provided with a dual-porosity transport module for handling nonequilibrium sorption in the transport of contaminants (*Gallo et al.*, 1996) . Field work at Lake Karachai has shown that in some cases this can be important.

- **Viscosity** The CODESA-3D code was born as a seawater intrusion code in which salt concentrations are not normally larger than 35 g/l. The Lake Karachai problem involves the simulation of brine transport (about 150 g/l of salt concentration). In such a case, the mathematical formulation must be modified to include the effect of solute concentration on brine viscosity and a term containing the concentration gradient in the flow equation which is currently neglected (*Kolditz et al.*, 1998) .

- **Radionuclide transport** Radionuclide transport is the focus of the RaCoS project. Several radionuclides are present in the contaminated plume, but only Sr (Strontium), which is the one at highest concentration and, then is the real problem for the site under study, will be modeled, at least at the first stage of the work. Strontium undergoes nonequilibrium and brine-dependent sorption, and its fate will be described by means of an *ad-hoc* set of computational module that will be integrated in the dual-porosity transport code.

- **Applications** The application of the CODESA-3D code described in this reporthas been done on a very simplified scenario. Further simulations will include the study of the Lake Karachai site using regional modeling (horizontal 2-D simulations) for reproducing the general pattern of the piezometry. This will be used as boundary conditions for 3-D simulations that will be run on a subdomain surrounding the radioactive disposal for which a better geological/hydrological characterization is available. These simulations will be aimed at evaluating the brine transport from the '60s to the '90s (current situation) and at calibrating model parameters. Then, on the basis of this calibration phase, different scenarios will be simulated, together with some possible interventions for evaluating the possibility to contain/deviate the radioactive plume.

- **GIS support** The application of CODESA-3D to real field scenarios will imply an intense use geographic information systems (GIS), particularly in relation with the transfer of "information" regarding soil characteristics, natural recharge, boundary conditions, etc. from the database into the numerical model. In this context, an interface between the GIS and CODESA-3D is under development.

Acknowledgements

This work has been financially supported by EC, Inco-Copernicus program (contract N^o ERBIC15CT960211).

Table 1. Test case A. Material and solute properties.

Zone	Range (m)	K_{xx} (m/day)	K_{yy} (m/day)	K_{zz} (m/day)	S_s (m^{-1})	α_L (m)	α_T (m)	n
1	0-20	0.1	0.1	0.05	10^{-5}	5.0	1.0	0.025
2	20-50	1.0	1.0	1.0	10^{-5}	5.0	1.0	0.025
3	50-100	0.3	0.3	0.05	10^{-5}	5.0	1.0	0.025

Table 2. Test case B. Material and solute properties.

K_{xx} (m/day)	K_{yy} (m/day)	K_{zz} (m/day)	S_s (m^{-1})	α_L (m)	α_T (m)	n
0.3	0.3	0.05	10^{-2}	5.0	1.0	0.025

Table 3. Test case B. Coarse mesh. Undershoot concentrations with different time steps.

Figure	N	C^*	$\%N$
??.a ($\Delta t = 50$ days, $t = 500$ days)	161	-1.7×10^{-2}	1.79
??.b ($\Delta t = 10$ days, $t = 500$ days)	224	-6.6×10^{-2}	2.49
??.c ($\Delta t = 1$ days, $t = 500$ days)	179	-4.6×10^{-2}	1.79
??.d ($\Delta t = 50$ days, $t = 4000$ days)	31	-1.7×10^{-2}	0.34
??.e ($\Delta t = 10$ days, $t = 4000$ days)	111	-5.4×10^{-2}	1.23
??.f ($\Delta t = 1$ days, $t = 4000$ days)	30	-9.9×10^{-2}	0.33
??.g ($\Delta t = 50$ days, $t = 8000$ days)	0	-1.4×10^{-4}	0.00
??.h ($\Delta t = 10$ days, $t = 8000$ days)	4	-2.1×10^{-2}	0.04
??.i ($\Delta t = 1$ days, $t = 8000$ days)	7	-7.6×10^{-2}	0.08

Table 4. Test case B. Undershoot concentrations from a comparison coarse mesh vs fine mesh. $\Delta t = 10$ days.

Figure	N	C^*	$\%N$
??.a (Coarse mesh, $t = 500$ days)	224	-6.6×10^{-2}	2.49
??.b (Fine mesh, $t = 500$ days)	1007	-3.65×10^{-2}	2.24
??.c (Coarse mesh, $t = 4000$ days)	111	-5.4×10^{-2}	1.23
??.d (Fine mesh, $t = 4000$ days)	0	-1.15×10^{-7}	0.00
??.e (Coarse mesh, $t = 8000$ days)	4	-2.1×10^{-2}	0.04
??.f (Fine mesh, $t = 8000$ days)	0	-5.56×10^{-8}	0.00

Table 5. Test case B. Undershoot concentrations from a comparison coarse mesh vs fine mesh. $\Delta t = 1$ days.

$Figure$	N	C^*	$\%N$
??.a (Coarse mesh, t = 500 days)	179	-4.58×10^{-2}	1.99
??.b (Fine mesh, t = 500 days)	279	-4.66×10^{-3}	0.62
??.c (Coarse mesh, t = 4000 days)	30	-9.92×10^{-2}	0.33
??.d (Fine mesh, t = 4000 days)	0	-1.00×10^{-30}	0.00
??.e (Coarse mesh, t = 8000 days)	7	-7.60×10^{-2}	0.08
??.f (Fine mesh, t = 8000 days)	0	-1.00×10^{-30}	0.00

Table 6. Test case B. Coarse mesh. Undershoot concentrations comparison with $\epsilon = 0.07$ and $\epsilon = 0.02$. $\Delta t = 1$ days.

$Figure$	N	C^*	$\%N$
??.a ($\epsilon = 0.07$, t = 500 days)	179	-4.58×10^{-2}	1.99
??.b ($\epsilon = 0.02$, t = 500 days)	187	-1.00×10^{-1}	2.08
??.c ($\epsilon = 0.07$, t = 4000 days)	30	-9.92×10^{-2}	0.33
??.d ($\epsilon = 0.02$, t = 4000 days)	129	-6.73×10^{-2}	1.43
??.e ($\epsilon = 0.07$, t = 8000 days)	7	-7.60×10^{-2}	0.08
??.f ($\epsilon = 0.02$, t = 8000 days)	116	-9.20×10^{-2}	0.00

Table 7. Test case B. Fine mesh. Undershoot concentrations with $\epsilon = 0.07$ and $\epsilon = 0.02$. $\Delta t = 1$ days.

$Figure$	N	C^*	$\%N$
??.a ($\epsilon = 0.07$, t = 500 days)	279	-4.66×10^{-3}	0.62
??.b ($\epsilon = 0.02$, t = 500 days)	1669	-6.82×10^{-2}	3.72
??.c ($\epsilon = 0.07$, t = 4000 days)	0	-1.00×10^{-30}	0.00
??.d ($\epsilon = 0.02$, t = 4000 days)	7	-1.24×10^{-3}	0.02
??.e ($\epsilon = 0.07$, t = 8000 days)	0	-1.00×10^{-30}	0.00
??.f ($\epsilon = 0.02$, t = 8000 days)	0	-3.750×10^{-4}	0.00

Table 8. Test case C. Material and solute properties.

Zone	Range (m)	K_{xx} (m/day)	K_{yy} (m/day)	K_{zz} (m/day)	S_s (m^{-1})	α_L (m)	α_T (m)	n
1	0-3	0.1	1.0^{-6}	1.0^{-6}	10^{-2}	50.0	10.0	0.025
1	3-40	0.1	1.0^{-6}	0.05	10^{-2}	50.0	10.0	0.025
2	40-70	1.0	1.0^{-6}	1.0	10^{-2}	50.0	10.0	0.025
3	70-85	0.3	1.0^{-6}	0.05	10^{-2}	50.0	10.0	0.025

References

[1] J. Bear. *Hydraulics of Groundwater.* McGraw-Hill, New York, 1979.

[2] A. Mazzia, and A. C. Bixio. Strongly coupled flow and transport problems: validation of the satc3d code and other numerical developments. Technical report, Dipartimento di Metodi e Modelli Matematici per le Scienze Applicate, Padua University, 1998.

[3] E. O. Frind. Simulation of long-term transient density-dependent transport in groundwater. *Adv. Water Resour.*, 5:73–88, 1982.

[4] G. Galeati, and G. Gambolati. On boundary conditions and point sources in the finite element integration of the transport equation. *Water Resour. Res.*, **25**(5), 847–856, 1989.

[5] G. Gambolati, C. Paniconi, and M. Putti. Numerical modeling of contaminant transport in groundwater. In D. Petruzzelli and F. G. Helfferich, editors, *Migration and Fate of Pollutants in Soils and Subsoils*, volume 32 of *NATO ASI Series G: Ecological Sciences*, pages 381–410, Berlin, 1993. Springer-Verlag.

[6] G. Gambolati, M. Putti, and C. Paniconi. Three-dimensional model of coupled density-dependent flow and miscible salt transport in groundwater. In J. Bear, A. H.-D. Cheng, I. Herrera, S. Sorek, and D. Ouazar, editors, *Seawater Intrusion in Coastal Aquifers: Concepts, Methods, and Practices.* Kluwer Academic, Dordrecht, Holland, 1999.

[7] S. M. Hassanizadeh and T. Leijnse. A non-linear theory of high-concentration-gradient dispersion in porous media. *Adv. Water Resour.*, 18(4):203–215, 1995.

[8] P. S. Huyakorn, P. F. Andersen, James W. M., and H. O. White. Saltwater intrusion in aquifers: Development and testing of a three-dimensional finite element model. *Water Resour. Res.*, 23(2):293–312, 1987.

[9] O. Kolditz, R. Ratke, H. J. G. Diersch, and W. Zielke. Coupled groundwater flow and transport: 1. Verification of variable density flow and transport models. *Adv. Water Resour.*, 21(1):27–46, 1996.

[10] O. Kolditz, R. Ratke, H. J. G. Diersch, and W. Zielke. Coupled groundwater flow and transport: 1. Verification of variable density flow and transport models. *Adv. Water Resour.*, 21(1):27–46, 1998.

[11] C. M. Oldenburg and K. Pruess. Dispersive transport dynamics in a strongly coupled, groundwater-brine flow system. *Water Resour. Res.*, 31(2):289–302, 1995.

[12] P. Perrochet and D. Bérod. Stability of the standard Crank-Nicolson-Galerkin scheme applied to diffusion-convection equation: Some new insights. *Water Resour. Res.*, 29(9):3291–3297, 1993.

[13] W.R. Souza. Documentation of a graphical display program for the saturated-unsaturated transport (SUTRA) finite-element simulation model. Technical report, U.S. Geological Survey, Water-Resources Investigations Report 87-4245, 1987.

[14] V. A. Mironenko, and V. G. Rumynin. *Field tests and ground water quality monitoring in aquifers.* , Nedra, Moscow, 1986.

[15] C. I. Voss and W. R. Souza. Variable density flow and solute transport simulation of regional aquifers containing a narrow freshwater-saltwater transition zone. *Water Resour. Res.*, 23(10):1851–1866, 1987.