

NUMERICAL SIMULATION OF THE IMPACT OF WATER-AIR FRONTS ON RADIONUCLIDE PLUMES IN HETEROGENEOUS MEDIA

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ABSTRACT

The goal of this paper is to investigate the interaction of water-air fronts with radionuclide plumes in unsaturated heterogeneous porous media. This problem is modeled by a system of equations that describes both water-air flow and radionuclide transport. The water-air flow problem is solved numerically by a mixed finite element combined with a non-oscillatory central difference scheme. For the radionuclide transport equation we use the Modified Method of Characteristics (*MMOC*). We present results of numerical simulations for heterogeneous permeability fields taking into account sorption effects.

Keywords: central difference scheme, Modified Method of Characteristics, mixed finite element method, radionuclide contamination, unsaturated porous media

NOMENCLATURE

c	radionuclide concentration, Bq/m ³
D	dispersion coefficient tensor, m ² /s
g	gravitational constant, m/s ²
h	length of an grid element, m
k	absolute permeability, m ² /s
k_d	distribution coefficient, m ³ /kg
k_{eff}	effective permeability, m ² /s
k_r	relative permeability, m ² /s
M	average absolute permeability, m ² /s
p	partial pressure, N/m ²
P	global pressure
R	retardation factor
s	saturation
S	strength of heterogeneity
t	time, s
u	flow velocity, m/s
x, y	cartesian coordinates, m

Greek symbols

β	edge of a grid element
γ	radionuclide decay constant, 1/s
θ	volumetric content
λ	total mobility
μ	fluid kinematic viscosity, m ² /s
ρ	density, kg/m ³
ϕ	porosity

Subscripts

a	air phase
s	solid matrix phase
w	water phase

INTRODUCTION

An important problem arises when a nuclear waste repository is located in subsurface media: the issue of an accidental release of radioactive material through the hydrological environment. Since this complex environment offers difficulties in the prediction of the movement of the radioactive contaminant, investigations concerning the radionuclide transport in unsaturated porous media must be performed to address adequately the problem.

We present a new numerical procedure developed for the problem of two-phase flow with radionuclide transport in an unsaturated two-dimensional porous medium. The immiscible and incompressible phases are water and air. The unsaturated flow and radionuclide transport problems can be described by two coupled sets of equations; one set models the two-phase flow and another set governs the contaminant transport within the water phase. This approach was used by Binning (1994).

The two-phase flow system is expressed in terms of an elliptic equation for a global pressure coupled to a nonlinear conservation law for the water saturation. We solve such system of equations using a precise numerical method which combines a mixed finite element method for the global pressure problem with a second order, non-oscillatory, central finite difference scheme for the conservation law associated with the water saturation; this numerical procedure was carefully validated by Aquino (2003). The two-phase flow solution displays a sharp water-air front that represents the transient water infiltration in the unsaturated zone.

Contaminant transport is typically described by convection-diffusion equations that are dominated by their hyperbolic term. Numerical solution of hyperbolic equations by first order finite difference schemes may introduce numerical diffusion near sharp fronts; some other methods, based on straightforward higher-order finite differences may introduce oscillations in the

numerical solutions. In the case of radionuclide transport, a linear convection-diffusion equation is associated to the radionuclide concentration. We solve this equation using the original *MMOC* procedure (Douglas and Russell, 1982) for the time discretization; a mixed finite element method is used in the approximation of the diffusive effect and domain decomposition iterations (Douglas *et al.*, 1993) are used to handle the resulting linear algebraic problems.

In this paper we perform numerical experiments in order to investigate the interaction of sharp water-air fronts with radionuclide plumes. We will apply our numerical procedure to simulate a hypothetical scenario of an accident involving nuclear contamination. In this accident the radioactive material is dissolved in water and this contaminated solution is transported by the water infiltration in the unsaturated zone. The numerical experiments consider heterogeneous media and sorption effects of the porous medium on the radionuclide.

GOVERNING EQUATIONS

We study a model that describes water and air flow with contaminant transport in unsaturated heterogeneous porous media. In this section the governing equations are presented. We make the following assumptions:

- The fluids are immiscible and incompressible.
- The media are isothermal.
- The water-air flow is independent of the contaminant concentration.

Water-air Flow System

Let us describe our model for the water-air flow problem. The equation for the water-phase flow in unsaturated porous media takes the following form (Douglas *et al.*, 2000):

$$\frac{\partial}{\partial t} (\phi s_w) + \nabla \cdot (\mathbf{u}_w) = 0 \tag{1}$$

where ϕ is the porosity and s_w the water-phase saturation. In Eq. (1) \mathbf{u}_w is the water-phase velocity, m/s, given by

$$\mathbf{u}_w = -k \frac{\nabla p_w}{\mu_w} - \alpha \mathbf{g} \tag{2}$$

where k is the absolute permeability, m²/s; \mathbf{g} is the gravitational constant, m/s²; p_w is the water-phase partial pressure, N/m²; and α is the soil depth, m. The total mobility is defined as $k_{ra}/\mu_a + k_{rw}/\mu_w$ where k_r is the relative permeability, m²/s; μ is the viscosity, m²/s; the relative mobility is defined as k_r/μ with w (water) or a (air). We assume that the medium is saturated by water and air, $s_a + s_w = 1$.

Thus, a bit of algebraic manipulation with Eq. (1) leads to the global pressure equation (Douglas *et al.*, 2000)

$$\nabla \cdot (\mathbf{u}) = 0 \tag{3}$$

where

$$\mathbf{u} = -k \frac{\nabla P}{\mu_a} - \alpha \mathbf{g} \tag{4}$$

where P is the global pressure. By neglecting capillary pressure forces, the saturation equation for the water phase can be written as

$$\frac{\partial s_w}{\partial t} + \nabla \cdot (\mathbf{u}_w k_a) - \gamma = 0 \tag{5}$$

Radionuclide Transport Equation

We assume that a non-volatile radionuclide is transported only by the water phase. Thus, the governing equation of the radionuclide transport depends on the mass balance statement for the water phase.

Let c_w be the concentration of the radionuclide in the water phase and let c_s be the concentration in the solid matrix. Then we can write two balance equations involving these concentrations (Bear, 1979). Within the water phase,

$$\frac{\partial}{\partial t} (\theta_w c_w) + \nabla \cdot (\mathbf{u}_w c_w) - \nabla \cdot (\mathbf{D} \nabla c_w) - \gamma c_w = f(c_w, c_s) \tag{6}$$

and on the solid matrix,

$$\frac{\partial}{\partial t} (\theta_s c_s) + \nabla \cdot (\mathbf{u}_s c_s) - f(c_w, c_s) = 0 \tag{7}$$

where θ_w is the water volumetric content $\theta_w = \phi s_w$; γ is the radioactive decay constant, 1/s; \mathbf{D} is the dispersion coefficient tensor, m²/s; θ_s is the solid matrix volumetric content $\theta_s = \phi(1 - s_w)$; ρ_s is the density of the solid matrix, kg/m³; and $f(c_w, c_s)$ is the sorption term.

Assuming that the radionuclide concentration in water phase and the solid matrix are in equilibrium, this implies on $c_s = k_d c_w$, where k_d is a distribution coefficient.

The two equations above may now be simplified by adding them:

$$\frac{\partial}{\partial t} (\theta_w c_w + \theta_s k_d c_w) + \nabla \cdot (\mathbf{u}_w c_w) - \nabla \cdot (\mathbf{D} \nabla c_w) - \gamma c_w = 0 \tag{8}$$

By expanding the derivatives in the first and second terms of the Eq. (8), one gets

$$c_w \frac{\partial \theta_w}{\partial t} + \nabla \cdot (\mathbf{u}_w c_w) - \nabla \cdot (\mathbf{D} \nabla c_w) - \gamma c_w + \theta_w \frac{\partial c_w}{\partial t} + \nabla \cdot (\mathbf{u}_w c_w) - \nabla \cdot (\mathbf{D} \nabla c_w) - \gamma c_w = 0 \tag{9}$$

By making use of the mass balance Eq. (1), the first and second terms in the above equation vanish, and then the radionuclide transport equation is given by

$$R \frac{\partial c_w}{\partial t} + \nabla \cdot (\mathbf{u}_w c_w) - \nabla \cdot (\mathbf{D} \nabla c_w) - \gamma c_w = 0 \tag{10}$$

where $R = \theta_w + \theta_s k_d$ is defined as the retardation factor.

NUMERICAL SOLUTION OF THE WATER-AIR FLOW PROBLEM

The water saturation equation (5) is solved by a conservative, non-oscillatory, central scheme. This scheme is able to solve problems that show sharp fronts in their solution, without solving Riemann problems. The global pressure equation (3) - (4) is discretized by mixed finite elements that are suitable to compute accurately the relevant fluxes in heterogeneous permeability fields. The resulting algebraic problems are solved by a preconditioned gradient conjugated method. An operator splitting technique is introduced allowing for a sequential solution of saturation and global pressure problems (see Douglas *et. al.* (1997); Douglas *et. al.* (2000)).

Operator Splitting

The development of our numerical method will start from Eqs. (3) - (5). As we do not consider here the diffusive part of the saturation equation (because capillarity is neglected), we will only split the pressure calculation from the saturation calculation and we will be led to time steps satisfying $t_p = t_s$, where the subscripts p and s refer to pressure and saturation transport, respectively.

We employ locally conservative mixed finite elements to discretize Eqs. (3) - (4). The discrete equations arising from the application of the lowest index Raviart-Thomas space over squares for one element are (see Douglas *et. al.* (1997); Raviart and Thomas (1977)):

$$u = 0 \tag{11}$$

$$u = \frac{k_{eff}}{h} [P - P] \tag{12}$$

where $=L,R,B,T$ indicate the four edges of an element, $h = x = y$ is the length of an element side, $k_{eff} = 2k^L k^R / (k^L + k^R)$ is the effective permeability between two elements. We use a preconditioned conjugate gradient (PCG) method to solve the global pressure equation. The symmetric successive over-relaxation (SSOR) is used for preconditioning the conjugate gradient iterations.

Now let us turn to the Eq. (5). We present a numerical method to handle the model problem

$$\frac{s}{t} - \frac{f}{x} s - \frac{g}{y} s = 0 \tag{13}$$

as we apply to solve Eq. (5). The numerical method developed by Nessyahu and Tadmor (1990) to solve this problem use a second-order, conservative, non-oscillatory, central finite differencing scheme. Such scheme satisfies both the Total Variation Diminishing (TVD) property and a cell entropy inequality. Since these properties guarantee the convergence to unique entropy solution, the numerical method is able to capture the physically correct solution of the problem. (See the related mathematical analysis in the above mentioned paper.)

After discretization this scheme takes the following predictor-corrector form

$$s_{j,k}^{n+1/2} = s_{j,k}^n - \frac{1}{2} x f_{x,j,k}^n - y g_{y,j,k}^n \tag{14}$$

$$s_{j,1/2,k}^{n+1} = \frac{1}{4} s_{j,k}^n + \frac{1}{4} s_{j,k-1}^n + \frac{1}{4} s_{j-1,k}^n + \frac{1}{4} s_{j-1,k-1}^n - \frac{1}{16} s_{x,j,k}^n - \frac{1}{16} s_{x,j,k-1}^n - \frac{1}{16} s_{x,j-1,k}^n - \frac{1}{16} s_{x,j-1,k-1}^n - \frac{1}{16} s_{y,j,k}^n - \frac{1}{16} s_{y,j,k-1}^n - \frac{1}{16} s_{y,j-1,k}^n - \frac{1}{16} s_{y,j-1,k-1}^n - \frac{x}{2} f_{j,k}^{n+1/2} - \frac{x}{2} f_{j,k-1}^{n+1/2} - \frac{x}{2} f_{j-1,k}^{n+1/2} - \frac{x}{2} f_{j-1,k-1}^{n+1/2} - \frac{y}{2} g_{j,k}^{n+1/2} - \frac{y}{2} g_{j,k-1}^{n+1/2} - \frac{y}{2} g_{j-1,k}^{n+1/2} - \frac{y}{2} g_{j-1,k-1}^{n+1/2} \tag{15}$$

where $x = t_s / x$, $y = t_s / y$, and the numerical derivatives of the grid-functions s, f, g , are

$$\frac{1}{x} s_{x,j,k}^n = \frac{s_{j,k}^n - s_{j-1,k}^n}{x} \tag{16}$$

$$\frac{1}{y} s_{y,j,k}^n = \frac{s_{j,k}^n - s_{j,k-1}^n}{y} \tag{17}$$

To guarantee the desired non-oscillation property of these approximations our numerical derivatives should satisfy for every grid-function (Nessyahu and Tadmor, 1990)

$$s_{x,j,k}^n = \text{MinMod} \left[\frac{s_{j,k}^n - s_{j-1,k}^n}{x}, \frac{s_{j,k}^n - s_{j-1,k}^n}{x} \right] \tag{18}$$

where MinMod stands for a nonlinear limiter; an analogous expression can be written for the y -direction; and the $\text{MinMod}[\dots]$ stands for the usual limiter

$$\text{MinMod} \left[x, y, \frac{1}{2} \text{sgn} x - \text{sgn} y \right] = \text{Min} \left[|x|, |y| \right] \tag{19}$$

At each time level, we first reconstruct a piecewise-linear approximation from $\bar{s}(x,y,t) = s_{j,k}^n$ for $x_{j-1/2,k} < x < x_{j+1/2,k}$ and $y_{j-1/2,k} < y < y_{j+1/2,k}$ (here the over-bar denotes the cell average for $x_{j-1/2,k} < x < x_{j+1/2,k}$ and $y_{j-1/2,k} < y < y_{j+1/2,k}$); next, we evolve in time the piecewise-linear approximation $s_{j,k}^{n+1/2}$ and, finally, the resulting solution is projected back into the space of original piecewise-constant grid-functions $s_{j,k}^{n+1}$.

NUMERICAL SOLUTION OF THE RADIONUCLIDE TRANSPORT

We split the linear convection-diffusion problem associated with the transport of the radionuclide into convective and diffusion parts, using the MMOC in the time discretization. Mixed finite elements are used in the spatial discretization of the diffusion problem; the linear algebraic problems are handled by a domain decomposition iteration (Douglas *et. al.*, 1995).

Modified Method of Characteristics

We rewrite the radionuclide transport equation (10) in the following form

$$R_w \frac{c_w}{t} \mathbf{u}_w \cdot \mathbf{v} = g c_w \quad (20)$$

where $\mathbf{v} = \nabla D c_w$ and $g c_w = R_w c_w$.

Considering that the flow is essentially along the characteristic associated with the convection $R_w c_w / t \mathbf{u}_w \cdot \mathbf{v}$, it is appropriate to introduce differentiation in this characteristic direction. Let

$$R^2 = \left| \mathbf{u}_w \right|^2 \quad (21)$$

and

$$\frac{d}{dt} = R_w \frac{d}{dt} \mathbf{u}_w \quad (22)$$

and note that the direction $\frac{d}{dt}$ is a function of the water volumetric content and the water volumetric flux, which vary in space and time. It follows easily that the radionuclide transport equation can be written in the form

$$\frac{c_w}{t} \mathbf{v} = g c_w \quad (23)$$

The MMOC procedure for approximating the directional derivative $\frac{c_w}{t}$ is based on

$$\left(\frac{c_w}{t} \right)^{n+1} \mathbf{x} = R_w^{n+1} \frac{c_w^n \mathbf{x} - c_w^n \bar{\mathbf{x}}}{t} \quad (24)$$

which allows us to write a discretized form for the radionuclide transport equation

$$R_w^{n+1} \frac{c_w^n \mathbf{x} - c_w^n \bar{\mathbf{x}}}{t} = \mathbf{v}^{n+1} \cdot \mathbf{g} c_w^{n+1} \quad (25)$$

where $\mathbf{v}^{n+1} = \nabla D c_w^{n+1}$ and

$$\bar{\mathbf{x}} = \mathbf{x} - \frac{t}{R_w^{n+1}} \mathbf{u}_w^{n+1}$$

In this numerical procedure it is not necessary to use extrapolations to obtain c_w^{n+1} and \mathbf{u}_w^{n+1} since their values have already been known from the solution of the water-air flow equations.

NUMERICAL EXPERIMENTS

Our numerical experiments are performed in a two-dimensional domain $0, L_x \times 0, L_y$, with boundary conditions $p = 0$ on $y = L_y$, $\mathbf{u} \cdot \mathbf{n} = 0$ on $x = 0, L_x$ where \mathbf{v} is a unit vector normal to the domain boundary. The value of $\mathbf{u} \cdot \mathbf{n} = 0$ on $y = 0$ has to be specified.

We present the results of numerical experiments for a physical domain having 16 m x 32 m discretized by a 64 x 128 computational grid. Water from rainfall penetrates uniformly into the domain at a constant rate of 1.035×10^{-8}

m/s from the top. The following data are held fixed in our experiments: air viscosity $\mu_a = 1.78 \times 10^{-5}$ Pa.s, water viscosity $\mu_w = 1.14 \times 10^{-3}$ Pa.s, air density $\rho_a = 1.23$ Kg/m³, water density $\rho_w = 1.0 \times 10^3$ Kg/m³, porosity $\phi = 0.37$, air residual saturation $s_{ra} = 0.2$ and water residual saturation $s_{rw} = 0.0716$. The expressions of the relative permeability function for air and water can be found in Touma and Vauclin (1986).

The radionuclide considered here is the cesium-237 with an initial concentration of 1.0 Bq/m³ in a 4 m x 4 m region in the physical domain. This initial condition corresponds to a situation where there has been a contaminant release from the radioactive waste repository below the soil surface. In the simulations we use the following parameters: decay constant for the cesium-237 $\lambda = 7.33 \times 10^{-10}$ s⁻¹ and a constant dispersion coefficient $D = 1.5 \times 10^{-9}$ m²/s.

The numerical experiments use stochastic permeability fields to model heterogeneous formations. These permeability fields were taken to be realizations of self-similar (or fractal) Gaussian random fields (see Glimm et al. (1993) and references therein for the generation of such fields). We consider x to be one realization of such random field, and a log-normal permeability field which is given by $k(x) = M e^{Sx}$, where S is the strength of the heterogeneity and M is the average value for the absolute permeability. In all simulations we have $M = 1.974 \times 10^{-14}$ m².

Mass Balance Error

It is well known that the MMOC procedure is not able to conserve mass (Douglas et al., 1997), however for miscible displacement problems it has produced accurate numerical solutions with small mass balance errors (Russell and Wheeler, 1983). In order to investigate the influence of water-air fronts on the mass balance of the radionuclide transport, we simulate three physical situations: homogeneous saturated medium and both homogeneous and heterogeneous unsaturated media. The computational domain for the simulations in unsaturated media contains initially 20% water and 80% air. We do not consider either sorption effects or radionuclide decay in this investigation.

The value of the relative mass balance error ($\hat{\alpha}$) as a function of the time is shown in the Fig. (1). Results for the homogeneous saturated medium show that the radionuclide mass is adequately conserved with a relative error close to zero. However, for the other situations considered here, the relative error has an increase initially, followed by a decrease in absolute value when the radionuclide plumes are far from the water-air fronts. After a long time this relative error converges to values less than 20%.

The oscillation in the relative mass balance error occurs due to the delay in the radionuclide plume position when compared to the water-air front position (such behavior was also observed by Yeh et al. (1993) and Douglas et al. (1997)). Since the radionuclide mass is computed as the product of the water content by the radionuclide concentration, delayed plumes cause large errors in the mass balance. Such a delay is a consequence of the failure of the MMOC to locate precisely the position of the plume.

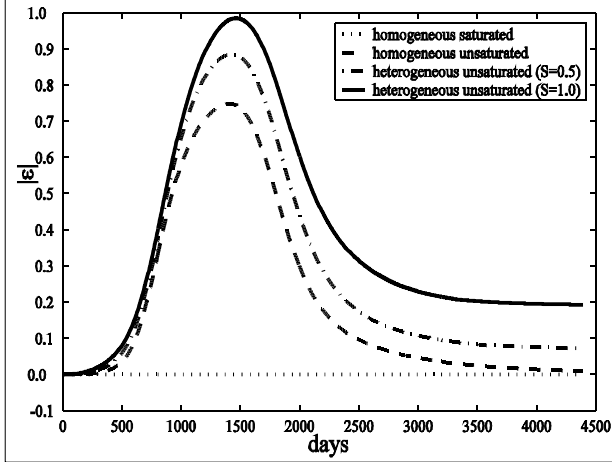


Figure 1. The relative mass balance error curves.

Weakly Heterogeneous Unsaturated Medium

We investigate the interaction of water-air fronts with radionuclide plumes by considering a permeability field with small variations by setting the strength of the heterogeneity to be $S = 0.5$. We also consider that the radionuclide decay is present in this situation. The pictures of water-air fronts and radionuclide plumes are illustrated in the Fig. (2).

Highly Heterogeneous Unsaturated Medium

Now we simulate our problem considering large variations in the permeability field. For this, the strength of the heterogeneity is set to be $S = 1.0$ and we maintain the radionuclide decay.

The relative mass balance errors produced by the *MMOC* procedure are higher in this heterogeneous medium. Such a medium generates a highly non-uniform velocity field that introduces some interpolation errors

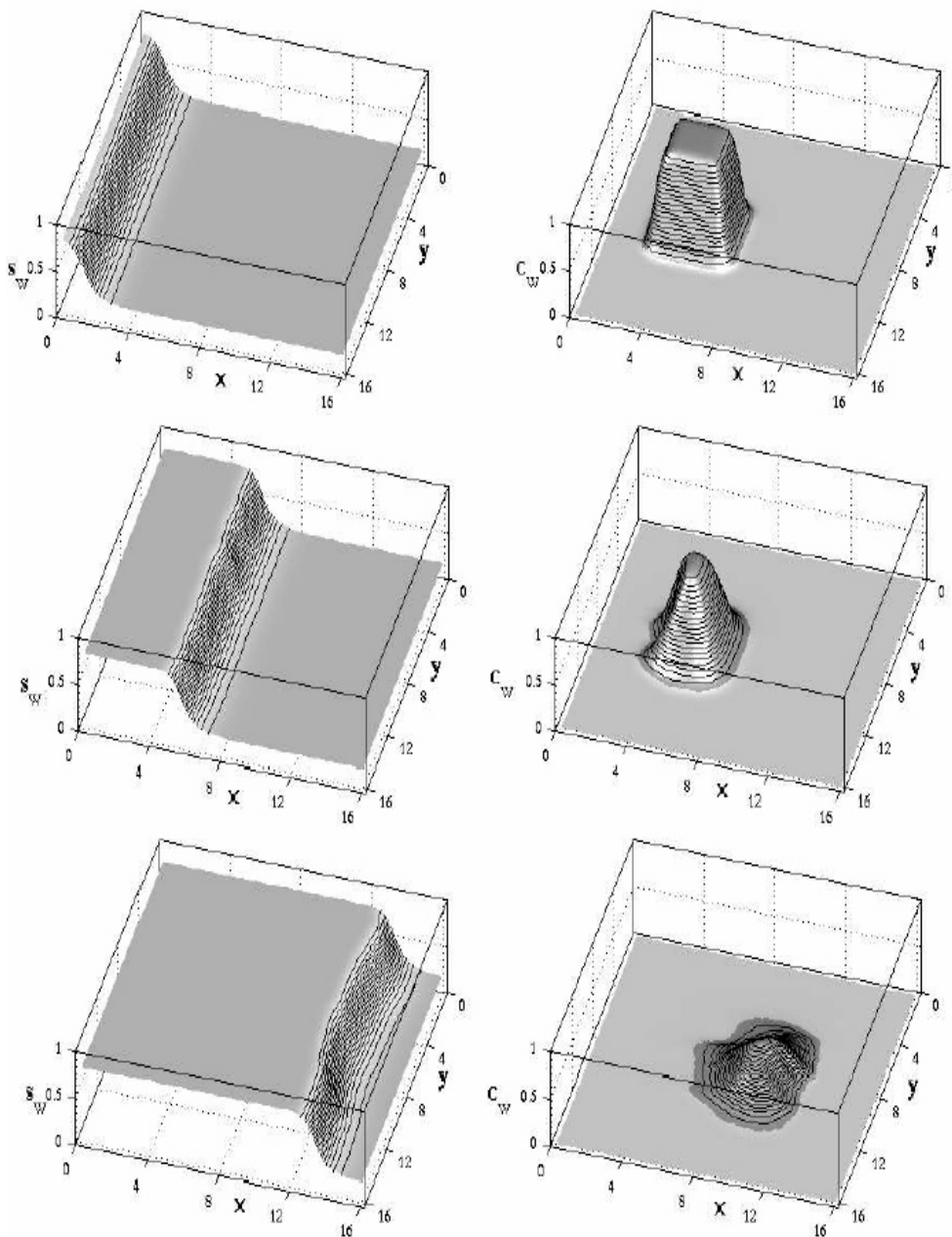


Figure 2. Numerical solution in a heterogeneous medium ($S = 0.5$) at 365, 1460 and 3285 days without sorption effects. Left: water-air front. Right: radionuclide plume.

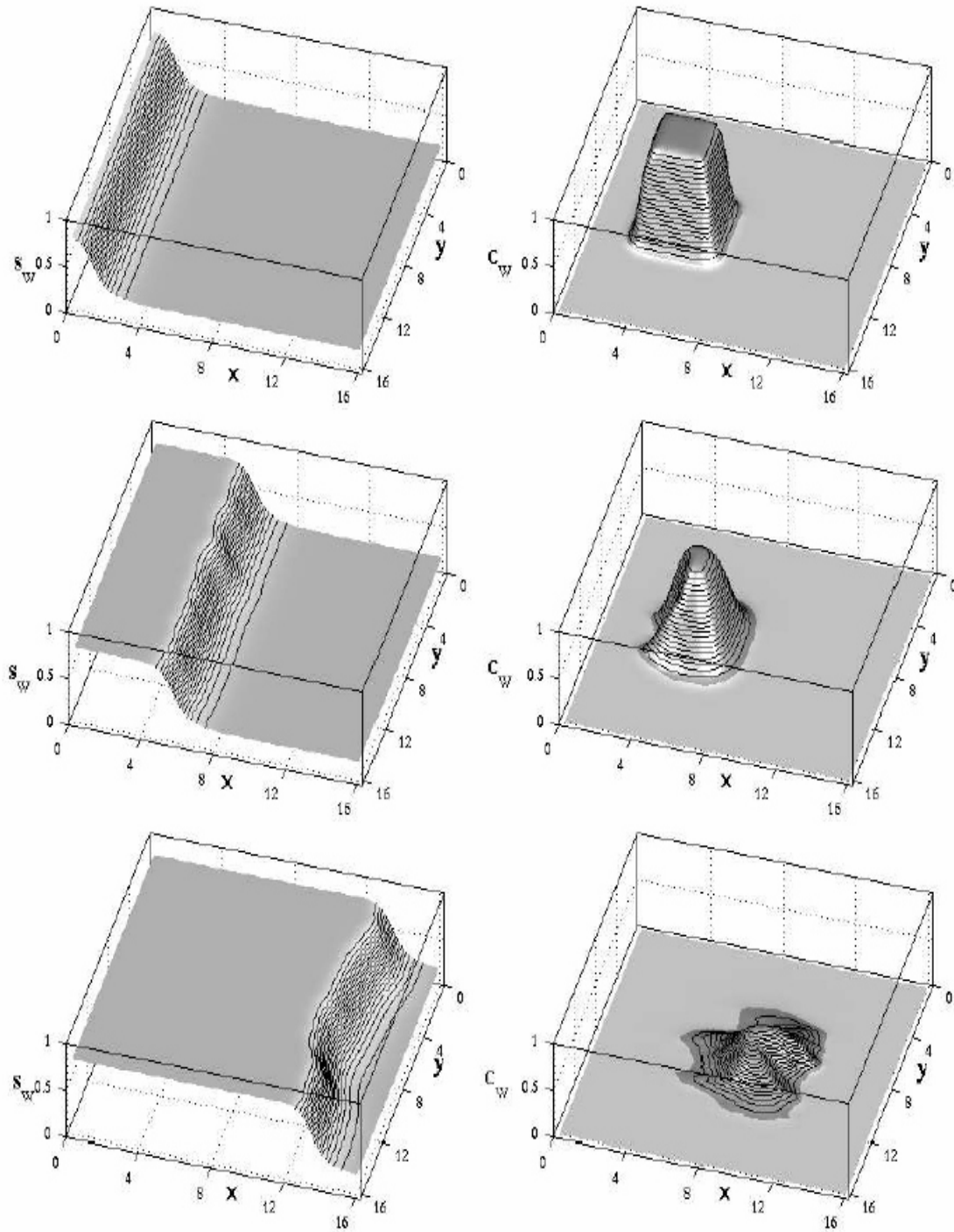
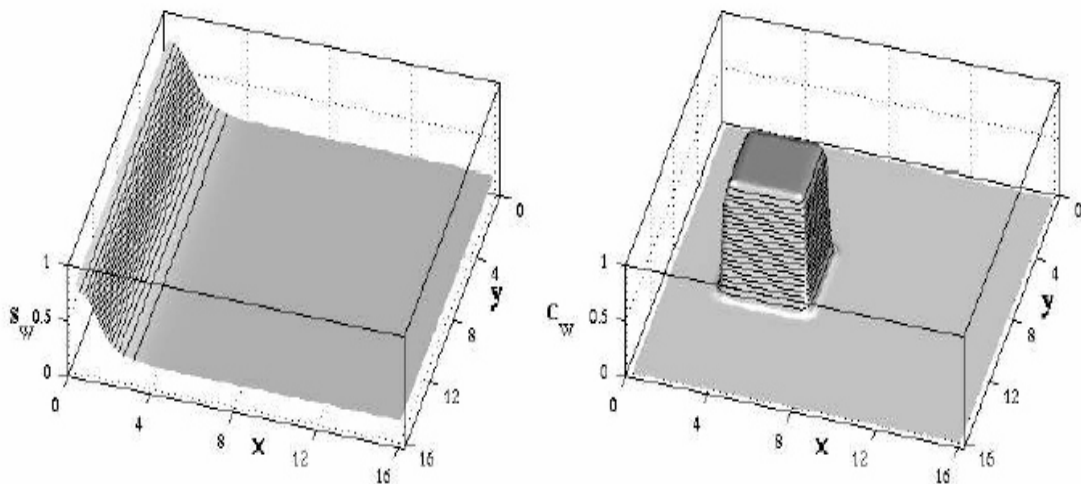


Figure 3. Numerical solution in a heterogeneous medium ($S=1.0$) at 365, 1460 and 3285 days without sorption effects. Left: water-air front. Right: radionuclide plume.



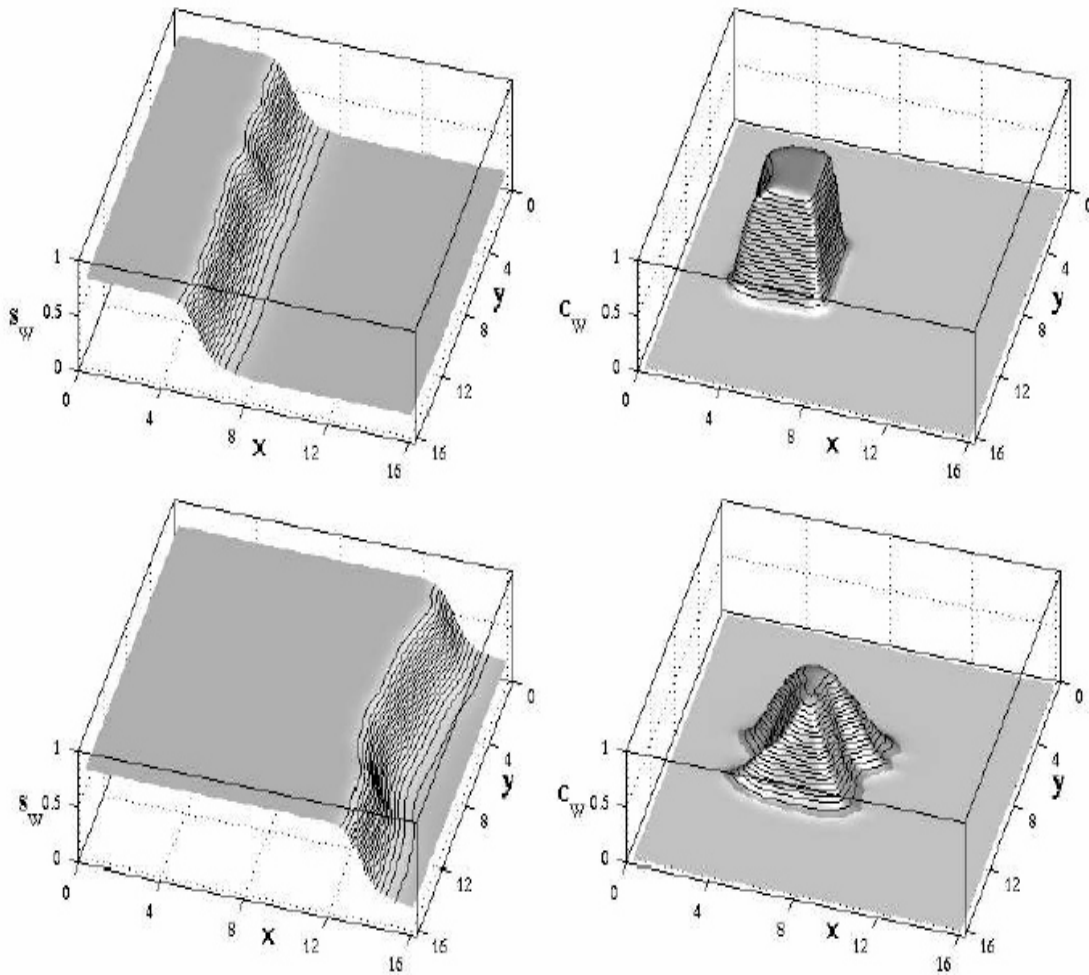


Figure 4. Numerical solution in a heterogeneous medium ($S = 1.0$) at 365, 1460 and 3285 days with sorption effects. Left: water-air front. Right: radionuclide plume.

during the *MMOC* back tracking. This causes numerical diffusion in the radionuclide plume simulation (Yeh *et al.*, 1993). Figure (3) displays the results of this study.

The Sorption Effect

Finally, we investigate the interaction of air-water fronts with radionuclide plumes considering heterogeneous unsaturated media, taking into account the sorption effect in the problem. In this simulation, the strength of the heterogeneity is $S = 1.0$ and the sorption term is $k_d = 0.5$. The result of the simulation is seen in the Fig. (4), where as expected we can observe that radionuclide plumes travel with a smaller velocity when the sorption effect is present.

SUMMARY AND CONCLUSIONS

A two-dimensional numerical method is described to simulate water-air flow and radionuclide transport in unsaturated porous media. Under transient water infiltration solutions of the water-air flow equations display sharp fronts which are propagated in the domain. As long as radionuclide plumes are reached by water-air fronts we find that the *MMOC* produces large mass balance errors.

To investigate the interaction of water-air fronts with radionuclide plumes numerical simulations were conducted considering heterogeneous permeability fields and sorption effects. Some conclusions based upon the various simulations can be summarized as follows:

- Our numerical scheme to solve the water-air flow problem is computationally efficient and is able to capture accurately sharp fronts in the solutions, even for large grid sizes (Aquino *et al.*, 2004). Thus, we can simulate sharp water-air fronts to investigate the behavior of radionuclide plumes;
- Under transient water infiltration the *MMOC* procedure produces large oscillations in the relative mass balance error due to the delay in the position of radionuclide plumes;
- We observed the highest relative mass balance errors for the simulations in highly heterogeneous media.

The authors are currently considering the following aspects of the problem at hand:

- The implementation of a Locally Conservative Eulerian-Lagrangian Method (Douglas *et al.*, 2000) for the purpose

of conserving the mass locally for the problem of radionuclide transport;

- The impact on the radionuclide plume of the stability-instability problem associated with water-air fronts subject to infiltration and evaporation; a stochastic framework is considered for heterogeneity modeling.

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