AN INTEGRAL TRANSFORM APPLICATION TO ENVIRONMENTAL IMPACT ASSESSMENT OF URANIUM MINING WASTE DISPOSAL

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Abstract. *In this paper we present hybrid numerical-analytical solutions for the transport of radioactive contaminant chains in the subsurface for environmental impact assessment related problems. The proposed model involves the advective-dispersive transport of multiple radionuclide species within separate but coupled saturated and unsaturated soil domains. The resulting partial differential equations are solved using the generalized integral transform technique (GITT) to yield analytical expressions for the concentration distributions versus distance, and analytical or numerical solutions as a function of time. The potential of the hybrid modeling approach is illustrated by means of an environmental impact assessment of an uranium mine liquid waste pond in Caetité, Brazil.*

Keywords*: Radionuclide Transport, Environmental Impact Assessment, Hybrid Methods, Integral Transforms*

1. INTRODUCTION

Regulatory bodies in most countries require a detailed environmental impact assessment when considering the disposal of solid and liquid wastes from industrial activities involving radioactive materials (IAEA, 1996; CNEN, 1998; Heilbron et al., 2000; Heilbron et al., 2004). The assessment typically includes an analysis of the transport of radionuclide decay chains through engineering barriers and the underlying soil and aquifer systems for both design and operation licensing purposes. Applications involve nuclear power production, electricity generation, petroleum and natural gas production, and mining and milling operations (Heilbron et al., 2002; Pontedeiro, 2006; Pontedeiro et al., 2007). Part of the environmental impact assessment generally pertains to the long-term effects to human health of the migration of the radionuclides in the geosphere through all possible pathways (soil, water, air). In order to simulate the transport of radionuclide decay chains in soils, one must solve the coupled system of transport equations for each species in the chain for the hydrologic and geochemical conditions of the specific site being considered. Several powerful and well-tested simulation tools are publicly or commercially available for this purpose (van Genuchten, 1985; Lung et al., 1987; Ungs et al., 1998; Simunek et al., 2005).

Classical integral transform methods during the past several decades have slowly evolved into a very flexible hybrid numerical-analytical approach that offers user-controlled accuracy and more efficient computational performance for a wide variety of *a priori* non-transformable problems (e.g., Cotta, 1993; Cotta & Mikhailov, 2006), including nonlinear mass transfer and heat and fluid flow applications related to environmental modeling (Cotta et al., 2003). Besides being an alternative computational method as such, the hybrid approach is particularly well suited for benchmarking purposes because of its automatic error control feature, and by retaining the same characteristics as a purely analytical solution. In addition to straightforward error control and estimation, an attractive aspect of the method is its straightforward extension to multi-dimensional situations with only a moderate increase in computational effort as compared to one-dimensional applications. Again, the hybrid nature of the solution process is responsible for this since the analytical part is employed for all but one independent variable, while the numerical part is used only for integration of a system of ordinary differential equations (ODE) in the one single independent variable. The hybrid method over the years has been applied to a large number of environmental problems (e.g., Cotta et al., 1998; Liu et al., 2000; Leal & Ruperti, 2001; Cotta et al., 2002; Heilbron et al., 2002; Cotta et al., 2003; Barros et al., 2006).

The Generalized Integral Transform Technique (GITT) is for these reasons used here in a hybrid numerical-analytical solution of the set of advection-diffusion equations describing the migration of radioactive decay chains in finite porous media, as implemented within the *Mathematica* 5.2 platform (Wolfram, 2005). The one-dimensional transient formulation is applicable to chains of any prescribed length, and to either imposed concentrations within the repository or as obtained from solution of the Bateman equations (van Genuchten, 1985). First- or third-type boundary conditions can be used for the leaching (inlet) boundary, and second- or third-type condition for the downgradient (outlet) side of the transport domain. The solution allows different transport and decay properties to be provided for each radionuclide. A simple filtering strategy can further be employed to homogenize the leaching boundary condition, whereas the NDSolve routine (Wolfram, 2005) is used to numerically solve the transformed ODE system. Each

transformed governing equation for the individual radionuclides may also be handled separately in a recursive implementation of the numerical solution of the ODE system. The present solution was tested against results by van Genuchten (1985) and Lung et al. (1987). Finally, an integral balance strategy is employed to enhance convergence behavior.

In this paper we apply the hybrid solution to four different scenarios for the migration of radioactive contaminants from liquid waste ponds at the Uranium mining facility of INB (Indústrias Nucleares do Brasil) located at Caetité, Bahia, Brazil. The solution procedure was selected because of our considerable experience with this class of methods, and because the approach is one of the recommended methods for radioactive waste safety assessment by the International Atomic Energy Agency (AIEA/ISAM) in 2004 (Heilbron et al., 2000, 2004).

2. ANALYSIS

Our conceptual model for the site involves a liquid waste pond, an underlying unsaturated soil zone through which water flows only vertically, a permanently saturated granular aquifer through which water flows horizontally, and a corresponding mixing zone where water from the unsaturated zone mixes with the saturated zone (Fig. 1). The recharge rate through the unsaturated zone into groundwater is defined by the long-term hydrologic characteristics of the site, thus disregarding operational conditions and the relatively short time period needed to fill the pond. Lateral flow through the aquifer is directly obtained from the corresponding hydraulic gradient and the hydraulic conductivity. The mixing zone is modeled assuming a certain penetration depth.

To solve the radionuclide transport problem, one-dimensional transient advection-diffusion equations are assumed for each of the N_r species in the radionuclide decay chain. The solution procedure is first described for the saturated medium. The governing transport equations for the individual species are given by:

$$R_i \frac{\partial C_i}{\partial t} + V \frac{\partial C_i}{\partial x} = D \frac{\partial^2 C_i}{\partial x^2} - \mu_i R_i C_i(x,t) + \mu_{i-1} R_{i-1} C_{i-1}(x,t), \; 0 \le x \le L, \; t > 0, \; i = 1,..,N_r \tag{1}$$

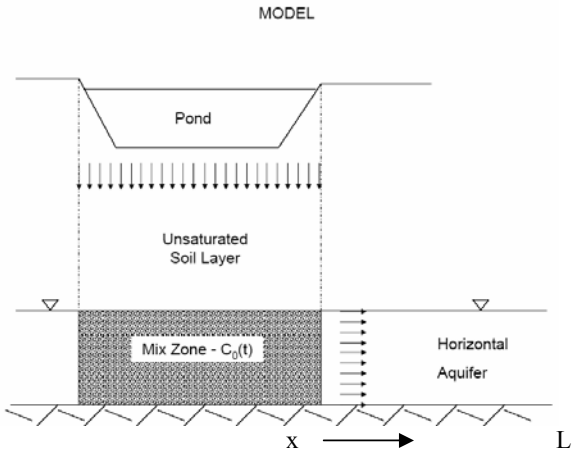


Figure 1 – Schematic representation of the adopted model for vertical infiltration in residual unsaturated soil coupled to the horizontal granular aquifer.

where C is concentration [Bq/m^3], t is time [year], x is distance [m], L is the length of the transport domain [m], V is the pore-water velocity [m/year], D is the dispersion coefficient [m^2/year], R is the retardation factor, μ is the decay rate [year^{-1}] (μ_0=0 for the first radionuclide), and the subscript i refers to the ith radionuclide in the decay chain. The associated initial and boundary conditions are:

$$C_i(x,0) = 0 \quad , \quad 0 \le x \le L \quad , \quad i = 1,..,N_r \tag{2}$$

$$-D \frac{\partial C_i}{\partial x} + V C_i = V f_i(t) \quad or \quad C_i = f_i(t) \quad , \quad x = 0 \quad , \quad t > 0 \quad , \quad i = 1,..,N_r \tag{3}$$

$$-D \frac{\partial C_i}{\partial x} + h C_i = 0 \quad , \quad x = L \quad , \quad t > 0 \quad , \quad i = 1,..,N_r \tag{4}$$

where $f_i(t)$ is the concentration of the ith radionuclide at the inlet. The boundary conditions at x=0 can be of a third or first type, while at the exit boundary conditions can be of a third or second type depending upon the selected value for the mass transfer coefficient h. Before applying the Generalized Integral Transform Technique (GITT), an analytical

filtering approach is first used to homogenize the boundary conditions at x=0, which will improve convergence of the proposed eigenfunction expansions. While several filtering methods are available for this purpose, here we use a very simple filter that employs the boundary source term concentration as follows:

$$C_i(x,t) = C_i^*(x,t) + f_i(t) \quad , \quad 0 \le x \le L \quad , \quad t > 0 \quad , \quad i = 1,..,N_r \tag{5}$$

The transient advection-diffusion problem with homogeneous boundaries can now be rewritten as:

$$R_i \frac{\partial C_i^*}{\partial t} + V \frac{\partial C_i^*}{\partial x} = D \frac{\partial^2 C_i^*}{\partial x^2} - \mu_i R_i C_i^* + \mu_{i-1} R_{i-1} C_{i-1}^* + g_i(t), \quad 0 \le x \le L, \; t > 0, \; i = 1,..,N_r \tag{6}$$

$$g_i(t) = -R_i \frac{df_i(t)}{dt} - \mu_i R_i f_i(t) + \mu_{i-1} R_{i-1} f_{i-1}(t) \quad , \quad t > 0 \quad , \; i = 1,..,N_r \tag{7}$$

$$C_i^*(x,0) = -f_i(0) \quad , \quad 0 \le x \le L \quad , \; i = 1,..,N_r \tag{8}$$

$$-D \frac{\partial C_i^*}{\partial x} + V C_i^* = 0 \quad , \quad x = 0; \qquad \frac{\partial C_i^*}{\partial x} = 0 \quad , \quad x = L \quad , \; t > 0 \quad , \; i = 1,..,N_r \tag{9}$$

To illustrate the remainder of the solution procedure, we use the above third type inlet and second type exit boundary conditions. The following auxiliary eigenvalue problem results:

$$D \frac{d^2 \varphi_n(x)}{dx^2} + \lambda_n^2 \varphi_n(x) = 0 \quad , \quad 0 < x < L \tag{10}$$

$$-D \frac{d\varphi_n(x)}{dx} + V \varphi_n(x) = 0 \; , \quad x = 0; \qquad \frac{d\varphi_n(x)}{dx} = 0 \; , \quad x = L \tag{11}$$

This Sturm-Liouville problem is readily solved for the eigenfunctions and the related eigenvalues and norms:

$$\varphi_n(x) = \cos\left(\frac{\lambda_n}{\sqrt{D}} x\right) + \frac{V \sin\left(\frac{\lambda_n}{\sqrt{D}} x\right)}{\lambda_n \sqrt{D}} \tag{12}$$

$$\lambda_n \sin\left(\frac{\lambda_n}{\sqrt{D}} L\right) = \frac{V \cos\left(\frac{\lambda_n}{\sqrt{D}} L\right)}{\sqrt{D}} \tag{13}$$

$$N_n = \int_0^L \varphi_n^2(x) dx = \frac{2V\lambda_n - 2V\lambda_n \cos\left(\frac{2\lambda_n}{\sqrt{D}} L\right) + \frac{\left(-V^2 + D\lambda_n^2\right) \sin\left(\frac{2\lambda_n}{\sqrt{D}} L\right)}{\sqrt{D}} + \frac{2L\lambda_n \left(V^2 + D\lambda_n^2\right)}{D}}{4\lambda_n^3} \tag{14}$$

The integral transform pair is next defined in terms of an eigenfunction expansion for the filtered concentrations, $C_i^*(x,t)$, by taking the eigenfunctions, $\varphi_n(x)$, as the basis for the expansions:

$$\overline{C}_{i,n}^*(t) = \int_0^L \varphi_n(x) C_i^*(x,t) dx \qquad \rightarrow \qquad \textit{Transform} \tag{15}$$

$$C_i^*(x,t) = \sum_{n=1}^{\infty} \frac{\varphi_n(x)}{N_n} \overline{C}_{i,n}^*(t) \qquad \rightarrow \qquad \textit{Inverse} \tag{16}$$

By implementing the above filtered problem using $\int_0^L \_\_\_\_ \varphi_n(x) dx$, the space variable x is eliminated, leading to the following transformed problem involving an infinite system of ordinary differential equations in time:

$$R_i \frac{d\overline{C}_{i,n}^*(t)}{dt} + V \sum_{m=1}^{\infty} a_{n,m} \overline{C}_{i,m}^*(t) = -\lambda_n^2 \overline{C}_{i,n}^*(t) - \mu_i R_i \overline{C}_{i,n}^*(t) + \mu_{i-1} R_{i-1} \overline{C}_{i-1,n}^*(t) + g_i(t) \overline{f}_n \quad , \tag{17}$$

$$t > 0 \quad , \quad i = 1,..,N_r \quad , \quad n = 1,2,...$$

where

$$a_{n,m} = \frac{1}{N_m} \int_0^L \varphi_n(x) \varphi_m'(x) dx \tag{18}$$

$$\overline{f}_n = \int_0^L \varphi_n(x)dx \tag{19}$$

and with the transformed initial conditions given by:

$$\overline{C}^*_{i,n}(0) = -f_i(0)\overline{f}_n = -\frac{\left[V - V\cos(\frac{Ll_n}{\sqrt{D}}) + \sqrt{D}\sin(\frac{Ll_n}{\sqrt{D}})l_n\right]f_i(0)}{l_n^2} \tag{20}$$

The above transformed system, while amenable of analytical treatment using the corresponding coefficient matrix eigensystem analysis, is most flexibly handled using the NDSolve routine (*Mathematica* 5.2) to generalize the solution procedure to unsaturated systems as will be discussed later. Once the transformed concentrations have been obtained numerically using *Mathematica*, an interpolating function is automatically generated to provide the time behavior in a continuous fashion. The inverse can then be recalled to explicitly provide the space dependence of the concentration fields.

For the more general situation of an unsaturated soil, the governing transport equations are given by:

$$\begin{aligned}&\frac{\partial[(\theta+\rho Kd_i)C_i]}{\partial t} + \frac{\partial(qC_i)}{\partial x} = \frac{\partial}{\partial x}(\theta D\frac{\partial C_i}{\partial x}) - \mu_i(\theta+\rho Kd_i)C_i(x,t)\\ &+\mu_{i-1}(\theta+\rho Kd_{i-1})C_{i-1}(x,t),\ 0\le x\le L,\ t>0,\ i=1,..,N_r\end{aligned} \tag{21}$$

where θ is the variable water content in the unsaturated zone, q is recharge rate given by the Darcy-Buckingham equation ($q=\theta V$), ρ is the soil bulk density, and Kd the distribution coefficient. For long-term simulations such as those considered here, in the order of thousands of years, a time invariable recharge rate can be assumed, in which case the water content will be a function of position only. The formulation then simplifies to:

$$\begin{aligned}&[\theta(x)+\rho Kd_i]\frac{\partial C_i}{\partial t} + q\frac{\partial C_i}{\partial x} = \alpha q\frac{\partial^2 C_i}{\partial x^2} - \mu_i[\theta(x)+\rho Kd_i]C_i(x,t)\\ &+\mu_{i-1}[\theta(x)+\rho Kd_{i-1}]C_{i-1}(x,t),\ 0\le x\le L,\ t>0,\ i=1,..,N_r\end{aligned} \tag{22}$$

where $\theta(x)$ is used to indicate that the water content is a function of the spatial coordinate, x. The corresponding initial and boundary conditions are:

$$C_i(x,0) = 0 \quad , \quad 0\le x\le L \quad , \quad i=1,..,N_r \tag{23}$$

$$-\alpha\frac{\partial C_i}{\partial x} + C_i = f_i(t) \quad or \quad C_i = f_i(t) \quad , \quad x=0 \quad , \quad t>0 \quad , \quad i=1,..,N_r \tag{24}$$

$$-\alpha\frac{\partial C_i}{\partial x} + h^* C_i = 0 \quad , \quad x=L \quad , \quad t>0 \quad , \quad i=1,..,N_r \tag{25}$$

in which we used the definition of the dispersion coefficient as the product of the dispersivity, α, and of the recharge pore-water velocity (i.e., $D=\alpha V$).

The integral transforms of the system of equations given by (22) to (25) are very similar to those of the saturated medium, and as such will not be repeated here. The only difference is that the integral transformation in this case generates other coupling terms, including the coupling matrix for the transient terms of the transformed concentrations. For an arbitrary distribution of the water content, $\theta(x)$, we may adopt a semi-analytical integration procedure to generate the coefficient matrix with substantial savings in computational effort. Also, the input profile for the water content depends on the solution of the nonlinear flow problem for the unsaturated zone as given by the Richards equation for steady-state flow. That initial profile was obtained using the HYDRUS-1D code of Simunek et al. (2005).

3. TESTS AND VALIDATION

The above hybrid numerical-analytical procedure was implemented in the *Mathematica* 5.2 system (Wolfram, 2005), including all of the symbolic calculations of the solution process. The resulting algorithm was tested against two codes developed previously for radionuclide decay chains: The LBL (Lawrence Berkeley Laboratory) computer code of Lung et al. (1987) and the CHAIN code of van Genuchten (1985). Solutions employed in these codes were derived using Laplace transform techniques as applied to semi-infinite media.

3.1. Comparison with the Lawrence Berkeley Laboratory code (Lung et al., 1987)

The LBL code deals with purely advective transport (no dispersion) of radionuclides in saturated semi-infinite media to allow for explicit analytical inversion of the Laplace transforms. We consider here the three element LBL chain example formed by U234, Th230, and Ra226 because of its similarity with the chain to be analyzed later for the Caetité site. The following parameter values were used: D = 50 m^2/year, θ= 3x10^{-3}, V = 1 m/year, R_1=120, R_2=1500,

R_3=300, μ_1= 2.806 10^{-6} year^{-1}, μ_2= 8.664 10^{-6} year^{-1}, and μ_3= 4.332 10^{-4} year^{-1}. The normalized concentrations at boundary x=0 are given by f_1=1, f_2=1, and f_3=10. Numerical results were obtained for the same time values (10 and 1000 years) as used by Lung et al. (1987), while we used a domain length L=200 m to avoid any effects of the exit boundary. Figure 2 compares calculated radionuclide concentration distributions for the two solutions at t=1000 years. Accounting for some loss in precision from reading the graphs in the report of Lung et al. (1987), the agreement is very good. While the GITT results were obtained for system truncation orders N<150, graphical convergence was attained much earlier. Figure 2 shows relatively rapid advancement of the contamination plumes for U234 and Ra226, which have much lower retardation coefficients in comparison with Th230. Table 1 illustrates the convergence behavior of the eigenfunction expansion for U234 at t=1000 years. The selected truncation orders were N=50, 75, 100, 125 and 150 for the whole solution domain. Convergence is overall very satisfactory, yielding five or six significant digits in the results. The GITT is then seen to be precise in predicting concentrations that are orders-of-magnitude smaller than those at the origin. This is important because utilities are usually required to satisfy environmental health criteria at the edge of their property, which in general involve much lower concentrations than at the source of contamination.

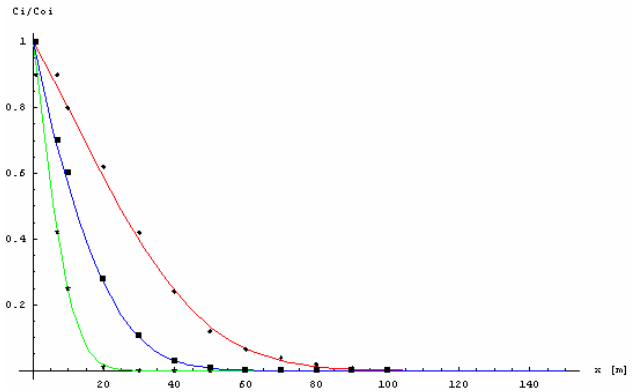


Figure 2 – Comparison of concentration distributions for three radionuclides, U234(right), Th230(left), Ra226(center), obtained with GITT (solid lines) and using Laplace transforms (symbols; Lung et al.,1987) at t=1000 years.

Table 1– Convergence of the GITT solution for the U234 concentration distribution at t=1000 years for the LBL test case of Lung et al. (1987) involving U234, Th230 and Ra226 (N=truncation order)

N	**50**	**75**	**100**	**125**	**150**
x [m]					
1	0.980879	0.980935	0.980955	0.980962	0.980963
10	0.797314	0.797306	0.797301	0.797300	0.797300
20	0.585814	0.585807	0.585811	0.585810	0.585810
30	0.393687	0.393696	0.393693	0.393694	0.393694
40	0.240582	0.240578	0.240579	0.240579	0.240579
60	0.0663583	0.0663609	0.0663615	0.0663618	0.0663619
80	0.0119769	0.0119747	0.0119757	0.0119756	0.0119755
100	0.00139205	0.00139003	0.00139092	0.00139085	0.00139068

3.2. Comparison with the CHAIN code of van Genuchten (1985)

The CHAIN code considers the advective-dispersive transport of a radionuclide decay chain of up to four species. For the present test we employed the four element chain with Pu238, U234, Th230, and Ra226, considered by van Genuchten (1985) using inlet boundary conditions obtained from the exact solution of the Bateman equations for radioactive decay in the waste site. We used the following parameter values: D = 10 m^2/year, V = 100 m/year, R_1 =10,000, R_2=14,000, R_3=50,000, R_4=500, μ_1=7.9x10^{-3} 1/year, μ_2=2.8x10^{-6} 1/year, μ_3=8.7x10^{-6} 1/year and μ_4=4.3x10^{-4} 1/year. Numerical results were obtained for t=10,000 years as presented by van Genuchten (1985), while a domain length, L, of 300 m was sufficient to warrant the semi-infinite medium assumption in our calculations. Table 2 illustrates the convergence behavior for Ra226 for this strongly advective case. The various columns show the GITT results for different truncation orders using increasing values of N. Also shown are the CHAIN results (last column) as obtained directly from the CHAIN output.

The results in Table 2 indicate that the convergence rate of the eigenfunction expansions is again very good, yielding four digit agreement with the CHAIN benchmark solution, except near the inlet boundary. Figure 3 shows the concentrations of the four elements at t=10,000 years. Notice that the Pu238 concentrations are not visible within the invoked log concentration range, and that Ra226 moves by far the fastest through the system because of its relatively low retardation factor.

Table 2– Convergence of GITT solution for the relatively concentration distribution of Ra264 at t=10,000 years (N=truncation order): Test case of van Genuchten (1985) for Pu238, U234, Th230, and Ra226.

N	50	75	100	125	150	CHAIN
x [m]						
10	1.3881 E-6	1.7003 E-6	1.6745 E-6	1.6573 E-6	1.6820 E-6	1.5111 E-6
20	3.5527 E-5	3.5934 E-5	3.5979 E-5	3.5977 E-5	3.5970 E-5	3.5898 E-5
30	8.9971 E-5	9.0234 E-5	9.0193 E-5	9.0203 E-5	9.0213 E-5	9.0103 E-5
40	1.4347 E-4	1.4352 E-4	1.4353 E-4	1.4353 E-4	1.4352 E-4	1.4343 E-4
50	1.9507 E-4	1.9499 E-4	1.9500 E-4	1.9500 E-4	1.9501 E-4	1.9491 E-4
60	2.4044 E-4	2.4036 E-4	2.4035 E-4	2.4035 E-4	2.4034 E-4	2.4029 E-4
80	2.6112 E-4	2.6117 E-4	2.6118 E-4	2.6118 E-4	2.6117 E-4	2.6109 E-4
100	2.4742 E-4	2.4744 E-4	2.4745 E-4	2.4745 E-4	2.4744 E-4	2.4736 E-4

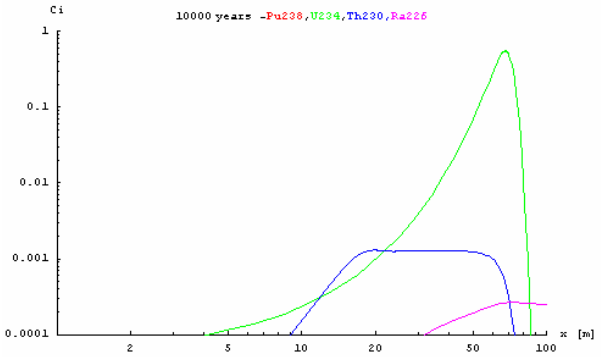


Figure 3 – Calculated concentration distributions for the four radionuclides , Pu238, U234 (top curve), Th230 (middle curve), Ra226 (bottom curve) ,at t= 10,000 years as obtained by GITT for the test case of van Genuchten (1985).

4. RESULTS AND DISCUSSION

Our code using the GITT approach is now demonstrated for the environmental impact assessment of an industrial installation that deals with liquid and solid waste containing radioactive contaminants. The case-study concerns possible pollution from an uranium mining facility operated by INB (Indústrias Nucleares do Brasil) in Caetité (Bahia, Brazil). Liquid wastes resulting from the uranium extraction process were deposited in ponds within the mining installation. Each cell of the ponds has an estimated operation period of four years. The Brazilian Nuclear Energy Commission (CNEN) required an analysis of the long-term transport of radioactive contaminants from the ponds into the subsurface, including an assessment of the total radioactive doses resulting from possible exposure to the radionuclides. Details of the analysis can be found in Orlande et al. (2006), including model development, code construction, identification of flow and transport properties, and dose calculations. Here we focus on the contaminant transport simulations of four cases (Table 3) that represent the most important scenarios for off-site migration.
The base case (Case No. 1) assumes a 6.9 m deep unsaturated zone below the pond, with recharge estimated from a hydrologic water balance of the Caetité region, and with water contents calculated with the HYDRUS-1D software package (Simunek et al., 2005) using soil hydraulic properties estimated from collected soil samples. The unsaturated zone was coupled to a horizontal granular aquifer of 1150 m length whose hydraulic and transport properties were estimated as part of the project. The flow rate through the horizontal aquifer was estimated from the hydraulic gradient and the hydraulic conductivity using Darcy's law. The other three cases (2, 3, 4) are sensitivity analyses relative to the base case. Of these, Case 2 assumes a shallower unsaturated zone (4.2 m), typical of wet seasons in the area, while keeping the remaining parameters unchanged. Case 3 considers a much higher recharge rate through the unsaturated zone by considering only precipitation and neglecting evapotranspiration. Finally, Case 4 illustrates the importance of accurately estimating the partitioning coefficients, *Kd*'s, by using reported literature values for clay soils (stronger retardation) rather than measured values (Table 4). Table 4 also gives estimates of the radioactive inventory of the simulated waste pond (Orlande et al., 2006). Results of our calculations are presented in Figures 4 through 7. In each plot we employed long dashed lines for U238, short dashed lines for U234, fine solid lines for Th230, medium solid lines for Ra226, and thick solid lines for Pb210.

The results for base-case 1 were first compared with a very conservative model, not further referred to in Table 3, which completely neglects the unsaturated zone below the pond. Figure 4a,b show for this scenario the calculated concentration distributions along the horizontal aquifer after 500 years. Clearly, including the unsaturated zone

considerably retards the transport process, while also significantly reducing the maximum concentrations. These effects are most pronounced for lead, radium and thorium, which for our soils have larger values of the retardation factors as compared with uranium. Figures 4c-f show the radionuclide concentration distributions at times t = 500, 1000, 1500, 2000, 2500, 5000, 7500, and 10,000 years, evolving from green to red in the plots. Results again illustrate the relatively rapid migration of uranium (lower *Kd* values), but also very low concentrations characteristic of thorium transport. At the same time, radium and lead become more abundant at later times despite their higher retardation factors

Table 3 – Simulated cases: Migration of radionuclides from uranium mine waste disposal pond.

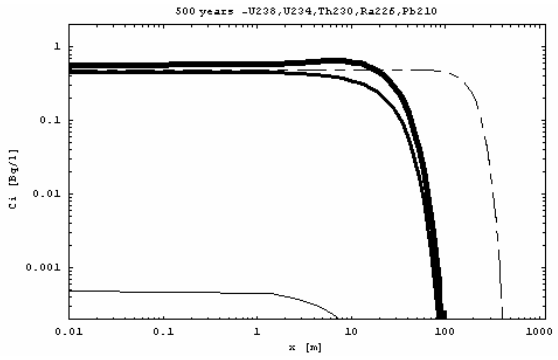
Case	Zones Considered	L1/L2 [m]	Infiltration	Water Content [%]	*Kd*´s
1	Unsaturated/Saturated	6.9 / 1150	q = P-ET / Darcy	θ = 0.308 /0.475	Table 4
2	Unsaturated/Saturated	4.2 / 1150	q = P-ET / Darcy	θ = 0.308 / 0.475	Table 4
3	Unsaturated/Saturated	6.9 / 1150	q = P / Darcy	θ =0.338 / 0.475	Table 4
4	Unsaturated/Saturated	6.9 / 1150	q = P / Darcy	θ =0.338 / 0.475	Clay / Table 4

(*)P= annual precipitation, ET=annual evapotranspiration, Darcy= estimated using Darcy's Law, Clay=*Kd*´s of clay soil.

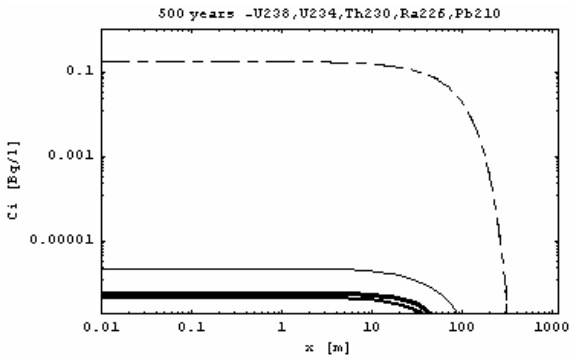
Table 4 – Partitioning coefficients, *Kd* [ml/g] and radioactive inventory, *A* [Bq]:

Element	*Kd* [ml/g]	*A* [Bq]
U238/U234	2985,6	5.34×10^{11}
Th230	44279,1	1.68×10^{10}
Ra226	198,31	5.51×10^{10}
Pb210	6772,8	6.16×10^{11}

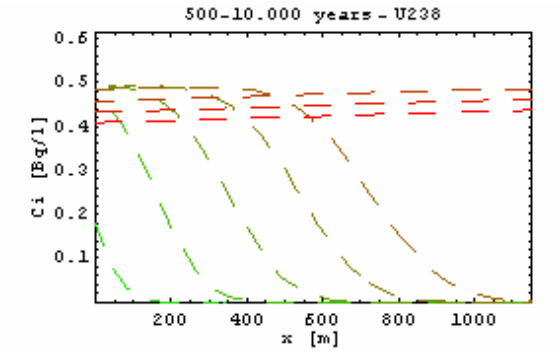
Figures 5 provides an interesting comparison between the base-case and Case No. 2 (shallower unsaturated zone). Figures 5a,b show concentration profiles in the unsaturated zone at t=100 years, while Figures 5c,d illustrate the migration along the horizontal aquifer at t= 500 years for the two cases. Results indicate that a thicker unsaturated zone (6.9 versus 4.2 m) only minimally decreases the transport rate in the aquifer, but significantly reduces the maximum concentrations in the aquifer at similar total migration times.



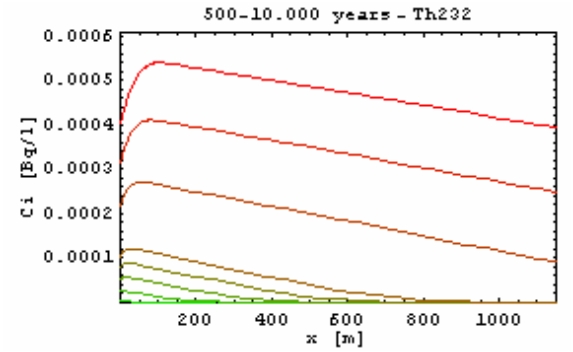
(a)



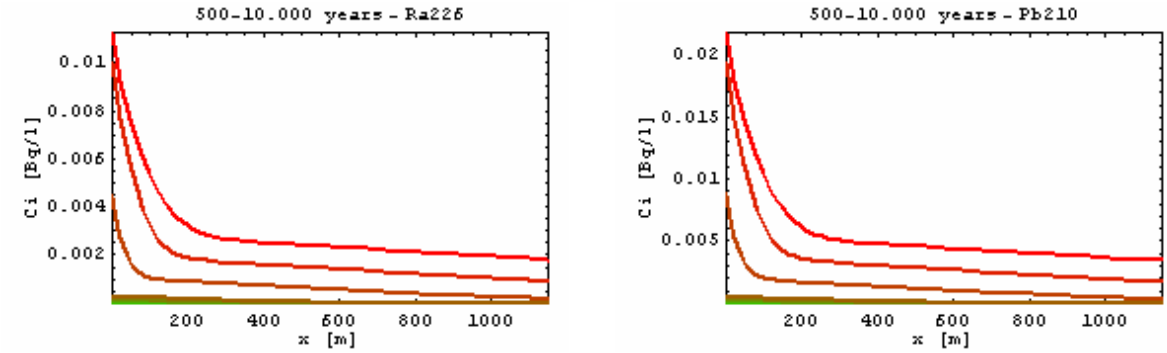
(b)



(c)

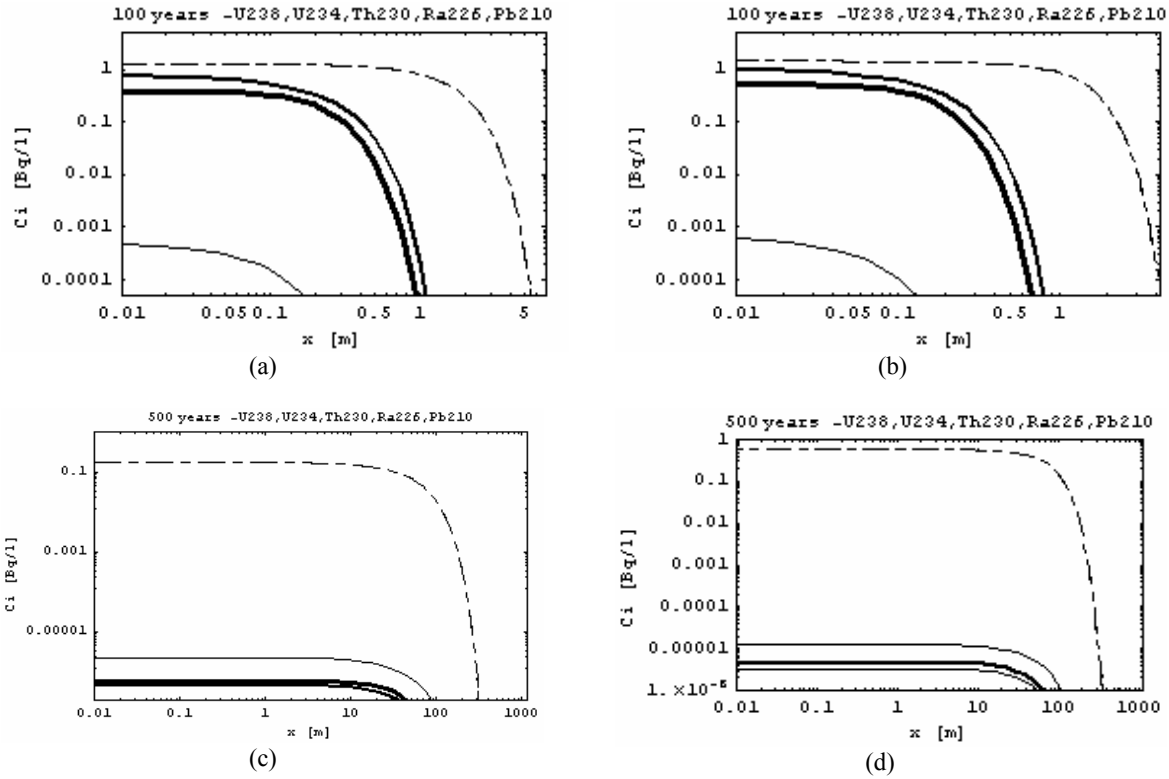


(d)



(e) (f)

Figure 4 – Radionuclides concentrations in the horizontal aquifer: a) Case 1 (all radionuclides, no unsaturated zone); b) Case 1 (all radionuclides, with unsaturated zone); c) Case 1 - U238; d) Case 1 - Th230; e) Case 1 - Ra226; f) Case 1 - Pb210.



(a) (b)

(c) (d)

Figure 5 – Radionuclide concentrations: a) Case 1 - all radionuclides in the unsaturated zone (6.9 m); b) Case 2 - all radionuclides in the unsaturated zone (4.2 m); c) Case 1 - all radionuclides in horizontal aquifer; d) Case 2 - all radionuclides in horizontal aquifer. (Long dashed lines for U238, short dashed lines for U234, fine solid lines for Th230, medium solid lines for Ra226, and thick solid lines for Pb210)

Figure 6 compares the base-case with Case No 3 (no evapotranspiration). Figures 6a,b show the concentrations in the unsaturated zone at t=500 years, while Figures 6c,d show the migration in the aquifer at t=10,000 years. As expected, the higher recharge rate for Case 3 causes more rapid transport of the contaminants, which now move much further into to aquifer at similar elapsed times, and with higher concentrations.

Figure 7 next compares Case 1 with the Case 4 (no evapotranspiration and higher *Kd*'s). Figures 7a,b present U238 concentrations in the unsaturated zone for times up to 1000 years, while Figures 7c,d illustrate the migration of the radionuclides in the aquifer at t=500 years. The higher *Kd* values typical of clay soils lead to much higher retardation factors and hence to markedly different distributions for the two cases. For example, the U concentrations after 1000 years in the unsaturated zone are much lower for case 4, while also barely reaching the horizontal aquifer after 10,000 years.

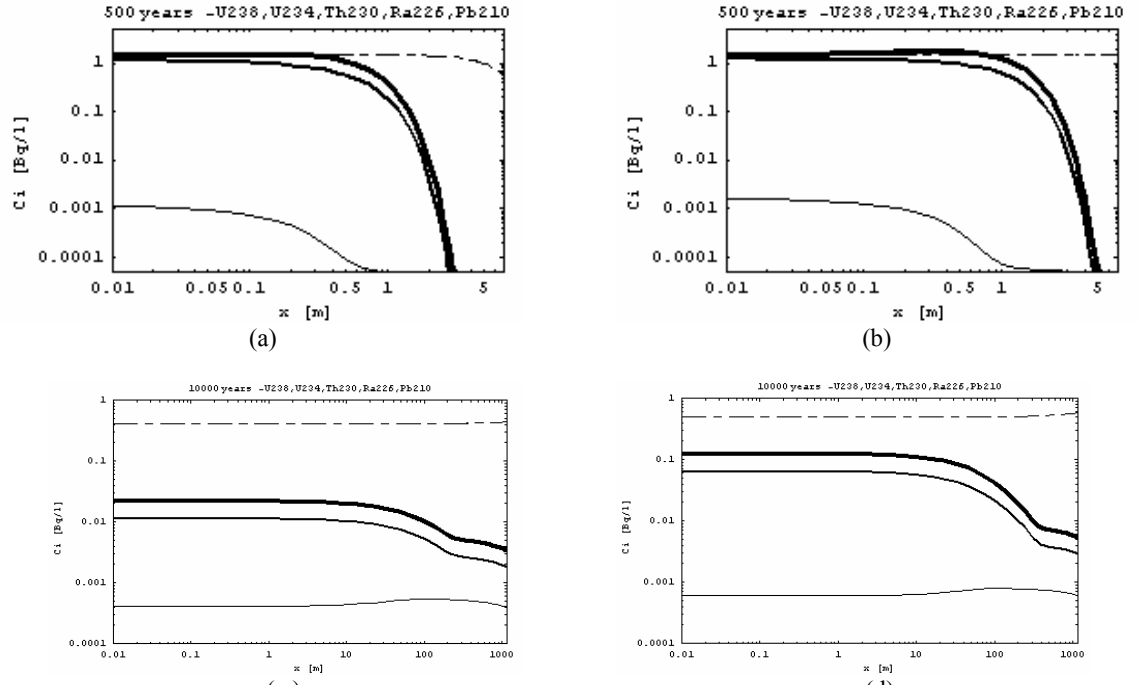


Figure 6 –Radionuclide concentrations: a) Case1 - all radionuclides in unsaturated zone; b) Case 3 - all radionuclides in unsaturated zone; c) Case 1 - all radionuclides in horizontal aquifer; d) Case 3 - all radionuclides in horizontal aquifer.

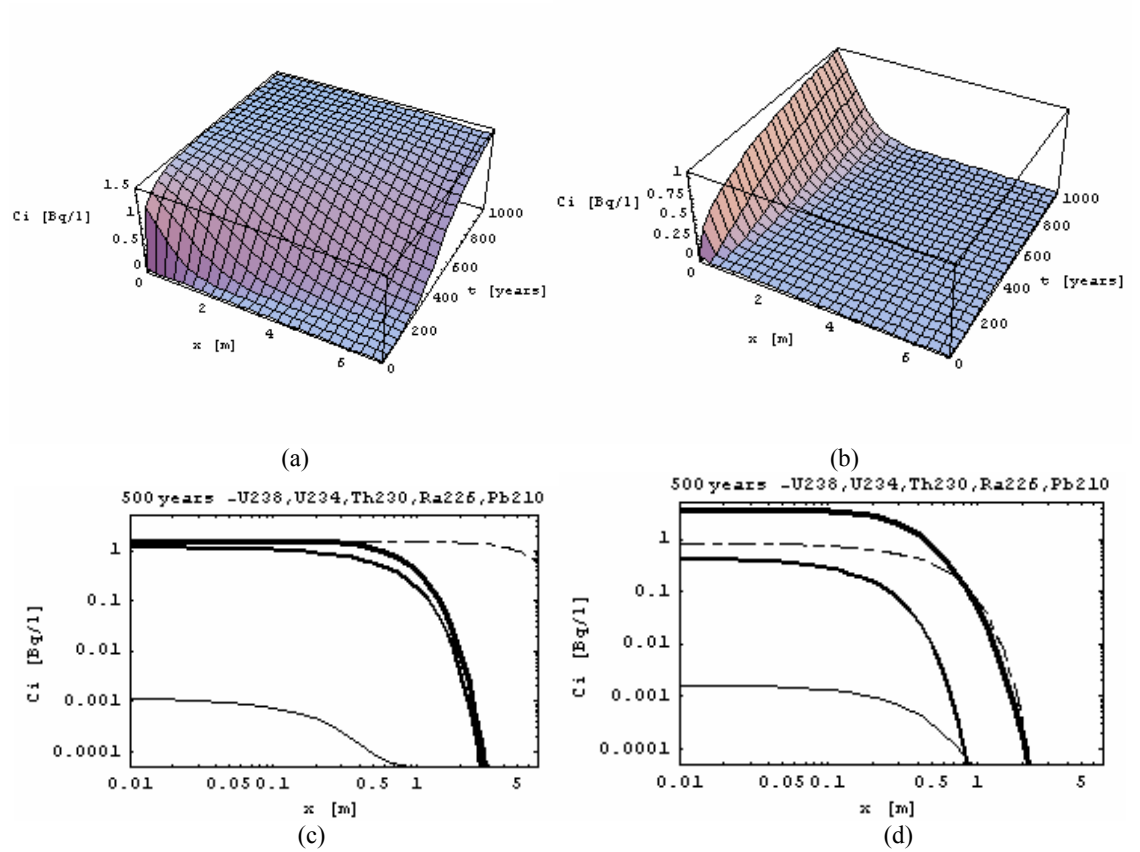


Figure 7 –Radionuclide concentrations versus depth in the unsaturated zone: a) Case 1 - U238 for t < 1000 years; b) Case 4 - U238 for t < 10000 years; c) Case 1 – all at t=10,000 years; d) Case 4 - all at t=10,000 years (*Kd*'s for clay).

The results above illustrate the considerable power of hybrid numerical-analytical solutions for evaluating relatively complex subsurface transport problems, in this case the transport of radioactive contaminant decay chains in a coupled saturated and unsaturated soil-aquifer system. The generalized integral transform technique (GITT) produced analytical expressions for the concentration distributions versus distance, and analytical or numerical estimates of the concentration as a function of time. We conclude that the method appears very useful for simulating a broad range of contaminant transport problems in soils and groundwater.

5. ACKNOWLEDGEMENTS

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