ENVIRONMENTAL FATE OF NATURAL RADIOACTIVE CONTAMINANTS IN FERTILIZERS AND PHOSPHOGYPSUM

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Abstract. *Agricultural soils often require the use of fertilizers and soil conditioners for optimal production. Phosphate fertilizers produced from igneous phosphate rock often contain small amounts of natural radionuclides (notably uranium and thorium), while the byproduct phosphogypsum (dihydrated calcium sulfate) is typically enriched in radium and lead. It is important to understand the long-term fate of these radionuclides when routinely applied via fertilizers (^{238}U and ^{234}U) and phosphogypsum as an amendment (^{226}Ra and ^{210}Pb) to agricultural lands. This study addresses the results of modeling their transport in a typical Cerrado soil profile. The HYDRUS-1D code was used to compare possible soil and groundwater pollution scenarios following the long term use of fertilizers and phosphygypsum in agricultural operations. Results using the equilibrium transport approach suggest that radionuclide concentrations originating from the use of phosphate fertilizers and phosphogypsum are relatively modest and will not pose a major risk to polluting underlying groundwater resources.*

Keywords*: radionuclide transport, numerical modeling, fertilizers, phosphogypsum, vadose zone, groundwater*

1. INTRODUCTION

Many Brazilian soils, including those in the Cerrado area, require the use of fertilizers and soil conditioners for optimal crop production. Soils in the area are typically highly weathered, slightly acidic, leached, and have a relatively low cation exchange capacity. As such, they are poor in nutrients, including P, Ca and Mg. Latosols in the area usually are also aluminum saturated. These features justify the use of phosphate fertilizers and phosphogypsum, the latter not only as a soil conditioner but also to supply Ca and S, and decrease Al toxicity to plants (Embrapa, 2005).

The main raw material for phosphoric fertilizer is apatite present in igneous phosphate rock. Phosphate fertilizers are obtained by treating such rock with concentrated sulfuric acid to produce phosphoric acid and phosphogypsum (dihydrated calcium phosphate) as a by-product. Phosphoric acid is the starting material for simple super-phosphate (SSP), triple super-phosphate (TSP), mono-ammonium phosphate (MAP) and di-ammonium phosphate (DAP). After desiccation, phosphogypsum (PG) is generally stockpiled as a waste because of elevated levels of impurities, including heavy metals and especially radionuclides. Each ton of phosphoric acid produces a considerable amount of PG, about 4.8 tons, which is generally stored nearby in the form of very large phosphogypsum stacks.

The natural U and Th decay series in phosphate rock are at equilibrium. During processing this equilibrium is disrupted and radionuclides migrate to the fertilizers and phosphogypsum. It is important to understand the natural radioactivity of different types of fertilizers and its by-products (Saueia and Mazzilli, 2006). Most of the uranium (^{238}U, ^{234}U) and thorium (^{230}Th) of the host rocks remains in the phosphoric acid during processing and then accumulates in the phosphate fertilizer; while radium (^{226}Ra), which chemically similar to calcium, and lead (^{210}Pb), favor the phosphogypsum (Mazzilli *et al*., 2000; Papastefanou *et al*., 2006).

Rather than merely stockpiling, some of the phosphogypsum could possibly be used as a soil conditioner in agriculture, similarly as gypsum (e.g., Oster, 1982, Embrapa, 2005). One area in Brazil that may well benefit from the use of PG is the Cerrado region where soils are generally slightly acidic (pH between 4.3 and 6.2) with high levels of exchangeable aluminum and low levels of phosphorus, calcium and magnesium. According to the Brazilian Agricultural Research Corporation (Embrapa, 2005), the use of PG is justified when there is a need to supply calcium and sulfur to plants, or when used as a means for decreasing toxic concentrations of exchangeable aluminum and increasing calcium to improve root growth.

Detailed analyses are needed of the advantages and risks related to the long-term use of fertilizers and phosphogypsum, especially the possible accumulations of radionuclides in the vadose zone and underlying or downgradient water resources, as well as their uptake by agricultural crops. In this paper we present results of modeling the subsurface transport of radionuclides resulting from the use of triple super-phosphate fertilizer (TSP) and phosphogypsum (PG) in a typical Cerrado soil profile. In particular, we evaluate the transport of ^{238}U, ^{230}Th, ^{226}Ra and

^{210}Pb when regularly applied via TSP fertilizer, and of ^{226}Ra and ^{210}Pb when applied via phosphogypsum (PG). The HYDRUS-1D flow and transport simulator (Simunek *et al.*, 2005) is used to predict their long-term transport through the vadose zone to groundwater. Rather than considering one single application, which should have very little impact on the environment (e.g., Rothbaum *et al.*, 1979; Zielinski *et al*., 1997), the scenario of concern in this study is the repeated application (in our example 24 years) of fertilizers and phosphogypsum to the same field, and the subsequent long-term fate and transport of the radionuclides in the subsurface. This scenario is similar to the simulations considered by Jacques et al. (2008a,c) using the more comprehensive HP1 multicomponent transport simulator of Jacques and Simunek (2010).

2. SIMULATION METHODOLGY

Here we consider the transport of radionuclides following the repeated application of both fertilizers and phosphogypsum to a typical soil profile in the Cerrado. Since contaminant transport is very much affected by the rates of water flow into and through the soil profile as a function of local weather conditions, we also consider variably-saturated flow as described with the standard Richards equation, which is given by (Simunek *et al*., 2008)

$$\frac{\partial \theta(h)}{\partial t} = \frac{\partial}{\partial x}\left[K(h)\left(\frac{\partial h}{\partial x} + 1\right)\right] - S(h) \tag{1}$$

where h is the soil water pressure head [L], θ is volumetric water content [L^3L^{-3}], t is time [T], x is the spatial coordinate (positive upward in this study) [L], K is the unsaturated hydraulic conductivity [LT^{-1}], and S is a sink term [$L^3L^{-3}T^{-1}$] that may be used to account for possible root water uptake.

The volumetric water content (θ) and the hydraulic conductivity (K) in Eq. (1) are strongly nonlinear functions of the pressure head, h. These functions are described here using the constitutive relationships of van Genuchten (1980) as follows

$$S_e(h) = \frac{\theta - \theta_r}{\theta_s - \theta_r} = \begin{cases} \left(1 + |\alpha h|^n\right)^{-m} & h < 0 \\ 1 & h \geq 0 \end{cases} \tag{2a}$$

$$K(h) = K_s S_e^l \left[1 - \left(1 - S_e^{1/m}\right)^m\right]^2 \tag{2b}$$

where S_e is effective saturation [L^3L^{-3}], θ_r and θ_s are the residual and saturated volumetric soil water contents, respectively [-], K_s is the saturated hydraulic conductivity [LT^{-1}], and where α [L^{-1}], n>1 [-], m=1-1/n [-] and l [-] are mostly empirical shape factors. Following van Genuchten (1980), the pore-connectivity parameter l was fixed at a value of 0.5.

Radionuclide transport in the soil profile was simulated using the standard advection-dispersion equation given by

$$\frac{\partial(\theta RC)}{\partial t} = \frac{\partial}{\partial x}\left(\theta D \frac{\partial C}{\partial x}\right) - \frac{\partial(qC)}{\partial x} - \mu C \tag{3}$$

where C is the solution concentration [ML^{-3}], R the retardation factor [-], D is the dispersion coefficient [L^2T^{-1}], q is the volumetric flux [LT^{-1}] given by the Darcy-Buckingham law, and μ is the decay coefficient [T^{-1}] for the particular radionuclide considered. The parameters R and D are given by

$$R = 1 + \frac{\rho_b K_d}{\theta} \tag{4}$$

$$D = D_d \tau + \lambda \frac{|q|}{\theta} \tag{5}$$

in which ρ_b is the bulk density of the soil [ML^{-3}] and K_d is the distribution coefficient for linear sorption [L^3M^{-1}], λ is the longitudinal dispersivity [L], D_d is the diffusion coefficient in free water [L^2T^{-1}], and τ is a porous medium tortuosity factor [-].

Equations (1) and (3) must be augmented with a set of initial and boundary conditions for variably-saturated flow and transport, respectively. For Eq. (1) we assumed an initial pressure head of -100 cm, the presence of atmospheric boundary conditions at the soil surface (i.e., given precipitation and calculated potential evapotranspiration data), and a free drainage condition (dh/dx=0) at the bottom of the profile. No root water uptake was considered in this study (i.e., S=0). For Eq. (3) we assumed an initially solute free soil profile, the annual application of radionuclides in a small pulse of water (10 mm) to the soil surface, and a zero concentration gradient (dc/dx=0) at the bottom of the soil profile at a depth of 1.3 m.

We also considered several steady-state flow simulations for which we simply used the long-term average recharge rate as a constant infiltration rate at the soil surface, while applying the radionuclides again as relatively small pulses in the infiltrating water or simply as averaged over the entire year, both subject to the constraints that in all cases the same amounts of radionuclide would be applied during the simulation.

The governing equations above are relatively standard in that they have been popularly used for the past 30 years or more in various forms, simplifications or extensions to simulate water and solute transport in soils and groundwater. For this study we used the HYDRUS-1D software, version 4.14 (Simunek *et al*., 2005), which provides finite element numerical solutions for simulating the one-dimensional movement of water, heat and multiple solutes (e.g., decay chains) in variably-saturated porous media. An overview and various applications of this code are provided by Simunek *et al.* (2008).

3. APPLICATION TO A CERRADO SOIL PROFILE

Simulations were performed to predict the fate of radionuclides present in fertilizers and phosphogypsum when applied routinely to a typical agricultural soil profile in the Cerrado area of Brazil. Meteorological data were obtained from a weather station in the western part of Minas Gerais state (latitude -17.2511, longitude -46,886, 625 m above mean sea level) for a 24-year period (1982-2005). Figure 1 shows for the first eight years observed precipitation and estimated potential evaporation data used in the simulations. Daily potential evapotranspiration rates were calculated with the Hargreaves equation (e.g., Jensen *et al*., 1997) from available weather data and the geographic location of the site. Since no root water uptake was considered, potential evapotranspiration was treated as potential evaporation from the soil surface. Potential precipitation and evaporation rates during the transient calculations with HYDRUS-1D were converted to actual infiltration and evaporation rates, respectively, depending upon the ability of the soil to allow infiltration or evaporation to occur at maximum or less than maximum rates (Simunek *et al*. 2005).

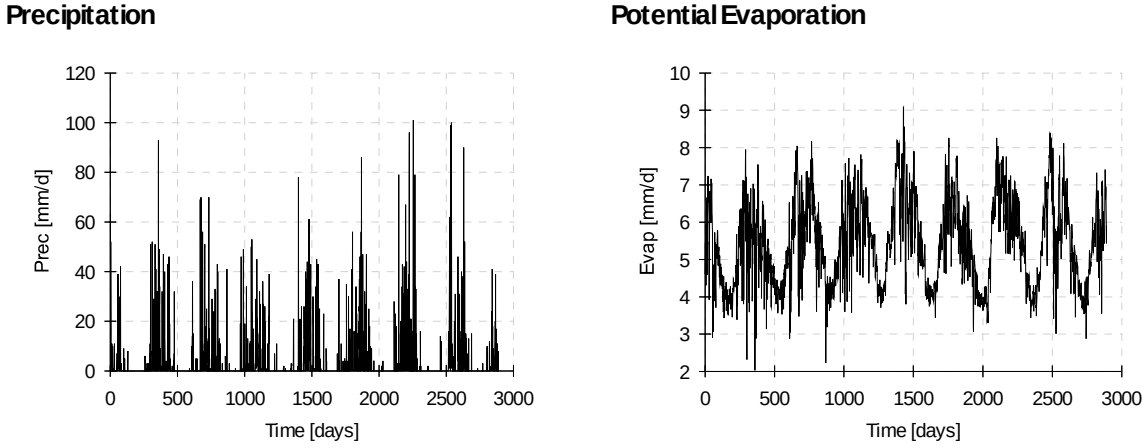

Figure 1. Measured daily precipitation (left) and estimated potential evaporation (right) rates at the Cerrado test site during 1982-1989.

Table 1 shows the soil texture of the selected soil, a red latosol containing 4 horizons within the 1.3 m deep profile considered in this study. Values of the soil hydraulic parameters in Eqs. (2a,b) needed for the water flow simulations were estimated from the soil textural data using pedotransfer functions that correlate the hydraulic properties with soil texture. We used for this purpose the Rosetta neural network based pedotransfer functions of Schaap *et al*. (1998) as implemented in HYDRUS-1D. Table 2 lists these parameters as well as values of the soil bulk density, ρ_b.

Table 1. Granulometric composition of the four soil horizons of the Latosol considered in this study.

Horizon	Depth (cm)	% Sand	% Silt	% Clay
A1	0 – 20	61.5	11.6	26.9
AB	20 – 50	52.6	10.3	37.1
BA	50 – 75	44.4	9.0	46.6
Bw1	75 – 130	37.5	10.6	51.9

Table 2. Values of the soil hydraulic parameters and bulk density used in the simulations.

θ_r (-)	θ_s (-)	α (m^{-1})	n (-)	K_s (m d^{-1})	ρ_b (kg m^{-3})
0.0682	0.3835	2.71	1.296	0.2323	1300
0.0791	0.3999	2.76	1.2397	0.2293	1300
0.0869	0.4219	2.93	1.1985	0.2518	1400
0.091	0.441	2.84	1.1881	0.2296	1400

For the solute transport simulations we used for all radionuclides a dispersivity (λ) of 0.2 m, a diffusion coefficient in free water (D_d) of 10^{-4} m^2d^{-1}, and a tortuosity factor (τ) of 0.5. Estimates of the distribution coefficients, K_d, were obtained using generic values for sand, silt and clay as provided by ISAM (2001) to obtain weighted K_d averages for the soil textural distributions listed in Table 1. Final K_d estimates used in the simulations are listed in table 3. Values of the decay coefficients were set to zero because of the relatively short simulation period considered in this study (200 years), relative to the half lives of the various radionuclides (mostly several orders of magnitude larger, except for ^{210}Pb).

Table 3. Values of the radionuclide K_d's used in the simulations.

Horizon	U (m^3 kg^{-1})	Th (m^3 kg^{-1})	Ra (m^3 kg^{-1})	Pb (m^3 kg^{-1})
A	0.280	2.03	1.00	0.256
AB	0.326	2.28	1.86	0.282
BA	0.418	2.51	2.56	0.306
Bw1	0.457	2.67	2.67	0.322

Very important to this study is correct definition of the amount of each radionuclide applied each year to the soil profile. This requires estimates of both the amount of fertilizer and phosphogypsum that would be applied using recommended agronomic practices, and the activity of the various radionuclides in the applied fertilizer and phosphogypsum. Table 4 shows results of measured radionuclide concentrations of triple super-phosphate (TSP) and PG. The measurements were carried out at the Poços de Caldas Laboratory of Brazilian Nuclear Energy Commission (CNEN) using procedures reported by Siqueira (2009). The results in Table 4 are consistent with literature data showing that during processing ^{238}U and ^{230}Th tend to accumulate in the fertilizer, while ^{226}Ra and ^{210}Pb tend to concentrate in the phosphogypsum. We note that ^{238}U and its daughter ^{234}U are always at secular equilibrium (in terms of their radioactivity). Hence, in this study we only consider the ^{238}U parent radionuclide.

Table 4. Measured ranges and average activities of ^{238}U, ^{230}Th, ^{226}Ra, and ^{210}Pb in TSP fertilizer and phosphogypsum

Radionuclides	TSP (Bq kg^{-1})	Average (Bq kg^{-1})	PG (Bq kg^{-1})	Average (Bq kg-1)
^{238}U	394 – 403	400	68 – 88	78
^{230}Th	420 – 430	425	82 – 93	88
^{226}Ra	110 – 120	115	226 – 255	240
^{210}Pb	120 – 130	125	204 – 256	125

The average concentrations in Table 4 were used to calculate amounts of radionuclide contaminants released each year with the fertilizers and phosphogypsum. We assumed annual applications of 180 kg/ha of P_2O_5 in 430 kg of TSP, and 1000 kg/ha of PG, applied once a year for 24 years as a short pulse in November, thereby mimicking local agronomic practices whereby applications are generally given in the rainy season Sousa et al. (2002).

After application for 24 years, the transport of the contaminants through the soil profile to the groundwater table is followed for a period of 200 years. To facilitate the long-term simulations, we assumed that radionuclides were added on top of the soil and dissolved in 10 mm of rainwater, similarly as was done in earlier simulations by Jacques *et al.* (2008a). This procedure agrees with Embrapa (2005) recommendations that PG should be applied to the soil surface and allowed to be transported to the subsurface by infiltrating rain or irrigation water. Separate results will be presented in the next section for triple super-phosphate (TSP) fertilizer and phosphogypsum (PG) transport.

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4. RESULTS

We first present selected results for the 24 year long transient simulations for which we used daily precipitation and evaporation data of the type shown in Fig. 1. Figure 2 (left) shows the calculated concentration distribution of fertilizer-derived ^{238}U in the soil after 24 years obtained with the transient simulations. Figure 2 (right) also shows results for steady-state flow, which will be discussed later.

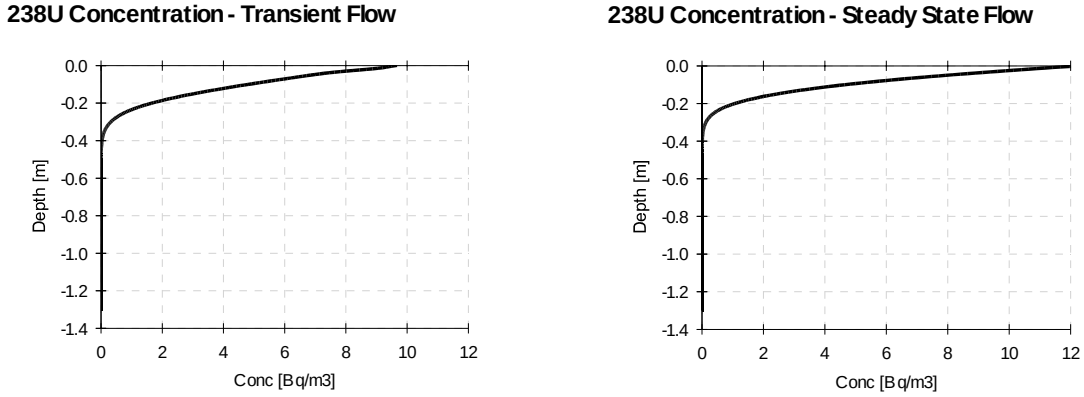

Figure 2. Calculated ^{238}U radionuclide distributions after 24 years of TSP application assuming transient flow (left) using daily precipitation and evaporation data, and steady-state flow (right) using the long-term average annual recharge rate of 0.546 m/year.

The transient results in Fig. 2 (left) indicate that ^{238}U moves only very slowly through the soil profile, a direct consequence of the very high K_d values of this radionuclide. If no sorption had occurred (i.e., K_d =0), the solute would have reached the bottom of the profile already after about 0.5 year. This is illustrated in Fig. 3 which shows concentration distributions versus depth for a non-adsorbing tracer during the first two years.

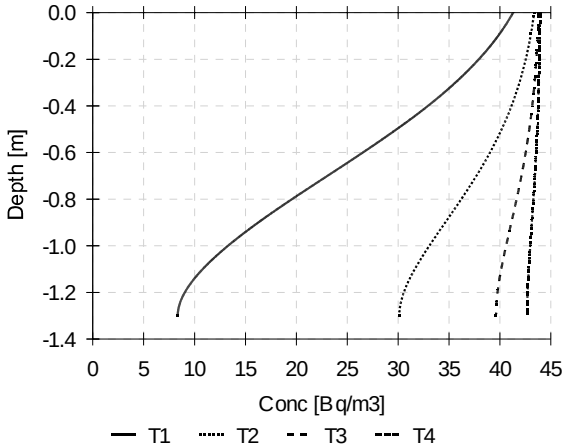

Figure 3. Transport of a non-adsorbing tracer (K_d=0) through the Cerrado soil profile during the first two years of the numerical experiment (T1, T2, T3 and T4 represent results after 6, 12, 18 and 24 months, respectively).

Figures 2 (left) and 3 were obtained assuming transient flow using daily precipitation and evaporation data. We next simulated the same ^{238}U radionuclide transport problem assuming steady-state water flow. For this we used the average recharge rate from the profile as calculated with the transient simulation. Figure 4 shows the calculated cumulative drainage flux from the 1.3-m deep soil profile (1982-2005). A long-term average recharge rate of 0.546 m/year could be calculated from the slope of the drainage flux plotted in Fig. 4.

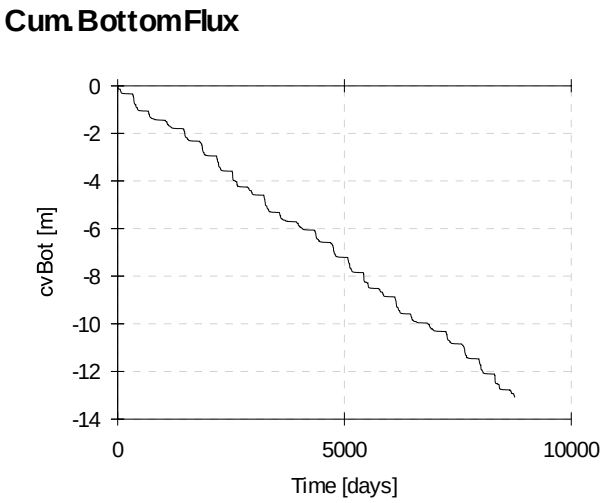

Figure 4. Calculated cumulative drainage flux obtained with 24 years of transient flow simulations using daily precipitation and evaporation data

Using the average recharge rate of 0.546 m/year, steady-state pressure head and water content distributions could be calculated. Results are shown in Figure 5. Notice that the pressure head profile (Fig. 5, left) is continuous as expected, whereas the water content distribution in the layered profile is discontinuous. The higher water content distributions in the bottom of the soil profile (Fig. 5, right) are a consequence of the more fine-textured horizons at depth (reflected by relatively small values of α, n and K_s in Table 2).

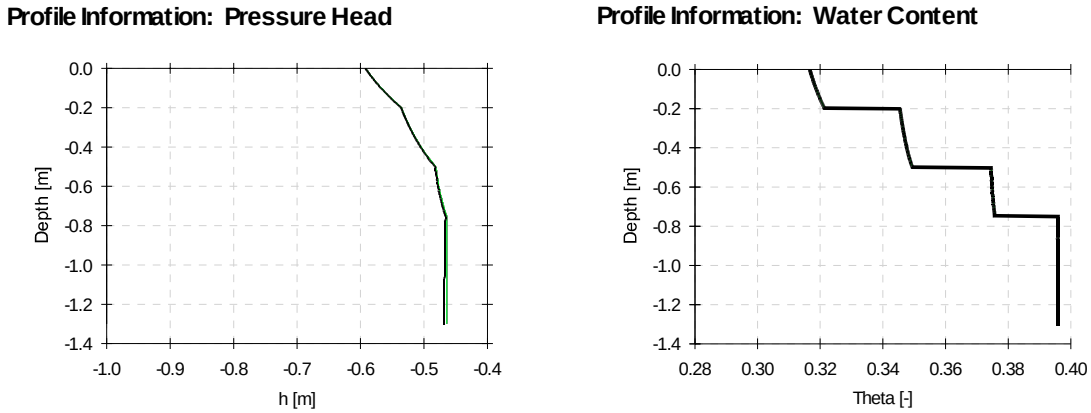

Figure 5. Calculated initial and steady-state pressure head, h (left) and water content, θ (right) distributions in the layered Cerrado soil profile.

Calculated concentration distributions of fertilizer-derived ^{238}U in the soil profile after 24 years using steady-state flow are compared with the steady-state results in Fig. 2. The two plots in Fig. 2 indicate very similar results for the transient and steady-state simulations, with the transient simulation results showing slightly more dispersion (and hence a somewhat deeper penetration) of the radionuclide in the soil profile. Using a higher dispersion coefficient for the steady-state simulation would have produced nearly exactly the same results as for the transient simulation. The results in Fig. 2 justify the use of steady-state flow regime for the long-term (200 year) radionuclide transport calculations.

Figures 6 presents calculated long-term radionuclide concentration distributions versus depth stemming from 24 years of fertilizer (TSP) application and subsequent leaching under natural weather conditions. The dotted lines are distributions in the soil profile after 100 years (76 after ceasing TSP fertilization), while the continuous lines represent distributions after 200 years. The results show only modest movement of all radionuclides through the soil profile, with U and Pb being transported the fastest. None of the radionuclides did reach the bottom of the 1.3-m deep soil profile within 200 years for the assumed conditions of linear equilibrium transport.

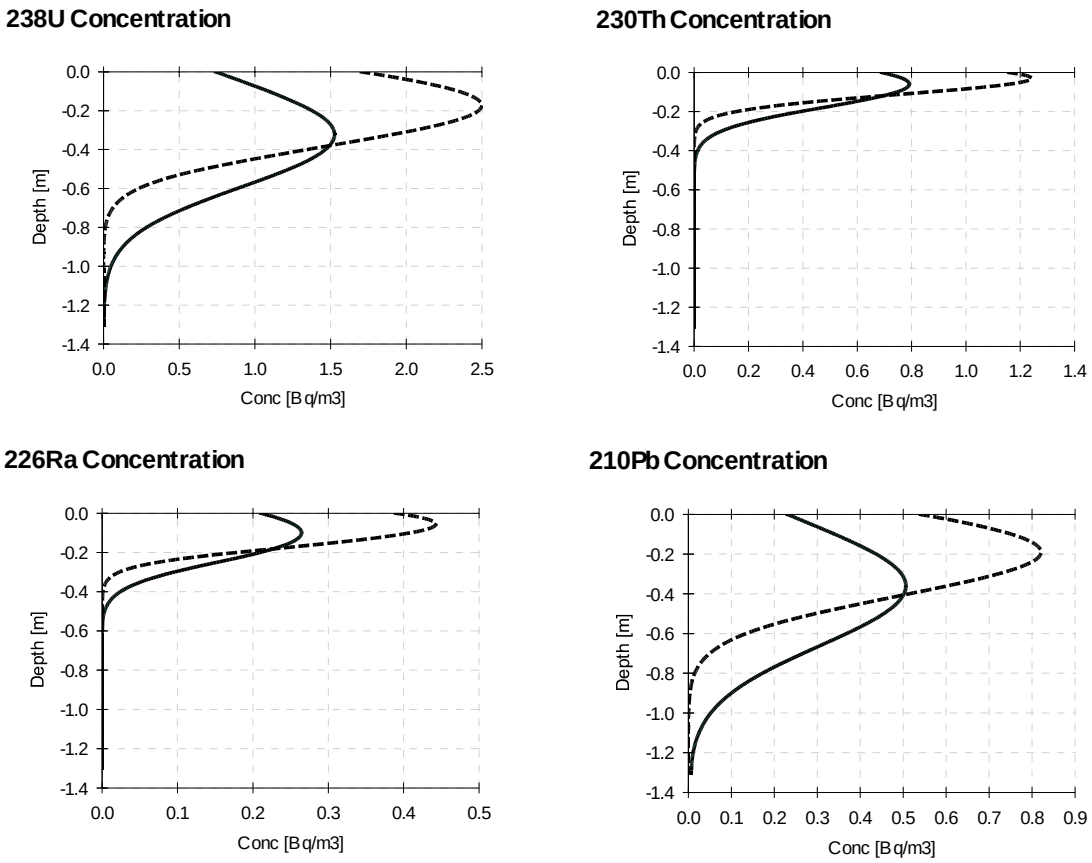

Figure 6. Concentration distributions in the soil profile of TSP derived uranium (top left), thorium (top right), radium (bottom left) and lead (bottom right) after 100 years (dashed) and 200 years (continuous lines).

Figures 7 present calculated distributions versus depth for the radionuclides resulting from 24 years of PG application. The dotted lines are again after 100 years (76 years after ceasing the use of PG amendments) and the continuous lines after 200 years. The long-term analysis for PG shows that while the ^{226}Ra remains in the upper 40 cm of soil, ^{210}Pb is transported much faster than Ra, albeit still not reaching the bottom of the soil profile. Except for a concentration scaling factor reflecting the different amounts (and hence equivalent concentrations calculated from these amounts) applied to the soil, the distributions in Fig. 7 are exactly the same as those in Fig. 6 for ^{226}Ra and ^{210}Pb (bottom left and right, respectively).

5. CONCLUSIONS

Simulations with HYDRUS-1D were performed to compare possible long-term (200 year) soil and groundwater pollution scenarios following 24 years on fertilizer and phosphogypsum application at a typical site in the Cerrado region of Brazil. The simulations indicate that assuming steady-state flow conditions will produce approximately the same results as transient simulations in which daily precipitation and potential evapotranspiration data were used. The transient simulations produced slightly lower maximum concentrations and a slightly deeper penetration of the solute fronts in the profile, which could be simulated using a higher dispersivity.

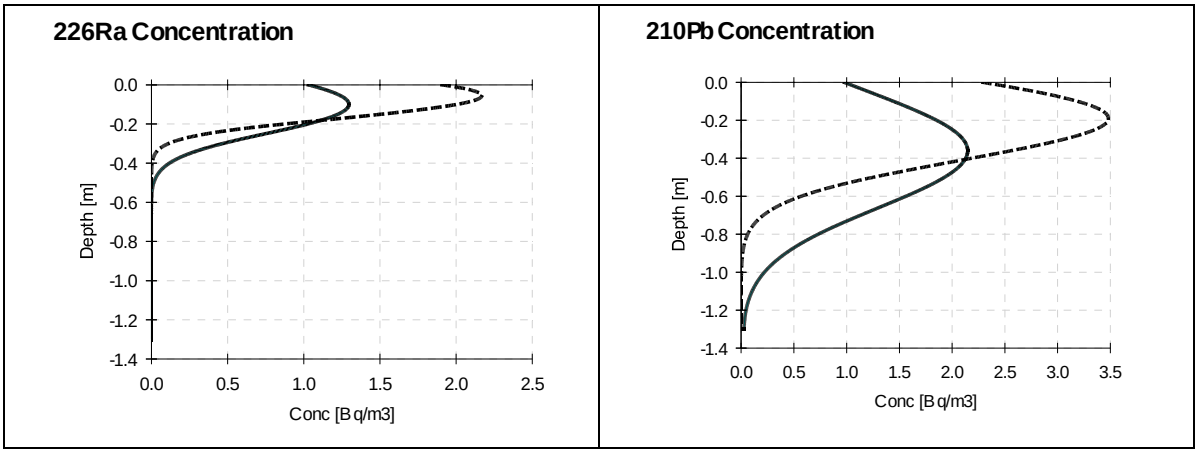

Figure 7. Concentration distributions in the soil profile of PG derived radium (left) and lead (right) after 100 years (dashed lines) and 200 years (continuous lines).

Phosphogypsum application produced less U pollution that the TSP fertilizer because of smaller radionuclide loadings in PG (results not shown here). However, ^{226}Ra and ^{210}Pb concentrations in the soil profile resulting from PG were about 5 times larger than those from TSP, thereby reflecting much larger amounts of Ra and Pb being added with PG as compared to TSP. Still, the calculated maximum concentrations were generally below background concentrations to be expected for uranium and thorium (e.g., Congé, 1993; Jacques *et al.*, 2008c).

Modeling efforts such as those in this study with HYDRUS-1D are essential for detailed environmental assessments of the long-term use of fertilizers and phosphogypsum containing normally occurring radioactive materials. They are especially needed for scenarios that cannot be easily monitored over relatively long time periods, or have largely unknown long-term consequences to human health or the environment.

The current simulations assumed linear equilibrium transport. Heterogeneities in the subsurface often will cause more rapid transport because of a number of physical and chemical nonequilibrium processes, including preferential flow and colloid-facilitated transport. These processes can have a major effect on the downward migration rate of the radionuclides (e.g., Simunek and van Genuchten, 2008; Pontedeiro *et al*., 2008). We also note that the simulations were carried out assuming a simple linear equilibrium K_d approach, thus ignoring the effects of geochemical processes including pH. As shown by Jacques *et al*. (2008a,b), more rapid transport may well be expected when a more comprehensive multicomponent transport modeling approach is invoked. These aspects are currently being studied in more detail for the subsurface transport of phosphogypsum-derived Ra and Pb.

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