MODELING THE TRANSPORT AND FATE OF CONTAMINANTS IN THE ENVIRONMENT: SOIL, WATER AND AIR

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Abstract: The environmental effects originated by uranium mining activities result mainly from the wastes generated by the ore processing. Large quantities of radioactive wastes are generated in this extractive process requiring a safe management. Besides the radioactivity these wastes may also hold different amounts of chemicals used in the extraction process, toxic pollutants associated with the mineralization and precipitates provoked by pH or Eh alterations.
The main concern of waste management and long term stabilization is to confine the residues in order to reduce the dispersion of contaminants to concentrations that not exceed the trigger values considered to be safe: there is thus a need to ensure that the environmental and health risk from these materials are reduced to an acceptable level. However, the confinement will always represent a potential source of environmental contamination to the air, soil, superficial water and groundwater, due to the contaminants release and transport in the environment, which may occur by natural erosion agents like rainfall or wind.

Keywords: Waste, disposal, release, dispersion, risk, environment.

1. Introduction

Uranium mining activities generates large volume of wastes composed by overburden, waste rock and tailings. Generally, these materials are deposited in waste rocks piles or dumps. Uranium mill tailings are the solid residues resulting from the leaching of the ore.
The Environmental effects resulting from the uranium mining activities are mainly derived from the wastes generated by the ore processing. Wastes constituents of concern include radionuclides (uranium, radium, radon and

Journal Information

thorium), arsenic, copper, selenium, vanadium, molybdenum, heavy metals and dissolved solids. The radionuclides in the tailings are more mobile and chemically reactive than in the original ore and may enter into the environment becoming a contamination source to the soil, air, superficial water and groundwater.

When modeling contaminant release and transport mechanisms for each environmental compartment, the major output chosen is the contaminant concentration in each exposition point selected (for instance, breathing air zone, superficial soil and well water). This will allow the assessment of doses, if an additional exposure scenery is created. The objective is to quantify the potential exposure levels of the hazard at the receptor location answering to these main questions: what is the contaminant concentration in the exposition point? How much of a contaminant do people inhale or ingest during a specific period of time? Is the situation acceptable?

This work proposes a generic exposure model that incorporates simultaneously an atmospheric and a hydrologic transport model. In the atmospheric transport a two-dimensional model is used for calculating the flux diffusion from a radioactive waste disposal, having as result the hazard concentration at a defined distance from the soil (the breathing or mixing height) which will be the starting point for the dispersion each can be considered either simultaneously in each wind octant direction or considering only a prevailing wind direction. For the hydrologic transport a two-direction model is proposed for simulating the contaminants release form the waste disposal and its migration process through the soil to the groundwater. The final result is the contaminant concentration in the groundwater as function of the elapsed time, at a defined distance from the waste disposal, generally the location where the exposition point is considered being represented by an hypothetical well.

2. Methods and Results

2.1. ATMOSPHERIC TRANSPORT MODEL

For the gaseous contaminants the release mechanism from the soil is generally based in the principles of diffusion across a porous medium. Basic diffusion equations are used for estimating the theoretical values of the gaseous flux from the waste material. The generic diffusion equation can be represented by a one-dimensional steady-state equation:

$$D\frac{\partial^2 C}{\partial x^2} - \lambda C + \frac{R\rho\lambda E}{\varepsilon} = 0 \quad (1)$$

In this equation D ($m^2.s^{-1}$) represents the radon diffusivity, λ the radioactive decay constant (s^{-1}), C ($Bq.m^{-3}$) the radon concentration in the pore space, R ($Bq.kg^{-1}$) the radium concentration in the material, ρ ($kg.m^{-3}$) the bulk density of the dry material, E (dimensionless) the radon emanation power coefficient for the pore spaces, ε (dimensionless) the total porosity and θ (dimensionless) the moisture. The solution of the diffusion equation for an homogeneous medium represents the flux release from the waste material to the surface, J_t ($Bq.m^{-2}.s^{-1}$). For a system without cover we obtain (Rogers, 1984):

$$J_t = R\rho E\sqrt{\lambda D_t}\,\tanh\left(\sqrt{\frac{\lambda}{D_t}}x_t\right) \quad (2)$$

The contaminant concentration released is estimated by a box model formulation which has implicit a mass balance formulation. The box volume (V) is defined by its length (L), width (W) and the mixing height (h). As a consequence of a steady state assumption, we have that the pollutant concentration (C) is constant in time, the mass flow rate entering (ϕA) into the box is equal to the flow rate leaving the box (uSC):

$$V\frac{dC}{dt} = \phi A - uSC \quad (3)$$

The atmospheric dispersion is modeled by a modified Gaussian plume equation which estimates the average dispersion of the contaminants released from the source in each wind direction. The Gaussian model of a plume dispersion accounts for the gaseous contaminant transport from the source area to a downwind receptor and is represented by the equation of Pasquill as modified by Gifford (Chacki, 2000):

$$C = \frac{Q}{2\pi\mu\sigma_y\sigma_z} e^{\left[-1/2\left(\frac{y}{\sigma_y}\right)^2\right]}\left\{e^{\left[-\frac{1}{2}\left(\frac{z-H}{\sigma_z}\right)^2\right]} + e^{\left[-\frac{1}{2}\left(\frac{z+H}{\sigma_z}\right)^2\right]}\right\} \quad (4)$$

This equation represents a Gaussian distribution, where C ($Bq.m^{-3}$) represents the radionuclide concentration, Q ($Bq.s^{-1}$) the source strength, and H (m) the corrected source released height. Dispersion parameters, σ_y (m) and σ_z (m), are the standard deviations of the plume concentration in the horizontal and vertical directions respectively. The atmospheric transport is done at wind-speed (height-independent), u ($m.s^{-1}$), to a sampling position located at surface elevation, z (m), and transverse horizontal distance, y (m), from the plume centre.

2.2. HYDROLOGIC TRANSPORT MODEL

A leaching model based on a sorption-desorption process is used for describing the contaminant release from the waste disposal. The leachate concentration, C_L ($Bq.m^{-3}$), is determined by the a distribution or a partition coefficient, K_d ($cm^3.g^{-1}$) which describes the relative transport speed of the contaminant to the water existing in the pores; soil properties such as bulk density, ρ ($g.cm^{-3}$), and water content, θ, affect the extent of contamination, described by the contaminated zone thickness, x_t (m), area, A (m^2) and the amount of contaminant in the source, I_t (Bq), (EPA 1996; Hung 2000):

$$C_L = I_t / \left[A \left(x_t \, \theta + x_t \, \rho \, K_d \right) \right] \quad (5)$$

The transport for the dissolved contaminants is considered to occur either in the vertical direction through the unsaturated zone until an aquifer is reached either in the horizontal direction, through the saturated zone flowing to an hypothetical well, where the contaminants become accessible to humans or other forms of life. The vertical flow is considered to be one-dimensional. It is assumed that there is retardation during the vertical transport that is estimated assuming that the adsorption-desorption process can be represented by a linear isotherm, which means that there is a linear relationship between the radionuclides concentration in the solid and liquid phases.

Movement and fate of radionuclides in groundwater follow the transport components represented by the basic diffusion/dispersion–advection equation. The following expression describes the basic equation for the advective and dispersive transport with radioactive decay and retardation for the radionuclide transport in the groundwater:

$$D \frac{\partial^2 C}{\partial x^2} - V \frac{\partial C}{\partial x} - R \frac{\partial C}{\partial t} - \lambda R C = 0 \quad (6)$$

In this equation, D represents the molecular diffusion coefficient ($L^2.T^{-1}$), C represents the solute concentration ($M.L^{-3}$), V represents the interstitial velocity ($L.T^{-1}$), R represents the retardation factor due the sorption phenomena. The component dispersive and diffusive is represented by $\partial^2 C/\partial x^2$, the advection is represented by $V \, \partial C/\partial x$ and the concentration gradient is represented by $\partial C/\partial x$.

In the analytical solution the term for the contaminant concentration, C, was replaced by the rate of radionuclide transport, Q'(t), (Hung 1986):

$$Q'(t) = Q_0 \left(t - \frac{R \, L}{V} \right) e^{\left[-(\lambda_d R \, L)/V \right]} \quad (7)$$

The rate of radionuclide transport to the well point is represented by Q'(t) ($Bq.yr^{-1}$), the rate of radionuclides transport at x = 0 is represented by Q_0,

($Bq.yr^{-1}$), t (yr) is the elapsed time and L (m) the distance from the disposal site to the well point.

2.3. A CASE STUDY

The model was applied to a specific case of contaminants from a uranium waste disposal. The contaminants of main concern were considered to be radon in air and radium in groundwater. The exposition point for the atmospheric model is a receptor located about 2 km from the source and for the hydrological model the exposition point is a hypothetical well located about 100 meters from the source.

The final output for the atmospheric model is the radon concentration at a defined distance from the source, in each wind direction and in the dominant wind direction, where is considered to be located the receptor. For the hydrological model the final outputs are the radium concentration in the well water and the corresponding cumulative rate of radium transported to the well after the time considered.

The necessary parameters for the simulation were adopted from some local measurements made in this particular site (Vicente 2005).

Local meteorological data, namely wind velocity and frequency, was used for simulating the dispersion in each octant direction. These data was obtained from a local automatic meteorological station (INAG 2004). The dominant wind direction is NW. The unknown parameters were estimated from available data.

The contaminated site is composed by $1,6x10^6$ ton of two different kind of wastes with a total area of approximately 75000 m^2. The total waste volume is about $1,5x10^3$ m^3. It was assumed in the simulation that there is no covering system although there is some natural vegetation partially covering the site.

The air breathing tallness, or mixing height, was defined as 1,0 m. The radon concentrations were calculated in each sector at this height, taking into account the average wind speed and respective frequency of blowing. The concentration at the breathing height in the dominant wind direction refers only to the boundary side that limits the respective sector. The dispersion results can be seen in the figures below.

Local hydro-geological conditions were considered for each zone where the radionuclides transport occurs, namely for the contaminated zone, for the unsaturated zone and for the saturated zone, using different densities, porosities, hydraulic conductivities, radionuclide distribution coefficients and thicknesses. The well is located at the down-gradient edge of the contamination source.

Meteorological data, precipitation and evaporation from the same automatic meteorological station were used for estimate the infiltrating water rate into the

contaminated zone. The figures 4 and 5 shows the results obtained for radium in groundwater.

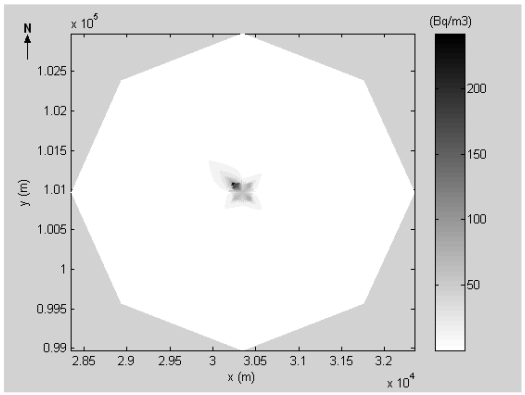

Figure 1: Radon dispersion in each wind direction, Bq.m^{-3}.

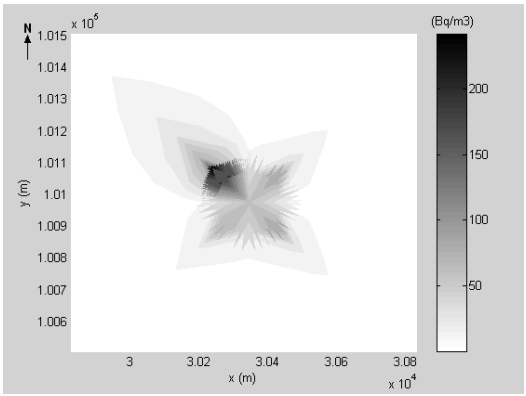

Figure 2: Radon dispersion in each wind direction, Bq.m^{-3}.

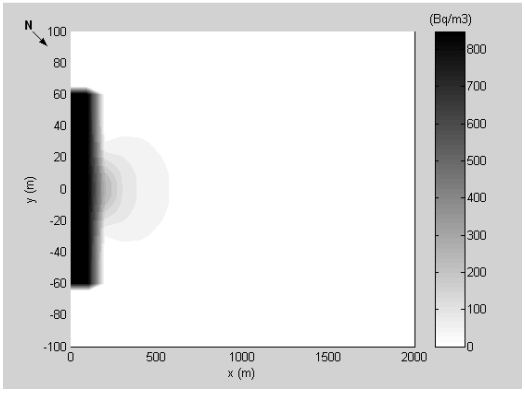

Figure 3: Radon dispersion, dominant wind direction, Bq.m^{-3}.

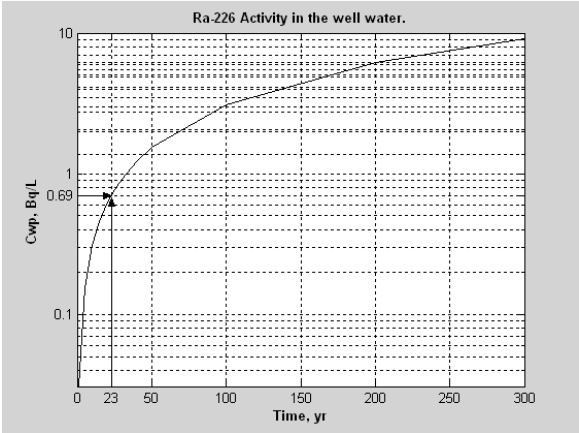

Figure 4: Radium activity concentration in the well water, Bq/L.

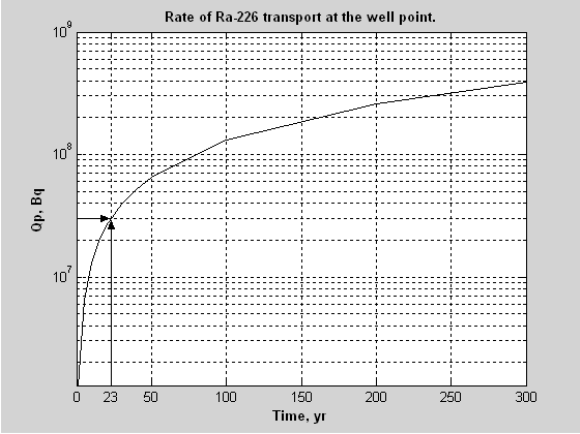

Figure 5: Cumulative rate of radium release, Bq.

3. Conclusions

Contaminants may travel through the atmosphere, soil, groundwater and superficial waters affecting the organisms that inhabit these media. The exposure modeling quantifies the impacts of contaminants as they travel through more than one of these compartments.

The atmospheric model describes the contaminant transport from the source area to a downwind receptor and the corresponding simulation quantifies the contaminant dispersion concentration in each wind direction.

The hydrologic model quantifies the movement of subsurface water and provides inputs to contaminant transport models. Its usage as a simulation tool allows previewing the contaminant behaviour in the groundwater al well as a

quantitative assessment for the concentration of the contamination at a particular exposition point.

The model limitations are mostly related with the application of an analytical solution which is limited by its specific form of boundary conditions. The boundary conditions are stipulated in order to solve the unknowns in the problem domain. This will generate some error which magnitude will depend on the conformity between the local conditions and those stipulated in the model. The forecast accuracy of models are often compromised since some key parameters are imperfectly known and may have to be estimated from literature references in the absence of actual site specific measurements.

Many soil specific parameters show a great variability both in space and in time. Also these parameters will vary over the year due to climatic changing. This will generate some difficulties in obtain the most appropriate data needed to characterize the contaminated site.

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