SIMULATION OF LIBERATION AND DISPERSION OF RADON FROM A WASTE DISPOSAL

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Abstract: Radon emissions from a radioactive waste disposal may constitute a major source of environment contamination and consequently a potential health hazard to the nearby population. Gaseous Radon-222 is generated from the radioactive decay of Radium-226 present in the tails. When it is formed, radon is free to diffuse along the pores of the residues to the surface and escape to the atmosphere.

Waste management and long term stabilisation has a major concern in reducing radon emissions to near-background levels. The common theoretical approach is done by calculating the cover thickness that allows a radon flux inferior to a stipulated and accepted value. The fundamentals of the conceptual model are based in the principles of diffusion across a porous medium, which allows the mathematical description of the radon transport through the waste and the cover.

The basic diffusion equations are used for estimating the theoretical values of the radon flux formed from the decay of the Radium-226 contained in the waste material. The algorithm incorporates the radon attenuation originated by an arbitrary cover system placed over the radioactive waste disposal.

Once the radon is released into the atmosphere, it is available for atmospheric transport by the wind. Radon atmospheric dispersion is modelled by a modified Gaussian plume equation, which estimates the average dispersion of radon released from a point source representative of one or several uniform area sources. The model considers the medium point release between all the areas contaminated. The dispersion can be simulated in different wind directions, with different wind velocities, as well as in the dominant wind direction.

Key words: Waste, disposal, radium, radon, flux, dispersion.

1. INTRODUCTION

Uranium milling tailings represent the highest potential source of environmental contamination for the great majority of uranium mining activities. In the ore milling, the uranium ore is grinded promoting liberation and thus increasing the possibility of radon to escape to the environment. The milling process generates large volumes of tailings being generally disposed in piles. The radionuclides presents in the tailings, ^{226}Ra, ^{230}Th and ^{222}Rn, are a major concern to the human health and to the environment. They are not dissolved during the leaching process, which breaks down the equilibrium chains of the ^{238}U and ^{235}U decay families.

The principal radon isotope, ^{222}Rn, formed from the ^{226}Ra radioactive decay, has a half-life of 3.82 days, which allows a large period of time for migration before it decays to another nuclide. Radon generation may continue for thousand of years due to the long decay periods of ^{226}Ra and ^{230}Th, present in the uranium tailings. Radon is an inert gas, which emanates from the solid tailings particles and is free to diffuse to the surface of the pile, escaping to the atmosphere. Coming up from the ground, it may become locally hazardous or it may be transported by the wind into the surrounding area, dispersing the potential damages.

The work proposes a two-dimensional model for calculating the ^{222}Rn flux diffusion from a radioactive waste disposal, having as a result the ^{222}Rn concentrations at a defined mixing height which will be the starting point to the atmospheric dispersion, as well as the bidimensional dispersion in the prevalent wind direction.

2. METHODS AND RESULTS

2.1 The ^{222}Rn diffusion

The basic equations of diffusion may be used for estimating the theoretical values of the radon flux from the ^{226}Ra content in the waste material. Radon migration to the surface is a complex process controlled mainly by porosity (ε) and moisture (θ), leading the cover efficiency in attenuating the radon flux. This efficiency depends on the capacity of the cover material for keeping the diffusion so slow that radon decays to another non-gaseous nuclide, becoming trapped by the cover system.

The movement of radon in soil is characterised by the diffusion coefficient, D, which can be measured, either in laboratory or in field, or be estimated by empirical correlation. These have the advantage of being simple and easy to use with a minimal amount of information needed. A correlation using the fraction of saturation, m, is recommended (Rogers, 1984):

$$D\left(cm^2.s^{-1}\right) = 0{,}07\, e^{\left[-4\left(m - m\varepsilon^2 + m^5\right)\right]} \quad (1)$$

Values for radon diffusivity in porous media may vary over a wide range of several orders of magnitudes depending on the porous material and particularly on its degree of water saturation. The generic diffusion equation can be represented by:

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

The diffusion process occurs in a multiphase system, where the porosity is either filled with air, either with water. If we apply the generic diffusion equation to each one of the phases (a-air, w-water, filled pore space):

$$D_a \frac{\partial^2 C_a}{\partial x^2} - \lambda C_a + \frac{R\rho\lambda E_a}{\varepsilon - \theta} + \frac{T_{wa}}{\varepsilon - \theta} = 0 \quad (3)$$

$$D_w \frac{\partial^2 C_w}{\partial x^2} - \lambda C_w + \frac{R\rho\lambda E_w}{\theta} - \frac{T_{wa}}{\theta} = 0 \quad (4)$$

In these equations, D ($m^2.s^{-1}$) represents the radon diffusivity, λ the radon 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, θ (dimensionless) the moisture and T_{wa} ($Bq.m^{-3}.s^{-1}$) the radon transfer rate from water to the air.

The solution of the diffusion equation for a homogeneous medium represents the flux release from the tailings to the surface, J ($Bq.m^{-2}.s^{-1}$). For a system without cover we obtain (Rogers, 1984):

$$J_t = R\rho E\sqrt{\lambda D_t}\, \tanh\left(\sqrt{\frac{\lambda}{D_t}}\, x_t\right) \quad (5)$$

In this equation, x_t represents the tailings thickness. If we consider a two media problem represented by the tailings (t) and a homogeneous cover material (c), the solution of the diffusion equation can be represented by (Rogers, 1984):

$$J_c(x_c) = \frac{2J_t e^{-b_c x_c}}{\left[1 + \sqrt{a_t/a_c}\,\tanh(b_t x_t)\right] + \left[1 - \sqrt{a_t/a_c}\,\tanh(b_t x_t)\right] e^{-2b_c x_c}} \quad (6)$$

In this solution, $b_i = \sqrt{(\lambda/D_i)}$ (m) (i = c or t) and $a_i = \varepsilon_i^2 D_i[1\text{-}0.74m_i]^2$ ($m^2.s^{-1}$), where m is the degree of saturation of the soil.

From the precedent equations it is possible to obtain a generic solution for the radon released in three main situations: flux directly diverted to the atmosphere without cover trapping; radon flux through a homogeneous cover and radon flux through a multilayer cover system.

Another possible approach consists in calculating the thickness of the cover which allows a value stipulated and accepted for the radon flux. This can be done directly using the waste and cover parameters, mainly the ^{222}Rn coefficient diffusion, porosity and moisture, the ^{226}Ra content in the waste material and ^{222}Rn emanation, rearranging the last equation (Rogers, 1984):

$$x_c = \sqrt{\frac{D_c}{\lambda}}\ln\left[\frac{2J_t / J_c}{\left(1 + \sqrt{a_t/a_c}\,\tanh(b_t x_t)\right) + \left(1 - \sqrt{a_t/a_c}\,\tanh(b_t x_t)\right)\left(Jc/Jt\right)^2}\right] \quad (7)$$

2.2 The ^{222}Rn atmospheric dispersion

The model uses a Gaussian model of plume dispersion to account for the transportation of ^{222}Rn from the source area to a downwind receptor, and is represented by the equation of Pasquill as modified by Gifford (Chacki, 2000):

$$X = \frac{Q}{2\pi\mu\sigma_y\sigma_z} e^{\left[-1/2\left(\frac{y}{\sigma_y}\right)^2\right]}\left\{e^{\left[-\frac{1}{2}\left(\frac{z-H}{\sigma_z}\right)^2\right]} + e^{\left[-\frac{1}{2}\left(\frac{z+H}{\sigma_z}\right)^2\right]}\right\} \quad (8)$$

This equation represents a Gaussian distribution, where X ($Bq.m^{-3}$) 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 standard deviations can be evaluated by the Pasquill-Gifford coefficients for flat rural areas or by Briggs method for urban areas. The atmospheric transport is done at wind-speed (height-independent), u ($m.s^{-1}$), to a sampling position located at surface elevation, z, and transverse horizontal distance, y, from the plume centre.

The concentration available for the dispersion is calculated from the radon flux released. The contamination source is defined as an emission area, or the sum of several areas, where the radon is diluted directly in the ambient air-breathing zone above the contaminated source zone. The wind speed for ^{222}Rn dilution should be matched to the average annual value through the mixing zone.

The contaminated area can be compared to an environment compartment in which the ^{222}Rn emission is uniform in all the area, characterised by its length and width. These values and the mixing height define the volume in which the concentrations are spatially homogeneous and instantaneously mixed.

The modified Gaussian plume equation estimates the average dispersion of radon released from a point source representative of one or several uniform area sources. This point represents the medium point release between all the areas contaminated, with the same mass flux as the entire affected zones, and it should be located in the weighted centre of the total contaminated area.

The ^{222}Rn concentration dispersion is calculated from the release point, at the mixing height, for defined distances in each wind direction. The release point source is located in the centre of a conceptual regular polygon area defined as the contaminated site. The concentrations along each direction are evaluated taking into account the respective mean wind velocity and the frequency of the occurrence. This will lead to different ^{222}Rn concentrations in the sectors defined by each direction in the collateral wind rose.

When we consider exclusively the dominant wind direction, we assume that all the ^{222}Rn concentration is being distributed in that direction, being inexistent in all other directions. Another assumption is that ^{222}Rn concentration is uniform in the volume defined by the central point source and the boundaries of the release area, and by the mixing height. Dispersion occurs outside this area.

2.3 A case study

The model was applied to a specific contaminate site, the Urgeiriça uranium tailings. The Urgeiriça uranium mine is located in the north of Portugal near Nelas (Viseu). The mine is surrounded by small farms and country houses, with most of the local population living in the village Canas de Senhorim within about 2 km of the mine.

The exploitation of the mine began in 1913 for radium extraction. The activity of the Urgeiriça mine was maintained until 1944 then exclusively dedicated to the production of radium. In 1951, a chemical treatment unit for the production of low-grade U_3O_8 concentrates was built, and in 1967 was transformed into a modern unit (Bettencourt *et al*, 1990). In 1991, local mining stopped but the facilities were still used for the treatment of ores from other mines, in the same region, until 2000.

The extensive exploitation and treatment of the uranium ore in the Urgeiriça mine, has led to an accumulation of large amounts of solid wastes (tailings). About $4x10^9$ kg of rock material was routed into natural depressions confined by dams that cover an area of about 0.11 km^2. The solid wastes are composed mainly of sand and silt particles, transported as a pulp to the site from the counter-current decantation. This operation was proceeded by grinding and acid leaching. The radon exhalation from the tailings is potentially one of the main sources of contamination for the nearby areas.

We tested the model in simple situations varying some of the parameters involved. The necessary parameters were adopted from some measurements made by ITN (Nuclear and Technological Institute) in the Urgeiriça tailings piles (Reis *et al*, 2000). Local meteorological data, namely wind velocity and frequency, was used for simulating the dispersion. The unknown parameters were estimated from available data.

The calculating procedure uses the following steps: (i) estimative of the ^{222}Rn diffusivity in the waste and in the cover, (ii) estimate of the ^{222}Rn flux release to the air in the breathing zone and (iii) calculation of the ^{222}Rn concentration dispersed in the atmosphere along each wind direction, taking into account for its intensity and frequency of blow.

The contaminated site is composed by four different tailings with a total area of approximately 110 321 m^2. We considered the inexistence of a covering system in this area. The medium point for the global area was defined by the arithmetic average of the medium point for each singular area and has the following coordinates x = 20612, y = 93326.

The ^{222}Rn flux was calculated for the global area, now conceptually defined as a regular octagon. Each sector has a characteristic average wind speed. The area contaminated is the same as the polygon area, and from it, one can estimate the length of the polygon sides and the radius of the circumscribed circumference. The air breathing or mixing height was defined as 1.7 m. The ^{222}Rn concentrations were calculated in each sector at this height. The total average flux estimated is 0.2042 Bq.m^{-2}.s^{-1}.

The wind speed data in each direction (N S E W NW NE SE SW) refers to the Nelas meteorological station. The values fit in the Pasquill stability class D (neutral) so this was chosen to estimate the typical dispersion coefficients. The dominant wind direction is NE.

The concentration at the breathing height in the dominant wind direction refers only to the polygon side that limits the respective sector.

The dispersed concentrations are different in each sector. We defined a 2-km distance from the centre for simulating the ^{222}Rn dispersion in each wind direction; the same distance was considered for the dominant wind direction (NE). The dispersion results can be seen in the figures below.

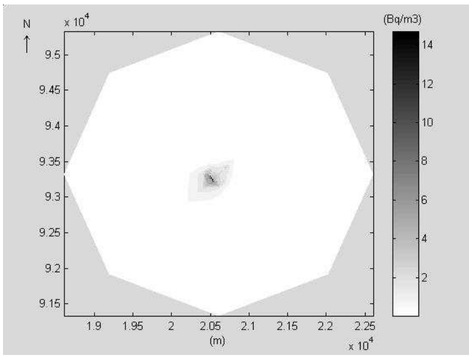

Figure 1: Radon dispersion in each wind direction, Bq.m^{-3}.

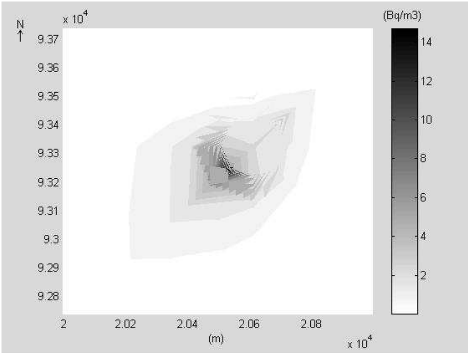

Figure 2: Radon dispersion in each wind direction, amplified, Bq.m^{-3}.

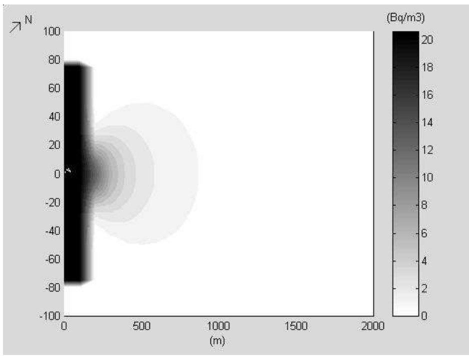

Figure 3: Radon dispersion in the dominant wind direction, Bq.m^{-3}.

A new simulation was done by estimating the radium content in the tailings assuming initial equilibrium in the two uranium series ^{238}U and ^{235}U. We have considered the same four contaminated areas (dams) but with different radium content from the previous case. We admitted that three of the tailings result from the treatment of an ore with an average grade of 1 kg.ton^{-1}, being leached at 90% and the other one results from the treatment of an ore with an average grade of 0.2 kg.ton^{-1}, being leached at 100%.

The values estimated for the radium content are respectively 10376 Bq.kg^{-1} and 2075 Bq.kg^{-1}. The ^{222}Rn flux was estimated for the global area also without cover. The value obtained was 5.96 Bq.m^{-2}.s^{-1}. The average concentration obtained inside the compartmental box was 70.2 Bq m^{-3}. All the others values from the first case study were assumed in this simulation. The results of the dispersion for this case can be seen in the figures below.

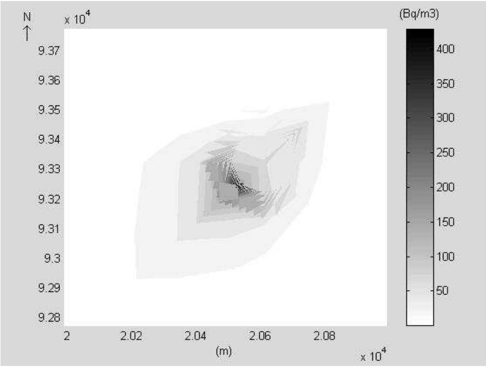

Figure 4: Radon dispersion in each wind direction, amplified (2^{nd} case), Bq.m^{-3}.

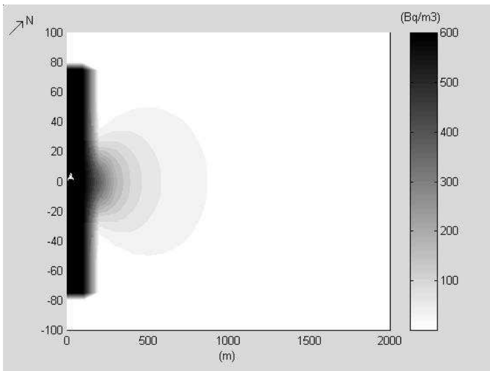

Figure 5: Radon dispersion in the dominant wind direction, (2^{nd} case), Bq.m^{-3}.

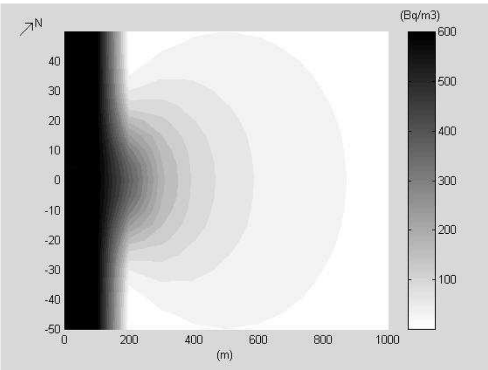

Figure 6: Radon dispersion in the dominant wind direction, amplified (2^{nd} case), Bq m^{-3}.

3. CONCLUSIONS

The work presented may be considered a first approach in modelling the dispersion of ^{222}Rn, thus allowing the assessment of exposure to uranium tailing piles. The obvious limitations are mostly related to the reduction of the source to a point. In addition, variations due to complex terrain topography cannot be modulated; our model assumes implicitly a flat plain. The wind velocity is constant with height and the dispersion is only two-directional, although this direction rotates horizontally.

We should to refer the difficulties that may be found when trying to obtain the data needed to characterise the contaminated soil and also the variability of these data, both in space and time. We stress that the radon flux depends on the diffusion coefficient, which greatly varies with the material moisture and porosity. These parameters will vary over the year, due to the climatic changing. They also depend on the radium content in the tailing which may be different in each pile and on the water retention which is also a seasonal parameter, with spread values at the Urgeiriça site (Reis *et al*, 2000).

Acknowledgements: This work has been carried out with the financial support of Foundation for Science and Technology (FCT-MCES).

4. REFERENCES

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