Environmental Modeling of the Intercompartimental Distribution of Low-Level Radioactive Wastes

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INTRODUCTION

The uranium mining in Portugal was active almost during one century in 62 different sites. One of these sites refers to the former Urgeiriça uranium mine, an underground mine which was considered to be the most important uranium exploitation in the country. This former mine is surrounded by small farms and country houses and most of the local population lives in a village located at 2 km from the mine.

The Urgeiriça mine was exploited between 1913 and 1944 for radium extraction and from 1951 to 1991 for the production of uranium concentrates. The ores from this mine, as well as from other uranium mines exploited in Portugal, were processed in the Urgeiriça uranium mill facility. About 2,5 million tones of low level radioactive wastes from this facility were disposed in an open air area originating the largest tailings pile of the mining area, named as the "Old Dam", with an average height of 14 meters covering 13,3 hectares in this region (Figure1).

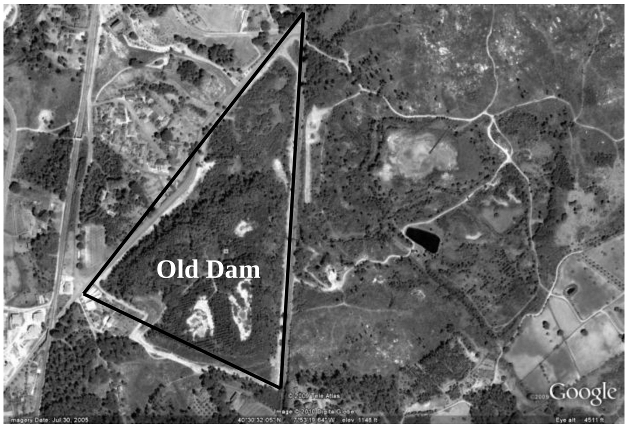

Fig. 1. Aerial photo of the Urgeiriça "Old Dam".

Since 1996 the Portuguese government had to deal with the decommissioning of the mines, mills and other facilities and the rehabilitation of the mining sites.

In particular, for the remediation of the Urgeiriça site, the reclamation program was announced in 2005. For this site the rehabilitation of the "Old Dam" was considered a key element of the overall environmental reclamation program [1].

Radioactivity measurements done at the "Old Dam" showed high concentrations for the radionuclides of the ^{238}U decay series [2]. It is also well known that these tailings were a source of external radiation and furthermore, a powerful source of radon originating higher radiation levels comparing to the background values. Due to precipitation, water, wind and soil erosion, radionuclides could be transported and redistributed in this region as well as in its vicinity. Therefore, people living in the nearby areas could be exposed to additional levels of radiation. This study intended to evaluate the environmental impacts from the "Old Dam" tailings disposal to the soil, water, and air.

DESCRIPTION OF THE ACTUAL WORK

The work developed involved an integrated approach in order to assess quantitatively the radionuclides dispersion in different environmental compartments, induced by the existence of uranium tailings disposals and storages of other low-activity mining wastes.

The important routes of contamination were identified and the radionuclides of major concern were selected, in each one of the compartments, based on their chemical, physical and radiological properties. Each compartment represents an environmental media: air, soil, water, vegetation and biota. A model for the radionuclides transfer to the food chain was also developed and incorporated allowing estimating the concentration of the different radioisotopes in all the environmental compartments.

Depending on the characteristics of the radioactive sources different types of possible releases were evaluated using phenomenological models. Models of environmental release, dispersion, transport and fate within each environmental compartment, as well as models of transfer between them, were applied to onsite available data.

The model outputs are the radionuclide activities in each environmental compartment, where relevant exposure end-points were selected. These data, complemented with an exposure scenario, allowed a dose estimate as well as a quantitative environmental risk assessment.

Radon Release and Wind Dispersion

Tailings samples were collected to determine radionuclides concentration [2]. In particular, the radium content in the tailings was extremely heterogeneous ranging from 3500 to 66500 Bq/kg [2]: a weighted average value of 12900 Bq/kg was adopted to simulate the radon generation.

The basic equations of diffusion across a porous medium were used for estimating the radon flux from the ^{226}Ra content in the tailings. It is also possible to describe radon transport through the tailings with or without a cover system, as the algorithm also incorporates the radon attenuation resulting from a cover system placed over the radioactive waste disposal. On the other hand, the thickness of the cover system could also be optimized to allow a radon flux inferior to a stipulated one.

The radon concentration released at a defined distance from the ground (1,5 meters) is estimated by a box model. This output will be the starting point for the dispersion with the wind, simulated either simultaneously in each wind direction or only in the prevailing wind direction. Radon dispersion is modeled by a modified Gaussian plume equation.

Transport and Dispersion in Groundwater and Superficial Waters

For the hydrologic transport a two-direction model simulates the release of contaminants from the tailings 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 tailings. A well with contaminated water was considered as an exposure point.

Radionuclides release from the tailings pile is simulated with a leaching model based on a sorption-desorption process. The leachate concentration is determined by the partition coefficient (describing the relative transport speed of the contaminant to the water existing in the pores), by the soil properties (bulk density and water content), by the the extent of contamination, (contaminated zone thickness and area) and by the radionuclide content in the source.

The radionuclides transport is considered to occur either in the vertical direction, through the unsaturated zone until an aquifer is reached, or in the horizontal direction, through the saturated zone, flowing to the considered well, where the contaminants could potentially become accessible to humans or other to forms of life.

Radionuclides transport and fate in groundwater is simulated with the generic diffusion/dispersion–advection equation with radioactive decay and retardation.

The hydrologic model also quantifies radionuclides transport in superficial waters; the liquid effluents from the uranium chemical treatment at Urgeiriça site used to be discharged, after treatment (neutralization and radium precipitation), into a stream near the contaminated site, which is also the main watercourse that drains the tailings area.

Transfer to Vegetation

Modeling the radionuclide transfer to vegetation contemplates several processes. They describe and quantify the radionuclides transfer mechanisms, transport, absorption and translocation to vegetation. The main purpose was to develop a radionuclide transfer model through the food chain.

The conceptual model is based on the assumption that each one of the transfer processes may have either origin in air and/or in soil. In the first case, the processes involved are deposition, interception and retention. In the second case, the radionuclide behavior in soil and its mobilization reflects the radionuclides physics and chemical properties, soil properties, the type of vegetation and local hydrology and geology characteristics. Both contamination pathways were combined in a global model that simulates the radionuclide transfer and estimates the total radionuclides concentration resulting from direct deposition, root uptake and/or irrigation with the well contaminated water.

Transfer to the Food-Chain

Contamination of the food-chain by radionuclides released into the environment may occur by primary ingestion of contaminated pasture by animals followed by ingestion of contaminated animal products (dairy or meat). A dynamic model was developed to describe mathematically the radionuclide behavior in the pasture-cow-milk exposure route and predict the activity concentration in each sub-compartment. It is also possible to determine potential long-term radionuclides accumulations in these sub-compartments.

The dynamic model is defined by a system of linear differential equations with constant coefficients describing a mass balance in different compartments. For each compartment, a transient mass balance equation defines the relationships between the inner transformations and the input and output fluxes. The output concentration within each compartment can then be transcribed to doses values based on a simplified exposure pathway and a pre-defined critical group.

RESULTS

For the model simulation, the tailings pile was considered to have an area of 133000 m^2 with a radium concentration of 12900 Bq/kg. This originates a radon flux of 5,29 Bq.m^{-2}.s^{-1}, if no cover-system is considered, leading to a concentration of 116,38 Bq.m^{-3} at the breathing height.

A multi-layer cover system of 5 meters was proposed in the rehabilitation plan for covering the tailings pile [3]. Model simulation, for this cover system, reduces the radon flux to 0,0012 Bq.m^{-2}.s^{-1} which originates a radon concentration of 0,0264 Bq.m^{-3} for the breathing height.

For radon dispersion with the wind it was possible to dilute radon concentration to negligible values at about 2 km from the release point (Figure 2). From the dispersion pattern with the distance it was observed that the dominant wind direction is towards Northeast (Figure 3).

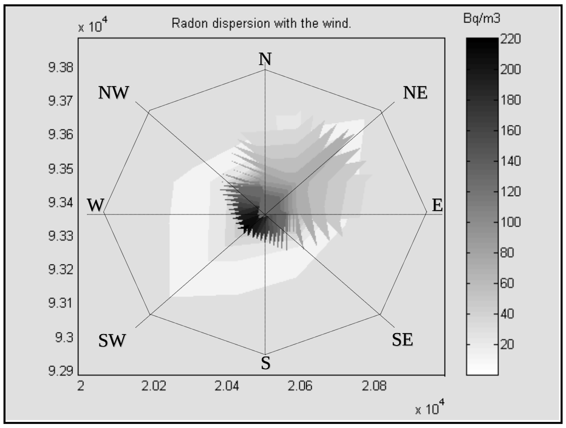

Fig. 2. Radon dispersion with wind (Bq/m^3).

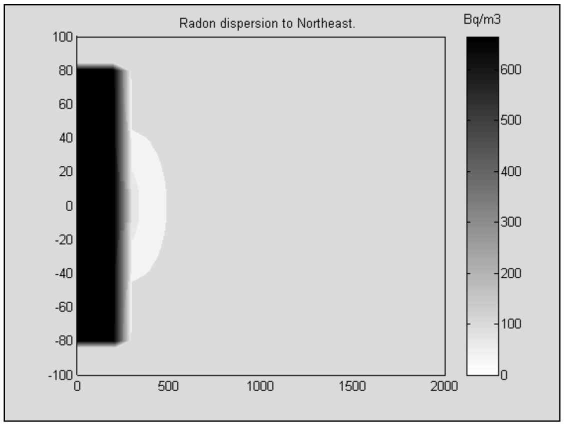

Fig. 3. Radon dispersion to Northeast (Bq/m^3).

For the radionuclides transport and dispersion in the groundwater the model was initially applied to uranium, thorium, radium, polonium and lead. However, only the results obtained for uranium and radium were relevant as the radionuclides with relatively long half-life (radium and uranium) are the ones of importance in the transport process due to the slow rate of contamination migration.

Total average values for uranium and radium concentration in the well water were: 1,6 Bq/L and 0,4 Bq/L, respectively. These values are achieved in the model within the first 30 years after the contamination reaches the aquifer [4].

It was also observed that there are two preferential directions for the underground contamination plumes, one for uranium and the other for radium. These two directions suggest that SW-NE direction is preferential for radium dispersion and that NW-SE direction is preferential for uranium dispersion. This means that, probably, there were two preferential contamination sources: at south for radium and at north for uranium.

To simulate the radionuclides transport in superficial waters, an effluent with 960 Bq.m^{-3} in ^{226}Ra [5] was considered to be discharged in the watercourse. The exposure point was considered to be located at 2750 m downstream from the discharge point. Radium concentration in the sediments was also estimated for the same exposure point.

The output showed a clear decrease in radium concentration downstream with the distance to the discharge point. The output value at the exposure point was 5,46 Bq m^{-3}. Radium concentration in the watercourse sediments follows the same pattern. The output value at the exposure point was 15,59 Bq.m^{-3}. Comparing model outputs with measured data for radium concentration in the exposure point it was observed that the highest values, both in water and sediments, do not correspond to the considered discharge point but to a distance comprised between 1650 and 2750 m away from it. Afterwards, radium concentration decreases with the distance. This suggests that probably there are other contamination sources beside the discharged effluent.

For radionuclide transfer through the food chain it was considered that pasture contamination is mostly due to deposition. Two compartments were considered for contamination transfer: soil and pasture. The endpoints are radionuclides concentrations in soil, pasture, cow and milk.

The model outputs represent the time variation for the considered radionuclide concentration in the pasture-cow-milk exposure pathway. It was observed that the general tendency is a slow decrease of radionuclides content in each compartment following the maximum value achieved within 30 days: 0,040 Bq/kg for ^{226}Ra; 0,023 Bq/kg for ^{210}Pb and 0,280 Bq/kg for ^{210}Po.

One year after achieving maximum values, radionuclides content in milk decreases as: 0,0012 Bq/kg for ^{226}Ra; 0,005 Bq/kg for ^{210}Pb and is null for ^{210}Po [6].

CONCLUSIONS AND DISCUSSION

The necessary parameters for model inputs were adopted from different sources: some parameters were adopted from measurements referring to the particular contaminated site, the Urgeiriça uranium tailings disposals, and others were adopted from published data.

Different radionuclides were considered in different sub-compartments due to their relevance in that particular environment.

The results of the global model for selected radionuclides, using as source data the results from the site survey, showed reasonable high concentrations in air, soil, underground and superficial waters, vegetation and transfer through the food-chain for nuclides of the ^{238}U decay series. The use of an integrated model contemplating different possible contamination pathways allowed to evaluate and quantify, preliminary, the extension of the site contamination by mining activities as well as the potential contribution from others sources to the overall contamination scenario.

Comparing radionuclides concentrations or activities determined by the model simulations with the legal limits helped to identify the sectors that required priority remediation measures.

For the Urgeiriça tailings disposal “Old Dam” and other mining waste facilities the remediation work was already implemented and monitoring is under way. There are still some other areas in this site, as well as other similar sites, where several remediation works are still going on.

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