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The effects of preferential flow and soil texture on risk assessments of a NORM waste disposal site

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ABSTRACT

This paper investigates the environmental fate of radionuclide decay chains (specially the ^{238}U and ^{232}Th series) being released from a conventional mining installation processing ore containing natural occurring radioactive materials (NORMs). Contaminated waste at the site is being disposed off in an industrial landfill on top of a base of earth material to ensure integrity of the deposit over relatively long geologic times (thousands of years). Brazilian regulations, like those of many other countries, require a performance assessment of the disposal facility using a leaching and off-site transport scenario. We used for this purpose the HYDRUS-1D software package to predict long-term radionuclide transport vertically through both the landfill and the underlying unsaturated zone, and then laterally in groundwater. We assumed that a downgradient well intercepting groundwater was the only source of water for a resident farmer, and that all contaminated water from the well was somehow used in the biosphere. The risk assessment was carried out for both a best-case scenario assuming equilibrium transport in a fine-textured (clay) subsurface, and a worst-case scenario involving preferential flow through a more coarse-textured subsurface. Results show that preferential flow and soil texture both can have a major effect on the results, depending upon the specific radionuclide involved.

1. Introduction

Much attention over the years has focused on the possible long-term effects of waste disposal on the environment and human health. Numerical models can be important tools for landfill designers and operators, as well as for environmental regulators, to predict the long-term subsurface transport of contaminants leached from solid waste disposal sites as a function of climatologic conditions and landfill design. Performance assessment is especially important for landfills containing radioactive mining wastes resulting from the processing of naturally occurring radioactive materials (NORM and TENORM wastes), often involving the U and Th radionuclide decay series [1–3]. TENORM refers to technologically enhanced naturally occurring radioactive material in which human activities (e.g., mining of ores or drilling of oil and gas) have increased, in comparison to the altered state, the concentration of naturally occurring radionuclides, and/or enhanced the potential for radiation exposure to humans and biota [4].

A large number of models are now available to predict the transport of aqueous contaminants from solid waste landfills or similar facilities in soils and groundwater. They range from relatively simple mass balance models designed specifically for landfills, such as the HELP code [5], to more comprehensive multi-dimensional models for more general variably-saturated flow and contaminant transport problems, such as the TOUGH [6] and HYDRUS [7] families of codes. While these models have seen application to a wide range of problems, including for performance studies of waste disposal sites and capillary barriers, they have been used only sparingly in combination with detailed risk assessments of NORM waste sites [1,8].

Safety or dose assessments for long time periods are essential for the management of long-lived NORM and TENORM contaminants. Since institutional controls may not last much longer than a few decades after site closure, the wastes are unlikely to remain safe. Natural leaching of radionuclides into the adjacent biosphere is almost certainly to occur within a few centuries or even earlier. Since NORM wastes in Brazil are typically deposited in industrial landfills, regulations require a safety assessment of the disposal facility using a leaching and off-site transport scenario.

In this paper we evaluate radionuclide transport from a NORM disposal site in Amazonia using the HYDRUS-1D numerical software package [9]. A sensitivity analysis is carried out of various factors affecting radionuclide transport, and indirectly the risk assessment, including soil texture and especially preferential flow

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through the waste layer, the vadose zone and the granular aquifer below and downgradient from the site.

2. Mining site and disposal facility

The radionuclide transport and safety assessment is applied here to a still operating NORM waste site located in the north of Brazil close to the equator. The analysis permits an evaluation of the potential consequences of slag disposal in a tropical forest. The exact location of the site is not given here for reasons of confidentiality. Ore extracted from an open pit mine at the site contains natural radionuclide decay chains, with the daughter products mostly in secular equilibrium with the parent. Different physical and chemical procedures (crushing/grinding, separation, concentration) are employed at the site to produce a final concentrate for pyrometallurgical processing. Pyrometallurgy involves treatment in a furnace at high temperatures to separate metals from large amounts of waste rock still present in the concentrate. The waste is most often removed in the form of slags in which all radioactive species have become more concentrated as compared to the original ore, except for radioactive lead which, together with its stable isotope, may be volatilized during the production process.

The radioactive components of the slag have long decay chains (14 daughters for the ^{238}U series) that end in stable lead. Modeling the transport of these contaminants hence should involve not only the parent nuclide, but also all of its daughter products. The calculations with the decay chains are not trivial since the daughter radionuclides formed during the decay process all have different physical and chemical properties. The daughters hence may move faster or slower than the parent in different layers of the disposal system and groundwater, while also having radiological toxicities that differ from those of the parent.

The concentrations of short-lived daughters were calculated from the activity of the parent nuclides (directly measured) using the assumption of secular equilibrium and applied to elements with half-lives of not more than 1 year. The general parent/daughter decay chain is of the form:

$$^{238}\text{U} \rightarrow {}^{234}\text{Th} \rightarrow {}^{234}\text{Pa} \rightarrow {}^{234}\text{U} \rightarrow {}^{230}\text{Th} \rightarrow {}^{226}\text{Ra} \rightarrow {}^{222}\text{Rn}$$
$$\rightarrow {}^{218}\text{Po} \rightarrow {}^{214}\text{Pb} \rightarrow {}^{214}\text{Bi} \rightarrow {}^{214}\text{Po} \rightarrow {}^{210}\text{Pb} \rightarrow {}^{210}\text{Bi}$$
$$\rightarrow {}^{210}\text{Po} \rightarrow {}^{206}\text{Pb(stableelement)}$$

In our analysis we limited ourselves to the chain $^{238}U \rightarrow {}^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra \rightarrow {}^{210}Pb$. Activities of the various isotopes are reported here in units of Bq/l for liquid-phase concentrations and Bq/g for solid-phase concentrations.

The facility containing the slags consisted of a modular disposal unit constructed of earth materials. An impermeable liner placed under the waste layer (a requirement of the regulatory agency) was not considered in our analysis. Average annual precipitation measured at a location close to site was 2430 mm, and the average evapotranspiration rate (as calculated with the Penman–Monteith equation) was 1610 mm. Adjusted for runoff, the long-term average recharge rate at the site was estimated to be 657 mm/y. This recharge rate was used as a surface flux boundary condition for the steady-state simulations of water flow through the site and into groundwater.

The dimensions of the disposal unit were 70 m wide by 100 m long and 6.0 m deep. Initial radionuclide concentrations were 71 Bq/g for ^{238}U and ^{234}U, 67 Bq/g for ^{230}Th, 63 Bq/g for ^{226}Ra and 4.8 Bq/g for ^{210}Pb. These values suggest that U is likely in secular equilibrium with ^{230}Th. Since ^{226}Ra is more mobile than the other radionuclides, there will be a loss of Ra by leaching. Because also some ^{222}Rn will escape through volatilization, both ^{226}Ra and ^{222}Rn will not be in equilibrium with ^{210}Pb during the pyrometallurgical process, and radioactive lead will volatize simultaneously with stable lead.

The slag waste layer had a measured bulk density of 1.89 g/cm^3. The unsaturated hydraulic properties of the waste layer and the underlying vadose zone were described using the constitutive equations [10]:

$$S_e(h) = \frac{\theta(h) - \theta_r}{\theta_s - \theta_r} = \frac{1}{[1 + (\alpha h)^n]^m} \quad \left(m = 1 - \frac{1}{n}\right) \tag{1}$$

$$K(h) = K_s S_e^{0.5} [1 - (1 - S_e^{1/m})^m]^2 \tag{2}$$

where S_e is effective saturation, h is the pressure head, θ is the volumetric water content, θ_r and θ_s represent the residual and saturated water contents, respectively, K_s is the saturated hydraulic conductivity, and α and n are empirical shape parameters [10]. Values of the hydraulic parameters of the waste layer were estimated from long-term drainage experiments on large lysimeters containing slags from the site, operated by the Poços de Caldas Laboratory of the Brazilian Nuclear Energy Commission (CNEN) in Minas Gerais, Brazil.

We also tried to estimate distribution coefficients (K_d values) of the radionuclides from the lysimeter experiments with the slags, but the concentrations (except those of ^{226}Ra) of the leachate were too low and too variable to obtain reliable estimates. Preliminary laboratory batch measurements at the Poços de Caldas Laboratory indicated radionuclide K_d values very close to those of a clay soil [11], except for Ra, which was closer to that of sand. For these reasons we used K_d values typical of clay for U, Th, and Pb (1.6, 5.8, and 0.54 m^3/kg, respectively), and the measured value (0.88 m^3/kg) for Ra. The same values were initially used for all layers (slag, vadose zone, aquifer). The assumed K_d values produced very high retardation factors (R) factors for all of the radionuclides involved (up to about 40,000 for Th), and hence very low effective transport rates. In another analysis we also used the hydraulic parameters and K_d values of a typical coarse-textured (sandy) soil to show the very significant effects of soil texture on the simulations.

The unsaturated zone below the waste layer consisted of reddish Belterra clay as described by Dennen and Norton [12] and Truckenbrodt and Kotschoubey [13]. Although Oxisols such as those found at the site can have very high clay contents (up to 90%), they often show relatively high infiltration rates more typical of coarse-textured soils, and considerable macroporosity. Soil texture (clay), bulk density (1.3 g/cm^3) and the saturated hydraulic conductivity (K_s = 21 m/y) of the vadose zone were locally measured. Other hydraulic parameters in Eq. (1) for Belterra clay were taken from Belk et al. [14], except for the value of α (we used 4.5 m^{-1}), which was incorrectly printed in [14]. The hydraulic parameters in the Belk et al. paper were otherwise very much consistent with generic values suggested by Hodnett and Tomasella [15] and Tomasella et al. [16].

The saturated zone consisted of red saprolite resulting from in-situ alterations of acid volcanic rocks, interspersed with lighter-colored (white, yellow) lenses derived from feldspars. The phreatic aquifer had an average thickness of 4.5 m, with a natural hydraulic gradient of 0.05625. Geotechnical essays of the aquifer produced values for the saturated hydraulic conductivity (K_s) between 4.22×10^{-5} and 7.9×10^{-4} cm/s. For our calculations we used a value of 1.7×10^{-4} cm/s for clay soil textural class as estimated with the pedotransfer function module of HYDRUS-1D, which is based on the hierarchical neural network (Rosetta) approach of Schaap et al. [17].

For the calculations we assumed a 6-m thick homogeneous waste layer (overlain by a negligibly thin layer of local soil), and a 5-m thick unsaturated zone consisting of Belterra clay. Radionuclides leached vertically from the vadose zone were assumed to mix with