

# Radon Diffusion Modeling for the Clive DU PA

Clive DU PA Model v1.4

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## 1.0 Radon Input Summary Distribution Summary

A summary of parameter values and distributions employed in the radon diffusion modeling component of the Clive Depleted Uranium Performance Assessment Model (the Clive DU PA Model) is provided in Table 1. Additional information on the derivation and basis for these inputs is provided in subsequent sections of this report.

For distributions, the following notation is used:

- Beta(  $\mu$ ,  $\sigma$ ,  $min$ ,  $max$  ) represents a generalized beta distribution with mean  $\mu$ , standard deviation  $\sigma$ , minimum  $min$ , and maximum  $max$ .

**Table 1. Summary of distributions for radon diffusion modeling**

| GoldSim Model Parameter           | Distribution or Value      | Units              | Notes           |
|-----------------------------------|----------------------------|--------------------|-----------------|
| free-air diffusivity of radon gas | 0.11                       | cm <sup>2</sup> /s | See Section 4.1 |
| E/P ratio for DU waste            | Beta( 0.290, 0.156, 0, 1 ) | —                  | See Section 5.2 |
| E/P ratio for other materials     | 1                          | —                  | See Section 5.3 |

## 2.0 Introduction

This white paper documents the modeling of diffusive transport of the radioactive noble gas radon-222 (<sup>222</sup>Rn) in the Clive Depleted Uranium (DU) Performance Assessment (PA) Model (the Clive DU PA Model, or the Model) for the EnergySolutions' radioactive waste disposal site at Clive, Utah. The Clive DU PA Model is dominated by a single waste form: depleted uranium. This material predominantly consists of <sup>238</sup>U. This uranium isotope is a parent of <sup>222</sup>Rn, its rate of production controlled by the decay rate of the <sup>238</sup>U and its escape from the solid waste form. Using a basic representation of fate and transport, radon emanation (production and escape) is accounted for, as is Henry's Law partitioning into water, and the gas is allowed to diffuse in pore air using GoldSim's internal diffusion processes, as corrected for unsaturated media and for numerical dispersion. Radon emanation and diffusion are discussed below.

The diffusion of radon is a special modeling problem, due to the coarse discretization (in time and space) of the Model relative to the diffusivity of radon and the short half-life of <sup>222</sup>Rn of 3.8 days. This combination causes the Model, which is set up as a finite difference model (i.e. a one-dimensional stack of compartments representing a column), to potentially over predict the diffusive flux of <sup>222</sup>Rn through the ground surface.

A solution is found in "calibrating" the GoldSim calculations to radon ground surface flux estimates resulting from an analytic solution presented in the Nuclear Regulatory Commission (NRC) Regulatory Guide 3.64 *Calculation of Radon Flux Attenuation by Earthen Uranium Mill Tailings Covers* (NRC 1989), and in NUREG/CR-3533 *Radon Attenuation Handbook for Uranium Mill Tailings Cover Design* (NRC 1984), referenced in NRC (1989). By varying the diffusivity of radon, the ground surface flux estimated by the Clive DU PA Model can be matched (assuming an initially uncontaminated cover and in the absence of other transport processes) to the NRC model calculation. Once that is done, other contaminant transport processes, such as advective water transport and biotically-induced transport, are enabled and the model runs as a fully-coupled system.

An additional parameter required to model the transport of radon is the escape/production (E/P) ratio (also known as the emanation factor and by other terms). In this white paper, factors influencing the magnitude of E/P ratios are discussed and the input parameter distributions for DU waste materials are presented.

## 3.0 GoldSim Diffusion of Radon

### 3.1 Background

Radon-222, a gaseous radioactive isotope of radon, is produced by the alpha decay of radium-226 ( $^{226}\text{Ra}$ ). The NRC models estimate the diffusive flux of  $^{222}\text{Rn}$  as a function of the concentration of  $^{226}\text{Ra}$  in a waste. A two-step method for estimating the cover thickness required to attenuate the radon flux from uranium mill tailings is described in NRC (1984 and 1989) and has been used for calculating radon flux at the Area 5 Radioactive Waste Management Site (RWMS) at the Nevada National Security Site (NNSS, formerly the Nevada Test Site) (Shott et al. 1998, p. 3-107). This approach provides an analytic solution for radon flux at steady state and is summarized in Appendix A. This approach was used in two RWMS PA models developed by Neptune and Company, and was readily applied at the NNSS sites, since the waste cover was a homogeneous monofill of local alluvium (Neptune, 2005). Radon transport through the waste and a clean cover was modeled in GoldSim using a series of diffusive flux links between cells. This approach is mathematically identical to a finite difference model of the system, as detailed in the GoldSim Contaminant Transport Manual, Chapter 4 (GTG, 2011).

The cover system for the portion of the Federal Cell that is allocated for disposal of DU (the Federal DU Cell), however, is more complex. Instead of using a single material in the cover, the layering above the DU waste is as follows, from the bottom up: Directly atop the DU waste lies generic Class A waste, which is represented in the DU model as Unit 4 material with no inventory. Above that is a specially-constructed radon barrier, made of two layers of compacted clay. Intact compacted clay is an especially effective barrier to radon diffusion, as it has a low permeability and high moisture content, and radon partitions between air and water. Above this clay is an evapotranspirative (ET) cover consisting of a frost protection layer, and an evaporative zone and surface layer made up of Unit 4 soils with varying admixtures of gravel in the surface layer. In essence there are three materials above the radon-producing DU waste, so the simple monolayer analytical models presented in the NRC Regulatory Guide (NRC 1989) are insufficient. For systems involving more than two layers, the practitioner is referred in that Guide to the previously published NRC Handbook (NRC 1984). This is the source of the analytical equations built into the Clive DU PA Model for purposes of calibration of the native GoldSim diffusion calculations.

While these analytical solutions are specifically acceptable to the NRC, use of this approach does not account for the coupling of other processes that may contribute to the radon flux at the ground surface. For example, diffusion of radium and other radon parents through the clay in the liquid phase is possible, and biota can translocate radon and its parents. These processes can be discretely disabled in the Clive DU PA Model for the purposes of examining their significance in determining radon ground surface flux.

## 3.2 Radon Diffusion

The transport of radon presents special modeling challenges. The diffusion of gas phase constituents follows concentration gradients, and these vary along the diffusive path. Local concentrations are subject to other transport processes, notably partitioning into water (governed the Henry's Law constant) and encountering sinks like the atmosphere. In the PA modeling of radioactive constituents, most radionuclides have relatively long half-lives, and the concentration gradients are not much affected by decay and ingrowth. Radon isotopes, however, have short half-lives relative to the rate of diffusive transport processes, can move quickly in pore air, and decay to a chain of radionuclides that can be significant in terms of dose and risk. In the Clive DU PA Model, the radon isotope of interest is  $^{222}\text{Rn}$ , with a half-life of about 3.8 days. With this short half-life,  $^{222}\text{Rn}$  decays away quickly enough that the decay alone can produce strong concentration gradients, causing additional challenges in numerical simulation.

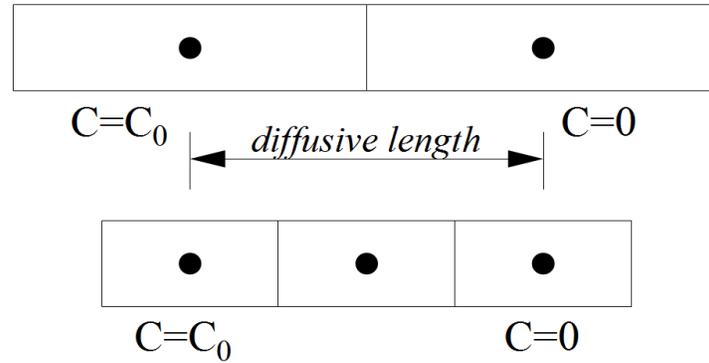
Chemical engineers are faced with similar issues in process plants, where chemicals in a process that moves through the plant are simultaneously undergoing chemical transformation to other substances. The quantification of this effect is called the Damköhler number. The value can be expressed in a number of different ways for different applications, and in the case of this Model, it is the ratio of the decay rate to the diffusive mass transport rate. For  $^{222}\text{Rn}$ , with its high rate of decay, the Damköhler number is also high, indicating that diffusive transport will be over-predicted in a coarsely-discretized model such as the Clive DU PA Model.

## 4.0 Diffusion Calculations in the Clive DU PA Model

### 4.1 Diffusion in Discretized Models

Compartment models (and all spatially-discretized models) tend to overestimate diffusive flux. The discrepancy in estimated flux is due to the discretized model's inability to accurately represent the fine spatial distribution of radon concentration using the model's coarse spatial discretization—a limitation shared by other finite difference models. Since the compartments are mathematically modeled as being fully mixed at all times, they tend to move radon through the system faster than it would in reality—a phenomenon known to modelers as numerical dispersion.

One approach for correcting or calibrating the flux is based on developing a ratio of the calculated flux to the exact flux (an analytical solution). The derivation for the exact flux and the ratio is given in Appendix B. The problem is illustrated for a simple case of a steady-state system with a one-dimensional diffusion pathway for radon in air with no porous media. For the "1 cell interface" problem the concentration in the first cell is held to a constant value and the last cell is advectively "flushed" to maintain a concentration of zero. Three GoldSim models were built using 1, 2, and 10 cell interfaces to model a 1-meter column. Configurations for the 1- and 2-cell interface models are shown in Figure 1.



**Figure 1. One-cell interface and two-cell interface diffusion model schematics.**

Steady-state fluxes were calculated for diffusivities of  $11 \times 10^{-6} \text{ m}^2/\text{s}$  (or  $0.11 \text{ cm}^2/\text{s}$ , the free air diffusivity of radon from Rogers and Nielson, 1991),  $1.1 \times 10^{-6} \text{ m}^2/\text{s}$ , and  $0.11 \times 10^{-6} \text{ m}^2/\text{s}$  and compared to the exact fluxes calculated from analytical solutions of the diffusion equation. These results are shown in Table 2.

**Table 2. Comparison of exact and GoldSim radon ground surface flux calculations.**

| No. of Cells | Diffusivity<br>( $\times 10^{-6} \text{ m}^2/\text{s}$ ) | Exact Flux<br>( $\times 10^{-8} \text{ g/yr}$ ) | GoldSim Flux<br>( $\times 10^{-8} \text{ g/yr}$ ) | Error<br>(percent) |
|--------------|--|---|---|--------------------|
| 1            | 11   | 3.499   | 3.614   | 3.3                |
| 1            | 1.1  | 1.095   | 1.479   | 35.1               |
| 1            | 0.11   | 0.0237  | 0.2142  | 803.8              |
| 2            | 11   | 3.724   | 3.756   | 0.9                |
| 2            | 1.1  | 1.484   | 1.618   | 9.0                |
| 2            | 0.11   | 0.04231   | 0.113   | 167.1              |
| 10           | 11   | 3.883   | 3.887   | 0.1                |
| 10           | 1.1  | 1.910   | 1.918   | 0.4                |
| 10           | 0.11   | 0.08764   | 0.09288   | 6.0                |

Comparison of exact and GoldSim fluxes in Table 2 shows that for any given number of cells representing the system, the error in the GoldSim flux increases as the diffusivity decreases. In addition, for a given value of diffusivity, the error decreases as the number of cells is increased.

The ratio of the linear flux calculated by GoldSim to the exact flux  $R$  (discussed in Appendix B) is

$$R = \frac{\sinh(bL)}{bL} \quad (1)$$

where

$L$  = diffusion path length and

$b$  = a function of the decay coefficient and the diffusivity.

For a one cell model, the exact flux is obtained from the GoldSim flux by dividing the GoldSim flux by  $R$ . Table 3 shows the close agreement between exact and corrected GoldSim fluxes for a one cell model.

**Table 3. Exact and corrected GoldSim flux comparisons.**

| No. of Cells | Diffusivity<br>( $\times 10^{-6}$ m <sup>2</sup> /s) | Exact Flux<br>( $\times 10^{-8}$ g/yr) | GoldSim Flux<br>( $\times 10^{-8}$ g/yr) | Error<br>(percent) |
|--------------|--|--|--|--------------------|
| 1            | 11   | 3.4980                                 | 3.5000                                   | 0.1                |
| 1            | 1.1  | 1.0948                                 | 1.0956                                   | 0.1                |
| 1            | 0.11   | 0.02368                                | 0.02370                                  | 0.1                |
| 2            | 11   | 3.7436                                 | 3.7532                                   | 0.3                |
| 2            | 1.1  | 1.5259                                 | 1.5499                                   | 1.6                |
| 2            | 0.11   | 0.04491                                | 0.04626                                  | 3.0                |
| 10           | 11   | 3.8491                                 | 3.8528                                   | 0.1                |
| 10           | 1.1  | 1.8043                                 | 1.8095                                   | 0.3                |
| 10           | 0.11   | 0.073252                               | 0.072097                                 | -1.6               |

To obtain a value of  $R$  for a multiple cell model,  $R$  is calculated using an effective length,

$$L_{eff} = \frac{L}{1.15n} \quad (2)$$

where  $n$  is the number of cells. The diffusive areas for each GoldSim cell are then reduced by dividing by  $R$ . Exact and corrected GoldSim fluxes using this method for two- and five-cell interface models are shown in Table 3. In all cases, the error is reduced to 3 percent or less.

A similar approach was applied to correct the radon fluxes for the NNSS Area 5 RWMS GoldSim Model without success. The ratios obtained from the simple problem described above with a constant known concentration at one end of the diffusion path were not applicable to the system at the RWMS where the radon source is distributed spatially throughout the depth of the waste. For such shallow land burial waste configurations, without correction, the flux calculated by GoldSim was 11 times the flux calculated using the NRC method. Reducing the diffusive areas by  $R$  resulted in only a 13 percent reduction in the flux calculated by GoldSim. The behavior of radon flux at the Clive facility would be expected to suffer the same limitation, and would result in an unrealistic over-prediction of the flux at the ground surface.

## 4.2 Scaling the GoldSim Calculations to the NRC's Analytical Method

An empirical approach was adopted in which a scaling factor was obtained that would reduce the value of the flux calculated in GoldSim to the value of flux calculated using various implementations of the NRC method. While this technique was originally developed for the NNSS RWMSs, it applies equally well for the Clive Facility. This approach is implemented in the Clive DU PA Model by multiplying the bulk diffusivity for radon by a scaling factor unique to each material in the cover system. The scaling factor is determined by calculating radon flux using both the radon flux estimated “natively” by GoldSim in the Model and a selected variant of the NRC method (coded into the Model itself) and adjusting the scaling factor until the time histories of ground surface flux matched sufficiently well. This is done using the Radon Calibration procedure (and the associated dashboard) built into the model, and comparing the resulting flux estimates graphically. In the Clive DU PA Model, there is a different effective radon diffusivity calibration factor for each of the different materials comprising the cover.

Calibration factors are developed for the various cover layers, making use of some special GoldSim functionality: In most GoldSim models the fluid properties of air, including the relative diffusivity of radon in air, are defined globally. In such a case, scaling the relative diffusivity would scale it everywhere in the model. To circumvent this global influence, local definitions of the fluid Air have been created for each modeled layer above the DU waste. This allows unique values of the bulk diffusion coefficient to be used for each layer (GoldSim Contaminant Transport Manual Chapter 4). Unique scaling factors for the generic Class A waste, clay radon barrier, and the upper cover materials are implemented and are to be calibrated independently.

## 5.0 Radon Escape/Production Ratio

### 5.1 Background

The fraction of  $^{222}\text{Rn}$  produced by decay of radium-226 ( $^{226}\text{Ra}$ ) that is released from the solid matrix is known as the E/P ratio, as well as the emanation coefficient, the emanation factor, or emanating power (Nielson and Sandquist, 2011). When  $^{226}\text{Ra}$  decays a small fraction of the decay energy, 0.1 MeV, is carried by the recoiling  $^{222}\text{Rn}$  atom. This is sufficient energy for the recoiling atom to travel about 45 nm in a mineral matrix, 0.1  $\mu\text{m}$  in water, and about 63  $\mu\text{m}$  in air. Recoiling atoms with just sufficient energy to stop in the air or water filled pore space will be released from the matrix and become available for transport. If there is too little energy available, the atom will remain trapped in the solid matrix. If there is too much energy, the atom will cross the pore space and be embedded in the solid matrix of a nearby grain. The E/P ratio describes that fraction of  $^{222}\text{Rn}$  that stops in the air or water-filled pore space and is free to diffuse. The E/P ratio can physically vary from 0, implying no escape, to 1, where all radon escapes.

Predicting the E/P ratio for a material is difficult as numerous factors have been identified that affect it: The E/P ratio is inversely related to grain size. The closer decaying atoms are to the surface of a grain, the more likely they will be released to the pore space. The adsorption or coprecipitation of  $^{226}\text{Ra}$  on surficial coatings increases emanation, as will cracks, fissures, or pitting of grains. In contrast, the E/P ratio is directly related to pore size. As the pore size increases, it is more likely that recoiling atoms will stop in the pore space, increasing emanation. The presence of water in the pore space also increases emanation, because the reduced particle range in water increases the likelihood that the recoiling atom will stop in the pore space. Predicting the E/P ratio of a material is particularly difficult because it requires detailed knowledge of the microscopic physical structure of the material, microscopic distribution of  $^{226}\text{Ra}$  in the material, and water content.

The E/P ratios for different types of common geologic materials have been reported. From geometrical considerations, the maximum emanation expected from a thick slab source is 0.5 and from a thin film, 1.0. The maximum E/P ratio of natural materials will lie somewhere between these two extremes. The maximum value reported for common materials is approximately 0.7 to 0.8. Reported E/P ratios for soils and rocks range from 0.02 to 0.7 (UNSCEAR 2000; NCRP 1988). The emanation factor of a single material may vary over a substantial portion of this range depending on the water content. Rock and uncrushed ores usually have lower emanation factors ranging from 0.02 to 0.26 (Nazaroff 1992). Concrete emanation factors may range from 0 to 0.3 (Rogers et al. 1994; Cozmuta et al. 2003).

## 5.2 Cover Materials

There are probably insufficient data to assign E/P ratios to  $^{226}\text{Ra}$  in earthen covers, and assigning a value of 1 takes no credit for attenuation of radon in the soil or clay materials. Radium-226 may be transported to the cover by liquid advection and diffusion, plant uptake, animal burrowing, and intruder activities. Radium transported by liquid advection and plant uptake, after sufficient time for degradation, may exist in surface coatings that are reported to have high E/P ratios. Radium transported by animal burrowing or intruder activities may be in forms similar to the original waste form.

## 5.3 Depleted Uranium Wastes

Nielson and Sandquist (2011) report that the E/P ratio of depleted uranium oxides should be expected to mimic that of natural western uranium ores, which are on average about 0.29. The bulk of the DU waste considered in the Clive DU PA Model is in the form of  $\text{U}_3\text{O}_8$ , which is similar to that of natural ores. Based on escape/production data presented in Table 1 of Nielson and Sandquist (2011), and the physical limitations of 0 and 1, a stochastic input parameter distribution to represent the E/P ratio of DU waste is fit with a beta distribution:  $\text{beta}(\mu = 0.290, \sigma = 0.156, \text{min} = 0, \text{max} = 1)$ .

## 6.0 Model Implementation

### 6.1 Calibration of Air Diffusion to Counteract Numerical Dispersion

In the Clive DU PA Model, where wastes and other materials with a variety of porous medium properties can be intermixed, calibration to analytical solutions is challenging. The most effective way to refine the solution is by refining the grid, which is the approach that is taken in the Model. This refinement reduces numerical dispersion, providing a more realistic simulation of the diffusion process. To refine the Top Slope and Side Slope columns, which integrate all the contaminant transport processes, would introduce an unreasonable computational burden, since most of the processes would not appreciably benefit from the finer discretization. By taking advantage of a clever side calculation, the model can benefit from the increased accuracy of the finer discretization, without significant computational effort.

Within the GoldSim container for the Class A South Cell (now the Federal DU Cell) Top Slope contaminant transport calculations, an additional container called RnCalibCalcs is added, with a switch for enabling and disabling its calculations. This container is devoted to determining the appropriate measure by which the radon diffusivity of  $0.11 \text{ cm}^2/\text{s}$  should be reduced in order to exactly counter the effects of numerical dispersion. The calculation consists of two columns: one coarsely and one finely discretized. The coarse discretization matches that of the main Top Slope column, with GoldSim Cell pathway elements representing layers in the cover of about 15 cm (6 in) thickness, and layers in the waste about 50 cm (20 in) thick. Each of the Cells is populated with air, water, porous media, and initial inventory just as is the main column, but the only processes represented include retardation, solubility, and air diffusion. This protects the coarse column from all the confounding factors of water advection and diffusion, biotic processes, etc.

The fine column is built the same way, but with 15 times as many Cells. That is, the cover cells in the fine column are about 1 cm thick, and those in the waste are about 3.4 cm. This fine column has significantly less numerical dispersion for the air diffusion calculations, and is used for calibration of the coarse column. The calibrated diffusion coefficients for radon in the coarse column, then, are also applied to the main Top Slope and Side Slope columns, thereby counteracting the effects of numerical dispersion there as well.

Both the coarse and fine columns are populated with porous materials that exactly match those in the Top Slope column. These reside in three distinct zones. From the bottom up, these zones are: 1) the waste zone, where radioactive wastes are disposed, and are of similar material properties as far as air diffusion is concerned, 2) the radon barrier clay layers, which consist of tightly compacted clays overlying the wastes, and 3) the upper cover materials, which are uncompacted fill materials. The radon fluxes (mass flow per area, with dimensions of  $M/L^2 \cdot T$  and units of  $g/m^2 \cdot s$  or  $pCi/m^2 \cdot s$ ) are recorded at the top of each of these three zones in both the coarse and fine columns. With less numerical dispersion, the finer column always has a lower rate of diffusion out the top of the zone.

If the coarse column's diffusion coefficient for radon is adjusted downward, it can be forced to match the finer column, producing a more accurate flux. This correction is performed sequentially, from the bottom up, and a different correction factor is applied for each material within the three zones. This results in one radon diffusion correction factor for the waste zone, one for the clays, and one for the upper cover layers. The correction factor for wastes is applied to the waste layers in the Top Slope and Side Slope contaminant transport columns. Likewise, the correction factors for the clay layers is applied to the radon barrier clay and liner clay layers in the Top Slope and Side Slope columns, and the correction factors for the upper cover layers is applied to both columns as well.

This radon calibration need be done only once, unless the layer geometries and/or material properties change significantly. Fortunately, performing the calibration in deterministic mode is sufficient, as it is robust and holds quite well even using stochastic inputs in probabilistic mode. Once the calibration has been completed, the radon calibration container may be disabled, so that it does not impose further computational burden on the model.

## 6.2 Implementation of the E/P Ratio

When an atom of  $^{226}\text{Ra}$  decays to  $^{222}\text{Rn}$ , it does one of two things: If it escapes, it can migrate away, subject to various contaminant transport processes. If it does not escape to a point where it may migrate away, then it will decay in place to polonium-218 and other short-lived progeny until it gets to lead-210 ( $^{210}\text{Pb}$ ) and eventually to stable lead. Of these progeny, only  $^{210}\text{Pb}$  is long-lived enough to be modeled for potential transport in the Clive DU PA Model.

In constructing the GoldSim Species element in the Model, the model developer defines decay chains according to branching fractions, or "stoichiometry", as GoldSim call it. Clever use of the branching fraction allows the Model to emulate the binary behavior of the decay of  $^{226}\text{Ra}$ , so that some decays to  $^{222}\text{Rn}$  and can migrate away, while the remainder does not travel as radon, since it did not escape, and decays in place to  $^{210}\text{Pb}$ . The decay of  $^{226}\text{Ra}$  is therefore assigned two branches: The first branch is decay to  $^{222}\text{Rn}$ , and the fraction of decays that follow that branch is

the E/P ratio, since these atoms have escaped. The second branch is decay directly to  $^{210}\text{Pb}$ , so that the atoms that have not escaped do not have the opportunity to migrate away as radon. The error in skipping the step of decay of  $^{222}\text{Rn}$  to  $^{210}\text{Pb}$  is negligibly small, since the half-life of  $^{226}\text{Ra}$  (1599 yr) and that of  $^{210}\text{Pb}$  (22.3 yr) are both much larger than the 3.8-day half-life of  $^{222}\text{Rn}$ .

In the DU waste, the E/P ratio is selected from the distribution presented in Section 5.3, and this fraction of decays is allowed to decay to  $^{222}\text{Rn}$ . The balance ( $1 - \text{E/P ratio}$ ) decays directly to  $^{210}\text{Pb}$ . Outside the DU waste, where the  $^{226}\text{Ra}$  is no longer in the crystalline matrix of the waste, the E/P ratio is 1, so all decays produce  $^{222}\text{Rn}$ , and none ( $1 - 1$ ) decay directly to  $^{210}\text{Pb}$ .

## 7.0 References

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## Appendix A: NRC Method for Calculating Radon Flux

The following discussion is a synopsis of the NRC method for calculating the ground surface flux of radon from a covered source of uranium mill tailings. The original notation has been preserved, and the units conversion factors in the original equations have been removed. As in any application, be sure to use a consistent set of units, such as the cgs system used here. The one-dimensional steady-state diffusion equation describing radon flux is given in NRC (1989) as

$$D \frac{d^2 C}{dx^2} = -\lambda C + \frac{R \rho E \lambda}{n} = 0 \quad (\text{A-1})$$

where

$D$  = diffusion coefficient for radon in the total pore space ( $\text{cm}^2 \text{ s}^{-1}$ ),

$C$  = radon concentration in the total pore space ( $\text{pCi cm}^{-3}$ ),

$\lambda$  = radon decay constant ( $2.1 \times 10^{-6} \text{ s}^{-1}$ ),

$R$  = specific activity of the parent material, radium-226 ( $\text{pCi g}^{-1}$ ),

$\rho$  = dry bulk mass density of soil or tailings ( $\text{g cm}^{-3}$ ),

$E$  = radon emanation coefficient (dimensionless), and

$n$  = soil or tailings porosity (dimensionless).

The radon flux is obtained from the radon concentration gradient by (NRC 1989):

$$J = -Dn \frac{dC}{dx} \quad (\text{A-2})$$

where

$J$  = radon flux ( $\text{pCi m}^{-2} \text{ s}^{-1}$ ).

The radon diffusion coefficient can be estimated using a correlation in (NRC 1989):

$$D = 0.07 \exp[-4(m - mn^2 + m^5)] \quad (\text{A-3})$$

where

$m$  = moisture saturation fraction, the volumetric fraction of saturation of pore space.

Alternatively, the correlation developed by Rogers and Nielson (1991) can be used:

$$D = D_0 n \exp(-6mn - 6m^{14n}) \quad (\text{A-4})$$

where

$D_0 =$  radon free air diffusion coefficient (0.11 cm<sup>2</sup> s<sup>-1</sup>).

In the first step of the calculation, the flux from the tailings source is calculated using (A-1) and assuming no cover material over the tailings. Boundary conditions are (Rogers and Nielson, 1984):

$$(a) \text{ zero radon concentration at the ground surface: } C(x = 0) = 0 \quad (\text{A-5})$$

$$(b) \text{ zero radon flux at the base of the source (tailings): } \frac{dC}{dx}(x = x_t) = 0 \quad (\text{A-6})$$

where

$x =$  thickness, and the subscript  $t$  refers to tailings.

The flux is given by NRC (1989) as

$$J_t = R_t \rho_t E_t \sqrt{\lambda D_t} \tanh(x_t \sqrt{\lambda/D_t}) \quad (\text{A-7})$$

This flux is used in the second step to solve for the surface flux of radon assuming continuity of flux and continuity of concentration across the tailings-cover interface:

$$J_c = \frac{2 J_t \exp(-b_c x_c)}{1 + \sqrt{a_t/a_c} \tanh(b_t x_t) + [1 - \sqrt{a_t/a_c} \tanh(b_t x_t)] \exp(-2b_c x_c)} \quad (\text{A-8})$$

where the subscript  $c$  refers to the cover. The inverse relaxation lengths are defined as

$$b_t = \sqrt{\lambda/D_t} \quad \text{and} \quad b_c = \sqrt{\lambda/D_c} \quad (\text{A-9})$$

and the interface constants are defined as

$$a_t = n_t^2 D_t [1 - (1 - k)m_t]^2 \quad \text{and} \quad a_c = n_c^2 D_c [1 - (1 - k)m_c]^2 \quad (\text{A-10})$$

where

$k$  = equilibrium distribution coefficient for radon in water and air (pCi cm<sup>-3</sup> water per pCi cm<sup>-3</sup> air).

This solution is presented without reference in NRC (1989) and Rogers and Nielson (1984). For sources thicker than 200 cm and thick covers (NRC, 1989), (A-8) reduces to

$$J_c = \frac{2 J_t \exp(-b_c x_c)}{1 + \sqrt{a_t/a_c}} \quad (\text{A-11})$$

For the Clive DU PA Model, porosities, diffusion coefficients, saturations, and distribution coefficients are the same for the waste and the cover such that  $a_c$  and  $a_t$  in (A-10) above are equal. For this case, the surface flux can be further simplified to

$$J_c = J_t \exp(-b_c x_c) \quad (\text{A-12})$$

This equation indicates that the radon flux decreases exponentially with distance through the cover.

## Appendix B: Ratio Method for Correcting GoldSim Radon Flux

Consider one-dimensional steady-state diffusion in a cell of length  $L$ . The concentration is held at one end at a value  $C_0$  and at the other end at zero. The concentration distribution is given by

$$D \frac{\partial^2 C}{\partial x^2} - \lambda C = 0 \quad (\text{B-1})$$

and the flux by

$$J = -DA \frac{\partial C}{\partial x} \quad (\text{B-2})$$

where  $A$  is the cross-sectional area.

Boundary conditions are

$C(x=0) = C_0$  (the initial concentration is maintained constant at  $x=0$ ), and

$C(x=L) = 0$  (the concentration is maintained at zero at  $x=L$ ).

Assume a solution for the concentration at any position  $x$  in the cell of

$$C(x) = C_1 \sinh(bx) + C_2 \cosh(bx) . \quad (\text{B-3})$$

Where  $b$  is some as yet unknown constant for which we intend to solve. Applying the first boundary condition,

$$C_0 = C_1 \sinh(0) + C_2 \cosh(0) .$$

Since  $\sinh(0) = 0$  and  $\cosh(0) = 1$ ,  $C_2 = C_0$ . Applying the second boundary condition,

$$0 = C_1 \sinh(bL) + C_0 \cosh(bL)$$

$$C_1 = -C_0 \frac{\cosh(bL)}{\sinh(bL)} .$$

Using B-2, B-3, and the values of  $C_1$  and  $C_2$  with

$$\frac{\partial C}{\partial x} = b C_1 \cosh(bx) + b C_2 \sinh(bx) ,$$

the flux at the end of the cell ( $x=L$ ) is

$$J = -ADb \left[ -C_0 \frac{\cosh(bL)}{\sinh(bL)} \cosh(bL) + C_0 \sinh(bL) \right]$$

$$\begin{aligned}
 &= -ADbC_0 \left[ -\frac{\cosh^2(bL) + \sinh^2(bL)}{\sinh(bL)} \right] \\
 &= ADbC_0 \left[ \frac{\cosh^2(bL) - \sinh^2(bL)}{\sinh(bL)} \right] \\
 &= \frac{ADbC_0}{\sinh(bL)},
 \end{aligned}$$

since  $\cosh^2 t - \sinh^2 t = 1$ . Substituting B-3 in B-1 gives

$$Db^2 [C_1 \sinh(bx) + C_2 \cosh(bx)] - \lambda [C_1 \sinh(bx) + C_2 \cosh(bx)] = 0$$

$$[C_1 \sinh(bx) + C_2 \cosh(bx)](Db^2 - \lambda) = 0$$

Since  $C_1 \sinh(bx) + C_2 \cosh(bx) \neq 0$ ,

$$b = \sqrt{\lambda/D}.$$

The value of  $b$  is now known. The linear flux as calculated in the Clive DU PA Model is therefore

$$J_0 = -\frac{AD(0 - C_0)}{L},$$

so the ratio of the linear flux to the exact flux is

$$\frac{J_0}{J} = \frac{\sinh(bL)}{bL}.$$