Radon Diffusion Through Multilayer Earthen Covers: Models and Simulations

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September 1981

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SUMMARY

Pacific Northwest Laboratory is investigating the use of multilayered earthen covers for controlling radon gas emissions from uranium mill tailings. A modeling study of radon diffusion through multiple cover layers was initiated to aid in the design of cover systems that can effectively control radon emissions to a prescribed flux limit. The design considers field conditions where compaction densities and variable moisture contents in the cover control the radon flux.

The study has been conducted using a four-phase approach. The first phase develops the solution to the steady-state radon-diffusion equation in one-dimension. The next phase develops the capability to model one-dimensional transient diffusion of radon gas and considers the effects of time-varying moisture content throughout the tailings pile. The third phase develops a multidimensional transient model that can be used to investigate the two- and three-dimensional aspects of radon diffusion. The final phase of the study used this analytical capability to investigate the radon flux and concentration profiles that develop in tailings piles and cover systems.

Modeling studies have helped us understand the complex, multidimensional, radon diffusion process that occurs in covered uranium mill tailings, and have yielded several design guidelines.

1. Radon diffusion rates and required cover thicknesses for multilayer covers can be determined successfully with the analytical or numerical solutions, or both, presented in this report. Cover thickness is not related in a simple manner to the bare tailings flux, except for single layer systems. Hence, detailed calculations are generally needed to determine a required cover thickness for a multilayer system. The computer codes RADON1, RNDI1D and RADMD can be used to conveniently solve problems of steady-state, transient, single- and multi-dimensional radon diffusion through multilayer cover systems.
2. Maximum reduction in radon emission is achieved if the top cover layer is engineered to exhibit the lowest radon diffusion of all of the cover

layers. However, in practice, factors such as requirements for plant cover, root zone depth, maintenance of layer moisture, and the need to prevent layer degradation by animal intrusion must be considered in a final cover design.

3. The variation of moisture content in the cover must be considered in order to assess variations in radon flux. Simulations of typical climatic conditions on an engineered cover produced moisture content changes that resulted in two-fold or greater changes in surface radon fluxes.
4. Multilayer systems generally require less total cover thickness than single cover layer systems to achieve the same flux reduction. This is true, provided that the multilayer system contains layer sequences having lower air-filled porosities than those found in the single cover system. The multilayer system described in this report contains a wet compacted clay/gravel layer, which is hydraulically isolated from the top cover soil by a rock layer that acts as a capillary barrier. When the clay/gravel is wet, radon diffusion is controlled for an extended period of time even under dry climate conditions. In contrast, a 35-fold increase in radon flux was calculated for a multilayer system containing a compacted clay/gravel layer that had drained from saturation to less than 40% saturation.

The preliminary multidimensional model results are very encouraging. We have successfully modeled radon diffusion for a discontinuous cover system and are able to match analytic solutions for concentration and fluxes with greater than 97% accuracy. Radon escape from the edge of a tight cover is shown to cause local, elevated radon flux levels, but the elevated levels are limited to a few meters from the edge.

CONTENTS

SUMMARY	iii
FIGURES	vii
TABLES	ix
NOMENCLATURE	xi
INTRODUCTION	1
CONCLUSIONS AND RECOMMENDATIONS	3
THEORY	5
GENERAL RADON DIFFUSION EQUATION	5
STEADY-STATE, ONE-DIMENSIONAL EQUATION AND ANALYTIC SOLUTIONS	7
TRANSIENT, ONE-DIMENSIONAL EQUATION	11
MULTIDIMENSIONAL ANALYSIS	17
APPLICATIONS	21
STEADY-STATE, ONE-DIMENSIONAL	21
TRANSIENT, ONE-DIMENSIONAL	25
TWO-DIMENSIONAL	47
REFERENCES	55

FIGURES

1	Schematic of Mill Tailings and Cover Layers	8
2	Finite Difference Spacing for Material Layers	12
3	Scaled Parameter Evaluation of "Infinite" Tailings	22
4	Influence of L_2 , $(P_2 - \theta_2)$ and Tailings Strength on Layer 2 Attenuation	22
5a	Surface Flux for a High Tailings Strength	26
5b	Surface Flux for a Medium Tailings Strength	26
5c	Surface Flux for a Low Tailings Strength	27
6a	A+ Clay/Gravel Mix Soil-Water Characteristic, Moisture Content Versus Suction Head	3D
6b	A+ Clay/Gravel Mix Soil Water Characteristic, Conductivity Versus Suction Head	31
7a	Moisture Content Distribution, Wet Year (1979 Climate Data) - 1st Year	32
7b	Moisture Content Distribution, Dry Year (1976 Climate Data) - 2nd Year	33
7c	Moisture Content Distribution, Dry Year (1976 Climate Data) - 3rd Year	33
7d	Moisture Content Distribution, Dry Year (1976 Climate Data) - 4th Year	34
7e	Moisture Content Distribution, Dry Year (1976 Climate Data) - 5th Year	34
8a	Moisture Storage for Top 100 cm, Wet Year (1979 Climate Data) - 1st Year	35
8b	Moisture Storage for Top 100 cm, Dry Year (1976 Climate Data) - 2nd Year	35
8c	Moisture Storage for Top 100 cm, Dry Year (1976 Climate Data) - 3rd Year	36
8d	Moisture Storage for Top 100 cm, Dry Year (1976 Climate Data) - 4th Year	36
8e	Moisture Storage for Top 100 cm, Dry Year (1976 Climate Data) - 5th Year	37
9	Moisture Storage in Clay Layer, Dry Year (1976 Climate Data) - 2nd, 3rd, 4th, and 5th Years	37

10a	Radon Gas Surface Flux, Wet Year (1979 Climate Data) - 1st Year	40
10b	Radon Gas Surface Flux, Dry Year (1976 Climate Data) - 2nd Year	40
10c	Radon Gas Surface Flux, Dry Year (1976 Climate Data) - 3rd Year	41
10d	Radon Gas Surface Flux, Dry Year (1976 Climate Data) - 4th Year	41
10e	Radon Gas Surface Flux, Dry Year (1976 Climate Data) - 5th Year	42
11a	A- Clay/Gravel Mix Soil Water Characteristic, Moisture Content Versus Suction Head	43
11b	A- Clay/Gravel Mix Soil Water Characteristic, Conductivity Versus Suction Head	44
12	Moisture Content Distribution, Dry Year (1976 Climate Data) - 2nd Year	45
13	Moisture Storage for Top 100 cm, Dry Year (1976 Climate Data) - 2nd Year	46
14	Radon Gas Surface Flux, Dry Year (1976 Climate Data) - 2nd Year	46
15	Analytical Versus Numerical Solution for 600-cm Thick Bare Tailings	48
16	Two-Dimensional Grid for Discontinuous Cover Simulation	50
17	Surface Flux Versus Surface Distance ($S = 0$ at midpoint of cover edge)	50
18	Concentration Profiles for Discontinuous Cover	51
19	Analytical Versus Numerical Solution Tailings (600 cm) with Cover (100 cm)	52
20	Analytical Versus Numerical Solution Bare Tailings (600 cm)	53

TABLES

1	Layer Characteristics	24
2	Surface Flux	24
3	Soil Profile Depth Nodes and Initial Conditions for Simulation of the 1979 Climate of Grand Junction, Colorado	28
4	Layer Characteristics A+ Clay/Rock Layer Case	29
5	Days Required for Plant Roots to Reach Various Depths	32
6	Water Balance for One Wet Year Followed by Four Dry Years (A+ clay/gravel layer case)	38
7	Layer Characteristics (A- clay/gravel layer case)	42
8	Water Balance for One Wet Year Followed by One Dry Year (A- clay/gravel layer case)	45
9	Layer Characteristics--Discontinuous Cover Case	47

NOMENCLATURE

A	An arbitrary coefficient
a	Leading coefficient in the diffusion coefficient relationship (typical value: 0.74)
B	An arbitrary coefficient or, the exponent in the diffusion coefficient relationship (typical value: 2.16)
C	Concentration of radon gas
c	Specific heat
D	Diffusion coefficient
E	Emanating power (typical value: 0.2)
G	Generation rate per unit volume
$\bar{J}$	Diffusive flux vector
k	Thermal conductivity
L	Thickness of a layer
M	Node number
P	Volumetric material porosity or void fraction
R	Radium content
T	Temperature
t	Time
$\bar{u}$	Velocity vector
x	Horizontal axis
z	Vertical axis (typically referenced to bottom of tailings pile)
α	Tortuosity
$\bar{\nabla}$	Del operator ($\bar{\nabla} = \hat{i} \frac{\partial}{\partial x} + \hat{j} \frac{\partial}{\partial y} + \hat{k} \frac{\partial}{\partial z}$)

- λ Decay constant
- ρ Density
- θ Volumetric moisture content

Subscripts

- a The property is for air
- BT The value corresponds to bare tailings
- e The property is an effective or bulk value
- L Refers to lower material layer
- M Node number M
- n Layer or interface number
- N The top layer or interface
- S The value corresponds to the surface
- U Refers to upper material layer

Superscripts

- $^{\circ}$ A reference value
- m Time index

INTRODUCTION

Uranium mill tailings consist of the waste or refuse left after uranium processing. The typical disposal method has been to slurry the tailings into large ponds that eventually dry out and leave a tailings pile. Since the tailings contain significant quantities of radium, they emit radon gas, which is a decay product of radium. Because exposure to radon gas has been linked to lung cancer, concern has been expressed over the health and environmental aspects of open tailings piles. In response to this concern, Pacific Northwest Laboratory (PNL)^(a) contracted with the Department of Energy's Uranium Mill Tailings Remedial Action Program (UMTRAP) to develop a capability to model and analyze the fundamental interactions that influence the diffusion of radon gas through uranium mill tailings and cover systems. The purpose of this study is to develop the theoretical basis for modeling radon diffusion and to develop an understanding of the fundamental interactions that influence radon diffusion.

This study develops the theoretical basis for modeling radon diffusion in one, two and three dimensions. The theory has been incorporated into three computer models that are used to analyze several tailings and cover configurations.

This report contains a discussion of the theoretical basis for modeling radon diffusion, a discussion of the computer models used to analyze uranium mill tailings and multilayered cover systems, and presents the results that have been obtained.

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CONCLUSIONS AND RECOMMENDATIONS

This study demonstrates that it is possible to model radon gas flux from bare tailings and covered tailings piles. Analyses can be performed in more than one dimension and can solve both the transient and steady-state problem. The effects of varying soil moisture content can also be taken into account. The models currently available--RADON1 (1D steady state), RND11D (1D transient), and RADMD (multidimensional transient or steady state)--can handle all the cover configurations studied to date. The models are also capable of simulating conditions that are much more complex. The analysis of covered uranium mill tailings using these models indicated the following:

- Radon gas flux can be attenuated more effectively by multilayer systems than by single layers of the same thickness, provided that the multilayer system contains layer sequences having lower air-filled porosity than those found in the single-cover system.
- The influence of varying soil moisture on radon gas flux has been demonstrated. Consequently, care must be taken when designing a cover system to ensure that the design will maintain a high soil moisture content over long periods of time.
- Radon flux, from covered tailings piles similar to those described in this report, will cycle by a factor of two or more in response to climatic variables that cause moisture accumulation and loss from the cover.
- An engineered moisture control barrier system such as the one considered in this report provides an effective method for maintaining a relatively high moisture content and, hence, results in effective attenuation of radon gas flux.
- The attenuation of radon flux through a given cover layer is not influenced by changes in the layers below. The attenuation will be influenced by changes in the layer or in layers lying above.
- The cover layer that is most resistive to the diffusion of radon should be placed at the top of the pile, if maximum radon attenuation is the only consideration.

- Usually, although not always, the maximum radon flux attenuation will be achieved if lower-resistance cover layers are always placed below higher-resistance cover layers. (A different ordering scheme may result in slightly greater attenuation of radon flux).

Future studies should make minor modifications to RADMD to allow the simulation of time and spatially varying moisture content in the cover materials. The moisture content values would need to be generated by a multi-dimensional, partially-saturated flow code. A likely candidate for performing the partially saturated flow analysis may be TRUST (Narasimhan and Witherspoon 1977; Key et al. 1981) since it uses a discretization and solution scheme that is similar to RADMD. Other studies may also use the convective transport capabilities of RADMD to investigate the effects that varying barometric pressure may have on the radon flux from tailings piles.

THEORY

The following four subsections discuss the theoretical background necessary to perform the analysis of radon diffusion through tailings and multilayer earthen covers. The first subsection discusses the general radon diffusion equation. The next three subsections present important aspects of steady-state and transient one-dimensional modeling of radon diffusion and multidimensional modeling of radon diffusion.

GENERAL RADON DIFFUSION EQUATION

The general equation that describes the diffusion of radon gas through a porous medium has been reported earlier by Nelson et al. (1980). Essentially, the equation is a mass balance for the radon gas and can be expressed as follows:

$$\frac{\partial}{\partial t} [(P-\theta)C] = \nabla \cdot D_e \nabla C - (P-\theta) \lambda C + R \rho_b \lambda E \quad (1)$$

where,

$(P-\theta)$ is the gas-filled pore space in the porous material,

P is the material porosity or void fraction on a volume basis,

θ is the moisture content on a volume basis,

C is the pore (gas-filled) concentration of radon (typical units: pCi-cm⁻³),

D_e is the effective or bulk diffusion coefficient (typical units: cm²-hr⁻¹),

λ is the decay constant for radon (typical units: hr⁻¹),

R is the radium (²²⁶Ra) content in the porous material (typical units: pCi-g⁻¹),

E is the emanating power of the material, which is slightly dependent upon the moisture content, particularly at very low moisture contents (see Tanner 1974).

The left term in Equation (1) accounts for the change in radon gas concentration (in the gas-filled pores) with time. The second term takes into account the change in radon concentration due to diffusion. The third term is a sink term that accounts for loss of radon by radioactive decay, and the final term is a source term that accounts for the generation of radon by radioactive decay of radium.

Equation (1) can be further generalized to include convective transport by replacing the time-derivative term (the left-most term) with the material derivative given by

$$\frac{D}{Dt}[(P-\theta)C] = \frac{\partial[(P-\theta)C]}{\partial t} + \bar{u} \cdot \bar{\nabla}[(P-\theta)C]$$

For this study, however, we have assumed that the velocity field is zero everywhere, leaving Equation (1) as the working equation.

The bulk diffusive flux vector, $\bar{J}$, can be defined based on Fick's law as follows:

$$\bar{J} = -D_e \bar{\nabla} C \quad (2)$$

A relationship describing the dependence of D_e on the air-filled pore space has been developed by Nelson et al. (1980) and is expressed by

$$D_e = a D_a^\circ (P-\theta)^B + D_w^\circ \alpha \theta \quad (3)$$

where,

a is an empirically determined coefficient (typical value: 0.740),

D_a° is the radon diffusivity in air (typical value: $360 \text{ cm}^2\text{-hr}^{-1}$),

B is an empirically determined coefficient (typical value: 2.16),
 D_w° is the radon diffusivity in water (typical value: $0.036 \text{ cm}^2\text{-hr}^{-1}$), and
 α is the tortuosity (typical value: 0.66).

Equations (1), (2), and (3) provide the framework for modeling diffusion of radon through tailings and cover layers. The following three sections will discuss simplifications that can be made to Equation (1) to allow the simulation of a variety of tailings pile configurations.

STEADY-STATE, ONE-DIMENSIONAL EQUATION AND ANALYTIC SOLUTIONS

Equation (1) can be simplified considerably if we consider only steady-state, one-dimensional diffusion of radon. These simplifications involve the elimination of the time dependent term and the assumption that the material properties ($P-\theta$) and D_e are constant throughout a given layer. The following equation results:

$$\frac{d^2C}{dz^2} - \frac{(P-\theta)\lambda C}{D_e} + \frac{R_p \lambda E}{D_e} = 0 \quad (4)$$

The z direction is taken to be the vertical distance from the base of the tailings, (see Figure 1 for a description of the notation). The analytic solution to Equation (4) for the n th layer can be written as follows:

$$C = A_n e^{W_n z} + B_n e^{-W_n z} + S_n; \quad z_{n-1} \leq z < z_n \quad (5)$$

and it follows from Equation (2), written in one-dimension, that

$$J = A_n - D_e W_n e^{W_n z} + B_n D_e W_n e^{-W_n z}; \quad z_{n-1} \leq z \leq z_n \quad (6)$$

where

FIGURE 1. Schematic of Mill Tailings and Cover Layers

$$w_n = \sqrt{\frac{(P_n - \theta_n)\lambda}{D_{e_n}}} \quad (7)$$

$$S_n = \frac{R_n \rho_{bn} E_n}{(P_n - \theta_n)} \quad (8)$$

and A_n and B_n are arbitrary coefficients dependent on the boundary conditions.

Note that the subscript, n , indicates a parameter that is associated with the n th layer or is associated with the top of the n th layer.

Equations (5) and (6) allow the problem to be solved, in conjunction with the following boundary conditions:

- flux is continuous across an interface
- concentration is continuous across an interface
- $J_0 = \text{a constant (usually 0)}$ or $C_0 = \text{a constant}$
- $C_N = \text{a constant (usually 0)}$ or $J_N = \text{a constant}$

The solution is obtained by solving the following matrix equation:

$$[K]\{V\} = \{R\} \quad (9)$$

where,

$$[K] = \begin{bmatrix} 1 & 0 & T_{11}^N & T_{12}^N & 0 & 0 \\ 0 & 1 & T_{21}^N & T_{22}^N & 0 & 0 \\ 0 & 0 & T_{31}^N & T_{32}^N & 1 & 0 \\ 0 & 0 & T_{41}^N & T_{42}^N & 0 & 1 \\ & & & & \ddots & \\ & & & & & 1 & 0 & T_{11}^n & T_{12}^n & 0 & 0 \\ & & & & & 0 & 1 & T_{21}^n & T_{22}^n & 0 & 0 \\ & & & & & 0 & 0 & T_{31}^n & T_{32}^n & 1 & 0 \\ & & & & & 0 & 0 & T_{41}^n & T_{42}^n & 0 & 1 \\ & & & & & & & & \ddots & & \\ & & & & & & & & & 1 & 0 & T_{11}^1 & T_{12}^1 & 0 & 0 \\ & & & & & & & & & 0 & 1 & T_{21}^1 & T_{22}^1 & 0 & 0 \\ & & & & & & & & & 0 & 0 & T_{31}^1 & T_{32}^1 & 1 & 0 \\ & & & & & & & & & 0 & 0 & T_{41}^1 & T_{42}^1 & 0 & 1 \end{bmatrix}$$

$$\{V\} = \begin{Bmatrix} C_N \\ J_N \\ A_N \\ B_N \\ C_{N-1} \\ J_{N-1} \\ \vdots \\ C_n \\ J_n \\ A_n \\ B_n \\ C_{n-1} \\ J_{n-1} \\ \vdots \\ C_1 \\ J_1 \\ A_1 \\ B_1 \\ C_0 \\ J_0 \end{Bmatrix}$$

$$\{R\} = \begin{Bmatrix} S_N \\ 0 \\ S_N \\ 0 \\ \vdots \\ S_n \\ 0 \\ S_n \\ 0 \\ \vdots \\ S_1 \\ 0 \\ S_1 \\ 0 \end{Bmatrix}$$

and

$$\begin{aligned}
 T_{11}^n &= -e^{W_n z_n} & T_{12}^n &= -e^{-W_n z_n} \\
 T_{21}^n &= D_{e_n} W_n e^{W_n z_n} & T_{22}^n &= -D_{e_n} W_n e^{-W_n z_n} \\
 T_{31}^n &= -e^{W_n z_{n-1}} & T_{32}^n &= -e^{-W_n z_{n-1}} \\
 T_{41}^n &= D_{e_n} W_n e^{W_n z_{n-1}} & T_{42}^n &= -D_{e_n} W_n e^{-W_n z_{n-1}}
 \end{aligned} \tag{10}$$

After substituting the bottom and surface boundary conditions, matrix Equation 9 becomes a system of $4N$ simultaneous equations. These equations can be solved by essentially two methods: 1) by algebraic substitution, or 2) by computer. A direct solution by algebraic substitution becomes tedious for more than two layers. Therefore, a computer program called RADON1 has been developed to solve the multilayer, steady-state, radon-diffusion equation in one dimension.

Certain analytic solutions to Equation (4) can be obtained. In the studies of uranium mill tailings, it is often of interest to determine the surface flux of radon from a bare tailings pile (i.e., a pile having no cover). This flux, J_{BT} , can be obtained analytically if it is assumed that the radon concentration at the surface of the tailings is zero ($C_N = 0$), and that the flux into the bottom of the tailings is zero ($J_0 = 0$). The following equation results:

$$J_{BT} = S_1 D_{e1} W_1 \tanh (W_1 L_1) \tag{11}$$

Using the same boundary conditions, it is also possible to obtain the following relationship for the concentration profile in the bare tailings:

$$C = S_1 \left[1 - \frac{\cosh W_1 z}{\cosh [W_1 (z_1 - z)]} \right]; \quad z_0 \leq z \leq z_1 \tag{12}$$

Another parameter of interest is the surface flux from a two-layer system (one tailings layer with one cover layer). If the previous boundary conditions

are assumed, and there is no radium in the upper layer, it can be readily shown that the surface flux, J_2 , is given by

$$\frac{J_2}{J_{BT}} = \frac{1}{\cosh[W_2(z_2 - z_1)] \left\{ \frac{D_{e1}W_1}{D_{e2}W_2} \tanh[W_1(z_1 - z_0)] \tanh[W_2(z_2 - z_1)] + 1 \right\}} \quad (13)$$

with J_{BT} computed by Equation (11). It can also be shown that the radon gas flux attenuation through the top cover of a multilayer system is given by

$$\frac{J_N}{J_{N-1}} = \frac{1}{\cosh[W_N(z_N - z_{N-1})]} \quad (14)$$

as long as the top layer does not contain radium ($R_N = 0$), and as long as the same boundary conditions used to develop Equations (11), (12) and (13) are assumed. The four analytic solutions (Equations 11, 12, 13 and 14) provide a starting point for the analysis of multilayered cover systems. They also can be used as valuable checks that the numerical solutions are correct. These analytical solutions have been compared with those of Roger et al. 1981. The calculated exit fluxes of radon from each layer of a six-layer test case were compared using both codes, and the results were found to differ by less than 1.4%.

TRANSIENT, ONE-DIMENSIONAL EQUATION

The diffusion boundary value problem, for which Equations (1) and (2) form the basic relationships, is solved numerically using the PNL-developed code, RNDI1D, i.e., RadoN Diffusion in 1 Dimension. The solution is obtained by using finite differences with a fully implicit time difference scheme.

The mathematical model consists of N layers of (possibly) distinct materials. Let n denote a layer index, $n = 1, 2, \dots, N$. The concentration will be computed at M_n uniformly spaced points or nodes within layer n . The first point is at the bottom boundary of layer n , and the M_{n-1} th point is located at

top node of layer n is located at the top boundary, and the first node of layer $n + 1$ is located at the same point, i.e., at the bottom boundary of layer $n + 1$ (Figure 2).

Material layer n is partitioned into M_n segments. Of these segments, $M_n - 2$ have equal lengths. The first and last segments are each only half as long as the others. To avoid notational difficulties, we drop the material index, n , unless it is essential for clarity. The values of C_i are defined at the top and bottom of the layer and at the center of each of the other segments. Thus, the C_i are uniformly spaced within the layer.

The diffusion coefficient, D_i , is defined at the lower boundary of the i^{th} cell. All other quantities are defined at the same points where the C_i 's are defined. We first consider only internal nodes.

The flux through the i^{th} cell lower face is

$$J_i = - D_i \left. \frac{\partial C}{\partial z} \right|_{z = z_i} \simeq D_i (C_{i-1} - C_i) / \Delta z; \quad i = 1, 2, 3, \dots, M \quad (15)$$

FIGURE 2. Finite Difference Spacing for Material Layers

The flux through the i^{th} cell upper face is

$$J_{i+1} = - D_{i+1} \left. \frac{\partial C}{\partial z} \right|_{z = z_{i+1}} \simeq D_{i+1} (C_i - C_{i+1}) / \Delta z; \quad i = 1, 2, \dots, M-1 \quad (16)$$

Since radon is the product of radioactive decay and is itself radioactive, the change in C due to decay is

$$\Delta z R \rho_b \lambda E - \Delta z (P - \theta) \lambda C \quad (17)$$

where R is the radium content in the material, ρ_b is the material bulk density, λ is the radon decay constant, E is the material emanating power, P is the material porosity, and θ is the volumetric moisture content. The total change in C in the cell with index i during the time period Δt is

$$(P - \theta) \Delta z \frac{\Delta C}{\Delta t} \simeq (P - \theta) \Delta z \frac{C_i^{m+1} - C_i^m}{\Delta t} \quad (18)$$

where the superscript m is the time index. The conservation of mass for cell i is obtained by combining Equations (15) through (18), dividing through by Δz and using a backward difference for the time derivative to yield

$$\begin{aligned} (P - \theta)_i^{m+1} \frac{C_i^{m+1} - C_i^m}{\Delta t} &= \frac{D_{i+1}^{m+1}}{\Delta z^2} (C_{i+1}^{m+1} - C_i^{m+1}) - \frac{D_i^{m+1}}{\Delta z^2} (C_i^{m+1} - C_{i-1}^{m+1}) \\ &\quad - (P - \theta)_i^{m+1} \lambda C_i^{m+1} + R \rho_b \lambda E_i^{m+1}. \end{aligned} \quad (19)$$

after defining the following variables:

$$A_i^{m+1} = - \frac{D_i^{m+1}}{(P - \theta)_i^{m+1}} \frac{\Delta t}{\Delta z^2} \quad (20)$$

$$C_i^{m+1} = - \frac{D_{i+1}^{m+1}}{(P-\theta)_i^{m+1}} \frac{\Delta t}{\Delta z^2} \quad (21)$$

$$B_i^{m+1} = 1 + \lambda \Delta t - A_i^{m+1} - C_i^{m+1} \quad (22)$$

$$D_i^{m+1} = C_i^m + \frac{\Delta t}{(P-\theta)_i^{m+1}} R_i \rho_{bi} \lambda E_i^{m+1} \quad (23)$$

Equation (19) can be rewritten as

$$A_i^{m+1} C_{i-1}^{m+1} + B_i^{m+1} C_i^{m+1} + C_i^{m+1} C_{i+1}^{m-1} = D_i^{m+1} \quad (24)$$

for $i = 2, 3, \dots, M-1$.

For the interface node between two adjacent material layers, we require that the flux be continuous at the interface. By introducing the following notation, in which L and U denote the lower and upper material layers, respectively, and i denotes the node index at the interface, we have

$$- D_L \frac{C_i^{m+1} - C_{i-1}^{m+1}}{\Delta z_L} = - D_U \frac{C_{i+1}^{m+1} - C_i^{m+1}}{\Delta z_U} \quad (25)$$

By defining

$$A_i^{m+1} = D_L^{m+1} / \Delta z_L \quad (26)$$

$$C_i^{m+1} = D_U^{m+1} / \Delta z_U \quad (27)$$

$$B_i^{m+1} = - A_i^{m+1} - C_i^{m+1} \quad (28)$$

$$D_i^{m+1} = 0 \quad (29)$$

then Equation (25) may be expressed in the form of Equation (24).

We now examine the boundary conditions. Two types are permitted at the bottom of material 1 and at the top of material N. The concentration, C , may be specified or the flux, J , may be specified at the boundary.

Consider first the bottom boundary node $i = 1$. If C_1 is specified, then no equation is needed for $i = 1$ and, for $i = 2$, we have the following modifications to (19) through (23):

$$A_2^{m+1} = 0 \quad (30)$$

$$C_2^{m+1} = - \frac{D_3^{m+1}}{(P-\theta)_2^{m+1}} \frac{\Delta t}{\Delta z^2} \quad (31)$$

$$B_2^{m+1} = 1 + \lambda \Delta t + \frac{D_2^{m+1}}{(P-\theta)_2^{m+1}} \frac{\Delta t}{\Delta z^2} - C_2^{m+1} \quad (32)$$

$$D_2^{m+1} = C_2^m + \frac{\Delta t}{(P-\theta)_2^{m+1}} R_2 \rho b_2 \lambda E_2^{m+1} + \frac{D_2^{m+1}}{(P-\theta)_2^{m+1}} \frac{\Delta t}{\Delta z^2} C_1^{m+1} \quad (33)$$

If the flux, J^{m+1} , is specified at the first node, $i = 1$, then, using a forward difference in z results in

$$J_1^{m+1} = - D_1^{m+1} \frac{C_2^{m+1} - C_1^{m+1}}{\Delta z} \quad (34)$$

This equation can be rewritten in the form of Equation (24) by letting

$$A_1^{m+1} = 0 \quad (35)$$

$$C_1^{m+1} = - D_1^{m+1} / \Delta z \quad (36)$$

$$B_1^{m+1} = - C_1^{m+1} \quad (37)$$

$$D_1^{m+1} = J_1^{m+1} \quad (38)$$

Consider the node at the earth's surface, $i = M_N$. If C_i is specified, then no equation is needed, but the equation for $i = M_N - 1$ is modified as follows:

$$A_i^{m+1} = - \frac{D_i^{m+1}}{(P-\theta)_i^{m+1}} \frac{\Delta t}{\Delta z^2} \quad (39)$$

$$C_i^{m+1} = 0 \quad (40)$$

$$B_i^{m+1} = 1 + \lambda \Delta t - A_i^{m+1} + \frac{D_{i+1}^{m+1}}{(P-\theta)_i^{m+1}} \frac{\Delta t}{\Delta z^2} \quad (41)$$

$$D_i^{m+1} = C_i^m + \frac{\Delta t}{(P-\theta)_i^{m+1}} R_i \rho_{bi} \lambda E_i^{m+1} + \frac{D_{i+1}^{m+1}}{(P-\theta)_i^{m+1}} \frac{\Delta t}{\Delta z^2} C_{i+1}^{m+1} \quad (42)$$

If the flux is specified at the surface then, using a backwards difference in z , we have for $i = M_N$

$$J_i^{m+1} = - D_i^{m+1} \frac{C_i^{m+1} - C_{i-1}^{m+1}}{\Delta z} \quad (43)$$

which can be written like Equation (24) by letting

$$A_i^{m+1} = D_i^{m+1} / \Delta z \quad (44)$$

$$C_i^{m+1} = 0 \quad (45)$$

$$B_i^{m+1} = -A_i^{m+1} \quad (46)$$

$$D_i^{m+1} = J_i^{m+1} \quad (47)$$

Using the above equations, we find that the entire system of equations written over all material layers can be expressed as

$$AC = D \quad (48)$$

where A is a tridiagonal matrix whose subdiagonal entries are the A_i^{m+1} , whose main diagonal entries are the B_i^{m+1} , and whose superdiagonal entries are the C_i^{m+1} . Vector D has the D_i^{m+1} for its components. The solution vector, C, has C_i^{m+1} as its components. Equation (48) is then solved by the Thomas algorithm (Ames 1977).

The accuracy of the numerical solution could be increased somewhat by using higher order difference approximations for the flux conditions at layer boundaries. This will entail a more complicated matrix, A, in Equation (48). For example, if a three-point difference approximation were used in the flux equation, the matrix, A, would be pentadiagonal (i.e., matrix A would have five diagonals instead of three). Algorithms exist for solving such systems (e.g., von Rosenberg 1969).

MULTIDIMENSIONAL ANALYSIS

To develop a multidimensional analysis capability for radon diffusion, we have used the mathematical similarity between heat conduction and gaseous

diffusion processes. The equations describing each process are mathematically identical except for the decay term that arises in the radon diffusion equation. The general radon diffusion equation (excluding convection) is given by Equation (1)

$$\frac{\partial}{\partial t} [(P-\theta)C] = \bar{\nabla} \cdot D_e \bar{\nabla} C - (P-\theta) \lambda C + R\rho_b \lambda E$$

In comparison, the general heat conduction equation (excluding convection) is given by

$$\rho c \frac{\partial T}{\partial t} = \bar{\nabla} \cdot k \bar{\nabla} T + G \quad (49)$$

where

- ρ is mass density,
- c is specific heat,
- T is temperature in absolute units,
- t is time,
- k is thermal conductivity, and
- G is heat generation per unit volume.

Now, if it is assumed that the gas-filled porosity $(P-\theta)$ is constant or nearly constant (at least during a given time step), Equation (1) can be rewritten as:

$$(P-\theta) \frac{\partial C}{\partial t} = \bar{\nabla} \cdot D_e \bar{\nabla} C - (P-\theta) \lambda C + R\rho_b \lambda E \quad (50)$$

which is identical in form to Equation (49) with the addition of the decay term, $-(P-\theta)\lambda C$. The source term, $R\rho_b \lambda E$, is analogous to the heat generation rate, G .

The multidimensional modeling capability is based on the solution scheme employed in the TRUMP computer code (Edwards 1972), which solves Equation (49)

in one, two, or three dimensions and in cartesian, cylindrical, or spherical coordinates. The new code, RADMD (for Radioactive Diffusion in MultiDimensions), uses the solution scheme employed by TRUMP modified to handle the additional decay term. The numerical formulation is based on an integrated, finite-difference representation of Equation (49). The code can, therefore, handle irregular grids and is not limited to the more traditional rectangular, finite-difference grids. Furthermore, it will be a simple matter to include convective transport if the need arises.

APPLICATIONS

STEADY-STATE, ONE-DIMENSIONAL

One parameter that is of particular interest in the study of uranium mill tailings is the flux of the bare tailings (i.e., tailings having no cover layers) expressed as J_{BT} . This value can be computed directly using Equation (11). A plot of J_{BT} divided by a measure of the tailings source strength versus the tailings thickness is shown in Figure 3. This plot demonstrates that, even for very porous material, tailings can be treated as if they were infinitely deep if their depth is greater than 4 meters.

The attenuation of the radon flux through the top cover layer for any system that consists of one or more cover layers is given by Equation (14). An interesting feature of Equation (14) is that the ratio of output and input fluxes for the top cover layer is not affected by changes in layers below. That is, the attenuation through layer N is a function of layer-N parameters (and the surface concentration boundary condition) only.

Based on an extension of this observation, a hypothesis is proposed regarding the attenuation through any individual cover layer. The hypothesis can be stated as follows:

- The attenuation through the n th layer will not be influenced by changes in the characteristics of lower layers (i.e., layers 1 through $n-1$), but will be influenced by changes in the n th layer and layers lying above (i.e., layers n through N). (This statement assumes that the n th and upper layers do not contain a radon source and that the surface concentration is fixed.)

We have not proven this hypothesis, but numerical experiments performed using RADON1 demonstrate that the hypothesis is correct. Figure 4 is a plot of the attenuation through the clay layer in a four-layer problem (i.e., three cover layers and one tailings layer). The bare tailings flux, J_{BT} , curves on Figure 4 indicate the design characteristics required for the clay layer so that a $2 \text{ pCi-m}^{-2}\text{-s}^{-1}$ surface flux will be achieved for the given bare tailings flux.

FIGURE 3. Scaled Parameter Evaluation of "Infinite" Tailings

FIGURE 4. Influence of L_2 , $(P_2 - \theta_2)$ and Tailings Strength on Layer 2 Attenuation

Note that the shape of the $P-\theta$ curves is the same regardless of the strength of the bare tailings, which indicates that the attenuation through the clay layer is not affected by a change in the lower layer.

To explain why the hypothesis holds true, consider the diffusion of radon through a multilayered cover. If the top layer is made more resistive to the diffusion of radon (e.g., if it is made thicker, given a higher moisture content, or a lower porosity), the diffusion of radon will be slowed down in lower layers. The slowdown occurs because the concentration gradient in lower layers is reduced as a result of the concentration buildup under the more resistive layer. Hence, more radon will decay in a given layer, and the attenuation through each layer will be increased. In a similar manner, if the top layer is made less resistive, the attenuation through all layers will be decreased.

From the previous analysis it is proposed that the layer most resistive to the diffusion of radon should be placed at the top of the pile, if maximum radon attenuation is the only consideration. An obvious generalization of this statement is that the multilayered cover should be designed such that the resistance of a given cover layer is always greater than the resistance of each cover layer below. While this is normally true, there are certain combinations of layer characteristics, order, and thicknesses for which the surface flux varies slightly from the rule.

To demonstrate these ideas, a numerical experiment has been performed in which all the parameters--except the order of the three cover layers--are fixed. Table 1 shows the layer characteristics. Since the moisture content in the three cover layers was fixed at zero, the only difference between Covers A, B and C is their porosity. Hence, the resistance of Cover A is the least, the resistance of Cover C is the highest, and the resistance of Cover B is between A and C. The resulting surface flux obtained for all possible cover configurations is presented in Table 2. These results demonstrate that, to achieve the maximum reduction in the surface flux, it is desirable to have the layer with the highest resistance at the top. The more general statement that the resistance of a given cover layer should be greater than the resistance of each cover

TABLE 1. Layer Characteristics

Layer	Thickness (m)	P-θ (m ³ -m ⁻³)	R (pCi-g ⁻¹)	ρ _b (g-m ⁻³)
A	2.0	0.459	0	1.46 E6
B	2.0	0.370	0	1.70 E6
C	2.0	0.259	0.	2.00 E6
Tailings	6.18	0.354	1600	1.23 E6

TABLE 2. Surface Flux

Case	Cover Configuration (Top → Bottom)	Surface Flux (pCi-m ⁻² -s ⁻¹)
1	C/A/B	1.637
2	C/B/A	1.697
3	B/C/A	2.058
4	B/A/C	2.108
5	A/C/B	2.460
6	A/B/C	2.617

layer below is generally upheld by the results in Table 2. A small departure from the rule is seen in the reversal of cases 1 and 2. The rule would have predicted that Case 2 would yield the lowest surface flux, while Case 2 actually yields only a slightly lower (3.5%) surface flux.

Design Considerations

The concepts presented thus far provide guidance for doing initial designs of multilayered uranium mill tailings covers. The designer should keep in mind the following guidelines:

- The radon attenuation through the nth layer is not influenced by changes in the characteristics of lower layers, but is influenced by changes in the nth or upper layers.
- The most resistive layer should be the top layer.

- Usually, it is desirable to place a lower-resistance cover layer below a higher-resistance cover layer.

The order of a given set of cover layers can be defined, based on these considerations, so that the surface radon flux is minimized. This design will have to be modified with consideration given to the

- requirement for plant cover
- root zone depth
- feasibility of construction
- maintenance of moisture in a layer
- prevention of layer degradation (e.g., by burrowing animals).

At this point in the design process, the multilayer, one-dimensional, steady-state diffusion equations will need to be solved to further refine the cover design. The analysis must be based on knowledge of the diffusion properties of the cover, tailings source strength, and cover thicknesses and cannot be approximated from measurements of bare tailings flux.

Figures 5a through 5c are plots of the computed surface flux for high, medium and low tailings strengths that result for a typical cover design. The plots have been normalized by dividing the surface flux by the bare tailings flux. The lower end point of the $P-\theta$ curves corresponds to the design characteristics of the second layer, which are necessary to achieve a $2 \text{ pCi-m}^{-2}\text{-s}^{-1}$ surface flux. Curves such as Figure 5 can be easily and rapidly generated for any proposed cover system using RADON1. Once these plots are available, it is a simple matter to select the air-filled porosity, $P-\theta$, and the layer thickness that will meet the surface flux criterion of $2 \text{ pCi-m}^{-2}\text{-s}^{-1}$.

TRANSIENT, ONE-DIMENSIONAL

Transient analysis of a uranium mill tailings pile becomes important when consideration is given to the effects of changing moisture content. Since the diffusion coefficient, D_e , is highly dependent on the gas-filled pore volume ($P-\theta$), changes in the moisture content can significantly affect the radon flux. Therefore, a realistic simulation of a tailings pile should account for all of the factors influencing the moisture content, θ .

FIGURE 5a. Surface Flux for a High Tailings Strength

FIGURE 5b. Surface Flux for a Medium Tailings Strength

FIGURE 5c. Surface Flux for a Low Tailings Strength

For the following simulations, the partially saturated flow code, UNSATV, developed by Simmons and Gee (1981) has been used. UNSATV is a version of an earlier code, UNSAT, developed by Gupta et al. (1978). The capability to model the effects of water vapor diffusion has been incorporated into UNSATV. The code solves the partially saturated flow equation in one dimension. The important factors considered include water infiltration from precipitation or irrigation, drainage, evaporation, and plant evapotranspiration. For a more detailed description of the code and applications of the code, the reader is referred to the two reference reports.

The test case simulated during this phase of the research is representative of the tailings cover test plots constructed at Grand Junction, Colorado. Table 3 indicates the initial conditions used for the partially saturated flow analysis. The initial conditions shown in Table 3 represent near-equilibrium conditions below the clay/gravel layer to the 763-cm-deep water table. The moisture conditions above the clay/gravel layer are the result of repeated simulations using the recorded climatic conditions and could represent the

TABLE 3. Soil Profile Depth Nodes and Initial Conditions for Simulation of the 1979 Climate of Grand Junction, Colorado

Soil Layer	Depth (cm)	Suction Head (cm)	Water Content
Grand Junction Clay	0.0	1450.0	0.170
	2.5	3167.0	0.172
	5.0	1293.0	0.175
	7.5	1230.0	0.177
	10.0	1176.0	0.179
	15.0	1090.0	0.183
	20.0	1025.0	0.187
	25.0	975.0	0.190
	30.0	935.0	0.192
	35.0	902.0	0.195
	40.0	874.0	0.197
	50.0	830.0	0.200
	60.0	796.0	0.203
	70.0	768.0	0.206
	80.0	746.0	0.208
	99.0	709.0	0.212
Gravel	100.0	709.0	0.008
	110.0	709.0	0.008
	120.0	709.0	0.008
	129.0	709.0	0.008
A+ Clay/ Gravel Mix	130.0	600.0	0.259
	132.0	600.0	0.259
	135.0	600.0	0.259
	140.0	600.0	0.259
	142.0	600.0	0.259
	144.0	600.0	0.259
Medium Tailings	145.0	651.0	0.141
	150.0	646.0	0.141
	160.0	629.0	0.142
	180.0	612.0	0.144
	200.0	590.0	0.145
	250.0	524.0	0.150
	300.0	467.0	0.155
	400.0	365.0	0.167
	500.0	265.0	0.185
	600.0	165.0	0.214
	650.0	155.0	0.238
	700.0	65.0	0.278
	750.0	15.0	0.370
	755.0	10.0	0.384
Water Table	760.0	5.0	0.399
	763.0	2.0	0.408
TOTAL INITIAL MOISTURE STORAGE 145.859 cm			

initial conditions from a long-term climatic history. The clay/gravel layer moisture content is initially at a nearly saturated state. Table 4 summarizes the layer characteristics used for the radon diffusion simulation. The soil-water characteristics reported by Simmons and Gee (1981) for the Grand Junction clay, rock, and tailings layers have been used for the partially saturated flow simulation. The soil-water characteristics for the A+ clay/gravel mix are represented by the curves shown in Figures 6a and 6b.

TABLE 4. Layer Characteristics for A+ Clay/Rock Layer Case

Layer	Thickness (cm)	Material	Porosity ($\text{cm}^3\text{-cm}^{-3}$)	Density (g-cm^{-3})	Radium Content (pCi-g^{-1})
4	100	Overburden (Grand Junction clay)	0.468	1.37	0
3	30	Clean rock	0.320	1.80	0
2	15	Clay/gravel mix (A+)	0.270	1.97	0
1	618	Medium tailings	0.458	1.47	1600

The moisture simulation assumes a plant cover crop consisting of cheatgrass. During the active growing season, the cheatgrass roots can extract moisture from the covers. Table 5 lists the time required for the roots to reach a given depth during the growing season. Plant growth began on day 120. The root zone moisture extraction has been handled in the same manner described by Simmons and Gee (1981).

Climatic parameters were obtained from monthly summaries of local climatological data for Grand Junction, Colorado, published by the U.S. Department of Commerce, National Climatic Center, Asheville, NC. The data from the summaries for the years 1976 and 1979 were provided in the following units: hourly precipitation (inches); temperature ($^{\circ}\text{F}$) in terms of daily maximum, minimum, average, and average dew point temperature; average wind speed (m.p.h.); percent of possible sunshine and sky cover in tenths; and maximum and minimum relative humidity. Hourly precipitation determined the water infiltration rate for the modeling. Other climatic parameters were used to estimate potential evapotranspiration on a daily basis.

FIGURE 6a. A+ Clay/Gravel Mix Soil-Water Characteristic, Moisture Content Versus Suction Head

Starting with the initial conditions shown in Table 3, UNSATV was run for one year using the 1979-climate data (wettest year on record). This simulation was then followed by four years using repeated 1976-climatic data (driest year on record). These simulations, therefore, are representative of a high rainfall year followed by a protracted dry spell.

Figures 7a through 7e are plots of the moisture content distributions for selected days from each of the five years simulated. The important feature to

FIGURE 6b. A+ Clay/Gravel Mix Soil Water Characteristic, Conductivity Versus Suction Head

note is that, although the moisture content in the surface layer varies dramatically, the lower layers exhibit very little change in moisture content. To better illustrate this, the moisture storage in the top layer and in the clay/gravel layer has been computed and plotted. Figures 8a through 8e are plots of the total moisture storage in the top cover layer, and Figure 9 is a plot of the moisture in the clay/gravel layer during the four dry years. The moisture storage in the top layer illustrates the seasonal cycle of moisture gain

TABLE 5. Days Required for Plant Roots to Reach Various Depths

Depth (cm)	Days	Depth (cm)	Days
0.0	5	30	20
2.5	6	35	25
5.0	7	40	35
7.5	8	50	40
10	10	60	50
15	11	70	60
20	15	80	70
25	16

FIGURE 7a. Moisture Content Distribution, Wet Year (1979 Climate Data) - 1st Year

FIGURE 7b. Moisture Content Distribution, Dry Year (1976 Climate Data) - 2nd Year

FIGURE 7c. Moisture Content Distribution, Dry Year (1976 Climate Data) - 3rd Year

VOLUMETRIC MOISTURE CONTENT
 FIGURE 7d. Moisture Content Distribution, Dry
 Year (1976 Climate Data) - 4th Year

VOLUMETRIC MOISTURE CONTENT
 FIGURE 7e. Moisture Content Distribution, Dry
 Year (1976 Climate Data) - 5th Year

FIGURE 8a. Moisture Storage for Top 100 cm,
Wet Year (1979 Climate Data) - 1st Year

FIGURE 8b. Moisture Storage for Top 100 cm,
Dry Year (1976 Climate Data) - 2nd Year

FIGURE 8c. Moisture Storage for Top 100 cm,
Dry Year (1976 Climate Data) - 3rd Year

FIGURE 8d. Moisture Storage for Top 100 cm,
Dry Year (1976 Climate Data) - 4th Year

FIGURE 8e. Moisture Storage for Top 100 cm,
Dry Year (1976 Climate Data) - 5th Year

FIGURE 9. Moisture Storage in Clay Layer,
Dry Year (1976 Climate Data) -
2nd, 3rd, 4th, and 5th Years

and loss as well as the short spikes of increased moisture due to individual rainfall events. When comparing Figures 8a through 8e, it is apparent that the top layer is drying out.

Figures 8d and 8e indicate that, during the last two dry years, the top cover approaches a steady-state value. The amount of drying that has occurred during the last two years is approaching zero. This trend is also indicated by the water balance summary for each of the years simulated (see Table 6). The initial storage values represent the amount of water stored in all four layers on day one for each of the five years. The infiltration value is that amount of water that soaked into the ground from precipitation. The amount of moisture transpired by plants is given as a transpiration value, and the total amount of water lost by evaporation is also given. The amount of water that drained out of the soil is also given, but there was no drainage during the simulation. The final storage values indicate the amount of moisture stored in all four layers on the last day of each year. The mass balance error is the difference that occurs when the change in water storage is computed as the difference in initial and final water storage or as the sum of the infiltration, transpiration, evaporation, and drainage (values must be added or subtracted as appropriate).

TABLE 6. Water Balance for One Wet Year Followed by Four Dry Years (A+ Clay/Gravel Layer Case)

Water (cm)	Year				
	1	2	3	4	5
Initial Storage	145.9	143.0	138.2	137.3	136.9
Infiltration	22.6	13.4	13.4	13.4	13.4
Transpiration	11.6	6.4	4.4	3.3	3.0
Evaporation	13.6	9.6	9.0	9.5	9.7
Drainage	0.0	0.0	0.0	0.0	0.0
Final Storage	143.0	138.2	137.3	136.9	136.7
Mass Balance Error	0.3	2.2	0.9	1.0	0.9

Figure 9 demonstrates that the clay/gravel layer is reasonably isolated from the effects of the varying climatic conditions. A nearly negligible amount of drying can be noted, and it is apparent that short-period rainfall events and seasonal moisture gain and loss do not cause an observable effect on the moisture storage in the clay/gravel layer. For this reason, a cover system like this is considered an engineered moisture-control system. The top cover layer provides a place for plants to grow and animals to burrow. The next layer (clean rock) forms a capillary barrier that serves to isolate the lower layers from the varying moisture content in the top layer. The capillary barrier helps to maintain a relatively high moisture content in the clay/gravel layer (refer to Figure 7a through 7e). The high moisture content in the clay/gravel layer results in a low value for the diffusion coefficient in that layer and, hence, can effectively control the emission of radon gas from the tailings pile.

To illustrate the effect of moisture content on radon diffusion, the transient radon diffusion model, RND11D, has been run using the time varying moisture contents predicted by the partially saturated flow analysis and the layer characteristics listed in Table 4. The diffusion coefficient for each layer was computed by Equation (3). The resulting surface fluxes are plotted in Figures 10a through 10e.

The radon diffusion simulations were started with a zero radon gas concentration throughout the tailings and covers. Hence, a period of time must pass before the surface flux becomes established. This is represented by the low surface fluxes during the first month of the simulation. As time progresses, however, the surface fluxes become larger and reach their highest value during the last year of the simulation. This is an expected result, since the gradual drying out of the cover system results in higher diffusion rates. The effect of rainfall events is also demonstrated by the downward spikes in the surface flux. These spikes coincide with the upward spikes on the moisture storage figures (Figures 8a through 8b) that are due to precipitation.

Another simulation of a nearly identical cover system has been performed. The only difference between this and the previous simulation is that a different clay/gravel mix (A- clay/gravel mix) was used for the clay/gravel layer. The

FIGURE 10a. Radon Gas Surface Flux, Wet Year (1979 Climate Data) - 1st Year

FIGURE 10b. Radon Gas Surface Flux, Dry Year (1976 Climate Data) - 2nd Year

FIGURE 10c. Radon Gas Surface Flux, Dry Year (1976 Climate Data) - 3rd Year

FIGURE 10d. Radon Gas Surface Flux, Dry Year (1976 Climate Date) - 4th Year

FIGURE 10e. Radon Gas Surface Flux, Dry Year (1976 Climate Data) - 5th Year

soil characteristics for the A- clay/gravel mix are represented by the curves shown in Figures 11a and 11b. When comparing Figure 6a with Figure 11a, it is apparent that the A- clay/gravel layer will be drier at a given suction head. It is, therefore, expected that the radon gas attenuation using the A- clay/gravel mix will not be as good. The layer characteristics are listed in Table 7 and are identical to the previous simulation with the exception of the clay/gravel layer.

TABLE 7. Layer Characteristics (A- clay/gravel layer case)

Layer	Thickness (cm)	Material	Porosity (cm ³ -cm ⁻³)	Density (g-cm ⁻³)	Radium Content (pCi/g ⁻¹)
4	100	Overburden (Grand Junction clay)	0.468	1.37	0
3	30	Clean Rock	0.32	1.80	0
2	15	Clay/gravel (A-)	0.241	2.05	0
1	618	Medium tailings	0.458	1.47	1600

FIGURE 11a. A- Clay/Gravel Mix Soil Water Characteristic, Moisture Content Versus Suction Head

The moisture content profile at the end of the first year from the previous simulation was used as an initial condition. Only one dry year was modeled for this simulation. Figure 12 is a plot of the resulting moisture content profiles for selected days during the simulation. Comparison of these profiles with Figure 7b indicates that the only difference is that the clay/gravel layer is significantly drier: it is only 40% as wet as the previous clay/gravel layer. The water balance for this simulation is listed in Table 8. A

FIGURE 11b. A- Clay/Gravel Mix Soil Water Characteristic, Conductivity Versus Suction Head

comparison of Table 8 with the first two years listed in Table 6 demonstrates that the A- clay/gravel layer is unable to maintain as high a moisture storage as the A+ clay/gravel layer. The final storage value for the second year is 2.3 cm of water less than the A+ clay/gravel layer case.

Figure 13 is the resulting moisture storage in the top layer. Not surprisingly, the moisture storage in the top layer is identical to the moisture storage in the previous simulation (Figure 13 and Figure 8b).

FIGURE 12. Moisture Content Distribution,
Dry Year (1976 Climate Data) - 2nd Year

TABLE 8. Water Balance for One Wet Year Followed by
One Dry Year (A- clay/gravel layer case)

Water (cm)	Year	
	1	2
Initial Storage	145.9	140.0
Infiltration	22.6	13.4
Transpiration	11.6	6.4
Evaporation	13.6	9.6
Drainage	0.0	0.0
Final Storage	143.0	135.9
Mass Balance Error	0.3	1.5

The surface flux (Figure 14) shows a dramatic increase over the previous results (Figure 10b). The surface flux with the 60% drier clay/gravel layer is roughly 35 times larger. This finding demonstrates the importance of maintaining a relatively high moisture content in the clay/gravel layer. A significant improvement in the control of radon emissions can be obtained if a

FIGURE 13. Moisture Storage for Top 100 cm,
Dry Year (1976 Climate Data) - 2nd Year

FIGURE 14. Radon Gas Surface Flux,
Dry Year (1976 Climate Data) - 2nd Year

properly engineered cover system is used. Consideration should be given to maintaining relatively high moisture content in the cover system.

TWO-DIMENSIONAL

Two-dimensional (and three-dimensional) analysis of radon gas diffusion becomes important whenever a spatially nonuniform geometry or boundary condition exists. A nonuniform condition will exist whenever there are spatial variations in thickness, porosity, moisture content, or radium content. Non-uniform boundary conditions arise near the edge of the tailings pile, near the apparatus used to measure flux and concentrations, and whenever the specified flux or concentration varies spatially. For these reasons, the capability to model multidimensional effects is very important.

The multidimensional radioactive diffusion code, RADMD, is capable of simulating the multidimensional effects of spatial variations for both steady state and transient problems. As a first step in verifying the code, a simple one-dimensional problem has been simulated and compared with the analytic solution. The problem solved deals with bare tailings having the layer characteristics shown in Table 9 (for Layer 1). The diffusion coefficient of the layer was selected to be $10^{-4} \text{ cm}^2 \text{ s}^{-1}$. A concentration of zero is imposed at the surface and the flux into the bottom of the tailings is assumed to be zero. The finite difference grid used to simulate this problem consists of a column of 48 cells that are 12.5 cm thick. The nodes are cell centered with an additional node placed at the surface of the tailings.

TABLE 9. Layer Characteristics for Discontinuous Cover Case

Layer	Thickness (cm)	Material	Total Porosity ($\text{cm}^3\text{-cm}^{-3}$)	Bulk Density (g-cm^{-3})	Moisture Content ($\text{cm}^3\text{-cm}^{-3}$)	Radium Content (pCi-g^{-1})
2	100	Overburden (clay soil)	0.48	1.404	0.433	0
1	600	Tailings	0.55	1.230	0.190	1600

The resulting concentration profile obtained with RADMD is shown in Figure 15, along with the analytic solution obtained by solving Equation (12). The results are in excellent agreement with the predicted concentrations; the results match the analytic solution with less than 3% error. The predicted bare tailings flux is $308.19 \text{ pCi-cm}^{-2}\text{-hr}^{-1}$, which matches the analytic solution obtained from Equation (11) with less than 0.2% error.

FIGURE 15. Analytical Versus Numerical Solution for 600-cm Thick Bare Tailings

To demonstrate the two-dimensional capability of the model, a tailings pile with a discontinuous cover has been simulated. The grid used for this case is shown in Figure 16. The variable grid size capability has been used to minimize the number of finite difference cells required and, at the same time, maintain sufficient resolution near the discontinuity. Based on the previous simulation, a 12.5-cm cell width has been used at the tailings and cover interface and along the surface of the cover and tailings. Since reasonable accuracy was obtained using this cell width for the one-dimensional case, it was assumed that reasonable accuracy could be obtained for the discontinuous cover simulation. All of the finite difference nodes are cell centered and have additional nodes on the surface midway between cell boundaries. The concentration at all surface nodes was specified as zero, as was the flux into the bottom of the tailings layer. The characteristics of each layer are listed in Table 9.

The surface fluxes predicted by RADMD are shown in Figure 17. As expected, the surface flux values over the covered region are much smaller than over the uncovered tailings. The flux over the cover is on the order of $1.1 \text{ pCi-cm}^{-2}\text{-hr}^{-1}$, reducing to $0.13 \text{ pCi-cm}^{-2}\text{-hr}^{-1}$ just before the abrupt edge. Because the covered region reduces the diffusion of radon gas, the concentration under the cover builds up, resulting in an increased vertical surface flux from the bare tailings near the edge of the cover. The value of the surface flux from the bare tailings far from the cover is $299.3 \text{ pCi-cm}^{-2}\text{-hr}^{-1}$. As expected, the flux from the edge of the cover is near zero near the top of the cover and increases as the tailings layer is approached.

The two-dimensional effect of the discontinuous cover is illustrated in Figure 18. Concentration profiles 450 cm to either side of the cover edge and to a depth of 550 cm from the cover top are shown. This plot shows the high radon concentration that occurs under the cover. At distances greater than 450 cm, the concentration profiles are essentially flat. This indicates that the two-dimensional influence of the abrupt edge is no longer being felt. It should be possible, therefore, to check the numerical results with the analytic calculations for one-dimensional radon diffusion. Figure 19 is the concentration profile through the center of the far left column of cells shown in

FIGURE 16. Two-Dimensional Grid for Discontinuous Cover Simulation

FIGURE 17. Surface Flux Versus Surface Distance
($S = 0$ at midpoint of cover edge)

FIGURE 18. Concentration Profiles for Discontinuous Cover (50 pCi-cm⁻³ interval)

Figure 16. Figure 20 is the concentration profile for the far right column of cells. The analytic solution shown in Figure 19 was computed using RADON1, while the analytic solution shown in Figure 20 is obtained from Equation (12). The match with the analytic solution for concentration is excellent. The surface flux from the cover predicted by RADMD is within 1.6% of the analytic solution computed using Equation (11). The bare tailings' surface flux computed by RADMD is within 3% of the analytic solution computed using Equation (13). This accuracy demonstrates that the multidimensional radioactive diffusion code can simulate steady-state, two-dimensional radon diffusion. Further applications are required to verify the accuracy of the transient solution, and the code should be verified against a two-dimensional analytic solution. In summary, the preliminary modeling results are extremely encouraging. Further development will allow us to account for varying moisture contents in two-dimensional tests. In addition, convective transport of radon gas can be readily simulated using RADMD when the need arises.

FIGURE 19. Analytical Versus Numerical Solution Tailings
(600 cm) with Cover (100 cm)

FIGURE 20. Analytical Versus Numerical Solution
Bare Tailings (600 cm)

FIGURE 2. Analytical versus numerical solution
Data taken from (100 cm)

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