
Validation of Methods for Evaluating Radon-Flux Attenuation Through Earthen Covers

Prepared by D. R. Kalkwarf, H. D. Freeman, J. N. Hartley

Pacific Northwest Laboratory
Operated by
Battelle Memorial Institute

Prepared for
U.S. Nuclear Regulatory
Commission

NOTICE

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, or any of their employees, makes any warranty, expressed or implied, or assumes any legal liability of responsibility for any third party's use, or the results of such use, of any information, apparatus, product or process disclosed in this report, or represents that its use by such third party would not infringe privately owned rights.

NOTICE

Availability of Reference Materials Cited in NRC Publications

Most documents cited in NRC publications will be available from one of the following sources:

1. The NRC Public Document Room, 1717 H Street, N.W.
Washington, DC 20555
2. The NRC/GPO Sales Program, U.S. Nuclear Regulatory Commission,
Washington, DC 20555
3. The National Technical Information Service, Springfield, VA 22161

Although the listing that follows represents the majority of documents cited in NRC publications, it is not intended to be exhaustive.

Referenced documents available for inspection and copying for a fee from the NRC Public Document Room include NRC correspondence and internal NRC memoranda; NRC Office of Inspection and Enforcement bulletins, circulars, information notices, inspection and investigation notices; Licensee Event Reports; vendor reports and correspondence; Commission papers; and applicant and licensee documents and correspondence.

The following documents in the NUREG series are available for purchase from the NRC/GPO Sales Program: formal NRC staff and contractor reports, NRC-sponsored conference proceedings, and NRC booklets and brochures. Also available are Regulatory Guides, NRC regulations in the *Code of Federal Regulations*, and *Nuclear Regulatory Commission Issuances*.

Documents available from the National Technical Information Service include NUREG series reports and technical reports prepared by other federal agencies and reports prepared by the Atomic Energy Commission, forerunner agency to the Nuclear Regulatory Commission.

Documents available from public and special technical libraries include all open literature items, such as books, journal and periodical articles, and transactions. *Federal Register* notices, federal and state legislation, and congressional reports can usually be obtained from these libraries.

Documents such as theses, dissertations, foreign reports and translations, and non-NRC conference proceedings are available for purchase from the organization sponsoring the publication cited.

Single copies of NRC draft reports are available free, to the extent of supply, upon written request to the Division of Technical Information and Document Control, U.S. Nuclear Regulatory Commission, Washington, DC 20555.

Copies of industry codes and standards used in a substantive manner in the NRC regulatory process are maintained at the NRC Library, 7920 Norfolk Avenue, Bethesda, Maryland, and are available there for reference use by the public. Codes and standards are usually copyrighted and may be purchased from the originating organization or, if they are American National Standards, from the American National Standards Institute, 1430 Broadway, New York, NY 10018.

Validation of Methods for Evaluating Radon-Flux Attenuation Through Earthen Covers

Manuscript Completed: August 1984
Date Published: October 1984

Prepared by
D. R. Kalkwarf, H. D. Freeman, J. N. Hartley

Pacific Northwest Laboratory
Richland, WA 99352

Prepared for
Division of Radiation Programs and Earth Sciences
Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
Washington, D.C. 20555
NRC FIN B2269

ABSTRACT

Field and laboratory measurements were made to test the validity of methods for calculating radon-flux attenuation through earthen covers as described in A Handbook for the Determination of Radon Attenuation Through Cover Materials, NUREG/CR-3533. Radon flux was measured at several sites on two, different, earthen-cover systems over uranium-mill tailings. Afterwards, the underlying materials were removed; and their properties which govern radon diffusion were measured. The validity of the diffusion equations presented in the handbook was established by the generally good agreement between the measured radon flux and the flux predicted when measured values of soil properties were used in these equations. Also, approximate values presented in the handbook for various soil properties were compared with those measured on samples collected at the test sites and were generally found to agree within a factor of four. When these approximate values were used in the diffusion equations, the predicted fluxes were larger than the measured values by factors of up to 31. This implies that the diffusion equations will overestimate the depth of cover soil required to attenuate radon flux to a prescribed level if these approximate values are used. However, investigation of the theoretical relationship between the radon flux from an earth-covered tailings pile and the thickness of that cover indicated that the latter would only be overestimated by a factor of up to 1.5 at field sites similar to those examined in this study. In addition, measurements of radon flux over the drill holes indicated that these cover defects, penetrating the entire thickness of the cover but accounting for only 0.07% of its surface area, had negligible effect on the average flux from the cover system.

CONTENTS

	<u>Page</u>
ABSTRACT	iii
ACKNOWLEDGMENTS	ix
EXECUTIVE SUMMARY	xi
1. INTRODUCTION	1
2. CONCLUSIONS	3
3. EXPERIMENTAL METHODS	4
3.1 Description of the Field Sites	4
3.2 Measurement of Radon Flux	4
3.3 Measurement of Bulk Density, Porosity and Moisture Saturation	4
3.4 Measurement of Radon Diffusion Coefficients	7
3.5 Measurement of Radon Emanation Coefficients and Radium Activity	7
4. RESULTS	8
4.1 Measured Values for Soil Properties	8
4.2 Comparison of Measured Soil Parameters with Approximate Values Presented in the HANDBOOK	9
4.3 Comparison of Measured Flux Values with Those Predicted from the Diffusion Equations and Measured Soil Parameters	12
4.4 Comparison of Measured Flux Values with Those Predicted from the Diffusion Equations and Approximate Values for Some of the Soil Properties	17
4.5 Comparison of Radon Flux Measurements Over Intact and Defective Cover Systems	18
5. DISCUSSION	21
6. REFERENCES	24
APPENDIX 1 - Dry Bulk Density and Moisture Contents of Cores Taken from the Test Sites	A.1
APPENDIX 2 - Specific Activities of ²²⁶ Ra and Emanation Coefficients of ²²² Rn in Samples of Tailings Collected at the Test Sites	A.4
APPENDIX 3 - Radon Flux Measurements at the Test Sites	A.5

FIGURES

	<u>Page</u>
1 Plan and Cross-Sectional Views of Field Sites	5

TABLES

	<u>Page</u>
1 Average Values for Dry Bulk Densities, Porosities and Moisture Contents of Soil Layers at the Test Sites	9
2 Measured Values for the Radon Diffusion Coefficients, Radium Activities and Radon Emanation Coefficients in Soil Samples from the Test Sites	10
3 Comparison of Diffusion Coefficients for Radon in Dry Dunite Sand (Porosity 0.44) Obtained by Different Methods	10
4 Dry Bulk Densities and Porosities of Soil Layers in the Cover Systems as a Function of Time at the Test Sites.....	11
5 Measured and Predicted Moisture Saturation in Soil Layers at the Test Sites	12
6 Measured Diffusion Coefficients for Radon in the Soil Columns and Values Predicted from Laboratory-Measured Moisture Saturation and Porosity	13
7 Radon Diffusion Coefficients Obtained by Extrapolating Laboratory-Measured Values to Field Conditions of Porosity and Moisture Saturation	14
8 Diffusion Coefficients for Radon in the Soil Layers Predicted from Meteorological Data, Clay Content and Nominal Porosity	15
9 Comparison of Measured Radon Flux Values with Those Predicted from the Diffusion Equations and Measured Values of Soil Properties	17
10 Comparison of Radon Flux Measurements with Those Predicted from the Diffusion Equations and Approximate Values for Some of the Soil Properties	18
11 Comparison of Radon Flux Measurements Over Intact and Defective Cover Systems	19

12	Overestimates of the Thickness of Cover Soil Corresponding to Overestimates of the Flux from Cover Systems at the Grand Junction Tailings Pile	22
----	------------------------------------------------------------------------------------------------------------------------------------------------------------	----

ACKNOWLEDGMENTS

The authors wish to acknowledge the helpful discussions with Dr. Glendon Gee of the Pacific Northwest Laboratory.

EXECUTIVE SUMMARY

The purpose of this study was to test the validity of methods for calculating radon-flux attenuation through earthen covers as described in A Handbook for the Determination of Radon Attenuation Through Cover Materials (Rogers and Nielson 1984). Radon flux was measured at several sites on the surface of two well-characterized cover systems prepared by Pacific Northwest Laboratory over the inactive tailings pile at Grand Junction, Colorado (Hartley et al. 1983). In one cover system, the major radon-diffusion barrier was a 1.2-m layer of compacted Mancos shale covered with 1.8 m of uncompacted adobe clay to retain moisture. In the other system, the major radon-diffusion barrier was a 1.2-m layer of compacted adobe clay that was also covered with 1.8 m of uncompacted clay.

At three sites in each cover system, the cover soil and tailings were cored, and the properties of these materials governing radon diffusion were measured. These measurements were used in the diffusion equations described in the handbook to calculate the radon flux to be expected at those sites, and the results generally agreed with the flux measurements. At four of the six sites, the calculated values were within the 95% confidence intervals for the measured values. At the other two sites, the equations predicted larger flux values; but these were within the site-to-site variation of surface flux across the cover system.

Radon fluxes at the test sites were also calculated using approximate values for soil properties that may be considered too costly or inconvenient to measure for the design of a cover system. These approximations were derived from empirical equations and field experience described in the handbook, and they were generally found to be within a factor of four from the measured values for the corresponding properties. Fluxes were calculated with two types of parameter sets. In one case, handbook approximations were used for radon diffusion coefficients and emanation coefficients, but measured values were used for all other soil properties. In the other case, handbook approximations were used for all values except those for radium activity in the tailings, clay content of the soils and annual precipitation and evaporation at the site. Use of these parameter sets overestimated the measured fluxes by factors of up to 31. In both cases, overestimation was mainly due to the fact that handbook approximations for the radon diffusion coefficients were larger than the corresponding measured values. These results imply that the thickness of cover soil required to achieve a prescribed flux would also be overestimated if handbook approximations were used for values of the soil properties. In this sense, they provided conservative estimates for the required thicknesses of cover soil.

Analysis of the theoretical relationship between the radon flux and the thickness of an attenuating soil cover indicated that the latter was less sensitive than flux to overestimation by the diffusion equations. Under conditions found at the Grand Junction test sites, analysis showed that cover thicknesses would only be overestimated by factors of up to 1.5 using the handbook approximations.

Measurements of radon flux over the drill holes showed that these cover defects, 0.15 m in diameter and penetrating the entire cover, had negligible effect on the average flux from the cover system. This is because the holes subtend only 0.07% of the system's surface area, and radon must still diffuse laterally through intact portions of the cover in order to reach the defect. In so doing, the radon flux is attenuated by radioactive decay along the diffusion path.

1. INTRODUCTION

Earthen covers have been recommended as effective means for reducing the radon flux from uranium-mill tailings to acceptable values (U.S. Nuclear Regulatory Commission 1980). The soil increases the diffusion path of radon gas to the atmosphere and provides time for radioactive decay of ^{222}Rn ($T=3.82\text{d}$) within the cover. The cover thickness required to reduce the radon flux to prescribed levels will be determined by calculation (U.S. Nuclear Regulatory Commission 1980). Methods for calculating this thickness have recently been reported in the document titled Radon Attenuation Handbook for Uranium-Mill Tailings Cover Design, NUREG/CR-3533 (Rogers and Nielson 1984). This handbook is an updated version of an earlier document titled A Handbook for the Determination of Radon Attenuation Through Cover Materials, NUREG/CR-2340 (Rogers and Nielson 1981). Both versions present equations based on diffusion theory and various soil parameters to calculate a cover thickness which will achieve the desired flux attenuation. Such parameters include the radium activity and radon emanation coefficient in the tailings, and the moisture contents, porosities and diffusion coefficients for radon gas in the tailings and cover soils. The handbooks also present empirical equations to estimate values for some of the parameters used in these equations. The empirical equations in the new version are derived from more-extensive experimental measurements of these parameters. The new version also provides modified radon-transport equations which include terms to account for advective flow in an earthen cover. Henceforth in this report, it will be referred to as the HANDBOOK.

The purpose of this study was to test the validity of HANDBOOK methods when applied to tailings piles in the field. Six sites were selected on two of the cover systems prepared by Pacific Northwest Laboratory over the inactive tailings pile at Grand Junction, Colorado (Hartley et al. 1983). In one system, the major radon-diffusion barrier was a 1.2-m layer of Mancos shale, whereas at the other system, the major barrier was a 1.2-m layer of adobe clay. An uncompacted, top-layer of adobe clay, 1.8 m thick, had been applied in both systems to inhibit evaporation of moisture. The tailings and cover soils had been carefully characterized during construction of the earthen covers; and radon flux, soil moisture and climate have been monitored for almost three years. Data for the validation test were obtained by measuring radon flux at selected locations on the surface of the covered tailings, drilling out samples of the underlying cover soil and tailings and measuring those soil properties that are parameters in the Handbook equations. Predicted values were then compared with measured values.

Four types of comparisons were made. First, measured values of radon flux were compared with those calculated with the diffusion equations, using measured values of soil properties at that site. This type of comparison was designed to test the validity of the radon diffusion equations presented in the HANDBOOK. Second, measured values of soil properties were compared with approximate values presented in the HANDBOOK or predicted by the empirical equations presented therein. Third, measured flux values were compared with those calculated with the diffusion equations using HANDBOOK approximations for some of the parameters. Such comparisons were designed to indicate the latitude

available in using these approximations instead of measured soil properties during the design of an adequate cover system. Finally, the radon fluxes measured directly over the drill holes were compared with those from the intact cover. This comparison was designed to test the effect of highly visible, cover-penetrating defects on the average flux from the cover systems.

2. CONCLUSIONS

It was concluded that the methods presented in the HANDBOOK will provide conservative estimates for the thickness of an earthen cover required to attenuate radon flux to a prescribed value. This conclusion is based on the fact that radon fluxes predicted from HANDBOOK equations were always equal to or larger than the fluxes measured at field locations in this study. It must be recognized, however, that tests were conducted at only six sites on two different cover systems.

The diffusion equations presented in the HANDBOOK were judged to be valid. When measured values were used for all soil properties, these equations predicted flux values that were within the 95% confidence interval for the mean of the measured values at four of the six sites. At the other sites, the equations predicted values which were within the site-to-site variation of surface flux across the cover system.

Approximate values for soil properties derived from empirical equations and field experience described in the HANDBOOK were generally found to be within a factor of four from the corresponding measured values. Radon diffusion coefficients calculated from the porosity and moisture content of the soil differed most significantly from the measured values. It was concluded that this HANDBOOK equation generally overestimates values for the radon diffusion coefficient.

When HANBOOK approximations for radon diffusion coefficients and the radon emanation coefficient were used in the diffusion equations, calculated fluxes exceeded measured fluxes at the test sites by factors of up to 31. It was concluded that this overestimation was mainly due to use of the larger, approximate radon diffusion coefficients in the diffusion equations.

Analysis of the theoretical relationship between radon flux and the thickness of an attenuating soil cover indicated that the latter was less sensitive than flux to overestimation by the diffusion equations. It was concluded that under conditions found at the Grand Junction test sites, the cover thickness would only be overestimated by factors of up to 1.5 using diffusion equations containing the HANDBOOK approximations.

Measurements of radon flux over the drill holes showed that these cover defects, 0.15 m in diameter and penetrating the entire cover, had negligible effect on the average flux from the cover system. This is because the holes subtend only 0.07% of the system's surface area, and radon must still diffuse laterally through intact portions of the cover in order to reach a defect. In so doing, the radon flux is attenuated by radioactive decay along the diffusion path.

3. EXPERIMENTAL METHODS

3.1 Description of the Field Sites

The field sites were selected in a group of cover systems prepared in June 1981 to test the abilities of compacted Mancos shale, compacted Bentonite clay, and compacted adobe clay to attenuate radon emission from the underlying tailings (Hartley et al. 1983). Plan and cross-sectional views of the systems are shown in Figure 1. A 0.2-m layer of uncompacted overburden had been applied initially to the tailings by the mill operators. In 1981, the area had been divided into test plots; and the tailings and overburden had been covered with 1.2 m (4 feet) of compacted test soil followed by 1.8 m (6 feet) of uncompacted adobe clay to inhibit loss of moisture. The dimensions of each plot were 19 m (62.5 feet) by 30.5 m (100 feet). Prior to cover application, measurements were made of the surface radon flux and the specific activity for ^{226}Ra in the underlying tailings as well as the bulk densities and moisture contents of the cover soils. Since then, measurements of radon flux at the upper surface of the covers have been made at intervals of about six months.

In the present study, sites for the field measurements were selected in the test plots containing compacted Mancos shale and compacted adobe clay. Surveyed areas, 9.1m by 9.1m (30 feet by 30 feet) were selected near the centers of the test plots for the measurements. These central areas were judged to have the most stable bulk density and soil moisture and thus provide the best test for the theoretical predictions of radon flux. Five sites were selected in each surveyed area for radon-flux measurements, and these were labeled NW (Northwest), SW (Southwest), C (center), NE (Northeast) and SE (Southeast), corresponding to their position on the test plot. The NW, C and SE sites in each area were selected for excavation, as shown in Figure 1. A prefix in front of the positional label indicates the underlying, compacted diffusion barrier, i.e., CMS for compacted Mancos shale and CAC for compacted adobe clay.

3.2 Measurement of Radon Flux

Flux measurements were made at each site during a 5-day period. The radon flux at the central site, C, in each cover system was measured for 24-hour periods with canisters of activated charcoal, 38 cm in diameter and with a PNL flow-through system (Freeman 1981). At the other four sites in each system, radon flux was measured with PNL flow-through systems for 4-hour intervals during the same 24-hour period. The 4-hour measurements were started in the morning and, again, around noon whereas the 24-hour measurements were made from one morning until the next morning.

3.3 Measurement of Bulk Density, Porosity and Moisture Saturation

After the radon flux measurements were completed, cores were drilled out of the systems at the Northwest, Center and Southeast sites. The uncompacted adobe layer on each of the covers was cored with shelby tubes, 7.1 cm in diameter. The cores were immediately cut into two 23-cm sections and one 46-cm section.

5

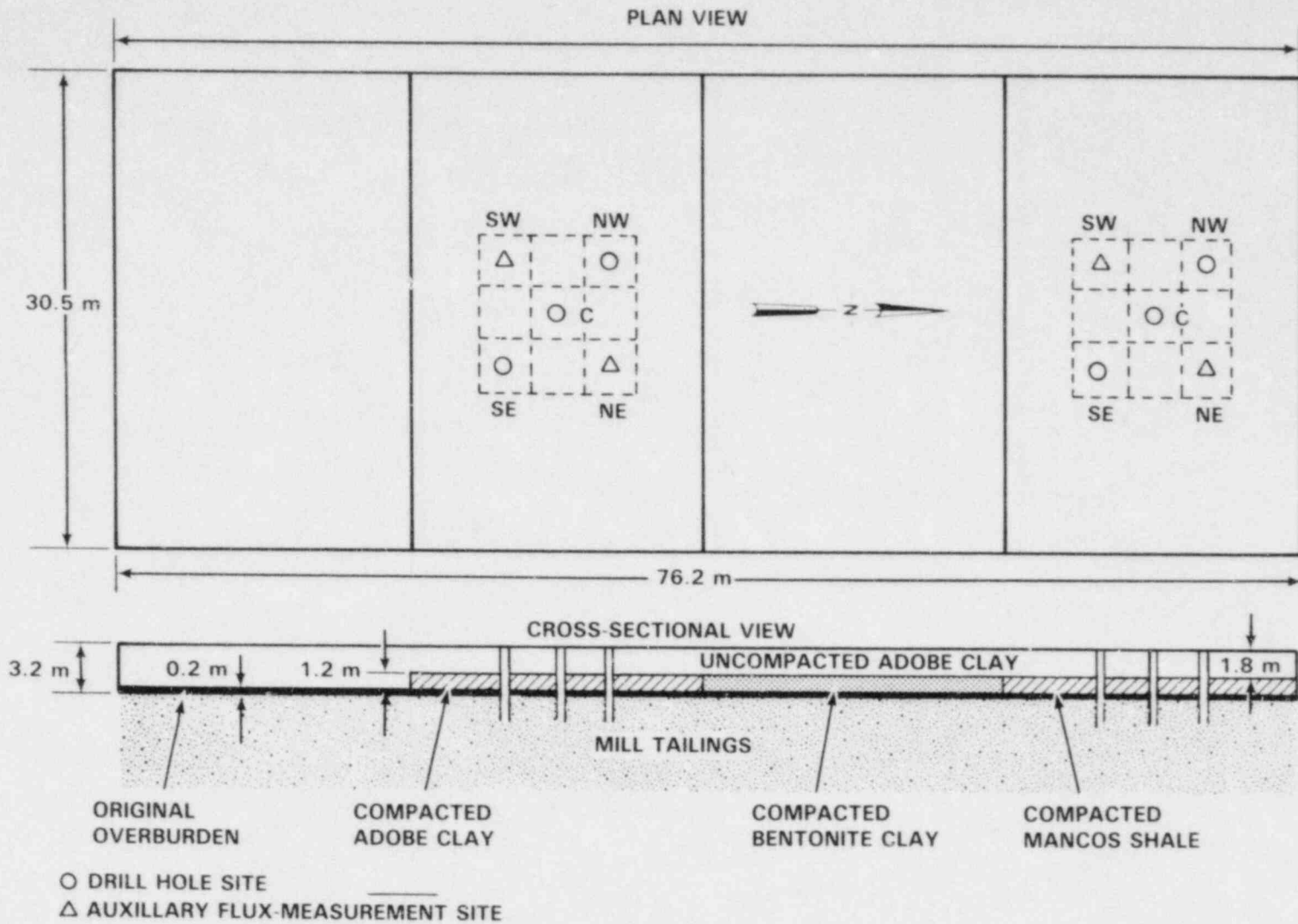


FIGURE 1. Plan and Cross-Sectional Views of Field Sites

The sections were weighed, their volumes were calculated, and these samples were stored in plastic bags to await measurements of their diffusion coefficients and moisture contents in the laboratory. The compacted adobe cover was treated in the same manner. Sampling the compacted Mancos shale layer required a split-spoon sampler because it was too hard to penetrate with a shelby tube. The weights and volumes of sections obtained with the split spoon sampler were measured in the field, and the samples were then sealed in plastic bags to await further measurements in the laboratory. After the cores of the earthen covers were obtained, the holes were augured out with a 15.2-cm (6-inch) auger. Shelby tubes were then used to core the tailings underlying the covers. The tailings removed with the shelby tubes were treated in the same way as the cover samples.

In the laboratory, portions of the core sections were used to measure their dry bulk densities, porosities and moisture contents. Each portion was dried at 110°C to constant weight; and the dry bulk density was calculated with the equation:

$$\rho = \frac{W}{V} \quad (1)$$

where ρ = the dry bulk density of the core section (g cm^{-3})
 W = the dry weight of the core section (g)
 V = the volume of the core section calculated from its wet dimensions (cm^3)

The dry porosity was calculated from the equation:

$$p = 1 - (\rho/g) \quad (2)$$

where p = dry porosity of the core section (dimensionless)
 g = the specific gravity of the earthen particles (g cm^{-3})

The specific gravities of the earthen particles were all assumed to be 2.7 g cm^{-3} in these calculations.

The dry-weight % moisture and the moisture saturation of a core section were used as measures of its moisture content. The dry-weight % moisture of a core section is defined as:

$$M = \frac{100(\text{weight of water in the section})}{\text{dry weight of the section}} \quad (3)$$

Moisture saturation is defined as the ratio of the water-filled pore space in a core section to the total pore space. It was calculated with the equation:

$$m = \frac{\rho M}{(100)p} \quad (4)$$

where m = the moisture saturation of the core section (dimensionless)

3.4 Measurement of Radon Diffusion Coefficients

Diffusion coefficients of radon in tailings, compacted covers and uncompacted covers were calculated from measurements of radon flux through cylindrical columns of each material. Each column was prepared by compositing core sections of material from a particular site and compacting them into a Lucite cell, 14 cm in diameter and 15 cm in length. These core sections had been stored in plastic containers so that their moisture contents stayed at field values. Also, the columns were compacted to wet bulk densities corresponding to those found in the field. Each cell was attached to a radon source manufactured by the Pylon Electronics Co., and a constant, known concentration of dry radon was passed underneath the cell. Any radon diffusing through the soil column was swept from the top of the cell with a stream of nitrogen gas and collected in convoluted tubes filled with charcoal. This charcoal was then analyzed for radon by gamma-ray spectrometry (Silker and Kalkwarf 1983). Diffusion coefficients were evaluated from the experimental parameters by means of the SEARCH computer program developed at PNL (Oster and Mayer 1982). Diffusion coefficients for tailings were corrected for radon formed in the column of tailings.

3.5 Measurement of Radon Emanation Coefficients and Radium Activity

Emanation coefficients for ^{222}Rn and specific activities of ^{226}Ra in the tailings were evaluated by the "sealed-can, gamma-only" technique (Austin 1975). Portions of the samples, at their field moisture contents, were sealed in aluminum cans and counted immediately for ^{214}Bi , a daughter of ^{222}Rn , with an intrinsic germanium diode coupled to a multichannel analyzer. The cans were kept sealed for at least 30 days to allow ^{222}Rn to achieve its equilibrium activity and the cans were recounted for ^{214}Bi . The specific activity of ^{226}Ra was computed from the counting rate at equilibrium and the emanating power was computed from the difference between the initial and equilibrium counting rates.

4. RESULTS

4.1 Measured Values for Soil Properties

Measured values for the dry bulk density, porosity and moisture content of soil layers at the cored sites are shown in Appendix I. Weighted averages of these values for the various layers are listed in Table 1. They were calculated with the equations:

$$\bar{\rho} = \frac{\sum_i t_i \rho_i}{\sum_i t_i} \quad (5)$$

and

$$\bar{M} = \frac{\sum_i t_i M_i}{\sum_i t_i} \quad (6)$$

where $\bar{\rho}$ = average dry bulk density of the layer (g cm^{-3})
 t_i = thickness of layer-section i (cm)
 ρ_i = dry bulk density of layer-section i (g cm^{-3})
 \bar{M} = average dry-weight % moisture in the layer (%)
 M_i = dry-weight % moisture in layer-section i (%)

Measured values for the radium activities and radon emanation coefficients of the tailings are listed in Appendix 2. Weighted averages of these values as well as measured values for the diffusion coefficients of radon in laboratory columns of soil from the site layers are shown in Table 2.

The accuracy of the method used to measure the radon diffusion coefficient was tested by applying it to samples of dry Dunite, an olivine sand. This material has been used to compare other methods for measuring radon diffusion coefficients (Nielson et al. 1983). The results are shown in Table 3, and validate the accuracy of the method.

The HANDBOOK recommends using an emanation coefficient, E , equal to 0.2, based on the average of measurements on tailings from various sites. The values found in this study were all higher than 0.2 and illustrate the variance of actual emanation coefficients from this recommended value. A recent review of emanation coefficients has reported values of E from less than 0.1 to greater than 0.5 (Freeman and Hartley, 1984).

TABLE 1. Average Values for Dry Bulk Densities, Porosities and Moisture Contents of Soil Layers at the Test Sites

<u>Site</u>	<u>Layer</u>	<u>Dry Bulk Density (g cm⁻³)</u>	<u>Porosity</u>	<u>Dry Weight % Moisture</u>	<u>Moisture Saturation</u>
CMS-NW	Uncompacted	1.51	0.44	7.76	0.27
CMS-C	Uncompacted	1.49	0.45	8.48	0.28
CMS-SE	Uncompacted	1.50	0.44	8.47	0.29
CMS-NW	Compacted	1.71	0.37	10.89	0.50
CMS-C	Compacted	1.51	0.44	11.40	0.39
CMS-SE	Compacted	1.64	0.39	12.10	0.51
CMS-NW	Tailings	0.75	0.72	20.49	0.21
CMS-C	Tailings	0.78	0.71	25.86	0.28
CMS-SE	Tailings	0.88	0.67	29.79	0.39
CAC-NW	Uncompacted	1.49	0.45	8.56	0.28
CAC-C	Uncompacted	1.53	0.43	8.70	0.31
CAC-SE	Uncompacted	1.58	0.41	7.74	0.30
CAC-NW	Compacted	1.63	0.40	14.76	0.60
CAC-C	Compacted	1.71	0.37	9.64	0.45
CAC-SE	Compacted	1.59	0.41	9.65	0.37
CAC-NW	Tailings	0.79	0.71	36.03	0.40
CAC-C	Tailings	0.80	0.70	32.99	0.38
CAC-SE	Tailings	0.85	0.69	32.02	0.39

4.2 Comparison of Measured Soil Parameters with Approximate Values Presented in the HANDBOOK

The HANDBOOK assumes physical stability in the tailings and cover system. This includes not only the absence of major land changes such as earthquakes and erosion but also less-noticeable actions such as changes in the dry bulk densities and porosities of the cover materials and tailings. Average values of these latter quantities for core sections taken at the two field sites are listed in Table 4 together with comparable values measured when the cover was installed two years previously.

TABLE 2. Average Values for the Radon Diffusion Coefficients, Radium Activities and Radon Emanation Coefficients in Soil Samples from the Test Sites

Site	Layer	Diffusion Coefficient ($\text{cm}^2 \text{s}^{-1}$)	Emanation Coefficient	Specific Activity of ^{226}Ra (pCi g^{-1})
CMS-NW	Uncompacted	0.009	--	----
CMS-C	Uncompacted	0.009	--	----
CMS-SE	Uncompacted	0.009	--	----
CMS-NW	Compacted	0.012	--	----
CMS-C	Compacted	0.030	--	----
CMS-SE	Compacted	0.008	--	----
CMS-NW	Tailings	0.035	0.33	1712
CMS-C	Tailings	0.007	0.31	1641
CMS-SE	Tailings	0.021	0.26	1838
CAC-NW	Uncompacted	0.005	--	----
CAC-C	Uncompacted	0.010	--	----
CAC-SE	Uncompacted	0.011	--	----
CAC-NW	Compacted	0.009	--	----
CAC-C	Compacted	0.035	--	----
CAC-SE	Compacted	0.008	--	----
CAC-NW	Tailings	0.050	0.47	2281
CAC-C	Tailings	0.013	0.38	2320
CAC-SE	Tailings	0.006	0.49	2297

TABLE 3. Comparison of Diffusion Coefficients for Radon in Dry Dunite Sand (Porosity 0.44) Obtained by Different Methods

Cohen Method ($\text{cm}^2 \text{s}^{-1}$)	Transient-Diffusion Method ($\text{cm}^2 \text{s}^{-1}$)	This Study ($\text{cm}^2 \text{s}^{-1}$)
$0.061 \pm 0.03^{(a)}$	$0.061 \pm 0.09^{(b)}$	$0.063^{(c)}$

(a) 95% confidence interval for mean, Silker and Kalkwarf 1983.

(b) 95% confidence interval for mean, Nielson et al. 1983.

(c) Average of two separate measurements.

TABLE 4. Average Dry Bulk Densities and Porosities of Soil Layers in the Cover Systems as a Function of Time

Site	Layer	Density	Density	Porosity	Porosity
		1981 (g cm ⁻³)	1983 (g cm ⁻³)	1981	1983
CMS	Uncompacted clay	1.38	1.50	0.49	0.44
	Compacted shale	1.52	1.62	0.44	0.40
	Tailings	1.46	0.80	0.46	0.70
CAC	Uncompacted clay	1.38	1.53	0.49	0.43
	Compacted clay	1.49	1.64	0.45	0.39
	Tailings	1.46	0.81	0.46	0.70

Comparison shows that the dry bulk density of the tailings decreased by 45% during the two-year period. No plausible reason can be given for this underground expansion, and it should be noted that the densities of both the compacted and the uncompacted covers remained constant during the two-year period. However, the results show that potential changes in the bulk densities of tailings and, perhaps certain cover materials, must be considered in estimating the accuracy of calculated radon flux, radon diffusion coefficients and cover thicknesses for attenuating radon emission.

The HANDBOOK indicates that the long-term moisture saturation in tailings and cover materials can be predicted with the equation:

$$m = \left[0.124P^{1/2} - 0.0012E - 0.04 + 0.156f_{cm} \right] \left[1 - \left(\frac{0.7 + f_{cm}}{H} \right)^2 \right] + \left(\frac{0.7 + f_{cm}}{H} \right)^2 \quad (7)$$

where: m = moisture saturation
P = annual precipitation (inches)
E = annual lake evaporation (inches)
f_{cm} = fraction of soil passing a U.S. Standard Sieve No. 200
H = depth of the water table (feet)

At the Grand Junction tailings pile, the water table was 24 feet below the top of the original cover. The measured values of f_{cm} were 0.85 for adobe clay and 0.187 for Mancos shale; f_{cm} was assumed to be 0.5 for tailings. Annual precipitation and lake evaporation have averaged 8.46 inches and 36 inches, respectively (NOAA 1979, NOAA 1981). Average measured and predicted values for moisture saturation in tailings and earthen covers at the two sites are compared in Table 5. They show that during the two-year period the moisture contents of the soil layers generally moved closer to the values predicted by Equation 7.

TABLE 5. Measured and Predicted Moisture Saturation in Soil Layers at the Test Sites

Site	Layer	m, 1981	m, 1983	m (predicted)
CMS	Uncompacted clay	0.22	0.28	0.41
	Compacted shale	0.40	0.47	0.31
	Tailings	0.27	0.29	0.36
CAC	Uncompacted clay	0.22	0.30	0.41
	Compacted clay	0.32	0.47	0.41
	Tailings	0.27	0.39	0.36

The HANDBOOK states that the diffusion coefficient for radon in tailings and earthen covers can be predicted from the equation:

$$D = 0.07 \exp [-4m (1-p^2 + m^4)] \quad (8)$$

where: D = diffusion coefficient of radon in the total pore space of the bulk soil ($\text{cm}^2 \text{s}^{-1}$)
 m = moisture saturation (dimensionless)
 p = dry soil porosity (dimensionless)

The diffusion coefficients for radon measured in laboratory columns of materials collected at the sites are listed in Table 6. Values predicted by Equation 8 are also listed for comparison.

The values of moisture saturation and porosity for these columns are also listed in Table 6 and are similar to, but not identical with, those at the field sites. Diffusion coefficients appropriate for the field sites were estimated by using Equation 8 to extrapolate from laboratory to field conditions as described elsewhere (Silker and Kalkwarf 1983; Kalkwarf and Silker 1984). These values are listed in Table 7.

Values for the diffusion coefficients of radon in the various soil layers were also calculated by using Equation 8 with values for m predicted by Equation 7 and the value of 0.15 for porosity, as suggested in the HANDBOOK. The results are shown in Table 8.

4.3 Comparison of Measured Flux Values with Those Predicted from the Diffusion Equations and Measured Soil Parameters

Predicted values of radon flux were calculated with the equations and procedures described in Section 2.6 of the HANDBOOK utilizing the measured values for soil parameters. Since the cover systems at the test sites consisted of three layers of soil above the tailings, the calculations involved several steps.

TABLE 6. Measured Diffusion Coefficients for Radon in the Soil Columns and Values Predicted from Laboratory-Measured Moisture Saturation and Porosity

Site	Layer	Moisture Saturation	Porosity	D	D
				(cm ² s ⁻¹) Measured	(cm ² s ⁻¹) Predicted
CMS-NW	Uncompacted	0.20	0.47	0.009	0.038
	Compacted	0.55	0.51	0.012	0.011
	Tailings	0.32	0.67	0.035	0.034
CMS-C	Uncompacted	0.20	0.47	0.009	0.037
	Compacted	0.30	0.42	0.030	0.026
	Tailings	0.28	0.70	0.068	0.039
CMS-SE	Uncompacted	0.20	0.47	0.009	0.038
	Compacted	0.37	0.43	0.008	0.020
	Tailings	0.34	0.67	0.020	0.032
CAC-NW	Uncompacted	0.23	0.45	0.005	0.033
	Compacted	0.36	0.36	0.009	0.020
	Tailings	0.32	0.69	0.050	0.035
CAC-C	Uncompacted	0.20	0.47	0.009	0.038
	Compacted	0.24	0.47	0.035	0.033
	Tailings	0.35	0.69	0.013	0.033
CAC-SE	Uncompacted	0.16	0.48	0.011	0.043
	Compacted	0.24	0.44	0.008	0.032
	Tailings	0.50	0.65	0.006	0.019

The first step was to evaluate the flux from the tailings. Because the depth of the tailings was greater than 10 m, the following equation could be used:

$$J_t = 10^4 R \rho_t E (\lambda D_t)^{1/2} \quad (9)$$

where: J_t = radon flux from the upper surface of the tailings (pCi m⁻² s⁻¹)
 ρ_t = dry bulk density of the tailings (g cm⁻³)
 R = specific activity of ²²⁶Ra in the tailings (pCi g⁻¹)
 E = radon emanation coefficient of the tailings (dimensionless)
 λ = disintegration constant for ²²²Rn (s⁻¹)
 D_t = diffusion coefficient of ²²²Rn in the tailings (cm² s⁻¹)

TABLE 7. Radon Diffusion Coefficients Obtained by Extrapolating Laboratory-Measured Values to Field Conditions of Porosity and Moisture Saturation

<u>Site</u>	<u>Layer</u>	<u>Field Moisture Saturation</u>	<u>Field Porosity</u>	<u>Extrapolated D (cm²s⁻¹)</u>
CMS-NW	Uncompacted	0.27	0.44	0.007
	Compacted	0.50	0.37	0.013
	Tailings	0.21	0.72	0.048
CMS-C	Uncompacted	0.28	0.45	0.007
	Compacted	0.39	0.44	0.022
	Tailings	0.28	0.71	0.069
CMS-SE	Uncompacted	0.29	0.44	0.007
	Compacted	0.51	0.39	0.004
	Tailings	0.39	0.67	0.018
CAC-NW	Uncompacted	0.28	0.45	0.004
	Compacted	0.60	0.40	0.003
	Tailings	0.30	0.71	0.054
CAC-C	Uncompacted	0.31	0.43	0.006
	Compacted	0.45	0.37	0.015
	Tailings	0.37	0.70	0.013
CAC-SE	Uncompacted	0.30	0.41	0.007
	Compacted	0.37	0.41	0.005
	Tailings	0.39	0.69	0.009

The next step was to calculate the flux of radon from the top of the first soil cover, the 0.2-m thick layer of overburden directly over the tailings. These calculations utilized the following equation:

$$J_1 = \frac{2J_t \exp(-b_1 x_1)}{1 + (a_t/a_1)^{1/2} + [1 - (a_t/a_1)^{1/2}] \exp(-2b_1 x_1)} \quad (10)$$

where: J_1 = radon flux from the top of the first soil cover ($\text{pCi m}^{-2} \text{s}^{-1}$)
 $b_1 = (\lambda/D_1)^{1/2}$
 x_1 = depth of the first soil cover (cm)
 $a_1 = p_1^2 D_1 [1-0.74m_1]^2$
 $a_t = p_t^2 D_t [1-0.74m_t]^2$
 p_1 = dry porosity of the first soil cover (dimensionless)

TABLE 8. Diffusion Coefficients for Radon in the Soil Layers Predicted from Meteorological Data, Clay Content and Nominal Porosity

Site	Layer	Moisture Saturation Predicted	D Predicted (cm ² s ⁻¹)
CMS-NW	Uncompacted	0.41	0.016
	Compacted	0.31	0.023
	Tailings	0.36	0.019
CMS-SE	Uncompacted	0.41	0.016
	Compacted	0.31	0.023
	Tailings	0.36	0.019
CAC-NW	Uncompacted	0.41	0.016
	Compacted	0.31	0.023
	Tailings	0.36	0.019
CAC-C	Uncompacted	0.41	0.016
	Compacted	0.41	0.016
	Tailings	0.36	0.019
CAC-SE	Uncompacted	0.41	0.016
	Compacted	0.41	0.016
	Tailings	0.36	0.019
	Uncompacted	0.41	0.016
	Compacted	0.41	0.016
	Tailings	0.36	0.019

p_t = dry porosity of the tailings (dimensionless)
 D_1 = radon diffusion coefficient in the first soil cover (cm² s⁻¹)
 m_1 = moisture saturation of the first soil cover (dimensionless)
 m_t = moisture saturation of the tailings (dimensionless)

and the other symbols have the same meanings as before. The values of m_t , p_t and D_t at the various sites have already been listed. Because of the small depth of the first soil cover, values for m_1 , p_1 and D_1 could not be discerned from the measurements. Therefore, they were assumed to be the same as the average of the corresponding measured values for compacted adobe clay in this cover system, i.e., $m = 0.47$, $p = 0.39$ and $D = 0.008$ cm² s⁻¹.

The third step was to calculate the radon flux from the top of the second soil cover. This was done with the equation:

$$J_2 = \frac{2J_1 \exp(-b_2 x_2)}{1 + (a_{t1}/a_2)^{1/2} + [1 - (a_{t1}/a_2)^{1/2}] \exp(-2b_2 x_2)} \quad (11)$$

where: J_2 = radon flux from the top of the second soil cover ($\text{pCi m}^{-2} \text{s}^{-1}$)
 $b_2 = (\lambda D_2)^{1/2}$
 x_2 = depth of the second soil cover (cm)
 $a_2 = p_2^2 D_2 [1-0.74m_2]^2$
 $a_{t1} = p_1^2 D_{t1} [1-0.74m_1]^2$
 p_2 = dry porosity in the second soil cover (dimensionless)
 D_2 = diffusion coefficient of radon in the second soil cover ($\text{cm}^2 \text{s}^{-1}$)
 m_2 = moisture saturation in the second soil cover (dimensionless)
 $D_{t1} = D_t \exp(-b_1 x_1) + D_1 [1 - \exp(-b_1 x_1)]$

and the other symbols have the same meanings as before. As explained in the HANDBOOK, section 2.6.2, the quantity, D_{t1} is an effective diffusion coefficient for radon through the combined system of tailings and first soil cover.

The final step was to calculate the flux of radon from the top of the third soil cover, the 1.8-m thick layer of uncompacted adobe clay. These values were calculated with the following equation:

$$J_3 = \frac{2J_2 \exp(-b_3 x_3)}{1 + (a_{t2}/a_3)^{1/2} + [1 - (a_{t2}/a_3)^{1/2}] \exp(-2b_3 x_3)} \quad (12)$$

where: J_3 = radon flux from the top of the third soil cover ($\text{pCi m}^{-2} \text{s}^{-1}$)
 $b_3 = (\lambda D_3)^{1/2}$
 x_3 = depth of the third soil cover (cm)
 $a_3 = p_3^2 D_3 [1-0.74m_3]^{1/2}$
 $a_{t2} = p_2^2 D_{t2} [1-0.74m_2]^{1/2}$
 p = dry porosity in the third soil cover (dimensionless)
 D_3 = diffusion coefficient of radon in the third soil cover ($\text{cm}^2 \text{s}^{-1}$)
 m_3 = moisture saturation in the third soil cover (dimensionless)
 $D_{t2} = D_t \exp(-b_1 x_1 - b_2 x_2) + D_1 [1 - \exp(-b_1 x_1)] \exp(-b_2 x_2) + D_2 [1 - \exp(-b_2 x_2)]$

and the other symbols have the same meanings as before. As explained in the HANDBOOK, Section 2.6.2, the quantity D_{t2} is an effective diffusion coefficient for radon through the combined system of tailings plus the first two soil covers.

Radon flux values predicted with Equation 12 using measured values of the soil properties are compared in Table 9 with the 95% confidence intervals for the mean radon flux measured with PNL flow-through systems at the test sites. Individual measured values of flux are listed in Appendix 3. Table 9 also lists the total flux attenuation of the cover system at each site, i.e., calculated flux from the tailings divided by the measured flux at the cover surface.

These data test the validity of Equations 9 to 12, the basic diffusion equations presented in the HANDBOOK. In each case, the predicted flux was either within the 95% confidence interval for the average measured flux at the site or was larger than the values in that interval.

TABLE 9. Comparison of Measured Radon Flux Values with Those Predicted from the Diffusion Equations and Measured Values of Soil Properties

Site	Measured Flux ^(a) from Cover (pCi m ⁻² s ⁻¹)	Predicted Flux from Cover (pCi m ⁻² s ⁻¹)	Calculated Flux from Tailings ^(b) Measured Flux from Cover
CMS-NW	0 to 2	3	1345
CMS-SW	0 to 10	--	--
CMS-C	0 to 3	10	1007
CMS-NE	0 to 2	--	--
CMS-SE	0 to 11	1	327
CAC-NW	0 to 14	0.3	407
CAC-SW	0 to 33	--	--
CAC-C	0 to 11	5	212
CAC-NE	0 to 15	--	--
CAC-SE	0 to 43	3	60

(a) 95% Confidence interval for a single measurement.

(b) Measured flux from cover site was taken to be the mid-point of its 95% confidence interval.

4.4 Comparison of Measured Flux Values with Those Predicted from the Diffusion Equations and Approximate Values for Some of the Soil Properties

Values for radon flux at the test sites were also calculated using approximate values for soil properties that may be considered too costly or inconvenient to measure for the design of a cover system. Flux values were calculated with two types of parameter sets. In one case, radon diffusion coefficients and emanation coefficients were approximated; but measured values were used for the other soil properties. The emanation coefficient for radon in the tailings was set equal to 0.2, as recommended in the HANDBOOK, and values for the diffusion coefficients of radon in the various soil layers were calculated with Equation 8, using measured values of moisture saturation and porosity in the layers. The results of using this set of parameters in the diffusion equations are shown in Table 10 for comparison with the measured flux values. Each calculated value exceeded the mid-point of the 95% confidence interval for the corresponding measured value by a factor that ranged from 1.6 to 17.

In the other case, HANDBOOK approximations were used for all values except those for radium activity in the tailings, clay content of the soils and annual precipitation and evaporation at the site. The values of moisture saturation in the soil layers were taken to be their long-term values as calculated from Equation 7. Measured values for the clay fractions in the soil layers and meteorological data for the Grand Junction site were used in this calculation.

TABLE 10. Comparison of Radon Flux Measurements with Those Predicted from the Diffusion Equations and Approximate Values for Some of the Soil Properties

Site	Measured Flux ^(a) from Cover (pCi m ⁻² s ⁻¹)	Predicted Flux ^(b) from Cover (pCi m ⁻² s ⁻¹)	Predicted Flux ^(c) from Cover (pCi m ⁻² s ⁻¹)
CMS-NW	0 to 2	13	31
CMS-SW	0 to 10	--	--
CMS-C	0 to 3	25	29
CMS-NE	0 to 2	--	--
CMS-SE	0 to 11	18	33
CAC-NW	0 to 14	12	33
CAC-SW	0 to 33	--	--
CAC-C	0 to 11	23	33
CAC-NE	0 to 15	--	--
CAC-SE	1 to 43	34	33

(a) 95% Confidence interval for a single measurement.

(b) Values calculated from Equations 9 to 12 using measured soil moisture contents and porosities, empirically derived values for the radon diffusion coefficients, measurements of radium activity, and a value of 0.2 for the radon emanation coefficient.

(c) Values calculated from Equations 9 to 12 using predicted long-term moisture contents in the soil layers, a bulk density of 1.5 g cm⁻³ and porosity of 0.35 for all soil layers, empirically derived values for the radon diffusion coefficients and a radon emanation coefficient of 0.2.

Dry bulk densities and porosities of the soil layers were set equal to 1.5 g cm⁻³ and 0.35, respectively, as indicated in the HANDBOOK; and the emanation coefficient was again set equal to 0.2. Radon diffusion coefficients were calculated from Equation 8, using the values for long-term moisture saturation and porosity described above. The results of using this set of parameters in the diffusion equations are also listed in Table 10 for comparison with measured flux values. Each calculated value exceeded the corresponding measured value by a factor that ranged from 1.5 to 31.

4.5 Comparison of Radon Flux Measurements Over Intact and Defective Cover Systems

After the cores were taken from the excavation sites, radon flux from the holes was measured in order to investigate the effect of these cover defects on the average flux from the cover system. Values for the flux measured with PNL flow-through systems directly over the holes are listed in Table 11. Average

TABLE 11. Comparison of Radon Flux Measurements Over Intact and Defective Cover Systems

Site	Measured Flux ^(a) from Intact Cover (pCi m ⁻² s ⁻¹)	Measured Flux from Defect (pCi m ⁻² s ⁻¹)	Average Flux from Intact System ^(b) (pCi m ⁻² s ⁻¹)	Average Flux from Defective System ^(c) (pCi m ⁻² s ⁻¹)
CMS-NW	1	3.2		
CMS-SW	5	--		
CMS-C	1.5	43.4		
CMS-NE	1	--		
CMS-SE	5.5	24		
CMS			2.8	2.8
CAC-NW	7	4		
CAC-SW	16.5	--		
CAC-C	5.5	6		
CAC-NE	7.5	--		
CAC-SE	21.1	25		
CAC			11.5	11.5

(a) Mid-point values for the 95% confidence intervals.

values were also calculated for radon flux over each cover system before and after the holes were drilled. The average flux before the holes were drilled was calculated from the equation:

$$J_{is} = \frac{1}{T} \sum_{i=1}^5 J_i \quad (13)$$

where J_{is} = average radon flux from the intact cover system (pCi m⁻² s⁻¹)
 J_i = radon flux at cover site i (pCi m⁻² s⁻¹).

After the holes were drilled, the average flux was calculated from the equation:

$$J_{ds} = [(A_s - A_h) J_{is} + A_h \sum_{h=1}^3 J_h] / A_s \quad (14)$$

where J_{ds} = average radon flux from the defective cover system (pCi m⁻² s⁻¹)
 A_s = surface area of the cover system (m²)
 A_h = cross-sectional area of drill hole (m²).

These flux values are also listed in Table 11, and comparison shows that the defects caused no detectable change in the average flux over either system.

5. DISCUSSION

Radon fluxes calculated for the Grand Junction test sites by the methods recommended in the HANDBOOK were either equal to or greater than the measured values. This implies that the HANDBOOK methods will prescribe thicknesses of soil in these cover systems that are not only sufficient to achieve the desired attenuation of radon flux but may be thicker than necessary. In this sense, the HANDBOOK methods provide conservative estimates for the required depths of cover soil in these systems. It must be recognized, however, that only six sites in two, different cover systems were examined.

The validity of the diffusion equations presented in the HANDBOOK was established by their ability to predict radon fluxes at the test sites. If measured values for all the soil parameters were used in the calculations, the predicted values were within the 95% confidence intervals for the measured values at four of the six test sites. At the other two sites, the predicted values were larger than the corresponding measured values by factors of up to six. However, in these cover systems, the measured flux at adjoining sites also differed by factors of up to six. In view of this variability in flux measured at nearby locations, the calculated values were considered to be in reasonable agreement with those measured.

Radon fluxes calculated with the diffusion equations and HANDBOOK approximations for values of the soil properties were much larger than the measured fluxes. Each calculated value exceeded its corresponding measured value by a factor that ranged from 1.5 to 31. Similar overestimations were reported when the RAMD computer model, developed at the Pacific Northwest Laboratory, was used to calculate radon flux emerging from these same cover systems at Grand Junction (Mayer and Gee 1983). In the present investigation, the overestimation of flux was mainly due to the fact that radon diffusion coefficients calculated with Equation 8 were generally larger than the corresponding measured values. A similar relationship was found in an earlier study when coefficients calculated for other soil columns with this equation were compared with their corresponding measured values (Silker and Kalkwarf 1983, Kalkwarf and Silker 1984). The larger diffusion coefficients indicate more rapid flow of radon through the tailings and earthen cover. In this present study, the enhancing effect of larger diffusion coefficients on the calculated flux even surpassed the diminishing effect of the HANDBOOK-suggested, but lower-than-measured, value for the radon emanation coefficient. These results suggest that thickness of earthen cover to attenuate radon flux will be less-severely overestimated if measured radon diffusion coefficients are used in the diffusion equations. Of course, these coefficients must be measured on columns of the candidate soil with the same porosity and moisture content as expected in the field.

Consideration of the relationship between radon flux from an earth-covered tailings pile and the thickness of that cover indicates that the latter would not be overestimated as severely by using HANDBOOK approximations for values of the soil properties in the diffusion equations. The exact relationship between the overestimated flux and overestimated cover thickness is difficult to

determine because the diffusion equations depend in a complex way on a variety of soil properties. Some indication of this relationship can be obtained by approximating the diffusion expression shown in Equation 10 by the simpler equation,

$$J_c = J_t \exp(-b_c x_c) \quad (15)$$

where J_c = calculated flux from the cover ($\text{pCi m}^{-2} \text{s}^{-1}$)
 J_t = calculated flux from the tailings ($\text{pCi m}^{-2} \text{s}^{-1}$)
 $b_c = (\lambda/D_c)^{1/2}$
 D_c = radon diffusion coefficient in the cover ($\text{cm}^2 \text{s}^{-1}$)
 x_c = thickness of tailings (cm)
 and λ = disintegration constant for ^{222}Rn .

Equation 15 can be derived from Equation 10 by letting $a_1 = a_t$.

If Equation 15 overestimates the true flux from the cover by a factor, F , the true value for b_c , namely b'_c , must be larger than the one used and is given by the equation,

$$b'_c = (1/x_c) \ln(J_t/J'_c) = (1/x_c) \ln(FJ_t/J_c) \quad (16)$$

where J'_c = the measured flux from the cover ($\text{pCi m}^{-2} \text{s}^{-1}$)

The correct thickness of cover soil, x''_c , required to achieve a desired radon flux, J_c , from the cover is then given by the equation,

$$x''_c = (1/b'_c) \ln(J_t/J''_c) \quad (17)$$

If the smaller value, b_c , had been used; the required thickness would be given by the equation,

$$x'_c = (1/b_c) \ln(J_t/J''_c) \quad (18)$$

Thus, the cover thickness would be overestimated by the factor, x'_c/x''_c , which is given by the equation,

$$\frac{x'_c}{x''_c} = \frac{(1/b_c) \ln(J_t/J''_c)}{(1/b'_c) \ln(J_t/J''_c)} = \frac{b'_c}{b_c} = \frac{(1/x_c) \ln(FJ_t/J_c)}{(1/x_c) \ln(J_t/J_c)} = 1 + \frac{\ln F}{\ln(J_t/J_c)} \quad (19)$$

and can be seen to be a function of both the factor, F , by which the flux was overestimated, and J_t/J_c , the flux attenuation factor predicted from Equation 15.

Table 12 lists calculated values for the thickness-overestimation factors, x'_c/x''_c , corresponding to the maximum flux-overestimation factors, F_{max} , found in the HANDBOOK predictions for the Grand Junction sites. Also, the corresponding flux-attenuation factors, J_t/J_c , presented previously in Table 9 are relisted in Table 12.

TABLE 12. Overestimates of the Thickness of Cover Soil Corresponding to Overestimates of the Flux from Cover Systems at the Grand Junction Tailings Pile

<u>Site</u>	<u>J_t/J_c</u>	<u>F_{max}</u>	<u>x'_c/x''_c</u>
CMS-NW	1345	31	1.5
CMS-C	1007	19	1.4
CMS-SE	327	6	1.3
CAC-NW	407	5	1.3
CAC-C	212	6	1.3
CAC-SE	60	1.6	1.1

This comparison shows that under conditions found at the Grand Junction test sites, cover thicknesses would be overestimated by factors of up to 1.5 if HANDBOOK approximations were used for values of the soil properties. Although this evaluation is an approximation due to its basis in Equation 15, it does show that overestimates of radon flux by the HANDBOOK equations correspond to much smaller overestimates of cover-soil thickness.

Negligible changes in the average radon flux at the surface of a cover system were produced by easily visible, cover-penetrating defects in the cover. This result depends on having an intact cover system over most of the tailings area and was predicted in the HANDBOOK on the basis of a theoretical and experimental study of radon diffusion from defective covers (Kalkwarf and Mayer 1983). Although defects provide a pathway for rapid diffusion of radon to the atmosphere, radon generated in the tailings must still diffuse laterally through intact portions of the cover in order to reach a defect. In so doing, the radon flux is attenuated by radioactive decay along the diffusion path.

6. REFERENCES

- Austin, S. R. 1975. "A Laboratory Study of Radon Emanation from Domestic Uranium Ores". In Radon in Uranium Mining, IAEA-Pl-565/8, pp. 151-163, International Atomic Energy Agency, Vienna.
- Freeman, H. 1981. An Improved Radon Flux Measurement System for Uranium Tailings Pile Measurement. PNL-SA-9215, Pacific Northwest Laboratory, Richland, Washington.
- Freeman, H. D. and J. N. Hartley. 1984. "Predicting Radon Flux from Uranium Mill Tailings." In: Proceedings of the Sixth Symposium on Management of Uranium Mill Tailings, Low-Level Waste and Hazardous Waste. pp. 221-233. Colorado State University, Ft. Collins, Colorado.
- Hartley, J. N., G. W. Gee, E. G. Baker and H. D. Freeman. 1983. 1983 Radon Barrier Field Test at Grand Junction Uranium Mill Tailings Pile, PNL-4539, (DOE/UMT-0213), Pacific Northwest Laboratory, Richland, Washington.
- Kalkwarf, D. R. and D. W. Mayer. 1983. Influence of Cover Defects on the Attenuation of Radon with Earthen Covers, NUREG/CR-3395, (PNL-4776), U.S. Nuclear Regulatory Commission, Washington, D.C.
- Kalkwarf, D. R. and W. B. Silker. 1984. "Diffusion of Radon in Candidate Soils for Covering Uranium-Mill Tailings". In Proceedings of the Sixth Symposium on Management of Uranium Mill Tailings, Low-Level Waste and Hazardous Waste, pp. 297-305. Colorado State University, Ft. Collins, Colorado.
- Mayer, D. W., C. A. Oster, R. W. Nelson and G. W. Gee. 1981. Radon Diffusion Through Multilayer Earthen Covers: Models and Simulations, PNL-3989 (UMT/0204), Pacific Northwest Laboratory, Richland, Washington.
- Mayer, D. W. and G. W. Gee. 1983. Multidimensional Simulation of Radon Diffusion Through Earthen Covers. PNL-4458 (DOE/UMT-0212), Pacific Northwest Laboratory, Richland, Washington.
- National Oceanic and Atmospheric Administration. 1979. Climatic Atlas of the United States. U.S. Department of Commerce, Washington, D.C.
- National Oceanic and Atmospheric Administration. 1981. Local Climatological Data: Annual Summary with Comparative Data, Grand Junction, Colorado. U.S. Department of Commerce, Washington, D.C.
- Nielson, K. K., D. C. Rich and V. C. Rogers. 1982. Comparison of Radon Diffusion Coefficients Measured by Transient-Diffusion and Steady-State Laboratory Methods. NUREG/CR-2875 (PNL-4370, RAE-18-3), U.S. Nuclear Regulatory Commission, Washington, D.C.

Rogers, V. C. and K. K. Nielson. 1981. A Handbook for the Determination of Radon Attenuation Through Cover Materials. NUREG/ CR-2340 (PNL-4084, RAE-18-1), U.S. Nuclear Regulatory Commission, Washington, D.C.

Rogers, V. C. and K. K. Nielson. 1984. Radon Attenuation Handbook for Uranium Mill Tailings Cover Design. NUREG/CR-3533 (PNL-4878, RAE-18-5). U.S. Nuclear Regulatory Commission, Washington, D.C.

APPENDIX 1. Dry Bulk Density and Moisture Contents of Cores Taken
From the Test Sites

<u>Location</u>	<u>Depth (cm)</u>	<u>Dry Bulk Density (g/cm³)</u>	<u>Dry Weight % Moisture</u>
CMS-NW Uncompacted Adobe Clay	0-34	1.32	6.8
	34-57	1.54	9.2
	57-80	1.57	9.0
	80-109	1.46	10.3
	109-132	1.56	7.5
	132-155	1.45	3.5
	155-178	1.43	6.4
	178-201	1.72	9.4
	201-224	1.60	7.5

Compacted Mancos shale	224-234	1.39	11.7
	234-249	1.66	9.1
	249-258	1.53	11.7
	258-268	1.60	10.2
	268-283	1.60	12.1
	283-291	1.46	11.8
	291-302	1.66	13.5
	302-315	1.64	9.8
	315-343	2.14	10.1

Tailings	343-366	0.88	27.6
	366-389	0.52	30.8
	389-423	0.73	10.2
	423-446	0.86	18.3
<hr/>			
CMS-C Uncompacted Adobe Clay	0-34	1.70	10.7
	34-44	1.60	9.4
	44-55	1.54	8.4
	55-65	1.58	7.6
	65-97	1.45	7.7
	97-127	1.27	7.5
	127-171	1.41	8.7

Compacted Mancos Shale	171-210	1.66	11.2
	210-262	1.51	11.1
	262-300	1.37	13.2

Tailings	300-333	0.68	12.1
	333-377	0.68	30.9
	377-418	0.88	21.1
	418-464	0.87	15.4
	464-486	0.76	39.7
	486-509	0.76	52.1

Location	Depth (cm)	Dry Bulk Density (g/cm ³)	Dry Weight % Moisture	
CMS-SE	0-28	1.64	8.1	
	28-51	1.57	7.9	
	Uncompacted Adobe Clay	51-74	1.37	7.3
		74-95	1.50	6.8
		95-118	1.68	7.7
	118-181	1.40	10.1	
Compacted Mancos Shale	181-192	1.40	11.0	
	192-230	1.72	12.6	
	230-271	1.21	13.1	
	271-293	1.61	13.5	
	293-326	---	9.7	
Tailings	326-349	0.72	16.2	
	349-385	1.08	28.9	
	385-408	1.04	25.1	
	408-431	0.94	25.7	
	431-456	0.81	27.9	
	456-479	0.72	34.2	
	479-502	0.71	49.6	
CAC-NW	0-23	1.57	9.5	
	23-47	1.55	10.8	
	47-70	1.46	9.3	
	Uncompacted Adobe Clay	70-93	1.39	8.7
		93-130	1.64	8.6
		130-165	1.51	7.1
		165-188	1.40	7.0
	188-211	1.41	8.0	
	211-226	1.32	8.8	
	Compacted Adobe Clay	226-249	1.79	9.4
249-272		1.79	10.5	
272-297		1.40	10.5	
297-320		1.47	12.5	
320-343		1.46	29.1	
Tailings	343-373	0.76	37.1	
	373-404	0.90	27.6	
	404-446	0.68	39.7	
	446-469	0.79	36.2	
	469-492	0.81	34.3	

<u>Location</u>	<u>Depth (cm)</u>	<u>Dry Bulk Density (g/cm³)</u>	<u>Dry Weight % Moisture</u>
CAC-C Uncompacted Adobe Clay	0-30	1.51	9.9
	30-53	1.58	9.8
	53-76	1.39	8.9
	76-105	1.51	8.7
	105-128	1.65	9.5
	128-151	1.55	7.7
	151-187	1.55	7.0
Compacted Adobe Clay	187-210	1.84	9.1
	210-232	1.60	8.9
	232-243	1.80	10.4
	243-265	1.76	10.0
	265-288	1.61	10.2
Tailings	288-342	0.83	33.7
	342-364	0.70	38.8
	364-398	0.86	29.7
	398-420	0.80	27.3
	420-443	0.75	36.1
CAC-SE Uncompacted Adobe Clay	0-36	1.48	9.0
	36-58	1.58	8.1
	58-84	1.63	8.5
	84-107	1.79	7.5
	107-130	1.60	7.1
	130-152	1.49	6.6
	152-179	1.51	6.7
Compacted Adobe Clay	179-216	1.72	6.8
	216-239	1.85	9.6
	239-262	1.59	7.6
	262-297	1.39	16.4
Tailings	297-328	1.45	7.0
	328-351	1.03	21.6
	351-373	0.77	42.1
	373-399	0.86	25.4
	399-422	0.75	33.5
422-444	0.84	39.1	

APPENDIX 2. Specific Activities of ^{226}Ra and Emanation Coefficients of ^{222}Rn in Samples of Tailings Collected at the Test Sites

<u>Location</u>	<u>Depth (cm)</u>	<u>Specific Activity (pCi g⁻¹)</u>	<u>Emanation Coefficient</u>
CMS-NW	343-366	2264	0.26
	366-389	1829	0.27
	389-423	823	0.32
	423-446	2357	0.47
CMS-C	300-333	2140	0.31
	333-377	1547	0.35
	377-418	1409	0.23
	418-486	2150	0.32
	486-509	1170	0.34
CMS-SE	326-385	1831	0.16
	385-456	1939	0.37
	456-479	1902	0.31
	479-502	1482	0.11
CAC-NW	343-373	2017	0.53
	373-404	2489	0.52
	404-446	2494	0.49
	446-469	2401	0.39
	469-492	1835	0.35
CAC-C	328-399	2332	0.50
	399-444	2244	0.46
CAC-SE	288-342	2300	0.36
	342-364	2404	0.42
	364-398	2488	0.30
	398-420	2267	0.40
	420-443	2096	0.49

APPENDIX 3. Radon Flux Measurements at the Test Sites

<u>Location</u>	<u>6/15/83</u>	<u>6/16/83</u>	<u>6/17/83</u>	<u>6/18/83</u>	<u>6/19/83</u>
CMS-NW	0.77	0.25	0.76	----	----
	0.98	0.43	0.96	----	----
	1.18	----	----	----	----
CMS-SW	4.06	2.56	0.34	----	----
	4.77	4.10	5.87	----	----
	7.93	----	----	----	----
CMS-C	1.13	0.88	0.77	0.89	0.72
CMS-C(a)	0.41	2.71	0.22	1.49	1.58
CMS-NE	0.91	0.28	0.46	----	----
	1.19	0.33	0.80	----	----
CMS-SE	6.12	1.89	3.74	----	----
	8.85	3.68	6.02	----	----
CAC-NW	7.47	1.85	4.71	----	----
	10.7	3.41	5.56	----	----
CAC-SW	21.8	2.56	11.9	----	----
	16.4	11.3	22.0	----	----
CAC-C	8.85	6.89	3.73	4.26	4.12
CAC-C(a)	0.84	4.43	0.73	5.36	8.57
CAC-NE	8.87	3.04	6.62	----	----
	11.8	4.33	6.99	----	----
CAC-SE	----	15.1	18.1	----	----
	31.9	19.0	23.5	----	----

(a) Measured with canister of activated charcoal placed next to center location. An open-ended canister was used from 6/15 to 6/17, and a closed-ended canister was used on 6/15 and 6/19.

DISTRIBUTION

No. of
Copies

No. of
Copies

OFFSITE

U.S. Nuclear Regulatory Commission
Division of Technical Information
and Document Control
7920 Norfolk Avenue
Bethesda, MD 20014

George F. Birchard
U.S. Nuclear Regulatory Commission
1130-SS
Washington, DC 20555

V. C. Rogers
Rogers & Associates Eng. Corp
P. O. Box 330
Salt Lake City, UT 84110

K. K. Nielson
Rogers & Associates Eng. Corp.
P. O. Box 330
Salt Lake City, UT 84110

M. L. Matthews
Department of Energy
Albuquerque, NM 87115

P. J. Magno
ANR 460
U.S. Environmental Protection
Agency
402 M Street S.W.
Washington, DC 20460

P. T. Owen
Remedial Action Program
Information Center
Oak Ridge National Laboratory
P. O. Box X, Building 2001
Oak Ridge, TN 38830

Walter C. Barber
Jacobs Engineering Group, Inc.
5301 Central Ave., N.E., Ste. 1700
Albuquerque, NM 87108

Mark Jackson
Jacobs Engineering Group, Inc.
5301 Central Ave., N.E., Ste. 1700
Albuquerque, NM 87108

Ken R. Baker
Jacobs Engineering Group, Inc.
5301 Central Ave., N.E., Ste. 1700
Albuquerque, NM 87108

Peter Rafferty
Weston
5301 Central Ave., N.E., Ste. 1700
Albuquerque, NM 87108

John Nelson
Colorado State University
Fort Collins, CO 80523

Thomas Shepherd
Water, Waste & Land Co.
1311 So. College Ave.
Fort Collins, CO 80524

John Themelis
U.S. Department of Energy
5301 Central Ave., N.E., Ste. 1700
Albuquerque, NM 87108

Victor Haw
National Uranium Tailings Program
555 Booth St.
Ottawa, Ontario
K1A 0G1

Gordon M. Ritcey
Canada Centre for Mineral &
Energy Technology
555 Booth St.
Ottawa, Ontario
K1A 0G1

DISTRIBUTION

No. of
Copies

No. of
Copies

Desmond M. Levins
Australian Atomic Energy Commission
Research Establishment
Lucas Heights, New South Wales 2232
Australia

Kaye P. Hart
Australian Atomic Energy Commission
Research Establishment
Lucas Heights, New South Wales 2232
Australia

Hoyt Mitchell
Project Engineer
ARIX
1005 North 12th St., Suite 202
Grand Junction, CO 81501

J. F. Park
R. W. Perkins
S. R. Peterson
A. E. Reisenauer
T. W. Schrauf
R. E. Schirmer
J. A. Stottlemyre
R. L. Skaggs
V. W. Thomas
N. A. Wogman
Publishing Coordination (2)
Technical Information (5)

ONSITE

50 Pacific Northwest Laboratory

W. J. Deutsch
D. W. Dragnich/C. E. Elderkin
M. E. Dodson
J. L. Downs-Berg
R. M. Ecker
M. R. Elmore
W. D. Felix
M. G. Foley
5 H. D. Freeman
J. S. Fruchter
G. W. Gee
5 J. N. Hartley
P. O. Jackson
8 D. R. Kalkwarf
E. A. Lepel
I. C. Nelson
R. W. Nelson
J. M. Nielson

RC FORM 335 <small>(1-81)</small>		U.S. NUCLEAR REGULATORY COMMISSION BIBLIOGRAPHIC DATA SHEET		1. REPORT NUMBER (Assigned by DDC) NUREG/CR-3457 PNL-5092	
TITLE AND SUBTITLE (Add Volume No., if appropriate) Validation of Methods for Evaluating Radon-Flux Attenuation through Earthen Covers				2. (Leave blank)	
AUTHOR(S) D. R. Kalkwarf, H. D. Freeman and J. N. Hartley				3. RECIPIENT'S ACCESSION NO.	
PERFORMING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code) Pacific Northwest Laboratory P.O. Box 999 Richland, WA 99352				5. DATE REPORT COMPLETED MONTH YEAR August 1984	
SPONSORING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code) Division of Radiation Programs and Earth Sciences Office of Nuclear Regulatory Research U.S. Nuclear Regulatory Commission Washington, D.C. 20555				DATE REPORT ISSUED MONTH YEAR October 1984	
TYPE OF REPORT Topical Technical Report				6. (Leave blank)	
SUPPLEMENTARY NOTES				8. (Leave blank)	
ABSTRACT (200 words or less) <p>Field and laboratory measurements were made to test the validity of methods for calculating radon-flux attenuation through earthen covers as described in <u>A Handbook for the Determination of Radon Attenuation Through Cover Materials, NUREG/CR-3533</u>. The validity of the diffusion equations presented in the handbook was established by the generally good agreement between the measured radon flux at six field sites and the flux predicted when measured properties of soil underlying these sites were used in these equations. When approximate values presented in the handbook for various soil properties were used in the diffusion equations, the predicted fluxes were larger than the measured values by factors of up to 31. However, investigation of the theoretical relationship between the radon flux from an earth-covered tailings pile and the thickness of that cover indicated that the latter would only be overestimated by a factor of up to 1.5 at field sites similar to those examined in this study.</p>				10. PROJECT/TASK/WORK UNIT NO.	
KEY WORDS AND DOCUMENT ANALYSIS				11. FIN NO. B-2269	
17a. DESCRIPTORS Radon Flux Attenuation Handbook				14. (Leave blank)	
IDENTIFIERS: OPEN-ENDED TERMS				19. SECURITY CLASS (This report) Unclassified	
AVAILABILITY STATEMENT Unlimited				21. NO. OF PAGES	
				20. SECURITY CLASS (This page) Unclassified	
				22. PRICE \$	

UNITED STATES
NUCLEAR REGULATORY COMMISSION
WASHINGTON, D.C. 20555

OFFICIAL BUSINESS
PENALTY FOR PRIVATE USE, \$300

FOURTH CLASS MAIL
POSTAGE & FEES PAID
USNRC
WASH. D.C.
PERMIT No. G-67

120555078977 1 LANIRU
US NRC
ADM-DIV OF TLOC
POLICY & PUB MGT BR-PDR NUREG
W-501
WASHINGTON DC 20555

NUREG/CR-3457

VALIDATION OF METHODS FOR EVALUATING RADON-FLUX ATTENUATION
THROUGH EARTHEN COVERS

OCTOBER 1984