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# Comparison of Field-Measured Radon Diffusion Coefficients with Laboratory-Measured Coefficients

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Prepared by E. A. Lepel, W. B. Silker, V. W. Thomas, D. R. Kalkwarf

**Pacific Northwest Laboratory**  
Operated by  
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Prepared for  
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U.S. Nuclear Regulatory Commission  
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# Comparison of Field-Measured Radon Diffusion Coefficients with Laboratory-Measured Coefficients

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## ABSTRACT

Experiments were conducted to compare radon diffusion coefficients determined for 0.1-m depths of soils by a steady-state method in the laboratory and diffusion coefficients evaluated from radon fluxes through several-fold greater depths of the same soils covering uranium-mill tailings. The coefficients referred to diffusion in the total pore volume of the soils and are equivalent to values for the quantity,  $D/P$ , in the Generic Environmental Impact Statement on Uranium Milling prepared by the U.S. Nuclear Regulatory Commission. Two soils were tested: a well-graded sand and an inorganic clay of low plasticity. For the flux evaluations, radon was collected by adsorption on charcoal following passive diffusion from the soil surface and also from air recirculating through an aluminum tent over the soil surface. An analysis of variance in the flux evaluations showed no significant difference between these two collection methods. Radon diffusion coefficients evaluated from field data were statistically indistinguishable, at the 95% confidence level, from those measured in the laboratory; however, the low precision of the field data prevented a sensitive validation of the laboratory measurements. From the field data, the coefficients were calculated to be  $0.03 \pm 0.03 \text{ cm}^2/\text{s}$  for the sand cover and  $0.003 \pm 0.002 \text{ cm}^2/\text{s}$  for the clay cover. From the laboratory data, the coefficients were calculated to be  $0.021 \pm 0.002 \text{ cm}^2/\text{s}$  for the sand cover and  $0.0036 \pm 0.0004 \text{ cm}^2/\text{s}$  for the clay cover. The low precision in the coefficients evaluated from field data was attributed to high variation in radon flux with time and surface location at the field site.

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Experiments were conducted to compare radon diffusion coefficients determined for 0.1-m depths of soils by a steady-state method in the laboratory and diffusion coefficients evaluated from carbon fluxes through several-fold greater depths of the same soils covering uranium-mill tailings. The coefficients referred to diffusion in the total pore volume of the soil and are equivalent to values for the quantity  $D_p$  in the generic Environmental Impact Statement on Uranium Milling prepared by the U.S. Nuclear Regulatory Commission. Two soils were tested: a well-graded sand and an inorganic clay of low plasticity. For the flux evaluations, radon was collected by adsorption on charcoal following passive diffusion from the soil surface and also from air recirculating through an aluminum can over the soil surface. An analysis of variance in the flux evaluations showed no significant difference between these two collection methods. Radon diffusion coefficients evaluated from field data were statistically indistinguishable at the 95% confidence level from those measured in the laboratory; however, the low precision of the field data prevented a sensitive validation of the laboratory measurements. From the field data, the coefficients were calculated to be  $0.03 \pm 0.03 \text{ cm}^2/\text{s}$  for the sand cover and  $0.003 \pm 0.003 \text{ cm}^2/\text{s}$  for the clay cover. From the laboratory data, the coefficients were calculated to be  $0.051 \pm 0.002 \text{ cm}^2/\text{s}$  for the sand cover and  $0.0036 \pm 0.0004 \text{ cm}^2/\text{s}$  for the clay cover. The low precision in the coefficients evaluated from field data was attributed to high variation in radon flux with time and surface location at the field site.

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## EXECUTIVE SUMMARY

The purpose of this study was to compare radon diffusion coefficients evaluated for 0.1-m depths of soils under laboratory conditions with diffusion coefficients evaluated for several-fold greater depths of the same soils when used to cover uranium-mill tailings. These coefficients referred to diffusion in the total pore space of the soils and are equivalent to values for the quantity,  $D/P$ , in the diffusion equations presented in the Generic Environmental Impact Statement on Uranium Milling prepared by the U.S. Nuclear Regulatory Commission<sup>(1)</sup>. Two soils were tested: a well-graded sand with a dry porosity of 0.38 and containing 0.07 moisture by volume; and an inorganic clay of low plasticity with a dry porosity of 0.49 and containing 0.20 moisture by volume. Both were considered by the mill operators as candidate cover soils for attenuating radon emission from tailings at the field test site.

In the field, radon diffusion coefficient were evaluated from the radon fluxes emitted by bare and covered tailings and from the porosities, and depths of the soil covers as well as the  $^{226}\text{Ra}$  content and radon emanation coefficient of the underlying mill tailings. The sand cover was 1.14 m in depth and covered an area of 1.31 m<sup>2</sup>. The clay cover was applied over an equal-sized area but to a depth of only 0.44 m. Radon fluxes were measured by two independent methods, one based upon the passive diffusion of radon into an open canister of charcoal and the other utilizing recirculated air to sweep the radon flux collected by an aluminum tent into a charcoal trap. Analysis of variance in the flux measurements showed no significant difference in the measurement methods. The diffusion coefficient evaluated for the sand cover was  $0.03 \pm 0.03 \text{ cm}^2/\text{s}$  at the 95% confidence level, whereas that for the clay cover was  $0.003 \pm 0.002 \text{ cm}^2/\text{s}$ .

In the laboratory, diffusion coefficients were evaluated by a steady-state diffusion method applied to soil samples, 0.02 m<sup>2</sup> in area and 0.10 m in depth, with the same porosity and moisture content as in the field experiments. The diffusion coefficient measured for radon in the sand cover was  $0.021 \pm 0.002 \text{ cm}^2/\text{s}$  at the 95% confidence level, whereas that for the clay was  $0.0036 \pm 0.0004 \text{ cm}^2/\text{s}$  at the 95% confidence level. Both values were within the uncertainty ranges for the field-evaluated coefficients, and the best estimates for coefficients obtained by the two methods agreed within a factor of two. However, the 95% confidence intervals for the field-determined values were too broad to provide a sensitive check on the validity of the laboratory-measured values. The low precision of the field-determined coefficients was attributed to the high variation of radon flux with time and surface location at the field site, a phenomenon reported by other investigators.

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In the laboratory, diffusion coefficients were evaluated by a steady-state diffusion method applied to soil samples, 0.02 m in area and 0.10 m in depth, with the same porosity and moisture content as in the field experiments. The diffusion coefficient measured for radon in the sand cover was  $0.091 \pm 0.007$  cm<sup>2</sup>/s at the 95% confidence level, whereas that for the clay was  $0.008 \pm 0.004$  cm<sup>2</sup>/s at the 95% confidence level. Both values were within the uncertainty ranges for the field-evaluated coefficients, and the best estimates for coefficients obtained by the two methods agreed within a factor of two. However, the 95% confidence intervals for the field-determined values were too broad to provide a sensitive check on the validity of the laboratory-measured values. The low precision of the field-determined coefficients was attributed to the high variation of radon flux with time and surface location at the field site, a phenomenon reported by other investigators.

## INTRODUCTION

Uranium-mill tailings continually release  $^{222}\text{Rn}$ , a gaseous, radioactive decay product of  $^{226}\text{Ra}$ , which enters the air-filled voids of the tailings and diffuses into the atmosphere. This gaseous radon transport or flux can be reduced by covering the tailings with soil. The soil increases the diffusion path of radon to the atmosphere and provides time for radioactive decay of  $^{222}\text{Rn}$  ( $T_{1/2} = 3.82\text{d}$ ) within the cover. The soil depths required to keep the flux or atmospheric concentration of radon below prescribed limits will be determined by calculation. These calculations require knowledge of the radon diffusion coefficient through the soil and its variation with moisture content and compaction.

The purpose of this study was to compare the diffusion coefficients of radon in soils measured under field conditions with those measured in the laboratory under identical conditions of soil-moisture content and porosity. The latter measurements can be made rapidly and conveniently with a cylindrical column of soil, 0.1 m in height, 0.02 m<sup>2</sup> in area, and weighing 3 to 4 kg<sup>(2,3)</sup>. The method involves sealing a prepared column of soil in a chamber containing a  $^{226}\text{Ra}$  source, which continually generates  $^{222}\text{Rn}$ . The radon diffusion coefficient is evaluated from the steady-state radon concentration in the bottom chamber, the predetermined radon flux escaping the source with no soil in place, and the depth of soil in the test column. Because of the small height and amount of soil used, however, the validity of these coefficients in calculating required cover depths for mill tailings was uncertain. As a result, it was considered prudent to evaluate some diffusion coefficients for larger-scale soil samples in the field and to compare them with laboratory-measured coefficients.

Field experiments were conducted at a mill-tailings disposal site and both a well-graded sand and an inorganic clay were tested as barriers to radon diffusion. The well-graded sand covered a 1.31-m<sup>2</sup> area of tailings to a depth of 114 cm and weighed 2.68 Mg. The inorganic clay covered another 1.31-m<sup>2</sup> area of tailings to a depth of 44 cm and weighed 0.91 Mg. Radon diffusion coefficients were evaluated from the radon fluxes emitted from bare and covered tailings and from the properties and depths of the soil covers. Radon fluxes were measured by two independent methods, one based upon the passive diffusion of radon into an open canister of charcoal and the other utilizing recirculating air to sweep the radon flux collected by an aluminum tent into a charcoal trap. Diffusion coefficients were evaluated from both types of flux measurements, and the results were compared with those obtained by the laboratory method on samples of the same soils under identical conditions of soil-moisture content and porosity.

## EXPERIMENTAL

### Field Test Site Preparation

Field tests were conducted on an inactive section of the tailings disposal site at the Dawn Mining Company's mill at Ford, Washington. The test facility consisted of six aluminum caissons, each 1.29 m in diameter and about 3 m high, placed vertically in a trench that was excavated in the tailings. The trench

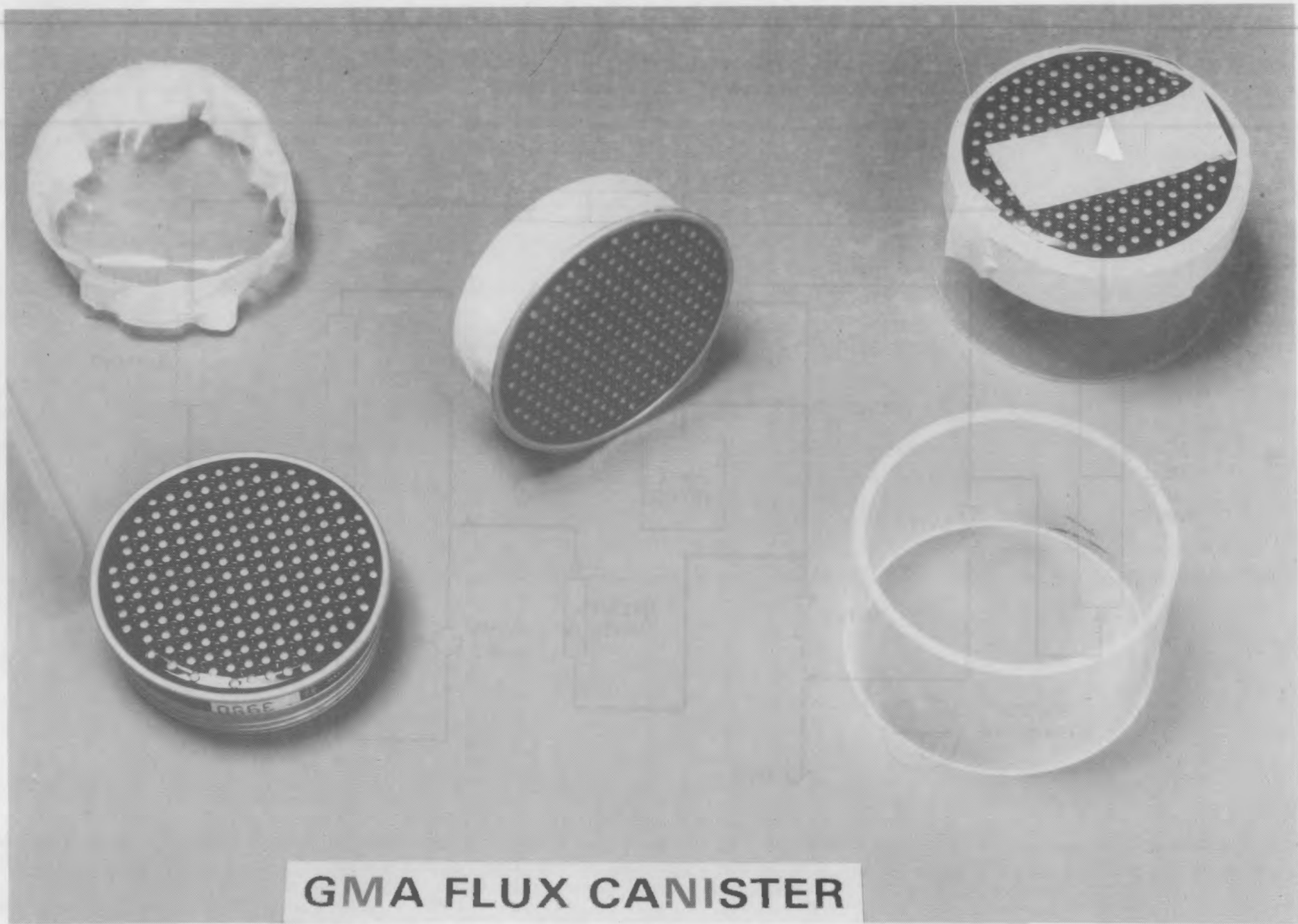
was then backfilled with tailings and compacted to replicate the surrounding landscape. Two meters of tailings material were then added in 20-cm lifts to the caissons and compacted to again replicate the undisturbed tailings. After sufficient time had elapsed to permit the radon to attain steady-state equilibrium, measurements were made to determine the radon flux from the bare tailings in each caisson. A cover of inorganic clay with low plasticity and weighing 911 kg was then applied in caisson No. 3 to a depth of 44 cm, using 8- to 10-cm lifts with compaction following each lift. A cover of well-graded sand weighing 2680 kg was applied in caisson No. 2 to a depth of 114 cm, utilizing the same compaction technique. After the field test, samples of the soils and tailings were packed in moisture-proof containers and taken to the laboratory for analysis. Physical properties of the tailings and each applied cover were determined by methods recommended by the American Society for Testing and Materials (ASTM).<sup>(4)</sup> The moisture content and specific gravity were determined by ASTM methods D2216 and D-854, respectively. The particle-size distributions of the soils were evaluated by ASTM methods D-422 and D-1140, and the soils were given engineering classifications according to ASTM method D-2487. The dry packing density was determined by taking a core of known volume and determining its dry mass.

#### Field Measurements of Radon Surface Flux

Surface fluxes of radon in the field were measured by two independent methods. These were 1) a passive charcoal-canister adsorption method, and 2) a tent method in which radon was removed from a recirculating air stream by adsorption on granular, activated charcoal.

The charcoal-canister method has been employed by several other investigators<sup>(5-8)</sup> and was reported to measure radon flux with a precision of  $\pm 15\%$ <sup>(7)</sup>. A commercial charcoal-filled gas mask canister (Mine Safety Appliance Company, Number GMA-459315) was used in this study. The larger end of the canister was covered with Tedlar® film, which is impervious to radon; the smaller end was pressed into a 7.5-cm diameter, 5-cm length of Lucite® cylinder and sealed with pressure-sensitive tape as seen in Figure 1. The assembled samplers were then placed on the soil surface and forced into the ground to a depth of from 1 to 2 cm to insure good contact. Each unit sampled an area of  $4.42 \times 10^{-3} \text{ m}^2$ . Several samplers were exposed simultaneously in each caisson for periods ranging from several hours to about 1 day. After exposure, the samplers were disassembled; and the charcoal canisters were removed, sealed in aluminum cans, and returned to the laboratory for radon analysis by gamma-ray spectrometry.

The tent method was developed at Pacific Northwest Laboratory,<sup>(8,9)</sup> and sampled a soil-surface area of  $0.225 \text{ m}^2$ . A pressure-balanced flow system recirculated air through an aluminum tent placed on the soil surface. Radon emitted from the soil under the tent was swept to an in-line trap of activated charcoal where it was adsorbed. The system is shown schematically in Figure 2. The U-shaped aluminum collection tent was sealed onto the soil surface, the pressure drop across the collection tent was balanced, and the system was flushed for a time to bring radon concentration within the tent to its steady-state value. Flow was then routed through the charcoal trap, and



**GMA FLUX CANISTER**

Figure 1. Steps in the Assembly of the GMA Radon Flux Canister

# REACTOR ASSEMBLY

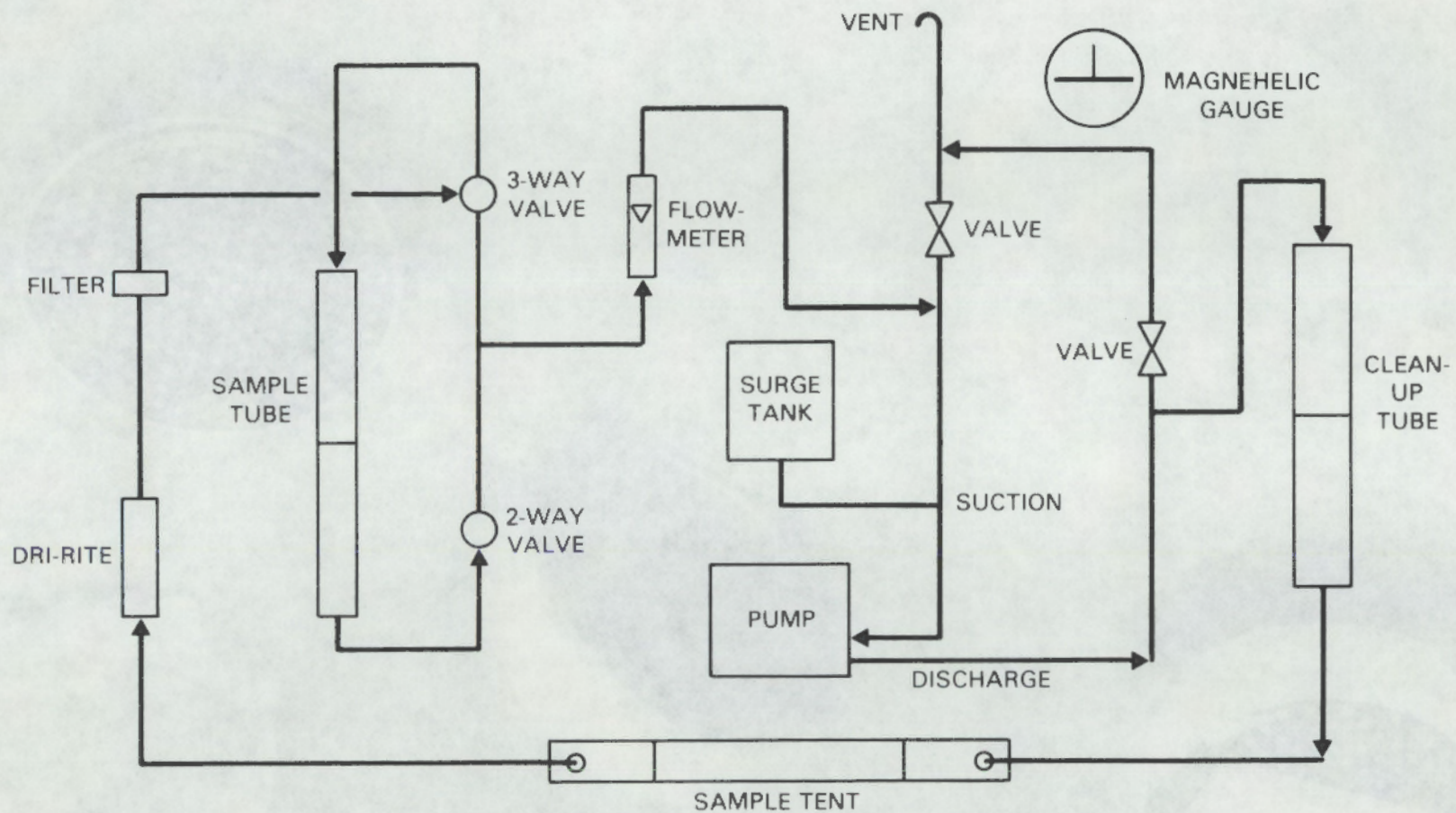


Figure 2. Schematic of Tent System Used to Measure Radon by a Pressure-balanced, Recirculating Pump System



radon was collected for 1/2 to 2 hours. Upon termination of sampling, the charcoal in the radon trap was transferred to a 2.5 x 15.2 cm diameter, plastic petri dish, which was then sealed with pressure-sensitive tape and returned to the laboratory for radon analysis.

The activity of  $^{222}\text{Rn}$  in each sealed sample of activated charcoal was measured by counting its  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  daughters with which it was in equilibrium. The counting rates were measured with multidimensional gamma-ray spectrometers that used NaI (Tl) detectors.<sup>(10)</sup> These spectrometers were calibrated with  $^{226}\text{Ra}$  standards that could be traced to NBS reference material and were contained in identical geometry as the samples to obtain a counter efficiency expressed as counts per disintegration. Radon fluxes were then calculated by the following equation:

$$J = \frac{C\lambda}{(2.22) EA(e^{-\lambda t_2}) (1-e^{-\lambda t_1})} \quad (\text{Equation 1})$$

where: J = radon flux (pCi/m<sup>2</sup>s)  
 C = net count rate (cpm)  
 λ = radon decay constant (s<sup>-1</sup>)  
 E = counter efficiency (counts/disintegration)  
 A = area sampled (m<sup>2</sup>)  
 t<sub>1</sub> = sampling period(s)  
 t<sub>2</sub> = time lapse between end of sampling and start of count(s).

Data were averaged for common collection periods from each caisson.

#### Evaluation of Radon Diffusion Coefficients by the Laboratory Method

Laboratory measurement of the diffusion coefficients for radon in tailings and the two cover soils was conducted with the Radon Attenuation Test Facility (RATF) described previously<sup>(2)</sup>. The facility employed columns of soil 0.1 m in depth and 1.54 x 10<sup>-2</sup>m<sup>2</sup> in area. Each sample was prepared to closely duplicate the moistures and compactions used for the field tests. The prepared soil columns were sealed over a constant source of  $^{222}\text{Rn}$  and allowed to equilibrate for 3 days to permit radon to attain its steady-state distribution throughout the bottom chambers and soil columns. The gas in the bottom chamber was then sampled and adsorbed onto activated charcoal, which was analysed for radon determination of the  $^{214}\text{Bi}$  daughter, using gamma-ray spectrometry.

Calculation of diffusion coefficients was accomplished using the following equation:

$$\frac{C_o}{J_o} = \frac{k (1-e^{-2kx})}{\lambda (1+e^{-2kx})} = \frac{k}{\lambda} \tanh(kx) \quad (\text{Equation 2})$$

where: C<sub>o</sub> = radon concentration in soil at the bottom of the column (pCi/cm<sup>3</sup>)  
 J<sub>o</sub> = radon flux with no soil in the facility (pCi/cm<sup>2</sup>s)  
 x = depth of soil (cm)  
 k = (λ/D)<sup>1/2</sup>

$\lambda$  =  $^{222}\text{Rn}$  decay constant ( $\text{s}^{-1}$ )

$D$  = diffusion coefficient for radon in the total pore space of bulk soil ( $\text{cm}^2/\text{s}$ ).

The quantity,  $C_o$  was evaluated from the radon gas concentration in the bottom chamber,  $C_g$ , by means of the equation<sup>(11)</sup>:

$$C_o = (P - 0.74\theta)C_g \quad (\text{Equation 3})$$

where:  $P$  = dry porosity of the soil (dimensionless)

$\theta$  = volume fraction moisture in the soil (dimensionless).

The diffusion coefficient,  $D$ , was then evaluated by iteration of Equation (2), using measured values for the other parameters. It is equivalent to the quantity "D/P" used in the equations presented in the Generic Environmental Impact Statement on Uranium Milling<sup>(1)</sup> prepared by the U.S. Nuclear Regulatory Commission.

#### Evaluation of Radon Diffusion Coefficients from Field Measurements

Radon diffusion coefficients for cover materials (as applied in the field test) were evaluated from the measured radon fluxes escaping the bare and covered tailings, using the following equation that describes flux attenuation as a function of the porosities of the tailings and cover soils and the diffusion coefficients for radon in these materials.<sup>(1)</sup>

$$J_c = \frac{2J_o \exp(-k_c x)}{\left[ 1 + \frac{P_t}{P_c} \left( \frac{D_t}{D_c} \right)^{1/2} \right] + \left[ 1 - \frac{P_t}{P_c} \left( \frac{D_t}{D_c} \right)^{1/2} \right] \exp(-2k_c x)} \quad (\text{Equation 4})$$

where:  $J_c$  = radon flux across the cover surface ( $\text{pCi}/\text{m}^2\text{s}$ )

$J_o$  = radon flux across the bare tailings surface ( $\text{pCi}/\text{m}^2\text{s}$ )

$D_t$  = diffusion coefficient for radon in the total pore space of bulk tailings ( $\text{cm}^2/\text{s}$ )

$D_c$  = diffusion coefficient for radon in the total pore space of bulk cover material ( $\text{cm}^2/\text{s}$ )

$P_t$  = dry porosity of tailings (dimensionless)

$P_c$  = dry porosity of cover material (dimensionless)

$x$  = cover depth (cm)

$k_c = (\lambda/D_c)^{1/2}$

$\lambda$  = radon decay constant ( $\text{s}^{-1}$ ).

The value for  $D_t$  could not be evaluated with Radon Attenuation Test Facility since the tailings themselves produced radon, and so did not meet the requirements of the measurement method. However, the diffusion coefficient for tailings was evaluated from other measured values by means of the equation<sup>(12)</sup>:

$$J_t = R_t E_t \rho_t (\lambda D_t)^{1/2} (10^{-4})^2 \tanh(x \lambda / D_t)^{1/2} \quad (\text{Equation 5})$$

where:  $J_t$  = radon flux from the tailings ( $\mu\text{Ci}/\text{m}^2\text{s}$ )  
 $R_t$  = specific activity of  $^{226}\text{Ra}$  in the tailings ( $\mu\text{Ci}/\text{g}$ )  
 $E_t$  = radon emanation coefficient of the tailings (dimensionless)  
 $\rho_t$  = dry packing density of the tailings ( $\text{g}/\text{cm}^3$ )  
 $D_t$  = diffusion coefficient for radon in the tailings ( $\text{cm}^2/\text{s}$ )  
 $\lambda$  = radon decay constant ( $\text{s}^{-1}$ )  
 $x$  = depth of tailings (cm)

In the field test,  $x > 300$  cm,  $D_t < 0.06$   $\text{cm}^2/\text{s}$  and  $\lambda = 2.1 \times 10^{-6} \text{s}^{-1}$  so that the quantity,  $\tanh(x^2 \lambda / D_t) \approx 1.00$ . Hence, Equation 5 reduces to:

$$J_t = R_t E_t \rho_t (\lambda D_t)^{1/2} (10^{-4})^2 \quad (\text{Equation 6})$$

or

$$D_t = \frac{1}{\lambda} \left( \frac{10^{-4} J_t}{R_t E_t \rho_t} \right)^2 \quad (\text{Equation 7})$$

The values for  $D_t$  were calculated from field measurements of  $J_t$  and  $\rho_t$ , gamma-ray spectrometric measurements of  $R_t$  (10), and the value,  $E_t = 0.28$ , estimated for uranium mill tailings containing a volume fraction moisture of 0.24 (13).

## RESULTS AND DISCUSSION

### Physical Properties of Tailings and Applied Cover Soils

Properties of the mill tailings and cover soils used in this investigation are listed in Table 1. They show that the two cover soils differed widely in type, moisture and compaction. Since soil-moisture content was only evaluated at the end of the field experiments, the values for weight fraction moisture and volume fraction moisture only apply with certainty at that time.

TABLE 1. Physical Properties of Tailings and Applied Covers

	<u>Field Samples</u>			<u>Laboratory Samples</u>	
	<u>Sand</u>	<u>Tailings</u>	<u>Clay</u>	<u>Sand</u>	<u>Clay</u>
Packing density, $\rho$ , ( $\text{g}/\text{cm}^3$ )	1.73	1.34	1.39	1.73	1.40
Soil-particle density, $d$ , ( $\text{g}/\text{cm}^3$ )	2.77	2.70	2.74	2.77	2.74
Dry porosity, $P = 1 - \rho/d$	0.38	0.50	0.49	0.38	0.49
Weight fraction moisture, $\omega$	0.04	0.18	0.14	0.04	0.14
Volume fraction moisture, $\theta = \rho \omega$	0.07	0.24	0.20	0.07	0.20

## Radon Flux Values from Field Measurements

Radon fluxes measured by the two methods are summarized in Table 2 and presented in detail in Appendices A and B. The values shown in Table 2 for fluxes obtained by the charcoal-canister method are averages of the several values measured simultaneously in each designated caisson. The individual measurements differed by factors of up to 3 during the same sampling period in the same caisson. Still larger variations in flux, ranging up to factors of 35, were found at the same caisson on different days. Radon fluxes measured by the tent method varied by factors of up to 15. This lack of precision was attributed to possible differences in meteorological conditions, changes in soil moisture and variations in the collection efficiencies of the two measurements.

TABLE 2. Radon Fluxes Evaluated by the Charcoal-Canister (C) and the Tent (T) Methods in the Field (All Values in  $\text{pCi}/\text{m}^2\text{s}$ )

Date (m/d/y)	Caisson											
	1		2		3		4		5		6	
	C	T	C	T	C	T	C	T	C	T	C	T
5/11/81	465	90	435	750	280	350	410	720	200	--	160	--
5/12/81	320	100	330	280	450	270	360	330	130	200	185	135
5/12-5/13/81	200	--	390	--	220	--	380	--	110	--	280	--
5/13/81	270	90	770	700	230	200	330	450	120	310	860	530
6/16/81	2750	790	680	1660	1110	160	650	890	840	750	4300	410
6/16-6/17/81	500	--	820	--	410	--	420	--	340	--	240	--
6/17/81*	80	60	90	200	350	350	150	380	60	50	60	90
8/5/81	160	110	30	20	155	150	270	220	120	110	90	170
9/2/81	160	140	65	4	200	120	290	340	135	170	80	100
9/3/81	320	--	60	--	180	--	290	--	110	--	190	--
11/9/81	--	280	70	80	30	150	--	340	--	310	--	560
11/9-11/10/81	250	--	97	--	43	--	210	--	190	--	500	--
11/10/81	--	230	270	140	60	150	400	420	--	490	--	1120
11/10-11/11/81	400	--	230	--	70	--	315	--	450	--	1420	--
11/11/81	810	--	380	--	110	--	620	--	1240	--	1690	--

\* After the measurements on this date, a 114-cm cover of well-graded sand was applied in caisson No. 2 and a 44-cm cover of inorganic clay was applied in caisson No. 3.

The data can be logically divided into two subsets, those measured on or before June 17, 1981 and those measured after that date when the cover of sand was in place in caisson No. 2 and the cover of clay was in place in caisson No. 3. An analysis of variance among the flux measurements made before June 17, 1981 showed no significant difference between caissons or between

flux-measurement methods. However, there was a highly significant difference in flux between dates of sampling. Similarly, an analysis of variance among the flux measurements after June 17, 1981 showed no significant difference between the uncovered caissons or between flux-measurement methods, but highly significant differences in flux between dates of sampling and between covered versus uncovered caissons. It was concluded that the diffusion coefficients for radon in the cover soils should be evaluated from Equation 4 using measurements of flux from bare and covered tailings for the same dates.

#### Values for Radon Diffusion Coefficients

Since soil moisture is an important factor in determining the diffusion coefficient of radon in a soil, only data taken under conditions of known moisture content were used in the evaluations. This limited the evaluation of diffusion coefficients from field measurements of radon flux to data collected during the period from November 9 to 11, 1981. These data and the computed diffusion coefficients are listed in Table 3.

TABLE 3. Evaluations of Radon Diffusion Coefficients from Field Data

Date (m/d/y)	Average Flux(pCi/m <sup>2</sup> s)			Diffusion Coefficients (cm <sup>2</sup> /s)		
	Bare Tailings	Sand Cover	Clay Cover	Bare Tailings	Sand Cover	Clay Cover
11/9/81	372	75	90	0.018	0.011	0.0037
11/9- 11/10/81	288	97	43	0.011	0.017	0.0019
11/10/81	532	205	105	0.038	0.028	0.0038
11/10- 11/11/81	646	230	70	0.056	0.029	0.0024
11/11/81	1090	380	110	0.158	0.040	0.0034

The values for the radon fluxes from bare tailings are the averages of all flux measurements in the four uncovered caissons, on the specified date. The diffusion coefficients for radon in bare tailings were calculated from Equation 7, the data shown in Tables 1 and 3 and the measured specific activity,  $R_t = 504$  pCi/g, for radium in the tailings. The diffusion coefficient in tailings measured for November 11, 1981 is clearly too high as it exceeds the coefficient in air,  $0.11$  cm<sup>2</sup>/s<sup>(14)</sup>. Nevertheless, it was used to evaluate the diffusion coefficients of radon in the cover soils on that date since all flux values were observed to be high at that time. Available meteorological data at the test site during this time period are shown in Table 4. They did not

TABLE 4. Meteorological Parameters at the Test Site From November 9 to 11, 1981

Date (m/d/y)	Air Temperature		Air Pressure	Precipitation	Max. Wind Velocity
	Max. °C	Min. °C	(mm mercury)	(mm.)	(km/hr, direction)
11/9/81	9	-8	772.7	0.6	0, ESE
11/10/81	4	-3	771.4	0.2	4, N
11/11/81	3	-3	768.2	0.4	2, WNW

suggest reasons for the variations in radon flux but may overlook short-term changes in air-temperature, atmospheric pressure or wind velocity during a day.

The diffusion coefficients evaluated from field data and those measured in the laboratory are compared in Table 5.

TABLE 5. Comparison of Field-Measured Radon Diffusion Coefficients with Laboratory-Measured Coefficients (95% Confidence Limits)

<u>Method</u>	<u>D (Sand Cover)</u>	<u>D (Clay Cover)</u>
Laboratory	0.021 ± 0.002	0.0036 ± 0.0004
Field	0.03 ± 0.03	0.003 ± 0.002

The values are presented as 95% confidence limits based on the data shown in Table 3 and on a relative standard deviation of ±5% for the laboratory method<sup>(10)</sup>. The best estimates of the diffusion coefficients evaluated by the two methods agreed to within a factor of two for both soil covers. This may be fortuitous, however, in view of the large 95% confidence intervals for the coefficients evaluated from field data. In any case, the diffusion coefficients evaluated by the two methods for each soil are indistinguishable at the 95% confidence level.

The low precision in the diffusion coefficients evaluated from field data were attributed to high variation in the radon flux with time and surface location at the field site. These variations occurred over relatively short time intervals and with only small differences in surface location of the radon collectors. Their exact cause is unknown, but variations of similar magnitude have been reported by other investigators for other tailings sites<sup>(5,15)</sup>. The higher precision of the laboratory-measured coefficients is attributed to the ability to better control influential parameters such as soil-moisture content and compaction as well as being relatively free from changes in meteorological conditions.

#### CONCLUSIONS

Laboratory-measured diffusion coefficients for radon in relatively small quantities of two widely different types of soils were indistinguishable, at the 95% confidence level, from coefficients evaluated from field measurements on much larger quantities of the same soils covering uranium mill tailings. The best estimates for coefficients obtained by the two methods agreed within a factor of two; however, the 95% confidence intervals for the field-determined values were too broad to provide a sensitive check on the validity of the laboratory-measured values. The low precision of the field-determined values was attributed to high variation in radon flux with time and surface location at the field site.

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APPENDIX A

Data Obtained With GMA Radon Flux Canisters

Area sampled was  $4.42 \times 10^{-3} \text{m}^2$

## GMA Radon Flux Canisters

## Caisson 1

Sample	On		Exposure		$\Delta t$ (min.)	Counted		(dpm)* Sample	J (pCi) (m <sup>2</sup> ·sec)
	Date	Time	Date	Time		Date	Time		
1A	5/11/81	1309	5/11/81	1810	301	5/14/81	1348	45320±500	260
1B	5/11/81	1309	5/11/81	1810	301	5/14/81	1348	115610±1200	670
1C	5/12/81	0728	5/12/81	1248	320	5/14/81	1410	74841±830	400
1D	5/12/81	1248	5/12/81	1800	312	5/14/81	1423	42726±435	240
1E	5/12/81	1802	5/13/81	0747	825	5/14/81	1457	92810±1200	200
1F	5/13/81	0747	5/13/81	1418	391	5/14/81	1445	61162±710	270
1A	6/16/81	1117	6/16/81	1710	353	6/18/81	1521	558000±4400	2750
1B	6/16/81	1710	6/17/81	0735	865	6/18/81	1536	240700±2100	500
1C	6/17/81	0735	6/17/81	1542	487	6/18/81	1549	22460±250	80
1A	8/5/81	0827	8/5/81	1515	408	8/6/81	1443	37100±160	160
1A	9/2/81	0756	9/3/81	0822	1466	9/4/81	1136	125560±1420	160
1B	9/3/81	0823	9/3/81	1140	197	9/4/81	1048	35782±600	320
1A	11/9/81	0906	11/10/81	1251	1665	11/14/81	0657	220317±1260	250
1B	11/10/81	1252	11/11/81	1014	1282	11/14/81	1511	275870±2220	400
1C	11/11/81	1014	11/11/81	1427	253	11/14/81	1513	118490±940	810

\* Decay corrected to end of sampling.

## GMA Radon Flux Canisters

## Caisson 2

Sample	Exposure					Counted		(dpm)* Sample	J (pCi) (m <sup>2</sup> ·sec)
	On		Off		Δt (min.)	Date	Time		
	Date	Time	Date	Time					
2A	5/11/81	1310	5/11/81	1810	300	5/14/81	1348	65690±690	380
2B	5/11/81	1310	5/11/81	1810	300	5/14/81	1354	84198±1030	490
2C	5/12/81	0728	5/12/81	1250	322	5/14/81	1411	61233±710	330
2D	5/12/81	1250	5/12/81	1800	310	5/14/81	1430	59314±780	330
2E	5/12/81	1802	5/13/81	0747	825	5/14/81	1459	178500±1900	390
2F	5/13/81	0747	5/13/81	1418	391	5/14/81	1447	172070±1660	770
2A	6/16/81	1118	6/16/81	1710	352	6/18/81	1524	137500±1300	680
2B	6/16/81	1710	6/17/81	0735	865	6/18/81	1538	398100±3100	820
2C	6/17/81	0735	6/17/81	0932	117	6/18/81	1551	6433±120	90
2A	8/5/81	0827	8/5/81	1515	408	8/6/81	1436	6390±40	30
2A	9/2/81	0754	9/2/81	2035	761	9/4/81	1034	2598±68	6
2B	9/2/81	0754	9/2/81	2035	761	9/4/81	1053	2802±69	7
2C	9/3/81	0749	9/3/81	1122	213	9/4/81	1034	7892±169	60
2A	11/9/81	0910	11/9/81	1655	465	11/14/81	0707	17220±230	70
2B	11/9/81	0910	11/10/81	1235	1645	11/14/81	0700	84070±770	90
2C	11/9/81	0910	11/10/81	1235	1645	11/14/81	0700	90460±730	100
2D	11/9/81	1655	11/10/81	1235	1180	11/14/81	0708	67212±798	100
2E	11/10/81	1236	11/10/81	1714	278	11/14/81	0710	42617±610	270
2F	11/10/81	1236	11/11/81	1017	1301	11/14/81	1514	170240±1080	240
2G	11/10/81	1236	11/11/81	1017	1301	11/14/81	0716	127380±1310	180
2H	11/10/81	1713	11/11/81	1017	1024	11/14/81	0717	151690±1310	270
2I	11/11/81	1017	11/11/81	1424	247	11/14/81	0721	50510±410	350
2J	11/11/81	1017	11/11/81	1424	247	11/14/81	0723	52540±410	370
2K	11/11/81	1017	11/11/81	1424	247	11/14/81	0725	59797±590	420

\* Decay corrected to end of sampling.

## GMA Radon Flux Canisters

## Caisson 3

Sample	On		Exposure Off		$\Delta t$ (min.)	Counted		(dpm)* Sample	J (pCi) (m <sup>2</sup> ·sec)
	Date	Time	Date	Time		Date	Time		
3A	5/11/81	1520	5/11/81	1826	186	5/14/81	1356	30196±350	280
3B	5/12/81	0730	5/12/81	1252	202	5/14/81	1416	59760±780	510
3C	5/12/81	1252	5/12/81	1804	312	5/14/81	1432	69640±780	390
3D	5/12/81	1805	5/13/81	0745	820	5/14/81	1500	98600±600	220
3E	5/13/81	0745	5/13/81	1417	392	5/14/81	1449	51650±620	230
3A	6/16/81	1117	6/16/81	1710	353	6/18/81	1529	226200±2200	1110
3B	6/16/81	1710	6/17/81	0735	865	6/18/81	1540	199100±2000	410
3C	6/17/81	0735	6/17/81	1145	250	6/18/81	1555	50720±500	350
3A	8/5/81	0827	8/5/81	1515	408	8/6/81	1448	31380±150	140
3B	8/5/81	0827	8/5/81	1515	408	8/6/81	1454	40143±165	170
3A	9/2/81	0803	9/2/81	2035	752	9/4/81	1119	80830±870	190
3B	9/2/81	0803	9/2/81	2035	752	9/4/81	1115	88401±1220	210
3C	9/3/81	0820	9/3/81	1122	182	9/4/81	1140	19256±290	180
3A	11/9/81	0912	11/9/81	1655	973	11/13/81	1325	15937±227	30
3B	11/9/81	0912	11/10/81	1242	1650	11/13/81	1325	48698±465	50
3C	11/9/81	0912	11/10/81	1242	1650	11/13/81	1325	63099±690	70
3D	11/9/81	1655	11/10/81	1242	1187	11/13/81	1336	8043±64	10
3E	11/10/81	1241	11/10/81	1715	274	11/14/81	1521	10007±188	60
3F	11/10/81	1241	11/11/81	1020	1299	11/14/81	1515	12644±200	20
3G	11/10/81	1241	11/11/81	1020	1299	11/13/81	1339	43899±152	60
3H	11/10/81	1712	11/11/81	1020	1028	11/13/81	1341	70661±190	130
3I	11/11/81	1022	11/11/81	1424	242	11/13/81	1501	2789±26	20
3J	11/11/81	1022	11/11/81	1424	242	11/13/81	1504	10163±48	70
3K	11/11/81	1022	11/11/81	1424	242	11/13/81	1505	33384±84	240

\* Decay corrected to end of sampling.

## GMA Radon Flux Canisters

## Caisson 4

Sample	On		Exposure Off		$\Delta t$ (min.)	Counted		(dpm)* Sample	$\frac{J}{(m^2 \cdot sec)}$ (pCi)
	Date	Time	Date	Time		Date	Time		
4A	5/11/81	1523	5/11/81	1827	184	5/14/81	1358	43710±445	410
4B	5/12/81	0730	5/12/81	1252	202	5/14/81	1417	39634±610	340
4C	5/12/81	1252	5/12/81	1804	312	5/14/81	1433	67874±730	380
4D	5/12/81	1805	5/13/81	0745	820	5/14/81	1436	173000±2000	380
4E	5/13/81	0745	5/13/81	1417	392	5/14/81	1451	73878±770	330
4A	6/16/81	1118	6/16/81	1710	352	6/18/81	1530	132200±1340	650
4B	6/16/81	1710	6/17/81	0735	865	6/18/81	1541	204100±2000	420
4C	6/17/81	0735	6/17/81	1542	487	6/18/81	1603	41070±400	150
4A	8/5/81	0827	8/5/81	1515	408	8/6/81	1438	62814±260	270
4A	9/2/81	0801	9/3/81	0818	1457	9/4/81	1121	226751±2470	290
4B	9/3/81	0818	9/3/81	1122	184	9/4/81	1128	31526±360	290
4A	11/9/81	0906	11/10/81	1248	1662	11/14/81	1458	184798±1330	210
4B	11/10/81	1249	11/10/81	1714	265	11/14/81	1458	61200±651	400
4C	11/10/81	1249	11/11/81	1027	1298	11/14/81	1458	222518±1820	320
4D	11/10/81	1713	11/11/81	1027	1024	11/14/81	1504	175404±1193	310
4E	11/11/81	1028	11/11/81	1426	238	11/11/81	1505	71675±777	520
4F	11/11/81	1028	11/11/81	1426	238	11/11/81	1507	98995±905	720

\* Decay corrected to end of sampling.

## GMA Radon Flux Canisters

## Caisson 5

Sample	On		Exposure Off		$\Delta t$ (min.)	Counted		(dpm)* Sample	$\frac{J}{(m^2 \cdot sec)}$ (pCi)
	Date	Time	Date	Time		Date	Time		
5A	5/11/81	1528	5/11/81	1830	182	5/14/81	1359	21080±300	200
5B	5/12/81	0848	5/12/81	1253	245	5/14/81	1419	15095±240	110
5C	5/12/81	1254	5/12/81	1806	312	5/14/81	1438	27460±470	150
5D	5/12/81	1807	5/13/81	0747	813	5/14/81	1502	51097±430	110
5E	5/13/81	0743	5/13/81	1416	393	5/14/81	1453	48819±350	220
5A	6/16/81	1117	6/16/81	1710	353	6/18/81	1532	170300±1600	840
5B	6/16/81	1710	6/17/81	0735	865	6/18/81	1543	163900±1200	340
5C	6/17/81	0735	6/17/81	1542	487	6/18/81	1604	18090±260	60
5A	8/5/81	0827	8/5/81	1515	408	8/6/81	1429	28017±120	120
5A	9/2/81	0751	9/3/81	0814	1463	9/4/81	1045	101192±1240	130
5B	9/2/81	0751	9/3/81	0814	1463	9/4/81	1048	105670±1343	140
5C	9/3/81	0815	9/3/81	1122	187	9/4/81	1123	11511±170	110
5A	11/9/81	0906	11/10/81	1247	1661	11/14/81	1523	171864±2080	190
5B	11/10/81	1247	11/11/81	1030	1303	11/14/81	1527	320419±1013	450
5C	11/11/81	1030	11/11/81	1426	236	11/14/81	1530	169771±2272	1240

\* Decay corrected to end of sampling.

## GMA Radon Flux Canisters

## Caisson 6

Sample	On		Exposure Off		$\Delta t$ (min.)	Counted		(dpm)* Sample	$\frac{J}{(m^2 \text{ sec})}$ (pCi)
	Date	Time	Date	Time		Date	Time		
6A	5/11/81	1530	5/11/81	1830	180	5/14/81	1408	16350±270	160
6B	5/12/81	0848	5/12/81	1253	245	5/14/81	1421	17830±285	130
6C	5/12/81	1254	5/12/81	1806	312	5/14/81	1439	43073±440	240
6D	5/12/81	1807	5/13/81	0743	816	5/14/81	1441	127500±1000	280
6E	5/13/81	0743	5/13/81	1416	393	5/14/81	1455	192584±1970	860
6A	6/16/81	1118	6/16/81	1710	352	6/18/81	1535	866000±5500	4300
6B	6/16/81	1710	6/17/81	0735	865	6/18/81	1545	118100±950	240
6C	6/17/81	0735	6/17/81	1542	487	6/18/81	1606	17260±220	60
6A	8/5/81	0827	8/5/81	1515	408	8/6/81	1431	21827±100	90
6A	9/2/81	0818	9/3/81	0813	1445	9/4/81	1115	62569±729	80
6B	9/3/81	0814	9/3/81	1122	188	9/4/81	6.3	21093±325	190
6A	11/9/81	0906	11/10/81	1246	1560	11/14/81	1532	439941±4592	500
6B	11/10/81	1245	11/11/81	1031	1306	11/14/81	1533	1009065±6889	1420
6C	11/11/81	1031	11/11/81	1426	235	11/14/81	1534	230393±2427	1690

\* Decay corrected to end of sampling.

APPENDIX B

Data Obtained With Radon Collection Tents

Area sampled was 0.225 m<sup>2</sup>



Radon Tent Samplers

Caisson 1

Sample	On		Exposure Off		$\Delta t$ (min.)	Counted		(dpm)* Sample	J (pCi) (m <sup>2</sup> ·sec)
	Date	Time	Date	Time		Date	Time		
C1-1	5/11/81	1305	5/11/81	1505	120	5/14/81	1544	369900±2800	90
C1-2	5/12/81	1031	5/12/81	1231	120	5/14/81	1558	418300±3400	100
C1-3	5/13/81	1230	5/13/81	1400	90	5/14/81	1615	293100±2400	90
C1-1	6/16/81	1126	6/16/81	1226	60	6/18/81	1422	1610000±7300	790
C1-2	6/17/81	0902	6/17/81	1002	60	6/18/81	1453	123200±1410	60
C1-1	8/5/81	1147	8/5/81	1247	60	8/6/81	1417	227430±500	110
C1-1	9/2/81	1528	9/2/81	1628	60	9/4/81	1147	283370±2580	140
C1-1	11/9/81	1421	11/9/81	1533	72	11/16/81	1230	675150±3260	280
C1-2	11/10/81	1828	11/10/81	1935	67	11/16/81	1239	517180±4020	230

\* Decay corrected to end of sampling.

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# Radon Tent Samplers

## Caisson 2

Sample	On		Exposure Off		$\Delta t$ (min.)	Counted		(dpm)* Sample	J (pCi) (m <sup>2</sup> ·sec)
	Date	Time	Date	Time		Date	Time		
C2-1	5/11/81	1305	5/11/81	1505	120	5/14/81	1546	3018000±13000	750
C2-2	5/12/81	1031	5/12/81	1231	120	5/14/81	1600	1135000±6800	280
C2-3	5/13/81	1230	5/13/81	1400	90	5/14/81	1617	2111000±12000	700
C2-1	6/16/81	1126	6/16/81	1226	60	6/18/81	1428	3370000±16000	1660
C2-2	6/17/81	0831	6/17/81	0902	31	6/18/81	1457	205800±2090	200
C2-1	8/5/81	0947	8/5/81	1047	60	8/6/81	1419	42581±140	20
C2-1	9/2/81	0936	9/2/81	1041	65	9/4/81	1153	6977±180	4
C2-1	11/9/81	1125	11/9/81	1251	86	11/16/81	1232	223490±1920	80
C2-2	11/10/81	1828	11/10/81	1935	67	11/16/81	1240	316650±2870	140

\* Decay corrected to end of sampling.

B-2

Caisson 2  
returned that day

Radon Tent Samplers

Caisson 3

Sample	On		Exposure Off		$\Delta t$ (min.)	Counted		(dpm)* Sample	J (pCi) (m <sup>2</sup> ·sec)
	Date	Time	Date	Time		Date	Time		
C3-1	5/11/81	1807	5/11/81	2007	120	5/14/81	1548	1397000±7600	350
C3-2	5/12/81	1330	5/12/81	1530	120	5/14/81	1601	1077000±5900	270
C3-3	5/13/81	1025	5/13/81	1155	90	5/14/81	1611	598300±3800	200
C3-1	6/16/81	1330	6/16/81	1430	60	6/18/81	1433	332400±3160	160
C3-2	6/17/81	1044	6/17/81	1144	60	6/18/81	1506	711500±3750	350
C3-1	8/5/81	0947	8/5/81	1048	61	8/6/81	1245	318640±650	150
C3-1	9/2/81	0936	9/2/81	1041	65	9/4/81	1158	258200±3820	120
C3-1	11/9/81	1125	11/9/81	1251	86	11/16/81	1234	442721±2870	150
C3-2	11/10/81	1639	11/10/81	1739	60	11/16/81	1243	313340±3000	150

\* Decay corrected to end of sampling.

B-3

Radon Tent Samplers

Caisson 4

Sample	On		Exposure Off		$\Delta t$ (min.)	Counted		(dpm)* Sample	J (pCi) (m <sup>2</sup> ·sec)
	Date	Time	Date	Time		Date	Time		
C4-1	5/11/81	1807	5/11/81	2007	120	5/14/81	1550	2911000±13000	720
C4-2	5/12/81	1330	5/12/81	1530	120	5/14/81	1603	1320000±7200	330
C4-3	5/13/81	1025	5/13/81	1155	90	5/14/81	1613	1381000±7600	450
C4-1	6/16/81	1330	6/16/81	1430	60	6/18/81	1441	1798000±9100	890
C4-2	6/17/81	1103	6/17/81	1144	41	6/18/81	1510	777700±4500	380
C4-1	8/5/81	1147	8/5/81	1247	60	8/6/81	1422	448400±980	220
C4-1	9/2/81	1528	9/2/81	1628	60	9/4/81	1200	695340±4680	340
C4-1	11/9/81	1421	11/9/81	1533	72	11/16/81	1251	827570±5250	340
C4-2	11/10/81	1639	11/10/81	1739	60	11/16/81	1245	853770±8370	420

\* Decay corrected to end of sampling.

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Radon Tent Samplers

Caisson 5

Sample	On		Exposure Off		$\Delta t$ (min.)	Counted		(dpm)* Sample	J (pCi) (m <sup>2</sup> ·sec)
	Date	Time	Date	Time		Date	Time		
C5-1	5/12/81	0736	5/12/81	0939	123	5/14/81	1553	667300±3800	170
C5-2	5/12/81	1615	5/12/81	1815	120	5/14/81	1605	944400±5200	230
C5-3	5/13/81	0820	5/13/81	0950	90	5/14/81	1608	943000±5300	310
C5-1	6/16/81	1523	6/16/81	1623	60	6/18/81	1443	1526400±7600	750
C5-2	6/17/81	1245	6/17/81	1346	61	6/18/81	1512	102100±1240	50
C5-1	8/5/81	1337	8/5/81	1437	60	8/6/81	1425	220840±580	110
C5-1	9/2/81	1738	9/2/81	1854	76	9/4/81	1202	422560±3440	170
C5-1	11/9/81	1619	11/9/81	1726	67	11/16/81	1254	692500±3380	310
C5-2	11/10/81	1445	11/10/81	1545	60	11/16/81	1247	985740±7390	490

\* Decay corrected to end of sampling.

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Radon Tent Samplers

Caisson 6

Sample	On		Exposure Off		$\Delta t$ (min.)	Counted		(dpm)* Sample	J (pCi) (m <sup>2</sup> ·sec)
	Date	Time	Date	Time		Date	Time		
C6-1	5/12/81	0736	5/12/81	0939	123	5/14/81	1553	203100±1900	50
C6-2	5/12/81	1615	5/12/81	1815	120	5/14/81	1607	886500±5800	220
C6-3	5/13/81	0820	5/13/81	0950	90	5/14/81	1610	1595000±10000	530
C6-1	6/16/81	1523	6/16/81	1623	60	6/18/81	1449	837400±5400	410
C6-2	6/17/81	1235	6/17/81	1335	60	6/18/81	1516	170500±2000	90
C6-1	8/5/81	1337	8/5/81	1438	61	8/6/81	1427	347710±660	170
C6-1	9/2/81	1738	9/2/81	1854	76	9/4/81	1204	258460±2040	100
C6-1	11/9/81	1619	11/9/81	1726	67	11/16/81	1256	1259980±5310	560
C6-2	11/10/81	1445	11/10/81	1545	60	11/16/81	1249	2259010±12750	1120

\* Decay corrected to end of sampling.

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