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Comparison of Radon Diffusion Coefficients Measured by Transient-Diffusion and Steady-State Laboratory Methods

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Operated by
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Rogers & Associates Engineering Corporation

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Office of Nuclear Regulatory Research
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SUMMARY

A method was developed and used to determine radon diffusion coefficients in compacted soils by transient-diffusion measurements. A relative standard deviation of 12 percent was observed in repeated measurements with a dry soil by the transient-diffusion method, and a 40 percent uncertainty was determined for moistures exceeding 50 percent of saturation. Excellent agreement was also obtained between values of the diffusion coefficient for radon in air, as measured by the transient-diffusion method, and those in the published literature.

Good agreement was also obtained with diffusion coefficients measured by a steady-state method on the same soils. The agreement was best at low moistures, averaging less than ten percent difference, but differences of up to a factor of two were observed at high moistures. The comparison of the transient-diffusion and steady-state methods at low moistures provides an excellent verification of the theoretical validity and technical accuracy of these approaches, which are based on completely independent experimental conditions, measurement methods and mathematical interpretations.

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INTRODUCTION

A new method has been developed and tested for measuring radon gas diffusion coefficients. The method is based on measurement of the non-equilibrium or transient movement of radon through a sample material, rather than on the more traditional steady-state transport of radon through the sample. The present application and evaluation of this method was conducted as part of a larger research and development project aimed at reducing radon emissions from uranium mill tailings piles. This project is being conducted for the U.S. Nuclear Regulatory Commission under a subcontract with Pacific Northwest Laboratory (PNL).^(a)

Due to the potential public health hazards from atmospheric radon (^{222}Rn) and its decay products, it is important to minimize its release into the atmosphere. Uranium mill tailings produce radon at nearly constant rates over periods of thousands of years; therefore permanent covers are being sought for tailings piles to reduce the fraction of the radon gas which reaches the atmosphere. The short (3.8-day) half-life of radon allows it to decay appreciably in the cover as long as its diffusion time through the cover is several days or longer. The radon diffusion coefficients of soils and other potential cover materials are therefore necessary to choose the proper tailings cover thickness and other design parameters to minimize radon release.

The present transient-diffusion measurement technique was developed and tested for two purposes. First, it could potentially provide improved capabilities over many existing methods, including lower cost, higher precision,

^(a) Operated by Battelle Memorial Institute

of both methods. Second, it could potentially provide improved capabilities over many existing methods, including lower cost, higher precision, shorter experiment time requirements and greater laboratory versatility.

The capabilities of the present transient-diffusion system are attractive in comparison with many traditional diffusion measurements. Typical equilibration times for large soil test columns in previous work⁽¹⁻³⁾ were one to two months or longer, and sample requirements were often on the order of hundreds of kilograms or more. Smaller-scale diffusion experiments have been proposed⁽⁴⁾ and recently developed and tested.⁽⁵⁾ These were equilibrium diffusion measurements, and typically utilized only a few kilograms of sample material. Because of the small sample size, equilibrium was quickly achieved (~ 3 days). The present transient measurements utilize samples of similar size, and can be completed over time intervals of one to two days for diffusion coefficients as low as $10^{-4} \text{ cm}^2/\text{s}$. Continuous data collection for the transient measurement provides high precision as well as a monitor of experimental variability.

Comparison of transient-diffusion coefficients for radon with those from steady-state measurements on the same materials is important for two reasons. First, agreement between these two independent measurements provides a check on their theoretical validity and their technical accuracy. Second, the nature of the diffusion process can be examined in greater detail. Steady-state diffusion measurements yield an effective radon diffusion coefficient which includes the effects of all experimental variables and mechanisms, such as soil structure and moisture effects, absorption and adsorption effects, temperature and pressure effects, and advective transport. The transient-diffusion measurements can potentially provide an extra degree of freedom in understanding the diffusion process by illustrating the effects of any parameters, such as absorption of radon by water, which may have very different time constants than the radon diffusion process.

The following sections compare the experimental parameters for transient and steady-state diffusion measurements, and describe the experimental details of sample preparation, data acquisition, system calibration, and data interpretation. The results of transient-diffusion measurements on natural soils are also presented and compared with steady-state measurements on the same soils. Transient measurements on several reference materials are also reported and discussed in terms of the precision and accuracy of the method.

PARAMETERS FOR RADON DIFFUSION MEASUREMENTS

Radon diffusion coefficients for homogeneous materials are usually measured by application of a radon concentration gradient across a sample and measurement of the resulting response in terms of steady-state radon flow, steady-state concentration gradients, or transient radon accumulation. For simplicity of interpretation, the experiments are designed so that one-dimensional diffusion equations are applicable, and occasionally, so that only one diffusion region needs to be considered. Although only the region defined by the sample is strictly of interest, it is often necessary to consider the air-filled source or detection regions at either end of the sample region to adequately interpret the experimental data.

Four main parameters can be measured in a radon diffusion experiment. Two of these four are generally adequate for the diffusion coefficient calculation. The four parameters are (a) the initial radon flux from the bare radon source, (b) the radon flux from the exit end of the sample column, (c) the radon concentration at the entrance end of the column and (d) the radon concentration at the exit end of the column.

Steady-state radon diffusion measurements have been conducted using parameters (a) and (b), parameters (a) and (c), and parameters (c) and (d). The

steady-state method used in the present comparisons with the transient technique utilized parameters (a) and (c). As indicated in References 4 and 5, the radon diffusion coefficient from these parameters can generally be determined from the one-region, one-dimensional equation

$$\frac{C_o}{J_o} = \frac{\tanh(k_s b)}{k_s D_e} \quad (1)$$

where

C_o = radon concentration in the pore space at the column entrance (parameter c) (pCi/cm³)

J_o = radon flux from bare source (parameter a) (pCi/cm²s)

k_s = $(\lambda P_s / D_e)^{1/2}$ (cm⁻¹)

λ = radon decay constant (2.1×10^{-6} s⁻¹)

P_s = soil porosity

D_e = effective diffusion coefficient of radon in the bulk soil (cm²/s)

b = thickness of soil layer (cm)

Equation (1) differs slightly from those reported in References 4 and 5 due to the present definition of C_o .

For the steady-state measurements of low diffusion coefficients, the volume of the source region beneath the sample becomes important due to radon decay in the source region. For general steady-state diffusion measurements, Equation (1) should be replaced by the two-region, one-dimensional equation

$$\frac{C_o}{J_o} = \frac{\tanh(k_s b)}{k_s D_e \cosh(k_A a) + k_A D_A \sinh(k_A a) \tanh(k_s b)} \quad (2)$$

where

k_A = $(\lambda / D_A)^{1/2}$ (cm⁻¹)

D_A = diffusion coefficient of radon in air (cm^2/s)

a = thickness of the air-filled source region (cm)

A similar phenomenon occurs in the transient diffusion measurement system. In this case, parameters (c) and (d) are used to determine the diffusion coefficient, with parameter (d) being measured continuously with time. There are two other experimental differences between the present equilibrium and transient measurements. First, the radon source concentration is maintained constant in the transient system instead of the source radon flux being constant. Second, the radon concentration at the exit end of the column increases with time in the sealed detection chamber rather than being kept at approximately zero as in the steady-state measurements. Due to the different boundary conditions, the thickness of the air-filled detection chamber becomes significant in the transient system rather than the thickness of the source region as in the steady-state system. In both systems, the thickness of the sealed air-filled regions is only significant when the radon diffusion coefficient of the soil being tested is low ($D_e/P_s < 10^{-3} \text{cm}^2/\text{s}$).

The basis for interpreting the transient radon diffusion data is the one-region, one-dimensional, time-dependent solution to the radon diffusion equation,

$$C(t) = C_0 \sum_{n=1}^{\infty} \frac{(-1)^{n-1}(2n-1)}{\pi(2n-1)^2/4 + \lambda b^2/(\pi D)} \left\{ 1 - \exp \left[-\lambda t - (2n-1)^2 \frac{\pi^2 D}{4b^2} t \right] \right\} \quad (3)$$

where

D = diffusion coefficient of radon in the soil pore fluid (cm^2/s) = D_e/P_s

t = time from radon source exposure to concentration measurement (s)

Since the transient-diffusion measurement system measures the alpha activity of the radon daughters ^{218}Po and ^{214}Po along with that of the radon, the

Bateman equations⁽⁶⁾ were coupled with Eq (3) in a computer program to calculate total alpha activities. This provided for calculating the radon daughter ingrowth with time for the varying radon concentrations which also increased with time. The coupled equations were analyzed by computer to calculate the total alpha activity at any time as a function of soil column length and radon diffusion coefficient.

Since the two-region transient diffusion problem is very complicated and analytical solutions are not available, the one-region analytical solution in Eq (3) was used with two correction factors to account for the air-filled detection region. One of these factors was the ratio of the radon concentration from a steady-state, two-region soil and air problem (C_2) to that from a steady-state, one region soil problem (C_1), and was calculated as

$$\frac{C_2}{C_1} = \left[1 + \left(\frac{D_A}{P_s D_e} \right)^{1/2} \tanh(k_s b) \tanh(k_A a) \right]^{-1} \quad (4)$$

This factor gave an exact correction for the final plateau region of the transient curves, and was multiplied by the source concentration in Eq (3). Its magnitude is near unity until diffusion coefficients of about $10^{-3} \text{ cm}^2/\text{s}$ or less are attained, and it approaches a value of 0.5 as the soil diffusion coefficient approaches $10^{-5} \text{ cm}^2/\text{s}$. Since the correction is a constant multiplier of any given transient activity curve, it does not directly affect the estimation of diffusion coefficients. Instead, it acts as a change in the detector efficiency calibration, which can even be treated as a variable in fitting transient activity curves.

The second correction factor had a direct effect on the value of the radon diffusion coefficient, and accounted for multidimensional effects near the boundary between the soil region and the air-filled detection region. The

decrease in radon accumulation rate in the detector region is related to the soil porosity, so that this correction is equivalent to using D_e in place of D in Eq (3). It should be noted that this correction was not applied to the transient-diffusion coefficients reported in Reference 7, so that those diffusion coefficients should be regarded as D_e . In the present work, the correction was applied so that the best fit to the measured data yielded the correct value of D .

EXPERIMENTAL METHOD FOR TRANSIENT MEASUREMENTS

Based on the foregoing time-dependent equations for radon diffusion, an experimental apparatus was designed to determine radon diffusion coefficients. The conceptual basis of the experimental measurements is as follows. A column containing the soil to be tested is exposed on one end at time zero to a large volume of air containing a known high radon concentration. A continuous alpha particle detector is sealed to the opposite end of the column to measure the alpha activity from radon and its daughters. As radon diffuses through the soil, the measured alpha activity increases to a constant maximum level which corresponds to an equilibrium radon distribution throughout the soil. The measured alpha activity buildup curve is then compared to theoretical curves calculated for various diffusion coefficients and the actual diffusion coefficient is inferred from the best fit. The following sections describe the experimental apparatus and procedure, the sample preparation procedure, the calibration procedure, and the data interpretation procedure.

Diffusion Apparatus

The experimental apparatus used for the transient radon diffusion measurements is illustrated in Figure 1. The radon source consisted of uranium mill

tailings obtained from the Vitro Tailings pile in Salt Lake City. These tailings have been found⁽⁷⁾ to contain about 1450 pCi/g ^{226}Ra , and to have a radon emanation coefficient of about 0.22. Approximately 150 kg of the tailings were placed in a 220-liter steel drum with five perforated tubes to facilitate radon diffusion. The large air volume at the top of the drum was sufficient to maintain a constant concentration radon source throughout the experiment. A 10-cm gate valve was located at the top of the drum to contain the radon between measurements and to allow unrestricted access of the radon gas to the test column entrance. By only opening the gate valve with a sample column sealed in position, the radon concentration in the drum accumulated to a steady-state concentration of about 2.8×10^5 pCi/L. A subsequent source was later utilized which reached 4×10^5 pCi/L. A sampling port located at the top of the drum

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FIGURE 1. TIME-DEPENDENT RADON DIFFUSION APPARATUS

facilitated sample collection for calibration purposes.

A double O-ring fitting was attached to the upper side of the gate valve for attachment of the sample column, which was made from SCH-80 PVC plastic pipe. A similar fitting was used on the detector assembly to provide a gas-tight seal to the sample column. The detector assembly consisted of a 10-cm diameter alpha scintillation detector, located 2.5-cm from a metal screen which rested on the top of the sample column. A 300-V negative bias was maintained on the detector face with respect to the screen to attract the positive radon daughters toward the detector as they were formed. A gas sampling port was also located in the detector assembly to allow collection of calibration samples.

The alpha scintillation detector was powered by a pre-amp/amplifier combination with adjustable threshold, discriminator, and gain setting. A scaler/timer and printer assembly provided continuous printouts of alpha activity over any selectable integration interval. Typical integration intervals were one, ten, or twenty minutes.

Sample Preparation and Measurement Procedure

Soil samples were prepared by first adjusting the moisture of the soil so the approximate desired level by addition of water or by permitting short drying periods. Once the water content was adjusted and equilibrated, the soil was packed into a 10-cm diameter PVC pipe in approximate 1-2 cm lifts. Packing was generally accomplished with a short metal rod, and the desired density could usually be attained in the first one or two attempts. Moist or highly compacted dry samples were self-supporting in the sample tube, but loose dry samples required a supporting screen at the bottom.

The compacted sample was then attached to the radon source and detector assemblies as illustrated in Figure 1. Background counts were then conducted

over approximately one hour, after which the diffusion experiment was started by opening the gate valve to the radon source. A small mixing fan located inside the source drum (Figure 1) was kept continually running, and served to quickly mix the air immediately beneath the sample column with that in the source drum. The data collection process was allowed to continue over the next 18-72 hours, after which the gate valve was closed, and the sample column was removed. The actual moisture and density of the soil sample were then determined by drying the entire sample at 105-110⁰ C until constant weight was attained.

Calibration

In order to interpret the transient alpha activity curves from the diffusion measurements, radon concentrations were required as a function of time at both ends of the column. For the source concentration, simple Lucas-cell samples were collected and found to remain constant with time. The continuous alpha scintillation detector was calibrated by allowing the radon in the detection chamber to reach equilibrium and then relating the observed count rate to the radon concentration measured from a Lucas cell grab-sample (10 cm³). The scintillation detector was found to have a total alpha detection efficiency of about 14 percent.

Individual efficiencies for radon gas and for ²¹⁸Po and ²¹⁴Po were also required to properly interpret the transient curves for cases of high diffusion coefficients. The individual efficiencies were determined by allowing a relatively high radon concentration to equilibrate in the detection chamber, and observing the decay rates as the chamber was opened and ventilated. The decay rates were monitored on a one-minute time scale, and clearly illustrated the radon gas contribution with an immediate drop in count rate as the chamber was ventilated. The contribution of the ²¹⁴Po daughter was determined from

the latter part of the decay curve (>25 min after ventilation) because the preceding nuclides had nearly all decayed by this time. The contribution of ^{218}Po was finally determined from the activity during the first ten minutes after correcting for the contribution of the ^{214}Po . The respective relative efficiencies determined in this manner for ^{222}Rn , ^{218}Po and ^{214}Po were 10 percent, 45 percent, and 45 percent, leading to corresponding absolute efficiencies of 4 percent, 19 percent, and 19 percent. The radon gas efficiency is lower than the daughter efficiencies because it is a volumetric source spread throughout the 2.5-cm thick detection chamber. The daughter efficiencies are higher because they are attracted by the 300-V bias to the detector surface, and therefore have a more favorable detection geometry.

Data Description and Analysis

The transient alpha activity curves which result from a diffusion experiment are characterized by an initial lag period, a transition or breakthrough region, and a final plateau region which corresponds to an equilibrium radon distribution. Figure 2 illustrates a family of characteristic alpha activity curves calculated for various diffusion coefficients for a 14.8-cm diffusion column. As illustrated, an empty, air-filled column having a diffusion coefficient of about $0.1 \text{ cm}^2/\text{s}$ would break through almost immediately, and would reach equilibrium within a few hours. A soil with a diffusion coefficient of about $10^{-3} \text{ cm}^2/\text{s}$ would only begin to break through after several hours, and would not reach equilibrium for more than a day. Materials with lower diffusion coefficients have even longer lag times, and reach plateaus at lower concentrations due to the significant decay which occurs in the sample column.

In order to provide greater flexibility in measuring a wide range of diffusion coefficients, the column length may also be varied. Longer columns are typically used for dry, porous materials with expected high diffusion

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FIGURE 2. TRANSIENT ALPHA ACTIVITY CURVES FOR VARIOUS DIFFUSION COEFFICIENTS

coefficients and shorter ones are used for moist, highly compacted clays.

Figure 3 illustrates the predicted alpha curves for columns of varying lengths and with a constant diffusion coefficient of $10^{-3} \text{ cm}^2/\text{s}$.

Experimental data were analyzed by a computer program which calculated the transient alpha activity curves as illustrated in Figures 2 and 3 using Eq (3) and (4) and the Bateman equations. The program utilized ten alpha activity data points spread primarily throughout the transition or breakthrough region of the curves, and determined by least-squares fit the diffusion coefficient which best fit the measured alpha activity data. The estimate of uncertainty in the diffusion coefficient was obtained as the standard deviation of ten diffusion coefficients determined from pairs of adjacent points taken from each of the ten locations on the curve used in the least-square fit. Typical relative standard deviations of the radon diffusion coefficients were calculated to be on the order of 5-12 percent.

RESULTS AND DISCUSSION

The transient-diffusion measurement technique was applied to several soil materials which had been previously analyzed by the steady-state diffusion measurement method.⁽⁵⁾ It was also used to measure the radon diffusion coefficient of air and of selected other reference materials whose diffusion coefficients were known. The following sections describe the results of the comparative soil measurements and the standard reference measurements.

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FIGURE 3. TRANSIENT ALPHA ACTIVITY FOR VARIOUS COLUMN LENGTHS

Radon Diffusion Through Soils

Transient radon diffusion measurements were conducted on five different soils at a variety of different moisture contents. The results of these diffusion measurements are summarized in Table I in terms of the soil porosity, its moisture content (volume fraction of saturation), and the radon diffusion coefficient. As indicated by the porosities, the compactions were held relatively constant for each soil, while the moisture contents were varied for the different diffusion measurements. As indicated, the diffusion coefficients generally decreased with increasing moisture content, as was expected from theoretical considerations⁽⁸⁾ and from previous experimental work and empirical correlations.^(2,3,9) Two exceptions to this trend are noted in Table I for the D clay and M shale materials at their highest moisture contents. The increases in these two diffusion coefficients were relatively small, and were attributed to the high variability in the value of the moisture content that occurs in preparing wet samples. Some of the uncertainty could also be associated with the value of the best fit diffusion coefficient to the data points.

Comparative steady-state radon diffusion measurements⁽⁵⁾ on the same soils are also listed in Table I. These data are part of a larger group of diffusion measurements conducted at PNL at varying soil moistures and compactions. Although the soil moistures and compactions used for the two types of diffusion measurements are not identical, they are sufficiently close to provide a valid comparison of results for many of the diffusion measurements. The ratios of coefficients from the two types of diffusion measurement indicate that the agreement between the two methods is within about 10 to 20 percent relative error for dry or relatively low-moisture soils (<20 percent saturation). The relative standard deviation among the five replicate transient diffusion measurements on dry dunite is 12 percent, suggesting that differences between the two methods for dry samples can largely be attributed to inherent uncertainties in the measurement procedure.

TABLE I
COMPARISON OF RADON DIFFUSION COEFFICIENTS FROM TRANSIENT-DIFFUSION
AND STEADY-STATE MEASUREMENTS

SAMPLE	TRANSIENT-DIFFUSION			STEADY-STATE			D Ratio* Trans/S.S.
	Porosity	Sat'n.	D (cm ² /s)	Porosity	Sat'n.	D (cm ² /s)	
D Clay	0.42	0.10	0.045				
	0.41	0.15	0.017	0.39	0.15	0.016	1.0
	0.43	0.25	0.0070				
	0.40	0.35	0.011				
DF Sand	0.41	0.23	0.045	0.36	0.05	0.030	1.4
WN Clay	0.38	0.54	0.011	0.39	0.22	0.024	0.4
	0.38	0.86	0.0012	0.37	0.67	0.00051	2.2
M Shale	0.34	0.55	0.020	0.33	0.54	0.022	0.8
	0.30	0.86	0.0013	0.28	0.67	0.0013	0.9
	0.30	0.88	0.0022				
Dunite	0.46	0.00	0.050				0.8
	0.45	0.00	0.056				0.9
	0.45	0.00	0.066	0.44	0.00	0.06	1.0
	0.45	0.00	0.068				1.0
	0.45	0.00	0.064				1.0
	0.41	0.41	0.014				
	0.40	0.56	0.0030				

*Ratios multiplied by 0.915 to correct for the atmospheric pressure difference between RAE (1286m elevation) and PNL (110m elevation).

Comparisons of the diffusion coefficients at higher moistures are more complicated. As shown in Table I, good agreement between the two methods was observed for M shale, despite significant moisture differences in one of the comparisons. For DF sand and WN clay, however, the diffusion coefficients differed by factors of 0.4 to 2.2. The diffusion through DF sand was measured at a much higher moisture in the transient case, which should have caused a lower diffusion coefficient, despite the partially compensating effect of the higher porosity with the transient measurement. The transient-diffusion measurements on WN clay showed a reasonably lower diffusion coefficient for the higher moisture content in the first case. However, the second case showed a 2.2-times higher diffusion coefficient in the presence of a slightly higher moisture content.

A likely reason for the higher D ratios at high moisture contents is in the technique used for the equilibrium diffusion measurements. Soil samples for these measurements were prepared with initially high moistures, and were used to determine diffusion coefficients at various lower moistures by allowing evaporation from the top surface of the soil column. The resulting moisture distributions were therefore non-uniform in the direction of diffusion and gave the effect of a lower diffusion coefficient than would occur if the measured moisture were uniformly distributed throughout the soil. Although the data did not permit a quantitative measure of this effect for each sample, it easily could have had sufficient magnitude to explain the high D ratios in Table I. Therefore, the reliability of the steady-state measurements is in question at high moistures.

Another cause of variation among the D-ratios at high moistures in Table I is the random error associated with packing homogeneous moist soils into the diffusion columns. This random error results from non-uniform moisture distributions, non-uniform compaction of the soils into the columns, and varying degrees of consolidation of the original soil crumbs being compacted. These

variations could affect both the transient and the steady-state measurements, and would cause decreased diffusion rates if they occurred in the direction of diffusion and enhanced rates if they occurred perpendicular to the direction of diffusion. The overall effect of soil or moisture inhomogeneity was thus an increase in the uncertainty of the diffusion measurements. Since moisture is a dominant parameter, the samples with high moisture contents tended to exhibit higher uncertainties in diffusion coefficients.

Evidence of radon absorption by the moisture in the soil was sometimes observed in the transient alpha activity curves. This phenomenon was usually expressed as a more gradual slope in the breakthrough region of the transient curves, and was readily detected by plotting the measured activity curve with the family of calculated curves for the given column length and source strength as shown in Figure 2. Ideally, the measured curve was parallel to the adjacent calculated curves. However, at high moistures, the measured curve sometimes crossed several of the calculated curves because of the time dependence of the moisture absorption and related effects. This phenomenon was usually minimized by using shorter diffusion columns. It was also generally possible to use the upper part of the breakthrough region of the curve to calculate the diffusion coefficient, as this region generally remained parallel to the expected curve shapes. The transient diffusion data were routinely plotted with the calculated curves as a check for systematic errors before computing diffusion coefficients.

Reference Diffusion Measurements

In order to further verify the accuracy of the transient-diffusion measurement technique, several diffusion measurements were conducted in dry air and certain other well-defined media. These diffusion measurements allowed comparison with theoretically-derived diffusion coefficients as well as with other measurement techniques, and avoided the uncertainties associated with

soil structures and moisture distributions. Table II summarizes these diffusion measurements. As indicated, a nine percent correction was applied to compensate for the elevation and resultant reduced pressure at the RAE laboratory.

The diffusion coefficients for dry air utilized an empty soil sample tube in the transient-diffusion apparatus. As in the measurements with soils, coarse filter papers, with porosities exceeding 0.9, were placed at the entrance and exit of the tube to avoid turbulence from the mixing fan in the radon source chamber and to define the tube boundary. The resulting diffusion coefficients in Table II are in excellent agreement with the theoretical diffusion coefficient predicted for radon in air from the Othmer-Chen equation,^(3,10) $0.105 \text{ cm}^2/\text{s}$. Good agreement is also noted with the experimental measurements referenced by Tanner,⁽¹¹⁾ which ranged from $0.10\text{-}0.12 \text{ cm}^2/\text{s}$.

In a subsequent experiment, the sample tube was packed with parallel wooden dowels. This provided a porosity of 0.25 in the diffusion tube, and a simple pore structure without tortuosity. Because of the much smaller size of the pores within the wood and the blockages caused by its cellulose structure, the porosity of the dowels was neglected. As indicated, the resulting diffusion coefficient was within experimental error of the values for dry air. Although the observed value may be slightly high due to the wood porosity, the magnitude of the bias is not significant.

Three additional measurements were conducted in which uniform glass balls and differential sieve fractions of dry sand were used as the diffusion medium. In these cases, the tortuosity was obviously not unity, and a lower value of D was expected. The measured D values verify that the diffusion coefficient for a straight air pathway was significantly lowered by the granular nature of these samples. If the standard D values in Table II are divided by the estimated tortuosities of these samples, the results should be comparable to the diffusion coefficient of air. Using the spherical-particle approximation reported

TABLE II
TRANSIENT-DIFFUSION MEASUREMENTS ON STANDARD REFERENCE MATERIALS

<u>SAMPLE</u>	<u>POROSITY</u>	<u>MEASURED D</u> <u>(cm²/s)^a</u>	<u>STANDARD D</u> <u>(cm²/s)^b</u>
Air	1	0.110	0.100
Air	1	0.120	0.109
1-cm diameter wood cylinders	0.25	0.124	0.113
4-mm diameter glass balls	0.33	0.078	0.071
Dry sand 16-30 mesh	0.47	0.060	0.054
Dry Sand 50-100 mesh	0.46	0.063	0.057

^a Measured at 1286m elevation, 0.908 atm. pressure.

^b Multiply measured D's by 0.908 to convert to Standard D at 1-atm pressure (sea-level).

previously⁽⁸⁾ for estimating tortuosities, the glass ball, coarse sand and fine sand samples were estimated to have respective tortuosities of 0.68, 0.66 and 0.66. Normalizing the respective standard diffusion coefficients from Table II by these tortuosities gives radon diffusion coefficients of 0.104, 0.082 and 0.086 cm²/s. The glass ball measurement is thus in excellent agreement with the expected value for radon in air, and the 20-25 percent relative errors in the normalized sand measurements could well be a result of the influence of the non-spherical sand grain shapes on the tortuosity estimate. The standard diffusion coefficients reported for the sieved sand fractions are thus reasonable despite being slightly lower than the glass ball sample. In all three granular samples, pore sizes were sufficiently large for diffusion to be in the molecular regime, and not in the Knudsen or transitional regimes.

Precision and Accuracy

The precision of the transient radon diffusion measurement technique was directly evaluated by the five replicate measurements on the dry dunite reported in Table I. The relative standard deviation among these diffusion coefficients was 12 percent (0.0608 ± 0.0076 cm²/s). This precision is probably representative of the uncertainties in the measurement process, since the dunite sample was dry and could readily be poured into a reproducible packing configuration and density.

The higher uncertainty associated with radon diffusion measurements through moist soil samples cannot be directly deduced from only the present measurements. This uncertainty was estimated from previous transient-diffusion measurements that were made on duplicate samples of moist clay-type soils. Moistures ranged from 50 to 100 percent saturation and moistures of the duplicate samples were within 1.0 wt percent of each other. In addition, the corresponding com-

pactions were nearly identical. Using a one-way analysis of variance on the log-transformed diffusion coefficients, the geometric standard deviation among the replicate analyses was estimated to be 1.43.

In evaluating the sources of random error in transient-diffusion measurements, uncertainties from alpha counting statistics and from the count timing sequence are minimal. The predominant components of the 12 percent relative variation observed for the dunite appear to be pressure and temperature variations and sample packing variations. Although the diffusion measurement system is sealed throughout the course of a diffusion measurement, the varying atmospheric pressures at the times of sample insertion could account for some of the observed variation. Random errors in excess of the 12 percent level are attributed to variations in sample characteristics.

The accuracy of the transient radon diffusion measurement technique was primarily evaluated from the diffusion coefficients in air and other well-defined materials in Table II. The variation of these coefficients from the theoretical value of $0.105 \text{ cm}^2/\text{s}$ is much smaller than the uncertainty associated with the precision of the method (12 percent). No significant bias was thus observed in these measurements.

In comparing the transient diffusion coefficients with the equilibrium measurements at PNL, the transient data fell within 10 to 20 percent of the equilibrium data for all but the three comparisons which involved extremely different soil moistures. The moisture differences, moisture gradients (in equilibrium measurements), and sample preparation uncertainties adequately account for these larger differences. The overall comparison of the two methods in Table I indicates very good agreement. The average ratio of all of the transient/equilibrium comparisons was 1.04.

It should be noted that both atmospheric pressures and laboratory temperatures can affect radon diffusion coefficients. The diffusion coefficients are inversely proportional to atmospheric pressure,⁽¹⁰⁾ and thus require a correction due to altitude differences when comparisons are made for different locations. These corrections have been implemented in Tables I and II, and amount to about nine percent in correcting for the 1286m elevation of the RAE laboratory to 1-atm sea level conditions. The temperature correction is ordinarily smaller, and was not applied to the present measurements. The transient-diffusion coefficients were measured at laboratory temperatures ranging from 19-23°C. Temperature effects on diffusion coefficients have been estimated⁽³⁾ to amount to 0.8 percent/°C, suggesting a three percent variation among the present measurements due to temperature differences. The temperature effects are thus small compared to diffusion measurement precisions. The calculated radon diffusion coefficient for air⁽³⁾ was based on a temperature of 25°C.

CONCLUSIONS

The measurement of radon gas diffusion coefficients under transient conditions provides a rapid and accurate alternative to traditional steady-state diffusion measurement techniques. Radon diffusion coefficients measured by the transient method show excellent agreement with the theoretically calculated diffusion coefficient for air as well as with previously measured coefficients for air. Measured diffusion coefficients also agreed with theoretical expectations for diffusion through a column packed with glass balls.

Comparative measurements on compacted soils also showed good agreement with steady-state diffusion measurements. The agreement was best at low moistures, where relative differences averaged less than ten percent. At

high moistures, the relative differences amounted to approximately a factor of two, although a significant part of this difference was attributed to non-uniform moisture in the steady-state measurements. Since the transient and steady-state diffusion measurements utilized completely different experimental conditions, measurement methods, and mathematical interpretations, the comparisons give an excellent verification of the theoretical and technical accuracy of both approaches.

Precisions were estimated from five replicate transient diffusion measurements with dry dunite. A relative standard deviation of 12 percent was observed. An estimate of the precision at soil moistures from 50 to 100 percent of saturation yielded a relative uncertainty for the moist soils that was about three to four times greater than that for the dry samples.

The transient-diffusion measurement technique utilizes small samples, facilitating greater control over sample characteristics. Since these appear to dominate experimental uncertainties, good precisions are attainable. The transient method also offers significantly shorter measurement times than steady-state methods.

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APPENDIX

DERIVATION OF RADON DIFFUSION EQUATIONS

DERIVATION OF RADON DIFFUSION EQUATIONS

Equation 1

Equation (1) was derived from the one-dimensional, steady-state diffusion equation without radon sources,

$$D \frac{\partial^2 C}{\partial x^2} - \lambda C = 0. \quad (A1)$$

The general solution to equation (A1) is

$$C_s(x) = E \exp(k_s x) + F \exp(-k_s x), \quad (A2)$$

where E and F are constants to be determined by the boundary conditions of the system and the subscript s refers to the soil medium in which the diffusion occurs. From Fick's law, the radon flux for the system is

$$J_s(x) = -D_e \frac{\partial C_s}{\partial x} = D_e k_s [-E \exp(k_s x) + F \exp(-k_s x)], \quad (A3)$$

and the boundary conditions imposed on this one-region problem are

$$J_s(0) = J_0 \quad (A4)$$

$$\text{and } C_s(b) = 0, \quad (A5)$$

where J_0 is the radon flux entering the soil at the source end ($x=0$) and $C_s(b) = 0$ is the negligible radon concentration at the exit end ($x=b$) of the system. Substituting the boundary conditions (A4) and (A5) into Equations (A2) and (A3), and defining the source concentration $C_0 = C_s(0)$, the ratio

for C_0/J_0 is found to be

$$\frac{C_0}{J_0} = \frac{\tanh(k_s b)}{D_e k_s} \quad (A6)$$

Equation 2

Equation (2) was also based on the one-dimensional, steady-state diffusion equation (A1), for which equation (A2) was defined to be the solution applying to the soil region. A second solution, applying to the air-filled source region, was also written as

$$C_A(x) = G \exp(k_A x) + H \exp(-k_A x), \quad (A7)$$

where the constants G and H are additional constants to be determined by the boundary conditions of the system. The radon flux defined by Fick's Law for the soil region is still given by equation (A3), and the corresponding equation for the flux in the air-filled source region was written as

$$J_A(x) = -D_A \frac{\partial C_A}{\partial x} = D_A k_A [-G \exp(k_A x) + H \exp(-k_A x)] \quad (A8)$$

Four boundary conditions were defined for the two-region system, which was defined to have its origin at the soil-air interface, and which had a soil thickness of b and an air thickness of a. The boundary conditions were

$$J_A(-a) = J_0 \quad (A9)$$

$$J_A(0) = J_s(0) \quad (A10)$$

$$C_A(0) = C_s(0) \quad (A11)$$

$$C_s(b) = 0 \quad (A12)$$

Applying the boundary conditions (A9-A12) to Equations (A2), (A3), (A7) and (A8), the constants E, F, G and H were determined, from which the ratio C_0/J_0 was determined to be

$$\frac{C_0}{J_0} = \frac{\tanh(k_s b)}{D_e k_s \cosh(k_A a) + D_A k_A \sinh(k_A a) \tanh(k_s b)} \quad (A13)$$

Again, the definition $C_0 = C_A(0) = C_s(0)$ was used.

Equation 3

The derivation of Equation (A3) for a single region comes from the one-dimensional, time-dependent diffusion equation

$$D \frac{\partial^2 C}{\partial x^2} - \lambda C = \frac{\partial C}{\partial t} \quad (A14)$$

Defining the origin as the source end of the diffusion column, the following three boundary conditions were employed:

$$C(x, 0) = 0 \quad (\text{no initial radon}) \quad (A15)$$

$$C(0, t) = C_0 \quad (\text{constant source concentration}) \quad (A16)$$

$$\left. \frac{\partial C}{\partial x} \right|_{x=b} = 0 \quad (\text{no leakage at detector end}) \quad (A17)$$

all t

Taking the Laplace transform of Equation (A14) with respect to t, and applying the boundary condition from Equation (A15),

$$D \frac{d^2 \bar{C}}{dx^2} - \lambda \bar{C} = S \bar{C} \quad (A18)$$

The boundary condition in Equation (A16) now becomes

$$\bar{C}(0,S) = C_0/S \quad (A19)$$

and that in Equation (A17) becomes

$$\left. \frac{d\bar{C}}{dx} \right|_{x=b} = 0 \quad (A20)$$

Defining $\alpha^2 = (S + \lambda)/D$, the solution to Equation (A18) is

$$\bar{C} = A \exp(-\alpha x) + B \exp(\alpha x) \quad (A21)$$

Applying the boundary conditions, the constants A and B are determined and the resulting solution for $\bar{C}$ is inverted and integrated to express the solution for $C(x,t)$ at the point $x = b$ as

$$C(b,t) = C_0 \sum_{n=1}^{\infty} \frac{(-1)^{n-1} (2n-1)}{\pi(2n-1)^2/4 + \lambda b^2/(\pi D)} \left\{ 1 - \exp \left[-\lambda t - (2n-1)^2 \frac{\pi^2 D}{4b^2} t \right] \right\}. \quad (A22)$$

Equation 4

The derivation of Equation (4) is based on a two-region solution of the one-dimensional, steady-state radon diffusion equation. The diffusion system being described is that of a steady-state radon distribution in a soil, one end of which is attached to a source of constant radon concentration, and the other end of which is sealed. An air gap is assumed between the soil and the sealed end of the column in the two-region case, and the thickness of the air region is set equal to zero for the one-region case. Equation (4) is simply the ratio between the radon concentrations at the sealed end in the two cases.

The general steady-state Equation (A1) is used, along with its solutions

for the soil and air regions, Equations (A2) and (A7). The prior definitions of radon flux from Fick's Law for the soil and air regions are also used, as given in Equations (A3) and (A8). The system is defined to have its origin at the interface, with the air region extending to +a and the soil extending to -b. The four boundary conditions are

$$C_s(-b) = C_o \quad (A23)$$

$$J_s(0) = J_A(0) \quad (A24)$$

$$C_s(0) = C_A(0) \quad (A25)$$

$$J_A(a) = 0 \quad (A26)$$

Applying the boundary conditions, solving for E, F, G, and H, and letting $C_2 = C_s(0)$,

$$C_2 = C_o \frac{D_e k_s}{D_e k_s \cosh(k_s b) + D_A k_A \tanh(k_A a) \sinh(k_s b)} \quad (A27)$$

For the one-region case, Equation (A2) can be similarly defined by letting $C_1 = C_s(0)$, $a = 0$, and simplifying to get

$$C_1 = \frac{C_o}{\cosh(k_s b)} \quad (A28)$$

The ratio of the concentration for the two-region case to that for the one-region case is therefore obtained by dividing Equation (A27) by Equation (A28) and simplifying with the definitions of k_s and k_A to obtain

$$C_2/C_1 = \left[1 + \left(\frac{D_A}{P_s D_e} \right)^{1/2} \tanh(k_s b) \tanh(k_A a) \right]^{-1} \quad (A29)$$

for the soil and air regions, Equations (A5) and (A7). The prior defini-
 tions of radon flux from Fick's law for the soil and air regions are also
 used, as given in Equations (A3) and (A8). The system is defined to have
 its origin at the interface, with the air region extending to $+z$ and the
 soil extending to $-z$. The four boundary conditions are

$$\begin{aligned} (A9) \quad C_2(-b) &= C_0 \\ (A4) \quad J_2(0) &= J_A(0) \\ (A8) \quad C_2(0) &= C_A(0) \\ (A5) \quad J_A(a) &= 0 \end{aligned}$$

Applying the boundary conditions, solving for E, F, G, and H, and letting

$$C_2 = C_2(0),$$

$$(A7) \quad C_2 = C_0 \frac{e^{k_2 z} \cosh(k_2 b) + J_A \tanh(k_2 a) \sinh(k_2 b)}{e^{k_2 z} \cosh(k_2 b) + J_A \tanh(k_2 a) \sinh(k_2 b)}$$

For the one-region case, Equation (A7) can be similarly defined by letting

$$C_1 = C_2(0), \quad a = 0, \text{ and simplifying to get}$$

$$(A8) \quad C_1 = \frac{C_0}{\cosh(k_2 b)}$$

The ratio of the concentration for the two-region case to that for the one-
 region case is therefore obtained by dividing Equation (A7) by Equation
 (A8) and simplifying with the definitions of k_2 and J_A to obtain

$$(A9) \quad C_2/C_1 = \left[1 + \left(\frac{D_A}{D_2} \right)^2 \tanh(k_2 b) \tanh(k_2 a) \right]^{-1}$$

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13. ABSTRACT A method was developed and used to determine radon diffusion coefficients in compacted soils by transient-diffusion measurements. A relative standard deviation of 12 percent was observed in repeated measurements with a dry soil by the transient-diffusion method, and a 40 percent uncertainty was determined for moisture exceeding 50 percent of saturation. Excellent agreement was also obtained between values of the diffusion coefficient for radon in air, as measured by the transient-diffusion method, and those in the published literature. Good agreement was also obtained with diffusion coefficients measured by a steady-state method on the same soils. The agreement was best at low moisture, averaging less than ten percent difference, but difference of up to a factor of two were observed at high moisture. The comparison of the transient-diffusion and steady-state methods at low moisture provides an excellent verification of the theoretical validity and technical accuracy of these approaches, which are based on completely independent experimental conditions, measurement methods and mathematical interpretations.			
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