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Measurement of Radon Diffusion from Uranium Mill Tailing Piles

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October 1979

Prepared for
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MEASUREMENT OF RADON DIFFUSION AND EXHALATION FROM URANIUM MILL TAILINGS PILES

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ABSTRACT

The concentrations of ^{226}Ra and ^{222}Rn (^{214}Pb) were measured as a function of depth within a uranium mill tailings pile by in-situ gamma-ray spectrometry. Radon diffusion and exhalation rates were determined from the concentration gradients by employing an integral solution of the diffusion equation that accommodates a non-uniform depth distribution of the parent radium. Radon diffusion coefficients of 0.0002 and 0.0017 cm^2/sec, and exhalation rates of 60 and 275 atoms/cm^2/sec were determined for two locations with differing soil moisture content.

1. INTRODUCTION

Release of ^{222}Rn* from uranium mill tailings to the atmosphere results in one source of radiation exposure to populations as a consequence of the uranium mining and processing industry. Radon is the daughter of ^{226}Ra† which, not being depleted by the uranium extraction process, is associated with the spent feed material which is disposed to a tailings pile. Being a noble gas, radon can be produced in or diffuse into the interstitial pores of the soil column, and diffuse upward through the soil surface, escaping to the atmosphere. Any assessment of the radiological impact of the exhaled radon requires knowledge of its rate of release and the influence of various physical and meteorological parameters on this rate.

A variety of approaches have been used to evaluate the quantities of radon released to the atmosphere from general soil areas or from specific sites. Most of these involve collection and subsequent analysis of radon or radon daughters from the air at or above the soil surface. A common method involves direct accumulation in a container that is sealed to the soil surface, followed by removal of an aliquot for radon analysis.(1,2) Countess(3) and Megumi and Mamuru(4) describe methods by which radon is adsorbed directly onto a layer of activated charcoal placed on the soil surface. By definition, disintegration rate of the radon in the air column above a soil surface is equal to the number of atoms crossing the soil-air interface. Determination of the vertical radon or radon daughter inventory as a measure of flux has been employed by many investigators, e.g.(5,6). While these techniques afford useful data on instantaneous radon emanation, they provide little or no information concerning the rates of radon exhalation or the distance below the soil surface from which the radon was derived. We took a more mechanistic approach, basically measuring the changes of concentration of radium and radon, as its (^{214}Pb) daughter, as a function of depth in the soil column by in-situ gamma-ray spectrometry. From the rate of change of parent-daughter concentrations with depth, the rates of radon diffusion and exhalation were calculable.

* hereafter called radon
† hereafter called radium

2. FIELD SITE AND MEASUREMENT METHODS

The tailings pile at the United Nuclear-Homestake Partners uranium mill at Grants, New Mexico was selected for this study. This mill employs a carbonate leach process to extract uranium from ore that has been crushed and milled so that 5% of the ore is +48 mesh and 38% is less than 200 mesh. The residual from the leach process, which contains essentially all of the radium originally present in the ore, is transported as a slurry to the tailings pile. Coarse particulates are removed by a cyclone separator and used to build the dike containing the pile; the remaining fines are disposed to the center of the tailings pile.

In the course of about twenty years of operation the pile is now approximately 600 m wide, 1400 m long and 25 m high. It is divided into roughly two halves, each of which alternately receives tailings material from the mill. This study was conducted in the inactive side of the tailings pile. Residual water covered about two percent of the total surface at the center of the inactive side, and soil moisture content decreased with distance to the sides of the pile.

Test holes were implanted at locations on the tailings pile that were selected as representative of the existing soil and moisture conditions. Each test hole consisted of a six inch (15.2 cm) diameter schedule 40 PVC pipe which was either forced into pre-drilled holes, or impacted to the desired sampling depth, usually between 1.5 and 3 meters below the soil. surface. Care was taken to insure good contact between the outside of the pipe and the soil to avoid any air voids that could allow flow channeling of soil gases, and the bottom of the pipe was sealed to prevent intrusion of water. Gamma-ray logging of the holes was accomplished by sequentially lowering a germanium diode detector to discrete depths and accumulating the resultant spectrum for a sufficiently long period at each depth to obtain the desired counting statistics, characteristically ± 15% and ± 4% for ^{226}Ra and ^{214}Pb, respectively. A 23% intrinsic germanium diode with 1.96 keV resolution, made by Princeton Gamma-Tech was used as the detector for this study. Signals were analyzed and stored in a Tracor Northern Econ II series analyzer, and the data were digitally recorded with a high speed printer. Field measurements were made, both with and without a 2.5 cm thick lead shield covering the top of the diode, which limited the sample viewed by the diode to a 2π configuration. Use of the shielding provided improved definition of the vertical radionuclide distribution. The counting system was calibrated in the laboratory, using uranium tailings of known concentration, for the different conditions of geometry, shielding, and soil composition and moisture content encountered in the field.

Concentrations of ^{226}Ra and ^{214}Pb were determined from the respective intensities of the 186 and 352 keV photo peaks. It is presumed that the 26.8 minute ^{214}Pb daughter was in equilibrium with ^{222}Rn with the recognition that conditions could occur over time scales comparable to the ^{214}Pb half life that would invalidate this tenet. The 185 keV photopeak from decay of ^{235}U, if present, would interfere with absolute determination of ^{226}Ra. For this work, it was assumed that the relative ^{235}U and ^{226}Ra concentrations were constant throughout the soil column, and any ^{235}U contributions were accommodated in the calibration procedure.

3. RADON DIFFUSION

One dimensional transport of radon through soil or other porous media is given by Junge(7) for the simplest case as:

$$C_z = \frac{S}{\lambda}\left[1 - \exp\left(-z\sqrt{\frac{\lambda}{D}}\right)\right] \qquad \text{Equation (1)}$$

where C_z = the radon concentration in soil air (atoms/cm^3)
S = the radon production rate (atoms/cm^3·sec)
λ = the decay constant (sec^{-1})
z = depth (cm)
D = the coefficient of diffusion (cm^2/sec)

The rate of exhalation (J) as atoms/$cm^2 \cdot$sec from the soil column to the atmosphere is:

$$J = S\sqrt{\frac{D}{\lambda}} \qquad \text{Equation (2)}$$

For these solutions, the radium concentration is assumed to be uniform with depth. This homogeneity does not exist in a tailings pile. As seen in Figures 1 and 2, dramatic changes of ^{226}Ra concentrations can occur with depth, due to variations of concentration in the feed stock processed at different times, leaching and vertical translocation of radium in addition to other factors. The net result is that high relative radon concentrations will occur at depths that lie in the direction of diffusion away from the location of high radium concentrations.

Gibbs et al.(8) provided a solution to the diffusion equation that accommodated a variable vertical source term, which describes the influence of radium within one depth interval on the radon concentrations in other depth intervals. The radon concentration $C_{(z)}$ occurring at a given depth is approximated by

$$C(z) \underset{-}{\sim} \frac{k\alpha}{2\lambda}\int_0^\infty S(z')\left[e^{-k|z-z'|} - e^{-k(z+z')}\right] dz' + (1-\alpha)S(z)/\lambda \qquad \text{Equation (3)}$$

where: α = fraction of radon available for diffusion

and $k = (\lambda/D)^{\frac{1}{2}}$

This value of D includes the porosity, or void fraction, e.g., $D = D_{eff}/v$,(8) and its use in the radon diffusion and exhalation equations (equations 1-4) is model dependent. However, using a different model may necessitate care in using the values in Table I.

Using assigned values for D and α, the predicted values of radon concentrations are calculated with the aid of a computer from the measured radium values. Best values for D and α are attained by iteration to provide the best least squares fit between the calculated and measured radon concentrations. The magnitude of the sum of squares provides an index for the reliability attained for a given data set.

Radon flux at the soil surface is described by:

$$J(z) \simeq \frac{\alpha}{2}\int_0^\infty S(z')\left[\operatorname{sgn}(z-z')e^{-k|z-z'|} - e^{-k(z+z')}\right] dz' . \qquad \text{Equation (4)}$$

4. RESULTS AND CONCLUSIONS

The radium data from two test holes depicted in Figures 1 and 2, were used for calculation of the radon diffusion coefficient and the fractional quantity of the total radon that was available for diffusion. Calculated vertical radon concentrations are compared with measured values in Figures 3 and 4. The validity of the integral solution of the diffusion equation is demonstrated by the agreement between calculated and measured radon concentrations. This provides a realistic evaluation of radon diffusion rates through soil columns in which radium is not homogeneously distributed with depth. Values of D and α derived from these calculations are given in the following table, together with other published values for these constants.

TABLE I

RADON DIFFUSION AND EMANATION COEFFICIENTS

Medium	Moisture Content, %	D cm^2/sec	α	Reference
U-tailings				
Hole A	5	0.0017	0.49	This work
Hole B	15	0.0002	0.24	This work
U-tailings		0.05	0.2	(9)
U-tailings	dry	0.004	0.2	(12)
Soil			0.1	(7)
Sandstone			0.03-0.12	(10)
Fine Quartz	0	0.068		(9)
" "	8	0.050		
" "	15	0.010		
" "	17	0.005		
Sand		0.07		(11)

The diffusable fraction of the total radon present in this tailings material was about four times higher than normally is found for soil and at least two times greater than values commonly applied to tailings material. Two factors are probably significant contributors. First, the ore for this plant is crushed and ground, and the test locations reported have received only the fines from this treatment. Thus the surface-to-volume ratio is greatly enhanced. The second point for consideration is the very nature of the ore itself as the uranium mineralization occurs on the surface of the sandstone matrix resulting in a dominantly surficial source of the radon precursor, with an enhanced probability of radon escapement to interstitial pores upon formation.

The other salient point afforded by these data is that the measured bulk diffusion coefficient for this tailings material was significantly lower than has been used for past calculations of radon diffusion and emanation from tailings material. This is quite probably a reflection of the small particle size and moisture content of the particular soil columns involved. Definitive explanation of this reduced diffusion is beyond the scope of this paper.

Radon fluxes calculated according to Equation 4, were 275 and 60 atoms/cm^2/sec for test holes A and B, respectively. Clements et al.(12) reported a mean flux of about 1000 atoms/cm^2/sec for dry alkaline leach tailings that were 2.5 m deep. The reduced rate of exhalation reported here again emphasized the effect of soil moisture on radon escapement shown by others.(9) Operating uranium mills maintain their tailings piles in a wet or moist condition, thereby reducing the quantities of radon escaping to the atmosphere, a fact that must be considered in assessment of radiological impact from these facilities.

In summary, the method of in-situ determination of the vertical concentration distributions of radium and radon progeny affords an accurate means for assessing the rates of emission of radon from a tailings pile to atmosphere. By employing a solution of the diffusion equation that accommodates the changing concentrations of ^{226}Ra that occur in tailings material, errors arising from assumption of an average radium distribution are not encountered. This method essentially evaluates the magnitude and extent of radon diffusion that has occurred at the measurement location for several days preceding the measurement, and thus tends to smooth out short-term variations resulting from external forces such as changing barometric pressure. It is theoretically possible that short term fluctuations of time scales of an hour or so can be evaluated by evaluating the ^{214}Bi concentrations relative to those of ^{214}Pb and ^{226}Ra at specific depths within the soil column.

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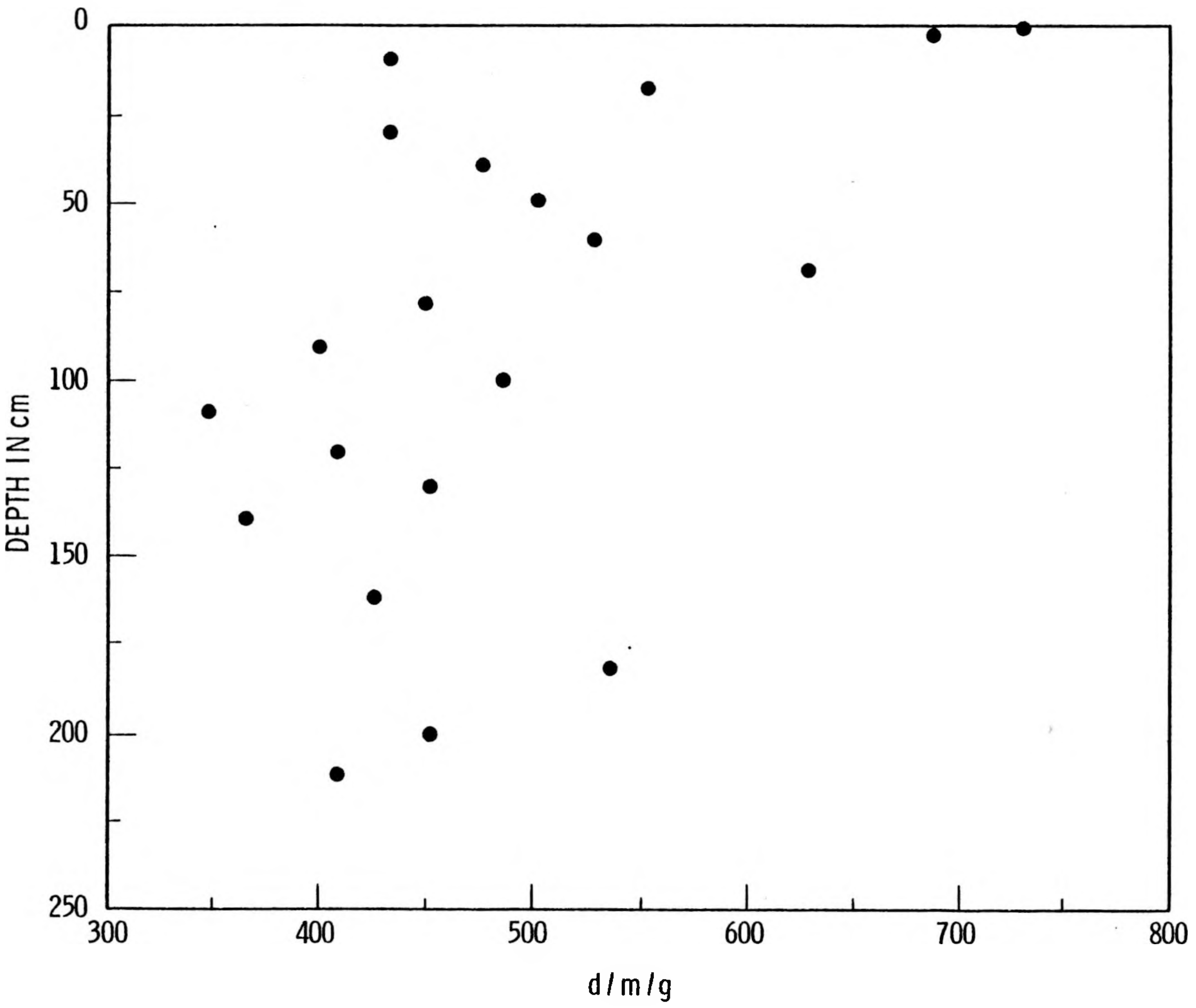

Figure 1. Radium-226 profile test hole A

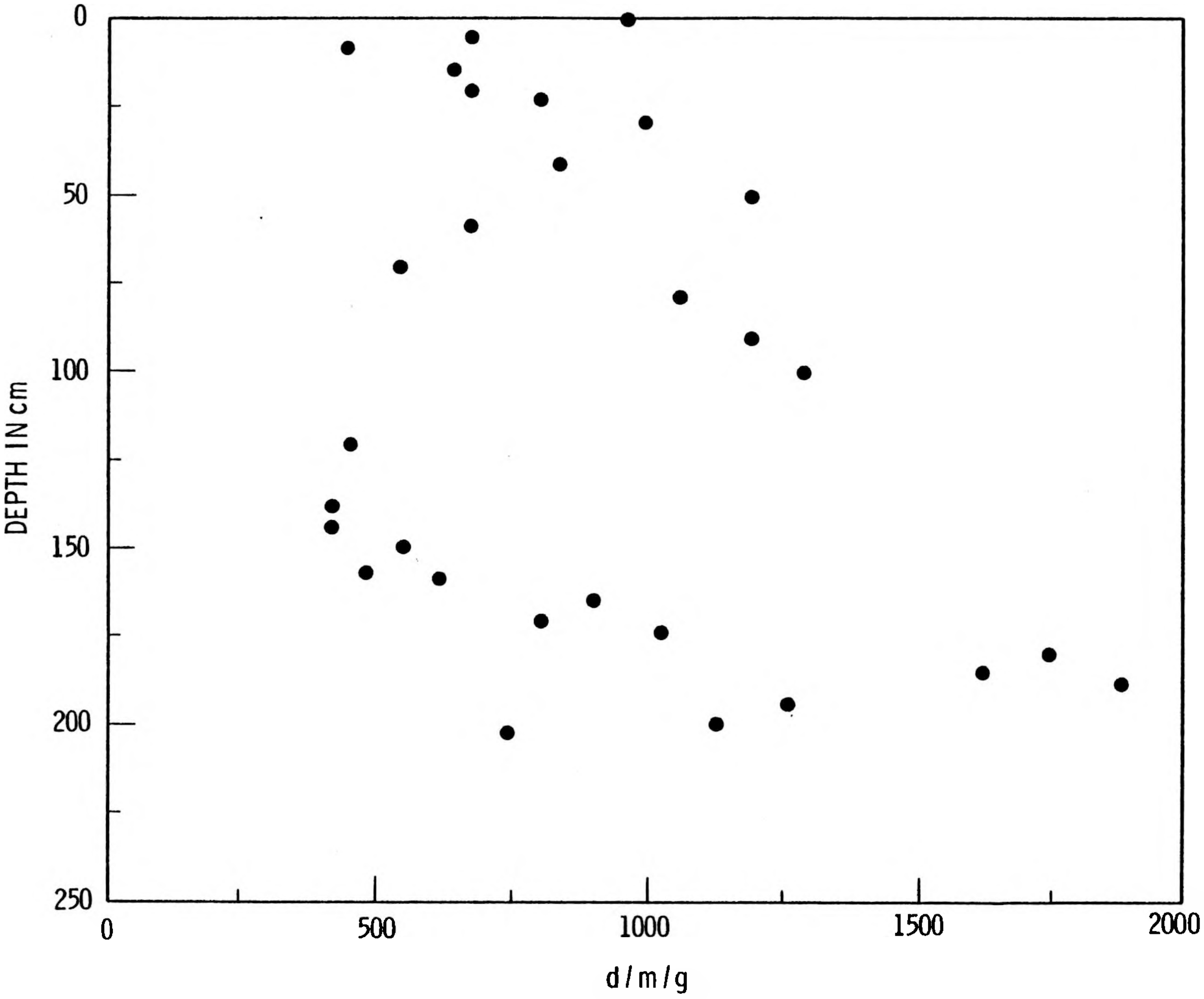

Figure 2. Radium-226 profile test hole B

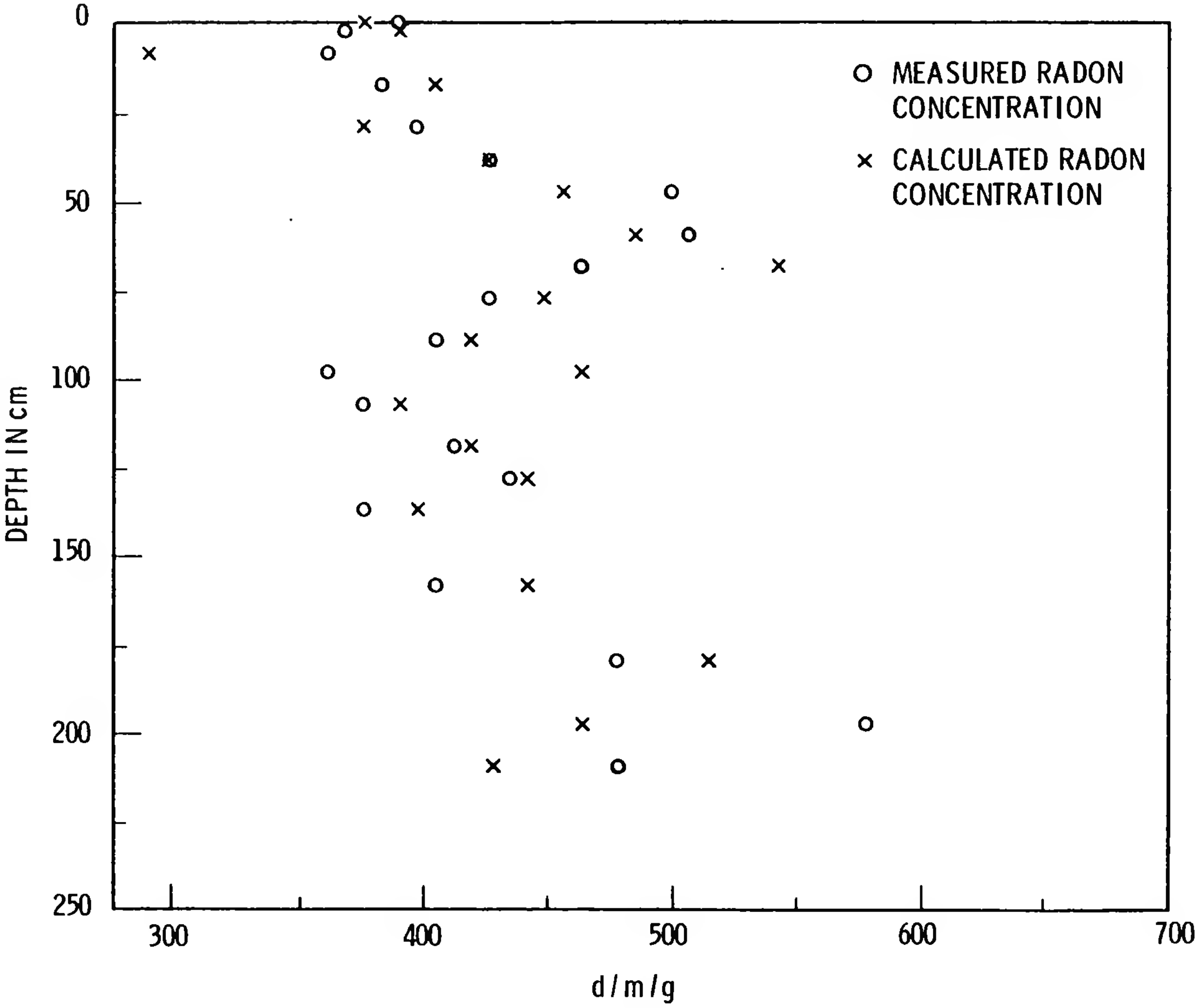

Figure 3. Radon-222 profile test hole A

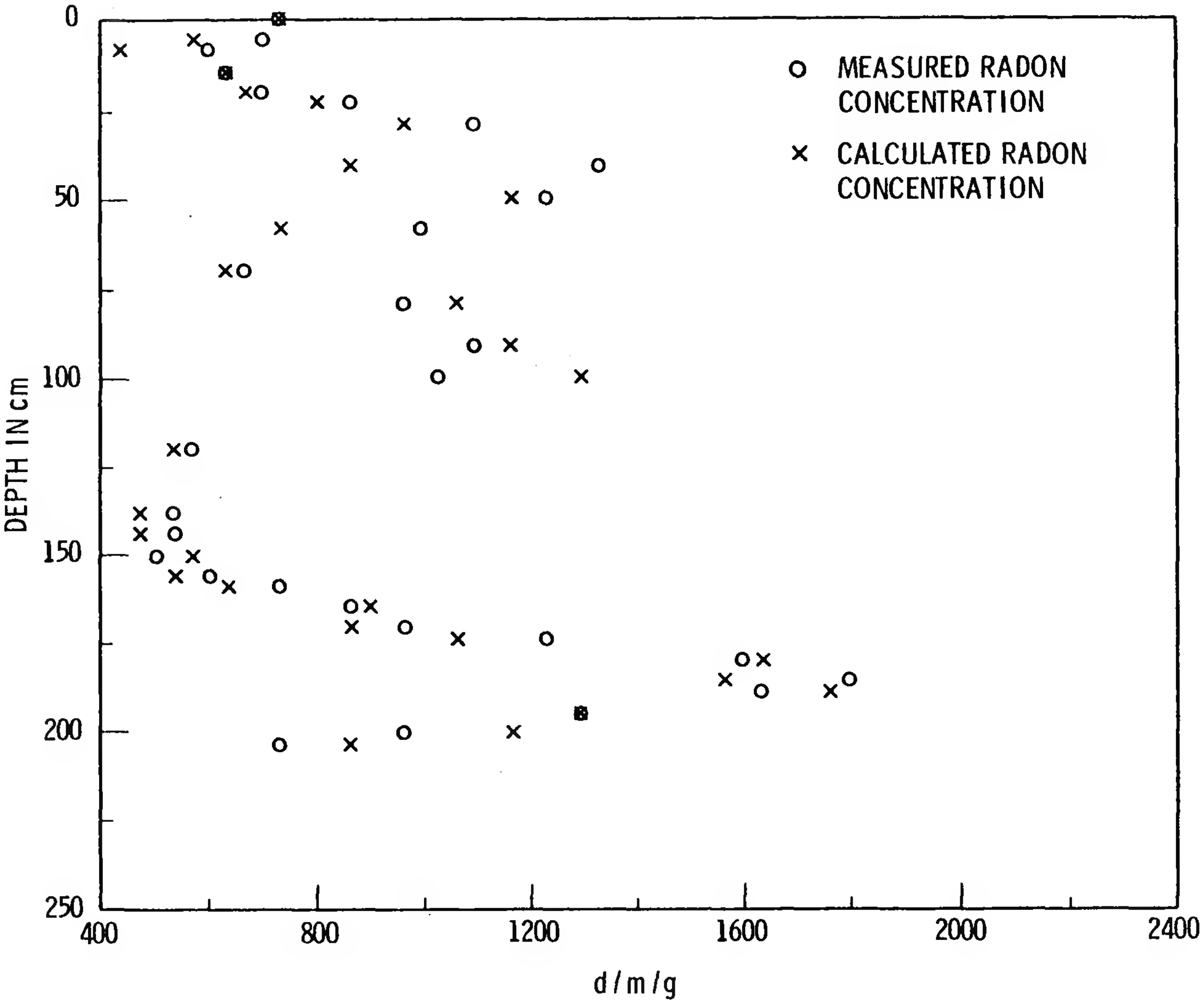

Figure 4. Radon-222 profile test hole B

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