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RADON EMANATION AND TRANSPORT IN POROUS MEDIA

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ABSTRACT

A unified model of radon emanation and transport has been developed that combines the RAECOM model for diffusive transport with new mathematical models of advective transport, moisture effects, and radon emanation. The model accounts for advective depletion in radon source regions, and for the effects of varying moistures on radon emanation, diffusion, and advective transport rates. Radon transport in gas- and water-filled pore space is characterized, and exchange between the phases is considered. Correlations are also given for diffusion and permeability coefficients. The model provides a comprehensive assessment of source potentials for indoor radon accumulation based on soil moistures, radium, emanation, and advection of soil gas.

INTRODUCTION

Radon emanation and transport in soils is illustrated schematically in Figure 1. Radon is emanated from radium-bearing minerals into the soil pore space, followed by diffusive and advective transport of the radon gas in both liquid and gas phases into a home, entering via cracks, sumps, porous building materials, water, and other routes. Advection becomes dominant with proximity to many home entry points, and exchange between air and water phases is continual.

Rogers and Associates Engineering Corporation (RAE) is performing a project for the Department of Energy to coordinate the eight key components of radon emanation and transport shown in Figure 1 into a unified, simplified model.

Previous RAE theoretial development on radon emanation, diffusion and advection in porous soils include a radon emanation computer model (1), diffusion coefficient model (2,3), and radon diffusion equations with analytical solutions for two-phase diffusion through porous materials (4). Although the two-phase diffusion model was restrictive and only treated the radon interactions in the water-air pore spaces, it has been used extensively in predicting radon migration through earthen materials (5,6) and for reducing diffusion and emanation coefficient data from laboratory measurements (7,8). The model was also extended to include both constant and cyclical advection (9).

In addition to the theoretical developments, RAE has developed several experimental procedures for determining the values of key input parameters (7,8) and has amassed an extensive comprehensive data base for a wide range of soils. This data base has been augmented and used extensively in the work associated with this project.

Theoretical developments for radon transport are presented in the next section, followed by the results of efforts to predict appropriate values for the diffusion, permeability, and emanation coefficients used in the model.

RADON EMANATION AND DIFFUSION IN A TWO-PHASE MEDIUM

Radon emanation and diffusion through a two-phase medium in the pore spaces of earthen materials involves several complex processes. There is diffusion through the air in the presence of a water layer. As shown in Figure 2, there is radon diffusion through the water in water-blocked pore regions, and there is radium and radon absorption in the water, as well as other mechanisms. The general objective of the present effort is to characterize these mechanisms which occur on a microscopic scale by a simple diffusion-type equation which can be used to describe these processes as if occurring through a simple homogeneous medium characteristic of the porous material.

It has been shown previously that radon diffusion through a porous material with moisture can be characterized by the standard diffusion equation in which the radon concentration is a pore average, and the emanation and diffusion coefficients are also appropriate pore averages (4). In order to examine the distinct mechanisms in more detail, rate balances have been constructed separately for the radon in the gas and liquid components of the pore space. These coupled equations, for the one-dimensional, steady-state condition, are:

$$D_a \frac{d^2C_a}{dx^2} - \lambda C_a + \frac{R\,\rho\,\lambda}{P(1-m)} [E_{air} - \frac{mp}{K_d\,\rho}] + T_{wa} = 0 \qquad (1)$$

$$D_w \frac{d^2C_w}{dx^2} - \lambda C_w + \frac{R\,\rho\,\lambda}{pm} [E_{water} - \frac{m(1-p)}{K_d\,\rho}] + T_{aw} = 0 \qquad (2)$$

where

D_a = radon diffusion coefficient in air, including the tortuosity factor

D_w = radon diffusion coefficient in water

C_a = radon concentration in the air-filled pore space

C_w = radon concentration in the water-filled pore space

λ = radon decay constant

R = radium concentration in the solid matrix

ρ = bulk dry density

p = total porosity

m = fraction of moisture saturation

E_{air} = component of emanation coefficient that is a direct pore air source of radon

E_{water} = Component of emanation coefficient that is a direct pore water source of radon

K_d = equilibrium distribution coefficient for radium in solid-to-pore-liquid

T_{wa} = transfer factor of radon from pore liquid to pore air

T_{aw} = transfer factor of radon from pore air to pore liquid

The second term in the brackets with E_{air} accounts for radon production in the water from radium dissolved in the water. The expression has a negative sign preceding it because measurements of R include the radium dissolved in pore water.

It follows, for negligible radon decay in the pore liquid,

$$T_{wa} = \frac{m}{1-m} T_{aw} \qquad (3)$$

and

$$T_{aw} = -\frac{R \rho \lambda}{pm} \left[E_{water} - \frac{m(1-p)}{K_d \rho}\right] \qquad (4)$$

The E_{air} and E_{water} components of the emanation coefficient are shown in Figure 3 and are given by

$$E_{water} = \frac{m}{m^*} E_w \qquad \text{for } m < m^* \tag{5}$$

$$= E_w \qquad \text{for } m \geq m^*$$

$$E_{air} = E_a (1 - m/m^*) \qquad \text{for } m < m^* \tag{6}$$

$$= 0 \qquad \text{for } m \geq m^*$$

where, m^*, E_a and E_w are constants of the emanation coefficient model (2). The term E_a is the emanation coefficient for no moisture, E_w is the emanation coefficient at saturation, and the plateau in the E curve starts at m^*.

Combining Equations (1) and (2), using Equations (3) and (4) and including advection in the gas phase, yields:

$$D \frac{dC_a}{dx^2} - K' \frac{dP}{dx} \frac{dC_a}{dx} - \lambda C_a + \frac{R \rho \lambda E'}{p} = 0 \tag{7}$$

where

D = pore average diffusion coefficient

$$K' = \frac{K}{(1-m)pu} \tag{8}$$

K = pore gas permeability

u = pore gas viscocity

P = pore gas pressure

$$E' = [E - m(1-P)/K_d \rho] / (1 - m + mk) \tag{9}$$

k = radon equilibrium distribution coefficient, water-to-air.

For no advection, Equation (7) is equal to the standard radon diffusion equation, using the pore average D and an adjusted emanation coefficient that decouples Equations (1) and (2). Consequently, standard measurements (7,8) and theoretical models (1,2) of D and E can be used with the new formulation, and previous analytical and numerical radon diffusion solutions are applicable. The computer code RAECOM (5,6) has been adapted to solve Equation (7) including advection, where the layer thicknesses are selected for constant (dP/dx). The development is readily extended to multi-dimensional and time-dependent formalisms.

DIFFUSION, PERMEABILITY, AND EMANATION COEFFICIENTS

Site-specific measurements of D and K are most desirable for obtaining input data for the radon transport models. RAE has accumulated an extensive soil data base including D and K measured values as well as values of many other nuclear, physical, and geological parameters. In addition, models have been developed to calculate D and K from basic data and first principles. The random-pore combination model that was developed earlier (2,3) was modified and extended to also include the calculation of soil permeabilities in a similar manner.

In the K calculations, soils were modeled to contain log-normal size distributions of cylindrical pores with uniform surface water films that could vary from dryness to saturation. Contiguous pore space contained all possible random combinations of the pore sizes, both in series and parallel configurations. Individual cylindrical pore permeabilities were computed from the radii (r) of the air-filled pore spaces as $r^2/8$, following the functional size-dependence used by Youngquist (10). The resulting permeabilities of individual pore segments were combined for the entire soil in the same way as pore diffusion coefficients were previously combined to estimate bulk soil radon diffusion coefficients. The resulting permeability calculations are illustrated by the black dots in Figure 4. Gas permeabilities varied slowly with moisture, increased rapidly with the geometric standard deviation of the grain-size distribution, and increased as the square of the average particle diameter.

In addition to field measurement, laboratory measurements or complex model calculations of D and K, empirical correlations have also been used. These correlations have the advantage of being simple and easy to use, with a minimum amount of information needed. One correlation frequently used for D is (5)

$$D = 0.07 \exp[-4 (m - mp^2 + m^5)] \tag{10}$$

Since that correlation was developed, RAE has measured over 1500 additional D measurements for the Department of Energy's Uranium Mill Tailings Remedial Action Program. Most of these D's were measured at high compactions. Consequently, Equation (10) has been modified as follows to include the effects of high compactions:

$$D = \frac{3p(1+p)d}{8(2+\pi d)} \exp[- \frac{7P(1+P)d}{2 + d} m - 7m^5] \tag{11}$$

where

d = geometric mean particle diameter (um).

A portion of the D values in the data base are plotted in Figure 5, along with correlation curves for a low compaction, large-grained material and a high-compaction, small-grained material. The higher compaction reduces the value of D at low moistures.

A similar correlation has been developed for soil permeabilities. It is

$$K = P\,(1+P)(d'/3)^2 \exp[-\,1.4m - 2m^5] \tag{12}$$

where

d' = geometric mean particle diameter (cm)
= 0.0001 d.

Equation (12) applies to soils whose particle distribution can be approximated by a log-normal distribution with a geometric standard deviation, S, of ten.

For other values of S, Equation (12) must be multiplied by

$$0.0034 \exp[0.264\ S^{4/3} - 0.6m\ (1 - 0.1S)] \tag{13}$$

The correlation values for K are also plotted in Figure 4 for several values of d'. Curves are given for an S of ten and of five.

Possible anisotropy of radon diffusion coefficients of soils was examined by conducting laboratory diffusion measurements on soils in both the direction of the compactive force and perpendicular to the compactive force. Six different soils were prepared by compacting to 95 percent of standard Proctor density, and then pressing a 10-cm diameter thin-walled diffusion tube into the soil either parallel or perpendicular to the direction of compaction. Eighteen time-dependent radon diffusion measurements (7) on the resulting soil cylinders gave an average diffusion coefficient ratio (perpendicular/parallel) of 1.2 ± 0.3, indicating only a marginal increase in diffusion along the bedding plane. This could still be greater in some undisturbed soils, but is probably insignificant in re-compacted soils in construction excavations.

Over 50 similar laboratory air-permeability measurements were conducted on five of the same soil samples. The average ratio of air-permeabilities (perpendicular/parallel to compaction) was 6 ± 6, indicating a somewhat more significant increase in permeability along the bedding plane. This ratio also may be greater in undisturbed soils, but may be important even in some re-compacted soils.

Emanation coefficients have been measured for several different soil types, because the RAE data base for E consisted mainly of values for uranium ores, uranium mill tailings, and phosphogypsum. The new E values were measured on soils documented in the data base to facilitate further development of the emanation coefficient model (1). The new values are shown in Figure 6. The fit to a relatively narrow normal distribution is encouraging, especially considering the widely diverse soils used for the measurements. These data indicate that a best-estimate default value of E for normal soils is 0.22, if site-specific data are not available.

SUMMARY

A simplified model has been developed for radon emanation and transport through a two-phase medium in the pore spaces of earthen materials. The model explicitely considers radon emanation into the water and the air pore regions, radon diffusion through the pore water and air regions, radon and radium absorption in the water, radon transfer between the pore water and air, and advection in the air region. The one-dimensional steady-state formalism is presented and has been encoded into the RAECOM computer code.

The DIFPERM code for calculating diffusion and permeability coefficients from basic principles is described and new correlations for D and K are presented. It is also shown that horizontal permeabilities are slightly greater than vertical permeabilities, but the difference is not as evident in the D measurements. Several new E measurements have also been made for a wide variety of soils. The geometric mean of the E measurements is 0.22.

ACKNOWLEDGMENT

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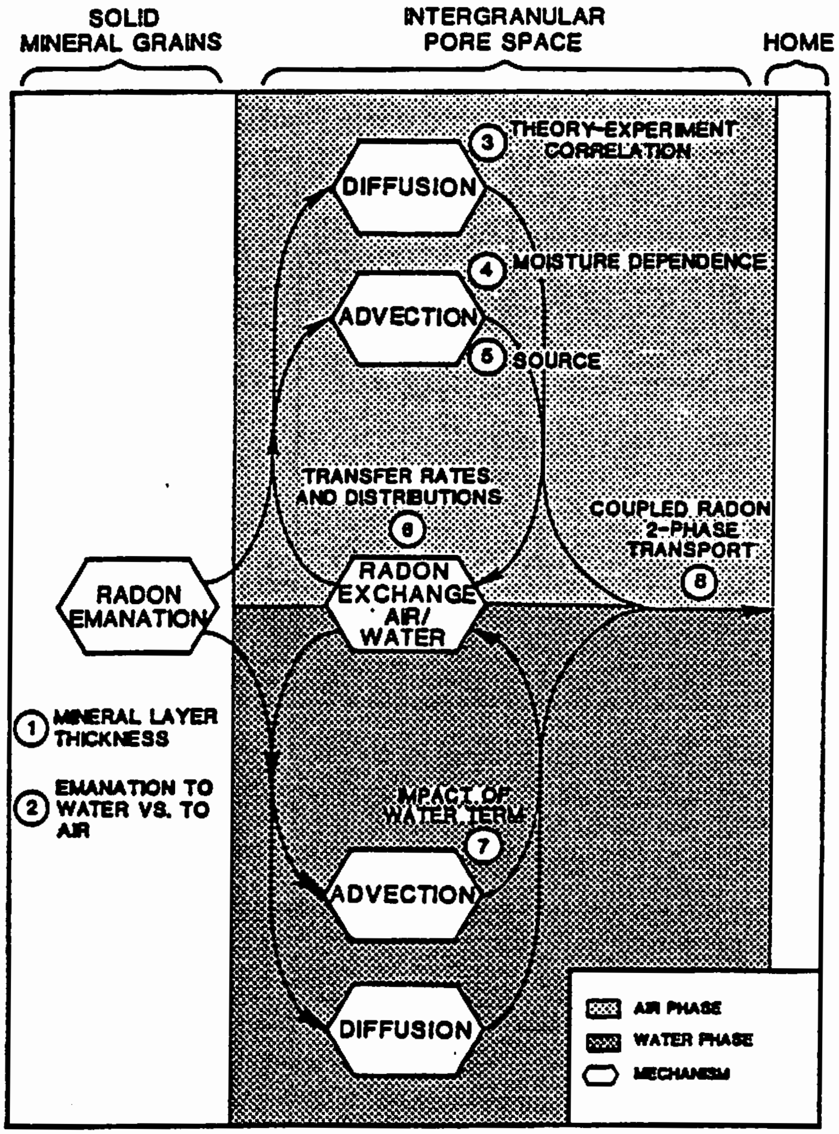

FIGURE 1. Physical mechanisms of radon generation and transport.

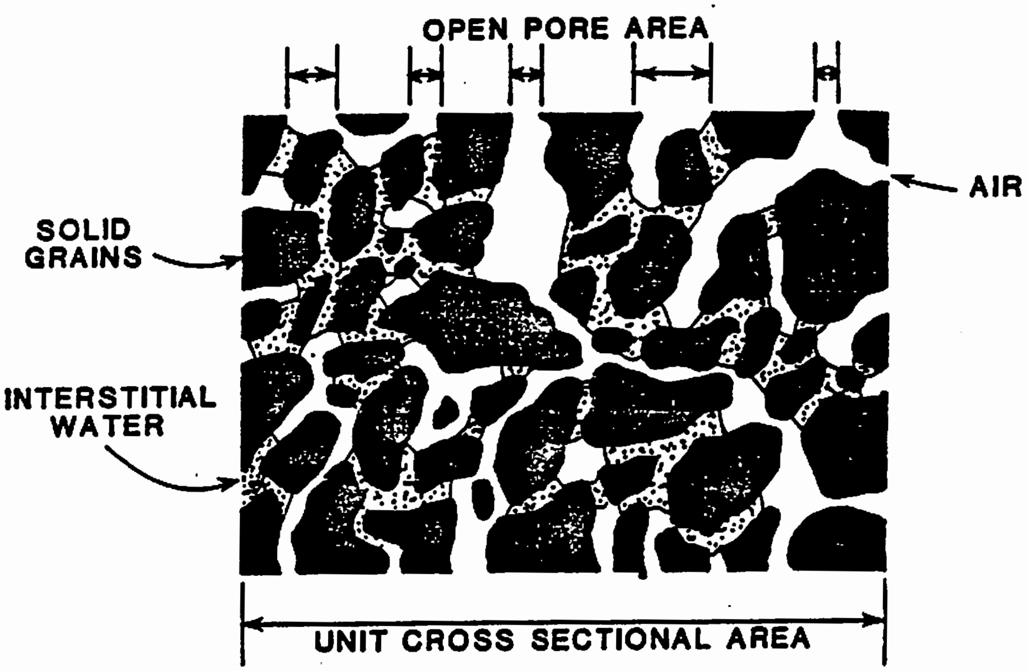

FIGURE 2. Volume of unsaturated porous material radium is in the solid grains and interstitial water.

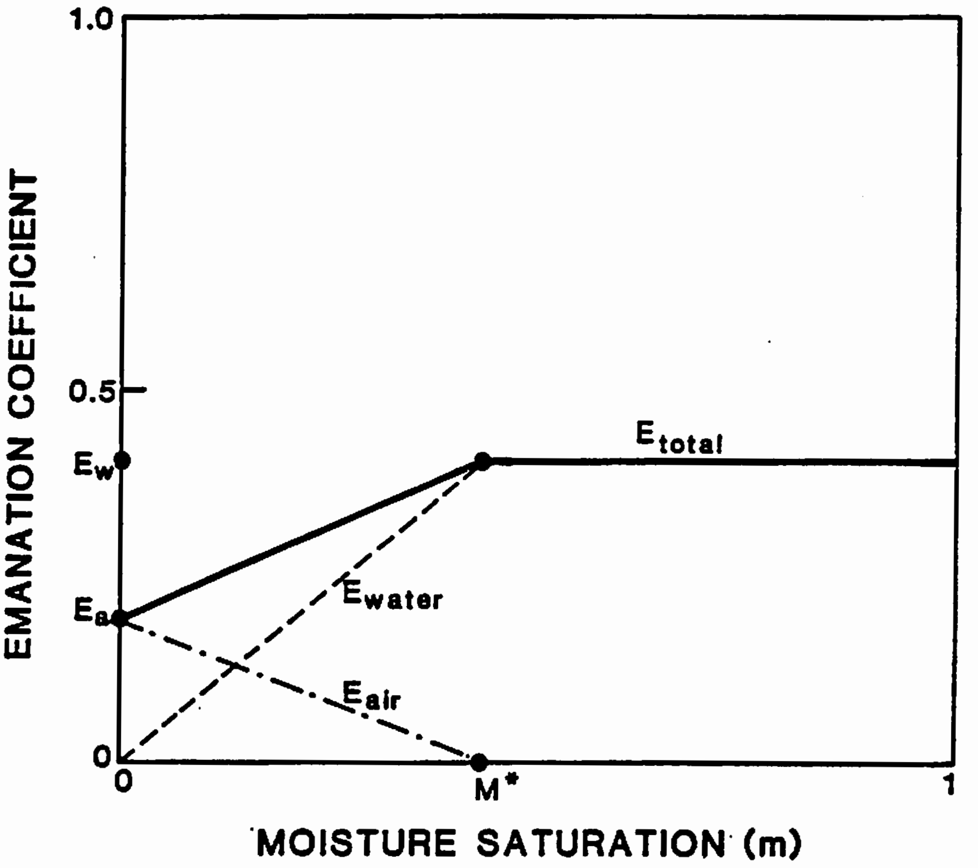

FIGURE 3. Components of the emanation coefficient as a function of moisture.

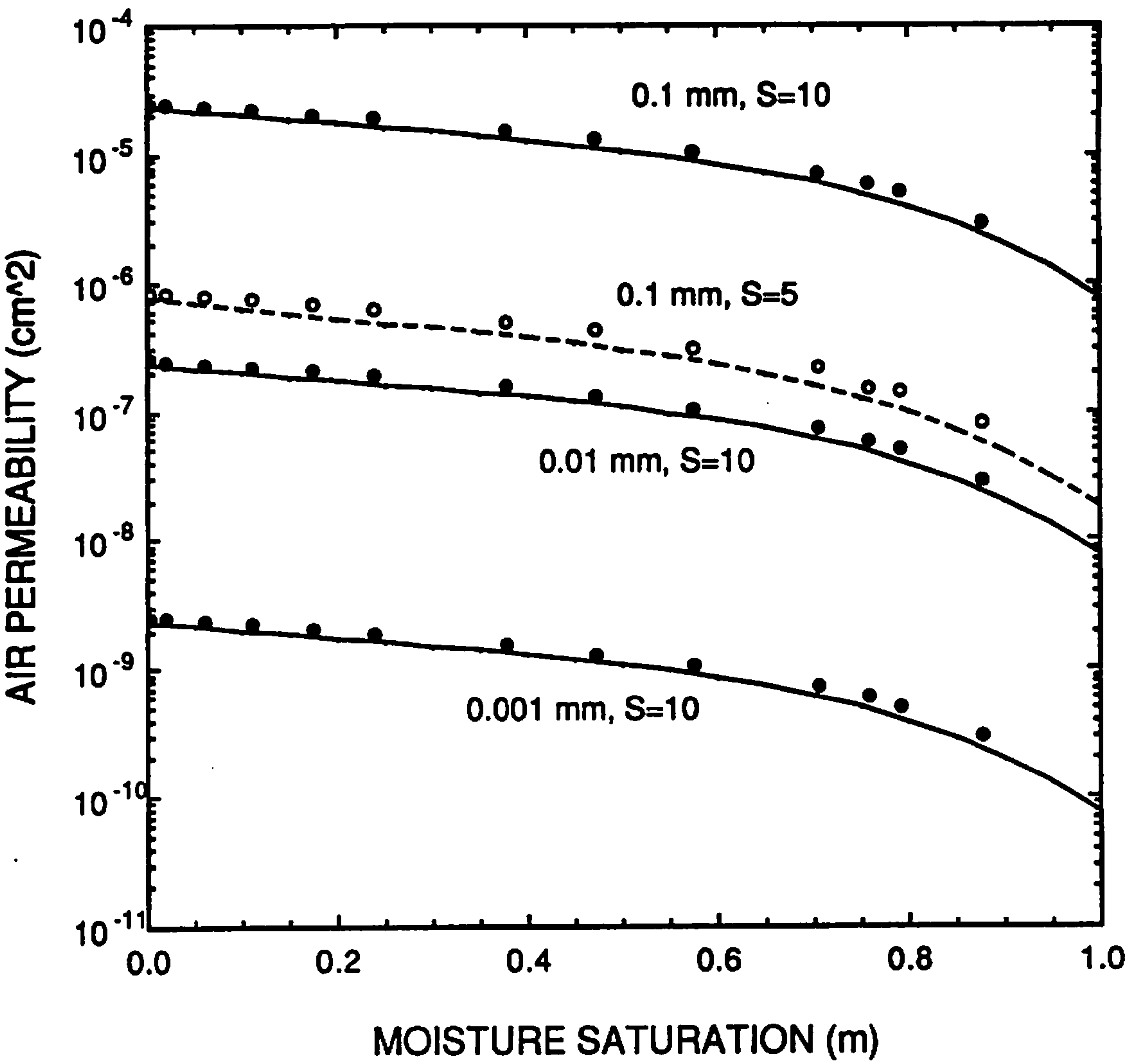

Figure 4. Air permeability for various soil types. Data points are model calculations. Curves are correlation values. Geometric mean particle diameters, and geometric standard deviations, S, are given for each soil type.

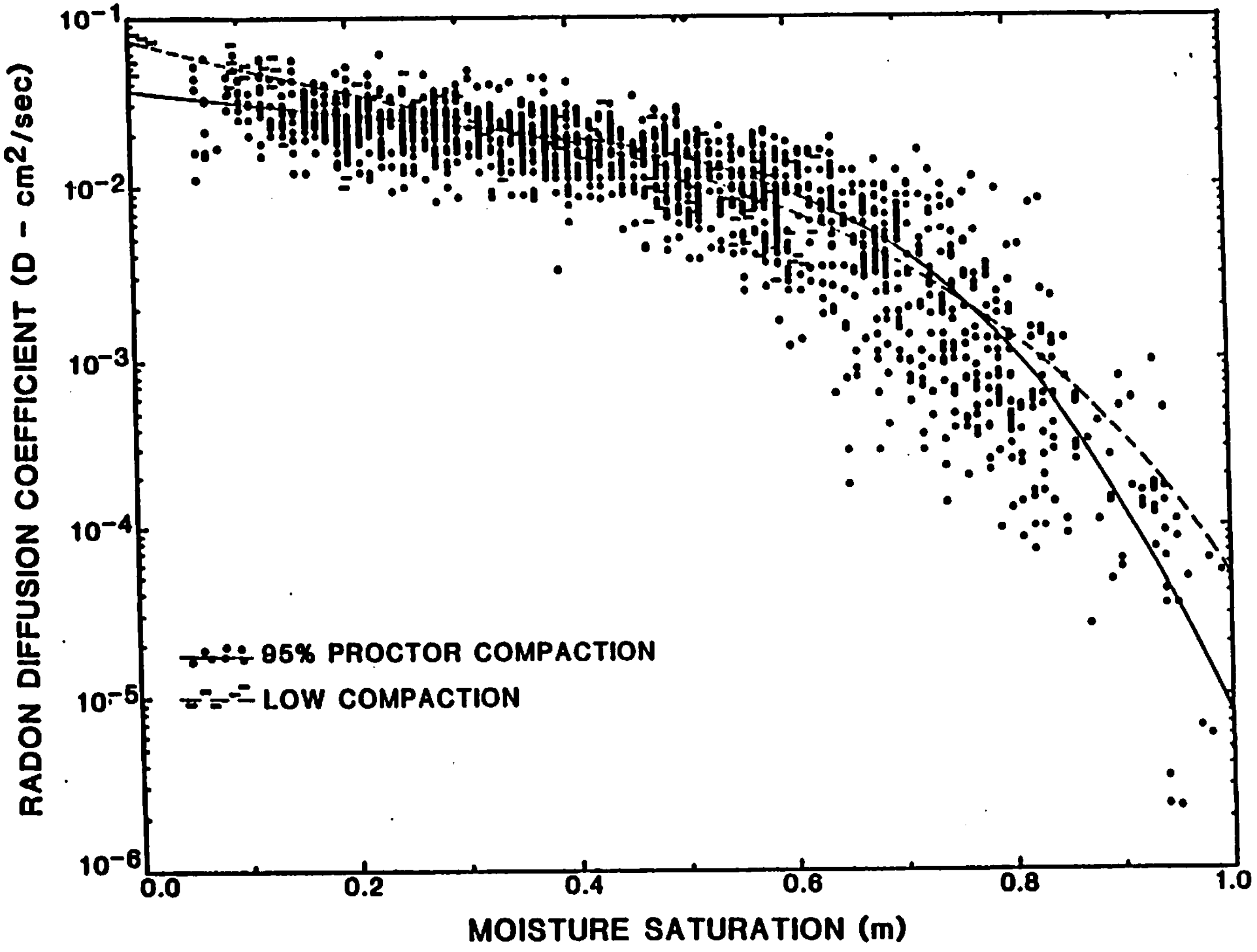

FIGURE 5. Measured radon diffusion coefficient and correlation predictions.

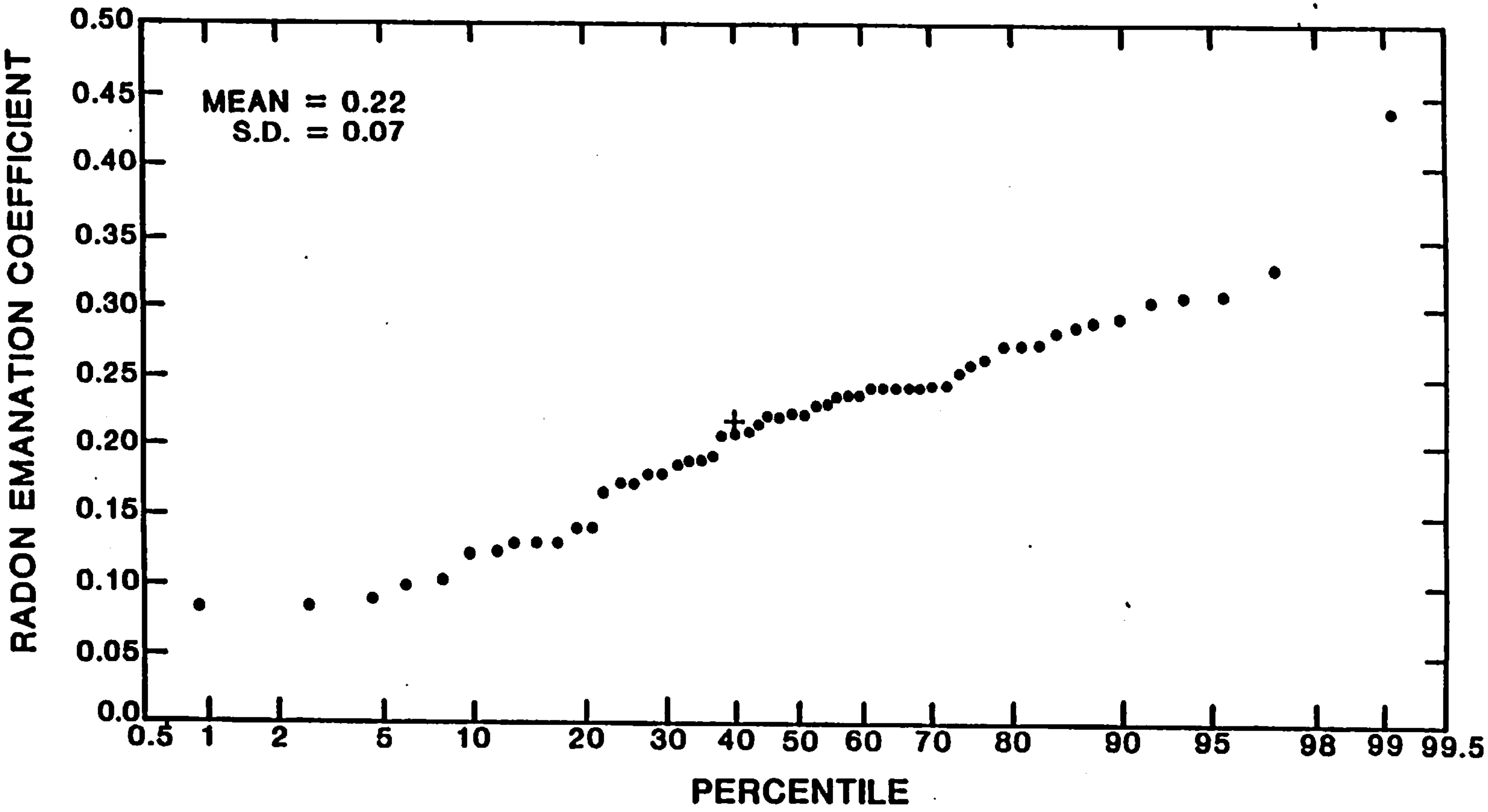

FIGURE 6. Probability plot of emanation coefficients for 56 soils.