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RADON TRANSPORT FROM THE SUBSURFACE: THE ROLES OF CERTAIN BOUNDARY CONDITIONS AT SUBSURFACE/ENVIRONMENT BOUNDARIES

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ABSTRACT

The effects wind on radon transport from its soil source to the environment are examined in two situations. In the first situation, the removal of radon from the soil-air interface was found to be partially rate limiting in conditions of nearly stagnant air (low wind). This gas-phase mass transfer limitation became especially important when high exhaling advective velocities or high diffusion fluxes to the air existed. A detailed mathematical formulation for one-dimensional steady-state radon transport from the soil to the air using an air side mass transfer coefficient was developed for this analysis.

In the second situation, the Rn3D computer code was used to estimate radon concentration profiles in soils beneath a two-dimensional slab-on-grade dwelling subjected to wind pressures. Of five generic dry homogeneous soils studied (gravel, sand, silt, loam and clay), only gravel showed significant changes in subslab concentrations as a result of wind pressures.

INTRODUCTION

This analysis is part of a larger study performed by the Pacific Northwest Laboratory (PNL) for the U.S. Department of Energy/Office of Health

and Environmental Research. The primary objectives of the larger study are to develop, evaluate, and apply a comprehensive model of radon transport within and from soils to the environment and structures.

In this paper, we examine two situations where atmospheric variables might affect the flux of radon from the subsurface to the environment. The atmospheric variables in question are surface winds and surface-induced pressure gradients that can exist both vertically within the soil and horizontally along the soil/environment interface. The two situations expressed as questions are: 1) Does the wind affect the transport of radon across soil/air boundaries, such as on open plains or in building crawl spaces? 2) Does the wind affect the concentration profiles at soil/building interfaces, and thus affect the amount of radon available for transport into the building? These questions are answered quantitatively with a one-dimensional analysis using analytical solutions to soil radon transport for the first question and a two-dimensional analysis using the the Rn3D code for the second question. The answers to these questions should help prevent some modeling pitfalls and experimental oversights when studying radon transport from soils.

ONE-DIMENSIONAL TRANSPORT OF RADON ACROSS SOIL/AIR BOUNDARIES

Previous modeling studies at PNL (Holford, et al. 1988, 1989; Owczarski, et al. 1990) have frequently examined the parameters controlling the flux of radon from soil to atmosphere. In these studies we have assumed that the soil/air boundary condition was type one where the radon surface concentration is specified; we specified zero concentration of radon at the soil surface. Specifying the surface concentration ignores the resistance to radon transport

in the gas phase above the surface. Technical details of the limitations of type one boundary condition where the effects of wind are ignored now follow.

The specific limitation of the type one boundary condition is that the radon transporting to the soil/air surface might build up to a sufficiently high level to make the zero concentration assumption invalid. Clearly this buildup can only happen if the transport processes (e.g., turbulent eddies and molecular diffusion) on the air side cannot remove the radon as fast as it flows into the boundary region. Transport processes that remove radon are a combination of molecular diffusion and turbulent eddies in the air. To examine these processes, we constructed a simple one-dimensional model of radon transport to the surface and subsequent removal of the radon by the wind using a type three boundary condition (defined below) where the radon transport resistance in the gas phase is included. We assumed steady state and ignored the dynamic nature of the wind, which, even when steady, produces rapid fluctuations in surface pressures that penetrate the soil and affect instantaneous surface fluxes. Our assumptions do not allow the wind to create pressure gradients within the soil.

The usual differential equation and boundary conditions for steady-state transport in homogeneous soils are

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial z^2} + \frac{V_z}{n} \frac{\partial c}{\partial z} - \lambda c + \phi = 0 \qquad (1)$$

Boundary conditions

a. $\frac{dc}{dz} \to 0$ as $z \to -\infty$

b. Type one or type three at $z = 0$.

where c = Rn concentration in soil gas, Bq/m^3

n = soil porosity

λ = Rn decay constant, $2.1 \times 10^{-6}/s$

Φ = Rn generation rate, $Bq/m^3/s$

D = Rn diffusivity in soil, m^2/s

V_z = superficial soil gas velocity, m/s

t = time, s

z = vertical distance into soil, m.

The type one boundary condition is expressed as

$$c(z=0) = c_0 = 0 \text{ or } c_a \text{ (ambient)}. \tag{2}$$

The type three boundary condition as used in this paper is defined as

$$\text{surface Rn flux} = k(c_0 - c_a)$$

where c_0 = surface Rn concentration

c_a = ambient Rn concentration

k = mass transfer coefficient, m/s.

The surface Rn flux is further defined as the sum of soil diffusive and advective fluxes at the surface, i.e.,

$$-D \left.\frac{dc}{dz}\right|_{z=0} + V_z c_0 = k(c_0 - c_a) \tag{3}$$

where $V_z = -\frac{\kappa}{\mu}\frac{dP}{dz}$

$\frac{dP}{dz}$ = soil pressure gradient, Pa/m

κ = soil permeability, m^2

μ = soil gas viscosity, kg/m/s.

The steady-state solutions of Equation (1) for soil Rn concentration and surface flux have been derived previously (Clements 1974) for the type one boundary condition:

$$\frac{c - c_a}{c_\infty - c_a} = 1 - \exp\left\{\left[\frac{V_z}{n} + \left(\frac{V_z^2}{n^2} + 4\lambda D\right)^{1/2}\right]\frac{z}{2D}\right\} \tag{4}$$

$$= 1 - \exp(\alpha z)$$

$$\text{flux}(z=0) = (c_\infty - c_a)\alpha D \tag{5}$$

where $c_\infty = \frac{\Phi}{\lambda}$, and V_z is constant for all z. (Note that z is negative in the soil.) The corresponding solution for the type three boundary condition is first presented here:

$$\frac{c - c_a}{c_\infty - c_a} = 1 - A\exp(\alpha z) \tag{6}$$

where $\alpha = \left[\frac{V_z}{n} + \left(\frac{V_z^2}{n^2} + 4\lambda D\right)^{1/2}\right] \Big/ 2D$

$A = \frac{1}{1 + a_1} - a_2$

$$a_1 = \frac{\left\{V_z + \left[V_z^2 + 4D\lambda n^2\right]^{1/2}\right\}}{2(k - V_z)}$$

$$a_2 = V_z \frac{c_a}{(c_\infty - c_a)} \Bigg/ \left\{\frac{V_2 + \left[V_z^2 + 4D\lambda n^2\right]^{1/2}}{2} + k - V_z\right\}$$

$$\text{flux}(z=0) = k(c_0 - c_a) = k\left[c_\infty - c_a - \frac{(c_\infty - c_a)}{(1 + a_1)} + a_2\right] \tag{7}$$

With careful algebra Equations (6) and (7) reduce to Equations (4) and (5) as k approaches infinity. This reduction is expected because a large mass transfer coefficient corresponds to rapid removal of surface radon and the resulting surface concentration is c_a. Thus, we conclude that the type three boundary condition is more general than the type one, where the latter ignores the resistance to radon transport in the gas phase above the surface.

The error in using the type one equations when type three should apply can be expressed as

$$\%\text{error} = 100\left[\frac{\text{flux (type 1)}}{\text{flux (type 3)}} - 1\right] \tag{8}$$

$$= 100\left\{\frac{n\alpha D(c_\infty - c_a)}{\dfrac{n\alpha D(c_\infty - c_a)k}{(1 + a_1)(k - V_z)} + ka_2} - 1\right\} \tag{9}$$

If the ambient concentration $c_a = 0$, Equation (9) reduces to

$$\%\text{error} = 100(V_z + n\alpha D)/k \ . \tag{10}$$

We now proceed to evaluate Equation (10) for a range of soil types, pressure gradients, and wind velocities.

First we evaluate the mass transfer coefficient or escape velocity k. A relationship for k can be derived from correlations for flow across a flat plate (Bird, Stewart and Lightfoot 1960). Here we use the expression

$$k = \bar{U} \frac{f}{2} (Sc)^{2/3} \tag{11}$$

where the Schmidt number $Sc = (\rho Da/\mu)$. Da is radon diffusivity in bulk air, ρ and μ are air density and viscosity, $\bar{U}$ is the mainstream wind velocity, and f is the friction factor. A rough surface friction factor of 0.01 is used here (this f is the turbulent flow limit in large pipes). For zero wind speed, k is diffusion-limited over a stagnant gas boundary layer, which we assume is 0.1 m thick, so k becomes $Da/0.1 = 1 \times 10^{-4}$ m/s. Table 1 shows values of k used in this paper for different wind speeds.

Table 2 lists the pertinent soil properties used in evaluating Equation (10). Five types of dry soils were used to limit the scope of this study.

Variations of the controlling parameters in Equation (10) are represented in Figures 1 and 2 and Table 3. In Figure 1 we vary the vertical soil pressure gradient and wind speed for dry sand. The other soils show similar patterns, with the lower permeability silt, clay, and loam showing lower and higher permeability gravel showing higher percent errors than sand. We note that surface flux values are always positive here (an overprediction that

results from using the type one boundary condition), and that positive pressure gradients and low wind speeds are conducive to larger errors than negative pressure gradients and high wind speeds. The overall result is that higher surface fluxes need higher escape velocities to maintain a low interface concentration as specified by a $c_a = 0$ boundary condition.

In Figure 2 we have eliminated the soil pressure gradient (effect of permeability) and have plotted percent error versus wind speed for all five soils. Again the wind speed has a similar effect as in Figure 1. However, Figure 2 shows the effect of the product $n\alpha D$, porosity times diffusivity. The higher $n\alpha D$ soils have higher percent errors at all wind speeds. Again, the higher surface flux (higher $n\alpha D$) cases need a higher escape velocity to maintain a $c_a = 0$ boundary condition. (Note: $n\alpha D$ for no pressure gradient is $n\sqrt{(\lambda D)}$.)

In Table 3 we show the effect of a stagnant condition (no wind) on the percent error. This is the condition of maximum error for all soils. The high permeability and high $n\alpha D$ soils give the highest errors. This table indicates that for stagnant conditions such as in a poorly ventilated house crawl space, the type one boundary condition with $c_a = 0$ would be a source of significant error in most soils. One method of reducing the error in both type one and type three boundary conditions would be to use the actual $c_a > 0$ if it is known, since an unventilated system would build up the radon concentration.

EFFECTS OF WIND ON RADON CONCENTRATION PROFILES BELOW A TWO-DIMENSIONAL SLAB-ON-GRADE DWELLING

To examine the effects of wind on subsurface radon concentrations around structures, we used the Rn3D code in two dimensions. This code is described in previous papers (Holford et al. 1989) and a companion paper in this symposium (Holford et al. 1990). To simplify the approach even further than the two-dimension limitation, we chose for the dwelling a simple slab-on-grade rectangle, 3 m high and 15 m wide, laid on a homogeneous dry soil (Figure 3). The same dry soils used in the previous section were used here. The radium activity was 110 Bq/kg with a radon emanation coefficient of 0.1. The following bulk dry soil densities (ρ_s, kg/m^3) were assumed: gravel 1275, sand 1934, loam 1494, silt 1583, and clay 1290. Here $\Phi = 110\ \rho_s\ (0.1)$ as in Equation (1).

As noted in Figure 3, the wind was assigned to be perpendicular to the infinite third dimension of the dwelling. A simple representation of the wind horizontal velocity was assumed, so we could calculate a surface pressure profile near the dwelling. The upstream and downstream surface pressures at the two building-soil-air points were based on actual measurements of wind pressure coefficients (Simiu and Scanlon 1978). A linear relationship for the horizontal pressure gradient between the above points along the dwelling-soil interface was assumed. Pressures as noted are in Pascals for wind velocities in m/s. A type one boundary condition with $c_a = 0$ at the air-soil interface and type two ($\left.\frac{dc}{dt}\right|_{z=0} = 0$) boundary condition under the slab were used.

Figure 4 is a plot of the radon concentrations at steady-state conditions (with $c_a = 0$ at the soil-air interface) for four wind speeds at the dwelling-dry sand interface. At zero wind the plot is symmetric, as one would expect.

As the wind speed is increased above zero, the profile becomes unsymmetric as shown. At the highest wind speed (25 m/s), the upstream half of the building-soil interface concentration is significantly depressed and only at the downstream edge is it slightly elevated over the zero wind profile. The nonsmooth shape of the profiles is due to a coarse grid space. If we average the concentration profile along the slab at 25 m/s, we find that this average is about half the zero wind average subslab concentration.

Figure 5 is a plot of this subslab average concentration versus wind speed for each of the dry soil types. Only in the case of the highly permeable gravel is the reduction of the subslab concentration significant. These results immediately raise the obvious question. Can buildings be laid over gravel to take advantage of its high permeability for passively reducing radon subslab concentrations using natural winds?

To partially answer this question we laid the above study dwelling on a gravel bed on top of 47.5% saturated clay (emanation coefficient 0.4, diffusivity 8.6×10^{-9} m^2/s, and permeability 2.8×10^{-16} m^2). Then the average subslab concentration of radon was calculated at three wind speeds for gravel bed thicknesses up to one meter thick. The calculated data are plotted in Figure 6. The surprising result is that, for all three wind speeds, the beneficial effects are realized at a bed depth of only 0.1 m (4 in.), with little benefit gained by increasing the bed depth beyond 0.1 m. The gravel (dry) had the same radium activity as that in Figure 5. For even the modest wind of 2.5 m/s, the reduction in subslab concentration was more than threefold for the 0.1-m gravel bed.

The next obvious questions are: What will an annual average wind spectrum do to the subslab concentration? Can a large leak path into the

dwelling through the slab seriously disturb the subslab concentration? The answers to these questions are forthcoming in future papers.

Another question that the reader might have at this time is: since we ran Rn3D in a steady-state mode, how long does it take for a steady-state profile to be established after a sudden change in wind velocity? For example, do wind-induced profiles have any meaning in the steady state, since the wind is highly variable? To answer these questions we now look at an advective diffusivity for each soil type and apply it to the study slab. This advective diffusivity (D_{Adv}) derives from the unsteady-state form of the flow equation and Darcy's Law:

$$D_{Adv} = P_o \kappa / 2n\mu \tag{12}$$

where P_o is the absolute pressure. Also in the flow equation a characteristic time, τ, arises if we assign some characteristic length, l, to flow path:

$$\tau = l^2 / D_{Adv} \ . \tag{13}$$

We assigned l = 7.5 m or half the dwelling width. For the soils in Table 2, we constructed Table 4. Inspection of the table shows the gravel τ = 0.0152 h or 1 min. Therefore, the steady-state concentration profiles under the study dwelling laid over gravel can be expected to be established in a few minutes for each minute-scale variation in the wind speed. This rapid response gives confidence that a steady-state analysis will suffice in analyzing wind effects

on houses laid over gravel beds. Also, with such a rapid response any hour-to-hour changes in wind direction on a given dwelling will result in a rapid adjustment in the gravel bed concentration profiles.

CONCLUSIONS

This paper has examined the effects of atmospheric variables, particularly wind, on the radon fluxes from the soil subsurface. For two distinctly different influences of the wind, one in sweeping radon away from soil-air surfaces and the other in causing pressure gradients around dwellings, we have arrived at the following conclusions:

- Wind can affect radon fluxes from soils.
- Representation of the soil/air boundary with a type one boundary condition is frequently adequate.
- The type one boundary condition can be inadequate for stagnant conditions and for high permeability soils that experience strong pressure gradients.
- The type three boundary condition can provide a suitable alternative to the type one boundary condition when the latter is unsuitable.
- The suitability of the type one condition can be estimated with the dimensionless number $(V_z + n\alpha D)/k$ if $c_a = 0$.
- Estimating the type three mass transfer coefficient is not always straightforward.
- Wind pressures on dwellings can alter the radon concentration profiles beneath the dwelling, especially if highly permeable soils surround the dwelling.

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Owczarski, P. C, D. J. Holford, H. D. Freeman, and G. W. Gee. 1990. "Effects of Changing Water Content and Atmospheric Pressure on Radon Flux from Surfaces of Five Soil Types." Geophysical Research Letters, 17(6):817-820.

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TABLE 1. Typical k Values Used In Evaluating Equation 10

$\bar{U}$, m/s	$\bar{U}$, mph	k, m/s
0	0	1×10^{-4}
1	2.24	4.38×10^{-3}
10	22.4	4.38×10^{-2}
25	55.9	0.11

TABLE 2. Dry Soil Properties Used in Evaluating Equation 10 (Owczarski et al. 1990)

Soil Type	Porosity	Permeability, m^2	Diffusivity, m^2/s
Clay	0.5131	1.0×10^{-15}	1.98×10^{-8}
Silt	0.4026	1.5×10^{-14}	2.57×10^{-6}
Loam	0.4362	2.0×10^{-13}	6.89×10^{-6}
Sand	0.27	3.4×10^{-12}	7.10×10^{-6}
Gravel	0.519	1.9×10^{-9}	7.72×10^{-6}

TABLE 3. Percent Error in Radon Surface Flux at Zero Wind

Soil Type	$\frac{dP}{dz}$(Pa/m) -50	-5.0	-0.5	0	0.5	5.0	50
Gravel	5300	530	53	2.1	0.083	0.0083	0.00077
Sand	9.6	1.6	1.1	1.0	1.0	0.67	0.11
Loam	2.0	1.7	1.7	1.7	1.7	1.6	1.4
Silt	0.96	0.94	0.93	0.93	0.93	0.93	0.91
Clay	0.11	0.10	0.10	0.10	0.10	0.10	0.10

TABLE 4. Values of Soil Advective Diffusivities and Characteristic Time for the Dwelling of Figure 3

Soil Type	D_{Adv}, m^2/s	τ, h
Gravel	1.030×10^{1}	0.0152
Sand	3.544×10^{-3}	4.41
Loam	1.291×10^{-4}	121
Silt	1.049×10^{-5}	1,490
Clay	5.485×10^{-7}	28,500

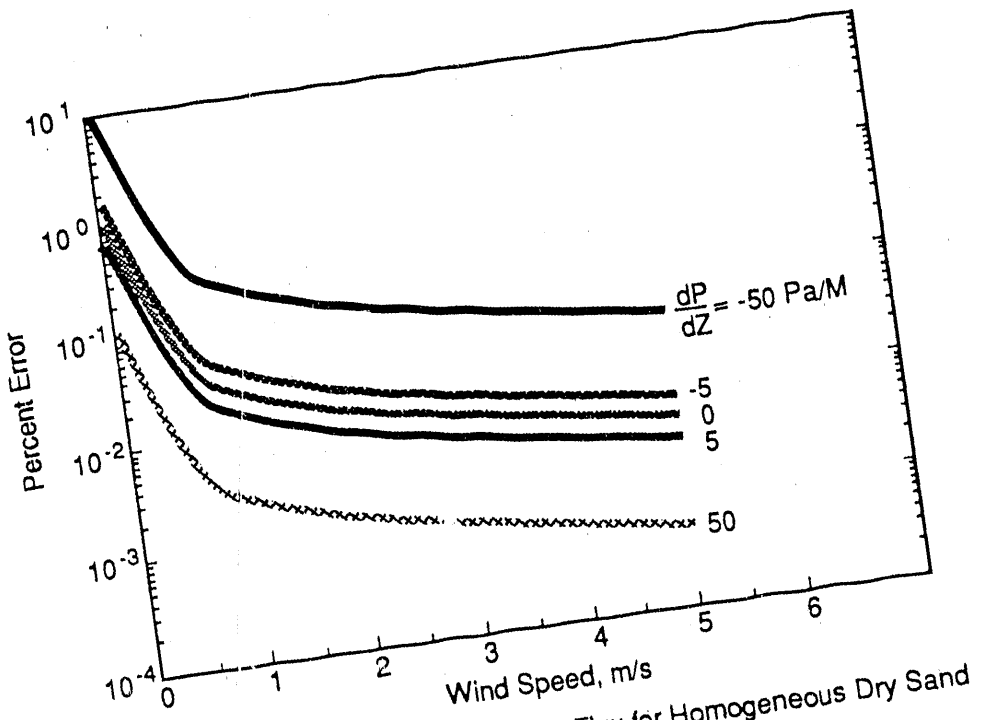

Figure 1. Percent Error in Radon Surface Flux for Homogeneous Dry Sand Using Type One Boundary Condition

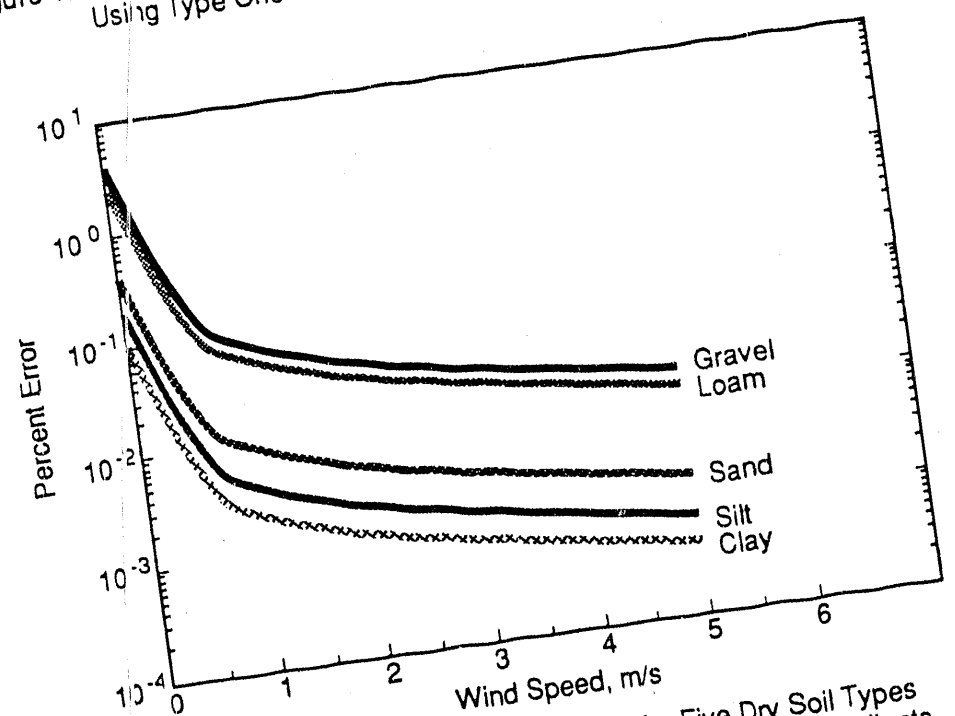

Figure 2. Percent Error in Radon Surface Flux for Five Dry Soil Types Using Type One Boundary Condition. No Pressure Gradients

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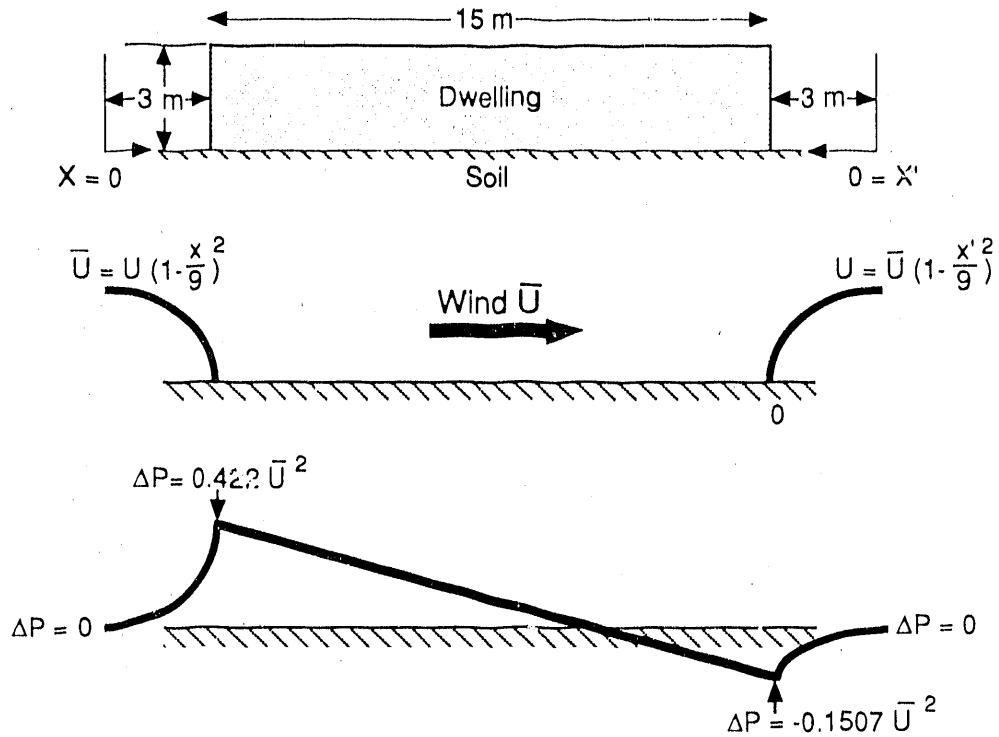

Figure 3. Schematic Diagram of Dwelling Wind and Pressure Profiles

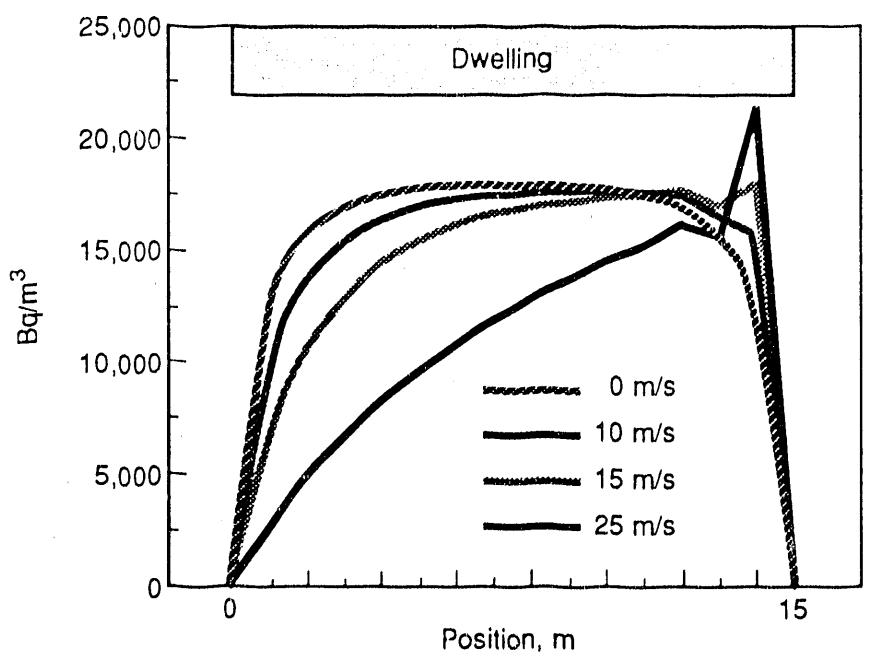

Figure 4. Steady-State Radon Concentration at Dry Sand / Dwelling Interface

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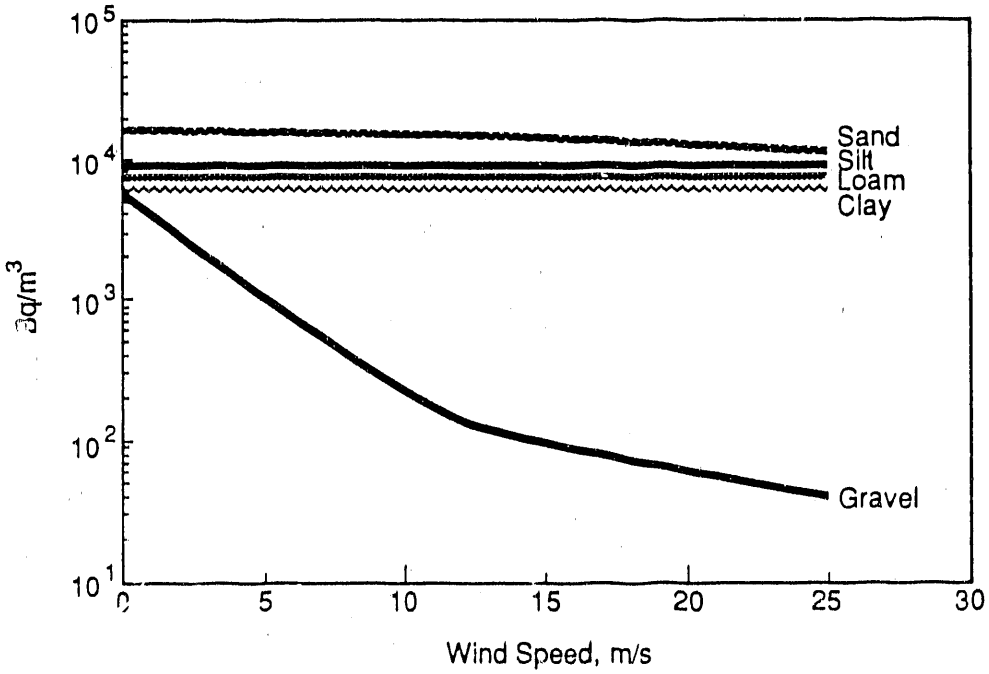

Figure 5. Average Dry Soil / Dwelling Interface Concentrations

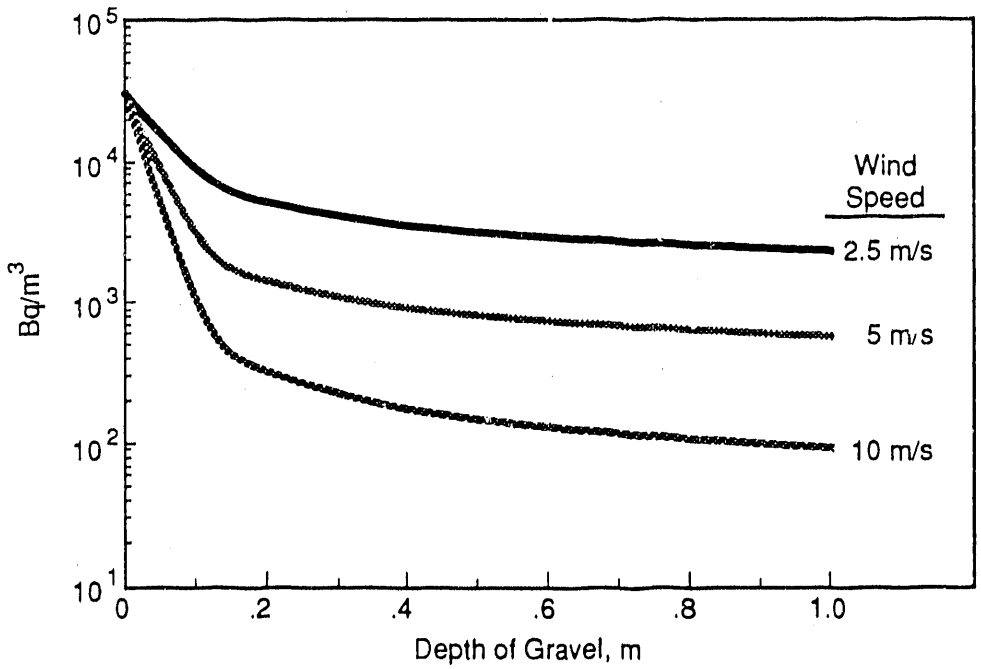

Figure 6. Average Concentration Under Dwelling Versus Gravel Depth and Wind Speed. 47.5% Saturated Clay Under Gravel

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