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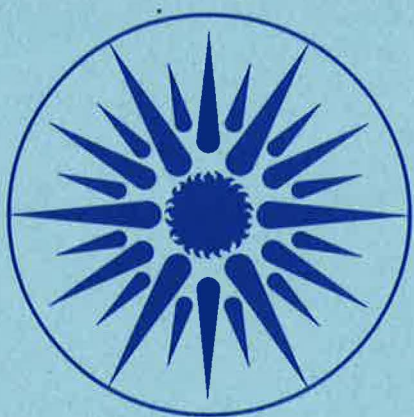
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Transport and Deposition of Indoor Radon Decay Products: Part 2 — Influence of Environmental Conditions

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**Transport and Deposition of Indoor Radon Decay Products:
Part 2 -- Influence of Environmental Conditions**

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TRANSPORT AND DEPOSITION OF INDOOR RADON DECAY PRODUCTS: Part 2 - Influence of Environmental Conditions

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ABSTRACT

The effects of indoor radon behavior on overall concentrations have generally been characterized using uniformly-mixed models, mathematical formulations based on steady-state macroscopic mass-balances, assuming uniform concentrations within the enclosure. The uniformly-mixed model parameterizes the deposition process as a constant volumetric removal rate, given different values for the free and attached progeny. The model requires prior knowledge of the deposition rates, and assumes them to be constant, independent of environmental conditions, and identical for all decay products. There has generally been little agreement regarding the actual values of the deposition rates, and the uncertainty in these required values presents an important limitation.

In response to the limitations of existing mass-balance models, a indoor radon mass-transport model, RADTRAN, was developed using a microscopic mass-balance. Deposition by molecular diffusion is accounted for through boundary conditions, and deposition velocity is calculated based on the concentration distribution near the wall. Parametric sensitivity studies using RADTRAN examined the sensitivity of the deposition of radon decay products to several factors: the size of the free progeny (measured by its diffusivity, D_f), particle concentration (using the attachment rate, X), and air motion. Deposition is described in terms of the deposition velocities of the free and attached progeny, u_f and u_a . The development of RADTRAN is described in a companion paper. This paper presents the results of the parametric sensitivity studies examining the influence of environmental conditions on radon progeny deposition. Results primarily focus on the influence on the free mode of the first radon decay product, ^{218}Po . RADTRAN is also used to examine the variations of deposition velocity between the decay products.

Keywords: radon progeny, indoor environment, deposition, diffusion, convection, modeling, prediction

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TRANSPORT AND DEPOSITION OF INDOOR RADON DECAY PRODUCTS: Part 2 - Influence of Environmental Conditions

G.E. Schiller¹, K.L Revzan²

1.0 INTRODUCTION

The behavior of indoor radon decay products is complex, involving radioactive decay, attachment to airborne particles, deposition on room surfaces, and other removal mechanisms. The deposition of radon progeny onto walls and other surfaces is particularly important at the low to moderate particle concentrations typically found indoors. The mechanisms of deposition depend on the mobility of the decay products and a driving potential, both of which are characteristic of the particular deposition mechanism. Diffusive deposition, for example, depends on both the diffusion coefficient of the decay product and also the concentration profile, or gradient, near the surface. This profile is determined by the environmental conditions, resulting in the deposition rate depending not only of the diffusion coefficient alone, but also on indoor conditions such as particle concentration and air flow patterns (Bruno, 1983). The concentration distribution of each decay product depends on the distribution of the parent concentration; consequently, one might also expect that deposition rates would vary for subsequent progeny in the decay chain.

The effects of indoor radon behavior on overall concentrations have generally been characterized using uniformly-mixed models, mathematical formulations based on steady-state macroscopic mass-balances, assuming uniform concentrations within the enclosure (Jacobi, 1972). However, the uniformly-mixed model does not examine the behavior of indoor radon decay products in a fundamental manner. In particular, the deposition process is parameterized as a constant volumetric removal rate, given different values for the free and attached progeny. The model requires prior knowledge of the deposition rates for these two modes, and assumes them to be constant, independent of environmental conditions, and identical for all decay products. There has generally been little agreement regarding the actual values of the deposition rates, and the uncertainty in these required values presents an important limitation. As a practical matter, the deposition rates depend on indoor particle concentration, diffusivity, enclosure geometry, and the effects of convective air flow patterns inside the space in a way that can only be characterized by taking account of the spatial distribution of radon progeny concentrations.

In response to the limitations of existing mass-balance models, an indoor radon mass-transport model, RADTRAN, was developed using a microscopic mass-balance. RADTRAN is based on differential equations describing the free and attached concentrations of each radon decay product, in terms of their generation, convective and diffusive transport, and source and removal terms including the radioactive decay rate, attachment rate, and recoil factor. Deposition by molecular diffusion is accounted for through boundary conditions. A mass

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transfer coefficient, or deposition velocity, is determined from the activity deposition using Fick's first law of diffusion, and the core concentration as the driving potential. With this approach, one can examine the "near-wall" behavior in greater detail to determine how the rate of deposition is influenced by environmental conditions, and what might account for the wide variability among experimentally observed deposition velocities. The development of RADTRAN is described in a companion paper by Schiller et al. (1989). This paper presents the results of parametric sensitivity studies examining the influence of environmental conditions on radon progeny deposition. Results focus on the influence on the free mode of the first radon decay product, ^{218}Po .

2.0 SIMULATIONS

Using RADTRAN, parametric sensitivity studies examined the sensitivity of the deposition of radon decay products to several factors: the size of the free progeny (measured by its diffusivity, D_f), particle concentration (using the attachment rate, X), and air motion. These parameters showed the greatest variation among published experiments, or seemed likely to have the greatest influence on concentrations and deposition rates. Deposition is described in terms of the deposition velocities of the free and attached progeny, u_f and u_a .

2.1 Diffusivity

As a measure of mobility, the diffusion coefficient has an important influence on deposition rates. Many researchers have investigated the influence of environmental conditions on the diffusion coefficients of radon decay products, with a particular emphasis on the more mobile free progeny (Chamberlain et al., 1956; Pörsendorfer, 1968; Raabe, 1968; Raghunath et al., 1979; Busigin et al., 1981; Knutson et al., 1983; Chu and Hopke, 1988; Phillips et al., 1988). Because of its strong dependence on diameter, the value of the free diffusion coefficient, D_f , depends on the decay products' physical and chemical nature. Busigin (1981) gives a good overview of previous work, discusses possible reasons for the wide range of values measured for D_f , and summarizes these values in the range of 0.005-0.1 cm^2/sec . The average, or commonly accepted, value of free progeny diffusivity is 0.054 cm^2/sec .

The parametric sensitivity studies examined three values of D_f : 0.01, 0.054, and 0.10 cm^2/sec . These values encompass the range found by the majority of researchers and are centered around the commonly used value. The studies examined several issues: whether experimentally determined values of deposition rates can be accounted for solely by molecular diffusion; to what extent the variations in experimental measurements might be explained by differences in the physical state of the free decay products; and the relationship between the ratios (u_f/u_a) and (D_f/D_a), used by researchers to interpret experimental data.

The diffusivity of the relatively immobile attached decay products can effectively be determined by the activity-weighted mean particle diameter. The average particle diameter found in typical indoor environments is about 0.1 μm (Sextro et al. 1986), with a corresponding activity-weighted average of 0.15-0.2 μm (Knutson et al., 1983; Mercer, 1976; Pörsendorfer, 1983). A commonly used value of attached progeny diffusivity is 4.5×10^{-6} cm^2/sec (Knutson et al., 1983). This value was used in the sensitivity studies and remained fixed.

2.2 Attachment Rate

The parametric sensitivity studies used different values of the attachment rate X to represent changes in particle concentration. Attachment rate increases with higher particle concentration, resulting in a smaller fraction of the progeny concentration existing in the free mode. The relationships between the total rate of activity deposition J , deposition rate constants q , activity concentrations A , and the free fraction f , can be summarized by the following expressions:

$$J_i = q_i A_i V = J_i^f + J_i^a = q_i^f A_i^f V + q_i^a A_i^a V \quad (1)$$

$$f_i = A_i^f / (A_i^f + A_i^a) = A_i^f / A_i \quad (2)$$

$$q_i = f_i q_i^f + (1-f_i) q_i^a \quad (3)$$

Researchers using the uniformly-mixed model have commonly assumed that X influenced only the value of f_i , and that values of deposition rate constants q^f and q^a (or deposition velocities u^f and u^a) were independent of variations in particle concentration, or attachment rate. However, the rate of deposition depends on the concentration profile near the surface. As a removal rate, X affects the boundary layer concentration profile and, as a result, may also influence the values of u^f and u^a .

The effect of particle concentration on u^f and u^a was examined by varying X over a range corresponding to typical particle concentrations found indoors. Seven values of attachment rate were examined - 0, 25, 50, 100, 150, 200, and 250 hr^{-1} . For a typical mean particle diameter of 0.1 μm , corresponding to an attachment coefficient of $4.3 \times 10^{-3} \text{ hr}^{-1}/(\text{particles}/\text{cm}^3)$, the range of $X = 0\text{-}250 \text{ hr}^{-1}$ corresponds to a maximum particle concentration of 60,000 $\text{particles}/\text{cm}^3$, representing the case of heavy smoking.

2.3 Air Motion

It is well accepted that air motion can have a significant influence on deposition rates, although its effect in room environments has generally been discussed only in a qualitative manner. Because of the lack of air flow measurements in the majority of experiments, and the inability of the uniformly-mixed model to account for airborne transport, the effects of air motion on radon progeny deposition have not previously been characterized quantitatively. A major contribution of RADTRAN is its ability to calculate concentration distributions and deposition rates for a specified flow field. The usefulness of the results are somewhat limited by the ability to characterize existing flows in real buildings or in the experiments, and also to accurately simulate a flow representative of those particular conditions. Given these limitations, however, RADTRAN still provides a useful theoretical framework for investigating the relative effects of air motion on predicted concentration distributions and deposition rates.

The parametric sensitivity studies examined several flow conditions, with D^f and X varying over the ranges previously discussed. The condition of pure diffusion was used as a reference for comparing the effects of air motion in the various flow configurations. Numerical simulations were performed for a stagnant enclosure (transport by molecular diffusion only), a recirculating enclosure flow (buoyancy-driven natural convection), and two boundary layer flows: free convection on an isolated vertical hot plate, and forced convection on a horizontal

flat plate. Descriptions of the significance of these flow regimes, and of RADTRAN's calculations of the velocity fields was presented by Schiller et al. (1989).

The magnitude of the buoyancy-driven enclosure flows was set by giving values to the representative Grashof number. For a $3 \times 3 \text{ m}^2$ enclosure, Grashof numbers were set equal to 3×10^9 and 3×10^{10} , corresponding to wall-to-wall temperature differences of 0.846 and 8.460 °C. (This corresponds to Rayleigh numbers of 2×10^9 and 2×10^{10} , since $Pr = 0.7$ for air). These two numbers represent the transition to turbulence, as quoted by various sources. The critical value of $Ra_{cr} = Gr Pr = 10^9$ is given by Bird et al. (1960) and Schlichting (1979), while the value of $Ra_{cr} = 10^{10}$ is given by Kaplan (1963), Gadgil (1980), Nansteel & Greif (1981), and Bohn et al. (1983). For flow inside an enclosure, transition to turbulence occurs at higher critical Rayleigh numbers than for isolated surfaces (Ruberg, 1978; Nansteel & Greif, 1981; and Bohn et al., 1983).

The free-convection boundary layer flow was compared to the enclosure by setting the wall-to-freestream temperature differences at half the wall-to-wall values, at 0.423 and 4.230 °C. To compare the forced convection boundary layer flow to the enclosure flow, the imposed free stream velocity corresponded to the nominal horizontal air velocities along the floor and ceiling in the enclosure, approximately 0.3 and 0.6 cm/sec for the two values of Gr . These flows are described in more detail by Schiller (1984). The laminar forced convection boundary layer flow was also used to examine the relative effects of free stream velocity ranging from 1-100 cm/sec.

3.0 RESULTS

Schiller et al. (1989) presented a comparison of RADTRAN's predictions of u^f and u^a to values obtained from experimental measurements. For experiments in which the flow conditions were most similar to those simulated, there was good agreement between experimental values of u^f and values predicted by RADTRAN. However, agreement was relatively poor for predicted values of u^a .

Equation (3) shows that the total deposition rate q is a function of q^f , q^a , and f . However, because the value of q^f is approximately two orders of magnitude greater than that of q^a , the contribution of the attached deposition rate to the total is relatively insignificant, unless the free fraction is less than about 10%. (For ^{218}Po , this occurs for particle concentrations greater than about 30,000 particles/cm³, or an attachment rate of $X = 130 \text{ hr}^{-1}$). For these reasons, the results of the parametric sensitivity studies presented in this paper focus on the influence of the parameters on only the free progeny deposition, u^f (or q^f).

3.1 Comparing Enclosure and Boundary-layer Flows

Simulations of the enclosure flows produce a large amount of information describing many complex interactions. In comparison, the simpler boundary layer flows enable one to perform a wide range of parametric simulations at considerably less expense, and the results provide a clearer picture of observable patterns in terms of the near-wall behavior. Comparing the velocity and concentration distributions in the different flow fields allows one to assess the adequacy of representing portions of the enclosure flow by simpler boundary flows.

The recirculating buoyancy-driven flow was calculated for a square two-dimensional enclosure with

isothermal hot and cold walls, and adiabatic floor and ceiling. Details of the velocity profiles in both the enclosure and boundary layer flows is given by Schiller et al. (1983) and Schiller (1984). Velocity profiles for the free and forced convection boundary layer flows showed a strong similarity between these simplified flows and the corresponding regions of the recirculating enclosure flow.

It is perhaps more useful to compare the predicted concentration profiles and values of deposition velocities in the different flows, since it is likely that the boundary layer treatments, based on integral methods, give generally less representative results for the profiles than for the transfer rates (Bird et al., 1960).

Figure 1 is a plot of u^f as a function of X , calculated from both the enclosure model for Grashof number = 3×10^9 , and from the representative free and forced convection boundary layer flows. The figure illustrates that there is close agreement between the predictions of u^f in these flows, especially for higher attachment rates. It also illustrates that u^f increases with particle concentration, a point that will be discussed in a later section. This comparison essentially validates the findings of these initial parametric studies, with regard to the boundary layer flows being representative of the enclosure flows for the deposition of unattached ^{218}Po . It also justifies further use of the simpler boundary layer flows for the parametric studies.

3.2 Effect of Air Motion on Deposition

The present formulation of RADTRAN accounts for deposition by molecular diffusion only, neglecting the mechanisms of turbulent diffusion and inertial impaction. Consequently, air motion influences the deposition velocities only indirectly, by affecting pollutant transport towards the wall and the thickness of the boundary layer across which diffusion occurs. The parametric sensitivity studies investigated laminar forced convection boundary layer flows to demonstrate the effects of a wide range of air velocity on progeny concentration profiles and deposition velocities.

Figure 2 plots u^f vs. freestream velocity U , for several values of attachment rate X . The value of U varies up to 100 cm/sec, representing the maximum influence of air motion for laminar flow. The figure shows that u^f increases with stronger air motion, although its sensitivity to air speed diminishes at higher values of X , corresponding to higher particle concentration. For X greater than 50 hr^{-1} , u^f is fairly insensitive to changes in air velocity below 10 cm/sec.

3.3 Effect of Free Diffusivity on Deposition

The sensitivity of u^f to D^f is expected to be influenced by air motion. This is because u^f is proportional to D^f , and inversely proportional to the boundary layer thickness δ^f . As the concentration gradient steepens with increasing air motion, δ^f decreases and the effect of changes in D^f is amplified. This effect is illustrated in Figure 3, where u^f is plotted vs. D^f for a range of air velocity, illustrating that the sensitivity of u^f to D^f increases slightly over the range of air velocity investigated.

Researchers have measured values of D^f varying over an order of magnitude. For this range of uncertainty, u^f will increase by a factor of 3-4.5, the minimum corresponding to stagnant conditions, and the maximum for the high range of laminar air velocity. The relationship between the ratios (u^f/u^a) and (D^f/D^a) is discussed in section 3.6 below.

3.4 Effect of Attachment Rate on Deposition

Deposition velocity can be considered a normalized deposition flux, in that it is equal to the deposition flux divided by the equilibrium core concentration. It is inversely proportional to the boundary layer thickness of the normalized concentration profile. Since removal of free progeny by attachment is proportional to concentration, as attachment rate X increases the equilibrium concentration outside the boundary layer decreases faster than the concentration near the wall. As the value of X increases, the concentration boundary layer thickens, but the normalized profile shows the opposite trend. The wall region of the normalized concentration profile becomes thinner and the concentration gradient at the surface correspondingly becomes steeper. This also results in an increase in the deposition rate constant q_f , and the deposition velocity u_f .

The relationship between deposition velocity and attachment rate for stagnant conditions is mathematically illustrated in the analytic equations produced by the one-dimensional diffusion model (Schiller et al. 1989). For ^{218}Po , the relationship between u_f and X in stagnant conditions can be expressed as:

$$u_f = [D_f (\lambda + X)]^n \quad (4)$$

where $n = 0.5$. Increasing X by an order of magnitude from 25-250 hr^{-1} will result in u_f increasing by a factor of 2.6, for ^{218}Po . The sensitivity of u_f to changes in X is reduced by increasing air motion, since the attachment rate has a relatively smaller influence on the thickness of the concentration boundary layer as air motion increases. This can be expressed mathematically by $n < 0.5$ in equation (4). This effect is illustrated in Figure 4, where it is seen that u_f increases for higher values of X , although the sensitivity is lessened for higher values of air velocity as shown by the increasingly flatter curves.

3.5 Variations Among Decay Products

Researchers have generally assumed that the values of u_f and u_a are identical for all the decay products, regardless of environmental conditions. Even if all the decay products were generated from a uniform source, the free or attached profiles of the various decay products differ due to differences in the radioactive decay rate constants. Furthermore, the concentration distribution of each decay product serves as the source distribution for the next. Thus the deposition velocities might vary among the decay product species.

Figure 5 shows the normalized concentration profiles of the first three decay products in stagnant conditions, illustrating the relative thickness of the boundary layer for each. Because of the non-uniform source, each subsequent decay product has a thicker normalized boundary layer and a broader gradient at the surface. This indicates that deposition velocity decreases for succeeding decay products in the decay chain. The variability of free deposition velocity is shown in Figure 6, where u_f vs. X is plotted, for the three progeny. This figure shows that, across the range of X , u_2^f is only 0.25 - 0.5 the value of u_1^f , and u_3^f is only 0.2 - 0.4 the value of u_1^f .

3.6 Ratio of (u^f/u^a)

An effective concentration film thickness, b , can be defined by assuming a linear concentration profile with a slope equal to the gradient at the wall. The deposition velocities, and their ratios, can then be expressed as:

$$u^f = D^f / b^f \qquad u^a = D^a / b^a \qquad (5)$$

$$(u^f / u^a) = (D^f / D^a) \cdot (b^a / b^f) \qquad (6)$$

The values of b^f and b^a for each decay product are descriptive of the shapes of the respective concentration profiles within the wall region. These profiles are influenced not only by the diffusion coefficients, but also the source distributions, the relative strengths of the source and removal terms, and the rate of convection which contributes to the transport of pollutants either towards or away from the wall. The overall influences of diffusivity can be mathematically described by expressing equation (6) as a proportional relationship in the following form:

$$(u^f / u^a) \propto (D^f / D^a)^m \qquad (7)$$

where the actual value of the power coefficient m is determined by the environmental conditions, and $0 < m < 1$.

This expression is significant in that it is often used by researchers to calculate one unknown parameter (e.g., u^f or D^f) in terms of assumed or measured values of the others (Jacobi, 1972; Knutson et al., 1983). Researchers have made various assumptions and used corresponding values of $m = 1/2$ or $m = 2/3$ in their calculations. These numbers appear to have come from two simplified cases that can be solved analytically. However, neither of these cases is accurate.

The first case ($m=1/2$) corresponds to two reactive components, each with a uniform source in stagnant conditions. For stagnant conditions the deposition velocity of the attached progeny is strongly influenced by its non-uniform source, the free progeny. Schiller et al. (1989) presented expressions describing radon progeny concentration distributions and deposition velocities in stagnant conditions, and equations (14a) and (14b) in that paper show that m will have a value of 1, and not $1/2$, in stagnant conditions.

The second case ($m=2/3$) corresponds to two non-reactive components (i.e., particles) in a forced convection flow. Although the relative influences of D^f and D^a will change when there is a convective flow contributing to pollutant transport, it is not clear that an analysis based on particle deposition in a convective flow can determine the appropriate value of m corresponding to reactive pollutants in conditions of faster air flow.

The parametric studies examined the extent of the influence of air velocity on the relationship between the ratios (u^f/u^a) and (D^f/D^a). Figure 7 shows results for a forced convection flow with freestream velocity U ranging from 0-100 cm/sec. The greatest sensitivity is seen for stagnant conditions (maximum value of $m=1$). A decreased sensitivity is shown for higher air speeds, and can be mathematically described by smaller values of m in equation (7). At the highest velocity, 100 cm/sec, the curve can be represented by the minimum value of $m=0.7$.

3.7 Comparing attached progeny and particle deposition

In experiments in which the removal rate of particles was determined (Pörstendorfer 1983; Sextro et al. 1986), it was assumed that u^a is equivalent to particle deposition velocity, u_p . For deposition by molecular diffusion, this may not be valid because the deposition rates depend on two factors: the diffusion coefficients (D^a and D_p), and also the concentration gradients at the surface. Because the decay products are more likely to attach to larger particles, there is a shift in their size distributions, resulting in a slight difference between D^a and D_p . Even more important, perhaps, is the potential differences between the concentration gradients of the attached progeny and the particles. This is because the attached progeny concentration distribution, A^a , depends on both the particle concentration distribution, N , and also the distribution of its free progeny source, A^f . This effect is illustrated in Figure 8, in which u^a is plotted as a function of air velocity, for various values of X , along with a curve for u_p calculated using the same diffusivity.

The difference between u^a and u_p is proportional to the extent to which the free progeny influence the distribution of the attached progeny. For low air velocities, the attached progeny distribution is dominated by that of the free progeny; as a result, the gradient at the surface will not be as steep as that of the particles, and u^a will be less than u_p . For higher air velocities, A^a is less influenced by A^f and u^a will approach the value of u_p . For higher particle concentrations (or higher values of X), the free fraction is reduced and A^f will again have a smaller influence on A^a . Therefore, u^a increases with increasing values of X and approaches the value of u_p . Figure 8 shows that u_p is as much as four times greater than u^a at low attachment rate and air velocity, but the values become nearly equivalent at higher values of X and U . It is not clear why u^a becomes greater than u_p at the highest values of velocity.

4.0 DISCUSSION

Simulations using RADTRAN have focused on stagnant and laminar flow conditions. Deposition velocities of $u^f = 0.014 - 0.079$ cm/sec have been computed for the range of D^f , X , and air motion investigated. These results suggest that experimental values of u^f greater than approximately 0.08 cm/sec cannot be accounted for by convective diffusion alone.

Researchers using the uniformly-mixed model have assumed that values of u^f and u^a were constant, independent of environmental conditions and identical for all decay products. The validity of this assumption for ^{218}Po was the focus of RADTRAN simulations. Results presented here show that u^f varied by an overall factor of 5.6 for the conditions investigated, and that values of D^f had the strongest influence on u^f for laminar conditions. When D^f increased by an order or magnitude, corresponding to the range of values measured in experiments, u^f increased by a factor of 3.2-4.2. The sensitivity of u^f to D^f was highest for higher air motion. For stagnant conditions, u^f increased by a factor of 2.5 when X was varied over an order or magnitude. The sensitivity of u^f to changes in X was reduced for higher air motion.

Because the present formulation of RADTRAN neglects the mechanisms of deposition by turbulent diffusion and inertial impaction, air motion influences deposition velocity only indirectly by affecting the thickness of the boundary layer across which diffusion occurs. For the case of zero particle concentration ($X = 0$), u^f is increased by a factor of 3.5 when air velocity in a forced convection flow is increased from zero to 100 cm/sec. This sensitivity is reduced as particle concentration increases, because the free progeny concentration profile will be predominantly influenced by the high rate of removal by attachment, and u^f becomes essentially independent of air velocity. These results suggest that, in general, the assumption of a constant value for u^f will be most appropriate for conditions of higher particle concentration and air speeds. For turbulent conditions often found in buildings, it is likely that the influence of X will be relatively insignificant, and u^f can be considered independent of particle concentration.

An important contribution of RADTRAN is its ability to examine the variations between the decay products, and it was found that u^f decreases for subsequent decay products. For the case of diffusion-only, the range of values predicted for u^f for ^{218}Po , were 2-4 times greater than the corresponding values for ^{214}Pb , and 2.5-5 times greater than those for ^{214}Bi . These differences are attributed to the fact that deposition velocity depends on both the value of D^f , and also the concentration gradient at the surface. The profile is, in part, determined by the source distribution, which is different for the various progeny in the decay chain.

The relationship between the ratios (u^f/u^a) and (D^f/D^a) is commonly used by researchers to interpret experimental data. The relationship is expressed in equation (7), in terms of the power coefficient m . RADTRAN simulations calculated values of $m = 0.7-1.0$, the minimum corresponding to higher air velocity. While this range is higher than that used by experimental researchers, $m = 0.5-0.67$, the calculated values may be reduced even further as velocity and turbulence increases.

RADTRAN also investigated the common assumption of equivalence of u^a and u^p , and found that u^p may be as much as four times greater than u^a . However, assuming $u^a = u^p$ is not likely to have a significant influence on predicted values of room concentration at low particle concentrations, since the total deposition rate is almost entirely determined by free progeny deposition for these conditions. The value of u^a only begins to have an influence on total deposition rates at higher particle concentrations, and for these conditions the value of u^a begins to approach u^p and the assumption of equivalence can be applied. This assumption also appears to be most accurate for flow conditions with moderate air velocities.

5.0 RECOMMENDATIONS FOR FUTURE WORK

The predictions and conceptual insights produced by RADTRAN have been used to indicate what parameters are most likely to account for the variability in observed deposition velocities, and to assess the validity of assumptions used in experiments. Schiller et al. (1989) compared the range of RADTRAN's predictions to measurements from experiments carried out under similar flow conditions. However, experimental methods used to date do not provide sufficient information to enable researchers to account for the variability of the free

and attached deposition rates, or to accurately characterize the flow conditions in their experiments. As a result, many of the detailed trends observed from the RADTRAN simulations cannot at this time be compared to experimental measurements. Based on the findings, recommendations are made regarding how experiments might be improved to investigate the sensitivity of free and attached progeny deposition, and increase our understanding of the behavior of indoor radon decay products. Recommendations for improving the capabilities of RADTRAN are also presented.

To determine the extent to which variations in experimental measurements are due to flow conditions, it is important for future studies to better characterize the air motion during the experiments. This can be done qualitatively using visualization techniques, or quantitatively by taking actual air speed measurements at various locations in the space, particularly near the surfaces. It might also be useful to do experiments in controlled flows, such as in a wind tunnel or controlled environment chamber.

Measuring free progeny concentration directly, and at various locations in the core and wall regions of the experimental space, would also provide useful information. The distributions predicted by the RADTRAN can then be compared to these measurements, and used to interpret experimental results. Experiments can also be designed to directly determine the values of u^f and u^a , rather than assuming constant values. If free and attached activity concentrations of each decay product could be measured independently, then the uniformly-mixed mass balance equations can be used to calculate the individual values of u^f and u^a for each decay product. By carrying out this procedure for experiments conducted under various environmental conditions, the variation of u^f and u^a can be determined experimentally.

To account for the variability among researchers' experimentally determined values of u^f greater than 0.08 cm/sec, an important next step for improving RADTRAN would be to simulate other deposition mechanisms such as electrostatic attraction, thermophoresis, inertial impaction and turbulent diffusion. More realistic flow patterns such as turbulence and enclosure flows generated by infiltration and ventilation should also be investigated.

6.0 CONCLUSIONS

The findings discussed in this paper illustrate one of the many advantages of RADTRAN over existing uniformly-mixed models. Specifically, RADTRAN is able to examine the dependence of radon progeny deposition on environmental conditions, and therefore provides a more realistic basis for understanding radon progeny behavior. These parametric sensitivity studies have demonstrated the relative influence of free progeny diffusivity, aerosol particle concentration, and convective air motion on free and attached deposition velocities. These parameters provide a partial description of the physical and environmental conditions likely to have the greatest influence on deposition rates. Results of the RADTRAN simulations provide a basis for evaluating the validity of assumptions often made by experimental researchers, and indicates which parameters are most likely to account for the variability in observed deposition velocities.

7.0 ACKNOWLEDGEMENTS

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8.0 REFERENCES

- Bird, R.B., W.E. Stewart, and E.N. Lightfoot, 1960, Transport Phenomena, John Wiley & Sons, New York.
- Bohn, M., D. Olson, and A. Kirkpatrick, 1983, "Experimental Study of Three-Dimensional Natural Convection at High Rayleigh Numbers", ASME Paper No. 83-HT-12.
- Bruno, Ronald C., 1983, "Verifying a model of radon decay product behavior indoors", *Health Physics*, Vol.45, No.2, pp.471-480.
- Busigin, A., et al., 1981, "The nature of unattached RaA (Po-218) particles", *Health Physics*, Vol.40, pp.333-343.
- Chamberlain, A.C., and E.D. Dyson, 1956, "The dose to the trachea and bronchi from the decay products of radon and thoron", *Br. J. Radiol.*, 29, pp.317-325.
- Chu, K.D., and P.K. Hopke, 1988, "Neutralization kinetics for polonium-218", *Environmental Science and Technology*, Vol.22, No.6, pp.711-717.
- Gadgil, A.J., 1980, "On convective heat transfer in building energy analysis", Ph.D. Thesis, Department of Physics, University of California, Berkeley. Also published as Report #LBL-10900, Lawrence Berkeley Laboratory, California.
- Jacobi, W., 1972, "Activity and potential alpha-energy of (222)radon- and (220)radon-daughters in different air atmospheres", *Health Physics*, Vol.22, pp.441-450.
- Kaplan, I., 1963, Nuclear Physics, 2nd ed., Cambridge Mass., Addison-Wesley Publishing Co.
- Knutson, E.O., A.C. George, et al., 1983, "Radon daughter plateout - II: Prediction model", *Health Physics*, Vol.45, No.2, pp.445-452.
- Mercer, T.T., 1976, "The effect of particle size on the escape of recoiling RaB atoms from particulate surfaces", *Health Physics*, Vol. 31, pp.173-175.
- Nansteel, M., and R. Greif, 1981, "Natural Convection in Undivided and Partially Divided Rectangular Enclosures", *ASME Journal of Heat Transfer*, Vol. 103, pp.623-629.
- Phillips, C.R., A. Kahn, and H.M.Y. Leung, 1988, "The Nature and Determination of the Unattached Fraction of Radon and Thoron Progeny," Radon and Its Decay Products in Indoor Air, W.W. Nazaroff and A.V. Nero, eds., John Wiley & Sons, Inc, pp.203-256.
- Pörstendorfer, J., 1968, "The diffusion coefficients and the mean free path of the neutral and charged radon decay products in air", *Zeitschrift für Physik*, Vol.213, pp.384-386.
- Pörstendorfer, J., 1984, "Behaviour of radon daughter products in indoor air", *Radiation Protection Dosimetry*, Vol.7, No.1-4, pp.107-113.
- Raabe, O.G., 1968, "Measurement of the diffusion coefficients of RaA", *Nature*, Vol.217, pp.1143-1145.
- Raghunath, B., and P. Kotrappa, 1979, "Diffusion coefficients of decay products of radon and thoron", *J. Aerosol Sci.*, Vol.10, pp.133-138.
- Ruberg, K., 1987, M.S. Thesis, Massachusetts Institute of Technology.

Schiller, G.E., A.V. Nero, and C.L. Tien 1989, "Transport and Deposition of Indoor Radon Decay Products: Part 1 - Model Development and Validation," (submitted to Atmospheric Environment).

Schiller, G.E., 1984, "A Theoretical Convective Transport Model of Indoor Radon Decay Products," Ph.D. Thesis, University of California, Berkeley. Also published as Report #LBL-20096, Lawrence Berkeley Laboratory, California.

Schiller, G.E., A.V. Nero, K.L. Revzan, and C.L. Tien, 1983, "Radon Decay-Product Behavior Indoor: Numerical Modeling of Convection Effects", Proceedings of the Air Pollution Control Association Annual Meeting, San Francisco, January, 1983.

Schlichting, H., 1979, Boundary-Layer Theory, 7th ed., McGraw-Hill.

Sextro, R.G., F.J. Offerman, W.W. Nazaroff, A.V. Nero, K.L. Revzan, and J. Yater, 1986, "Evaluation of Indoor Aerosol Control Devices and Their Effects on Radon Progeny Concentrations", Environment International, Vol. 12, pp.429-438.

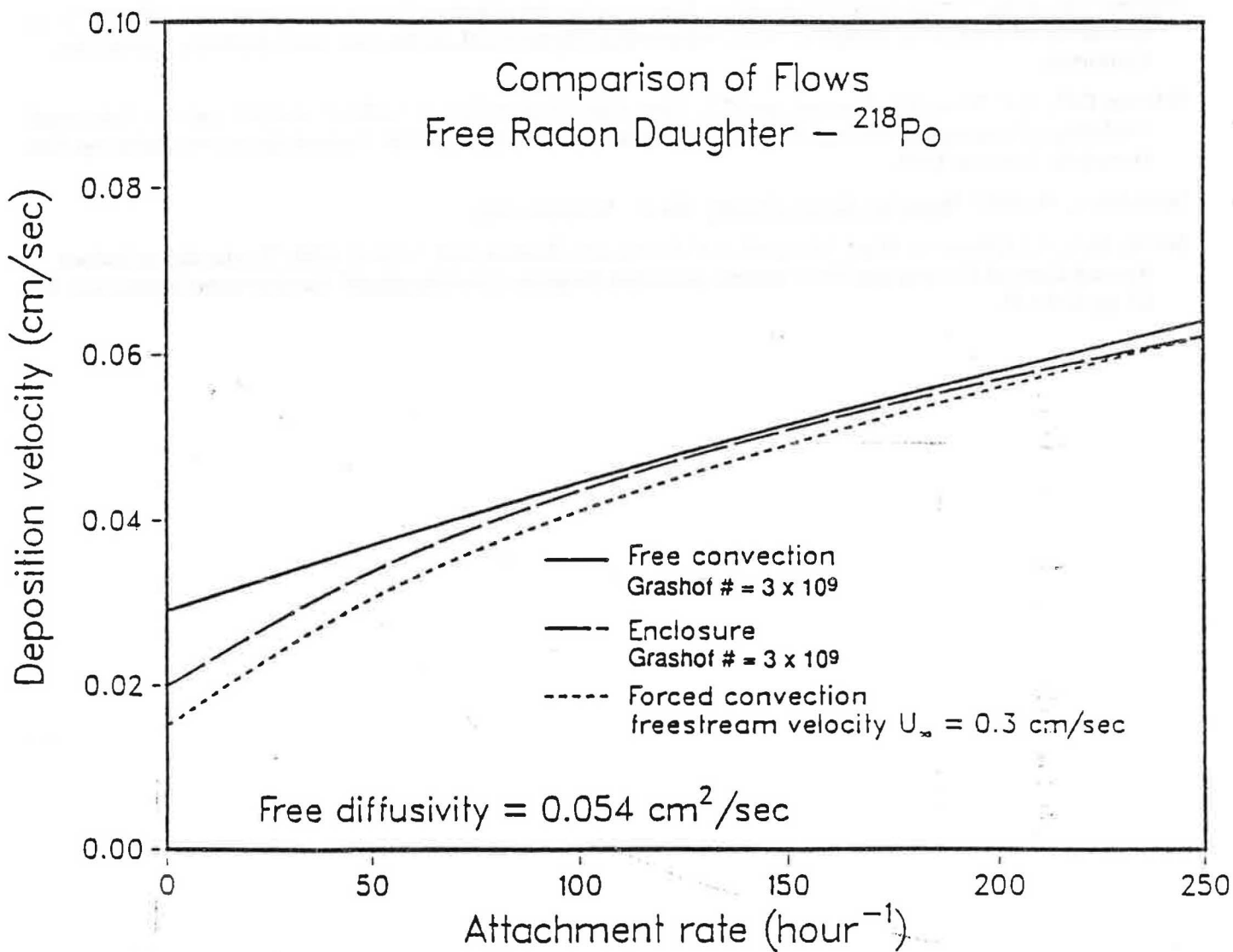


Figure 1. Variation of free deposition velocity with attachment rate, for ^{218}Po . Predictions from the Grashof number = 3×10^9 enclosure flow, and representative free and forced convection boundary layer flows are compared. Surface lengths are 3 m. The agreement between these predictions validates the findings of the parametric studies, with regard to the boundary layer flows being representative of the enclosure flows.

Forced Convection
Free Radon Daughter - ^{218}Po

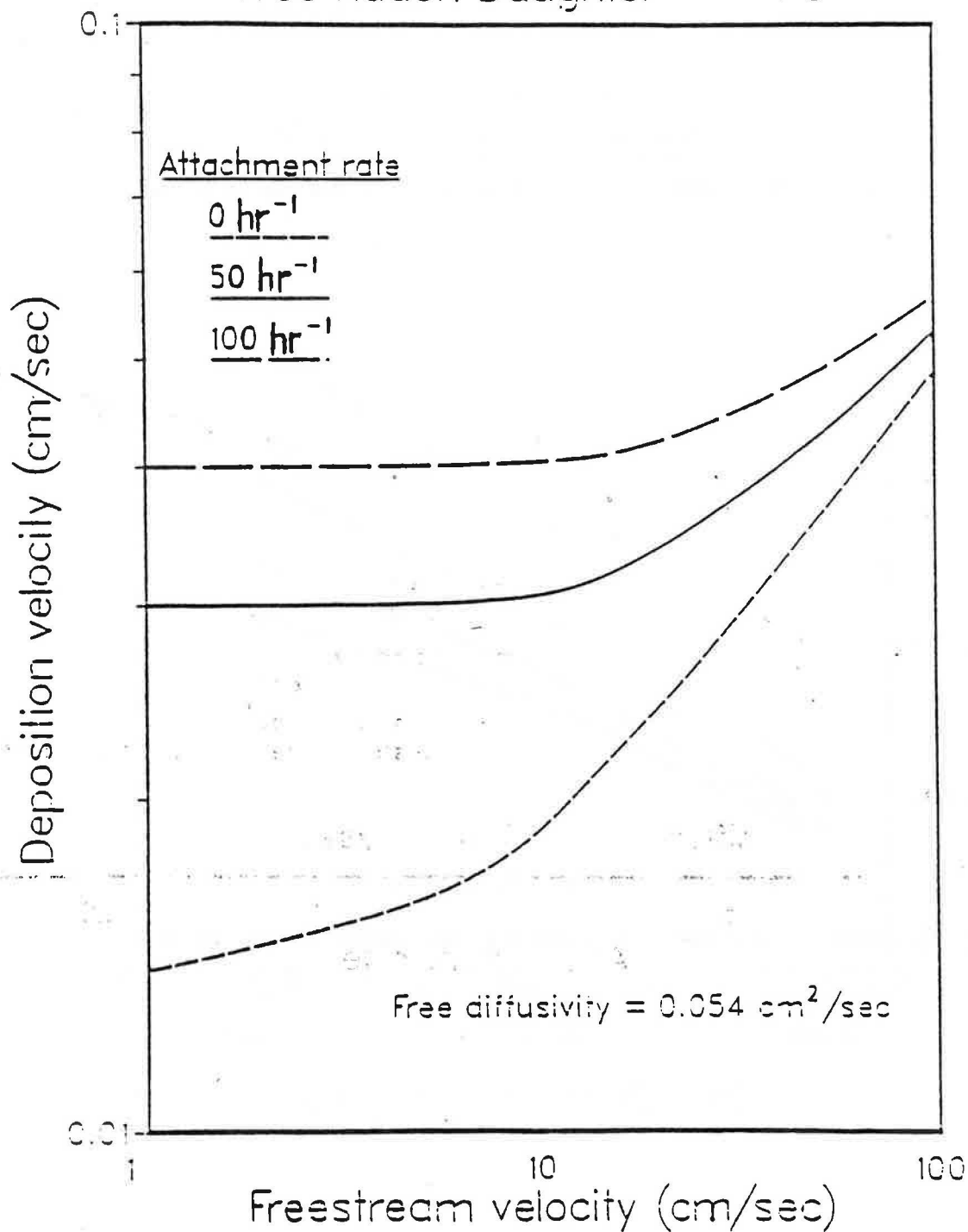


Figure 2. Variation of free deposition velocity with air velocity, for various values of attachment rate, for ^{218}Po . The sensitivity of deposition velocity to air motion diminishes at higher values of attachment rate.

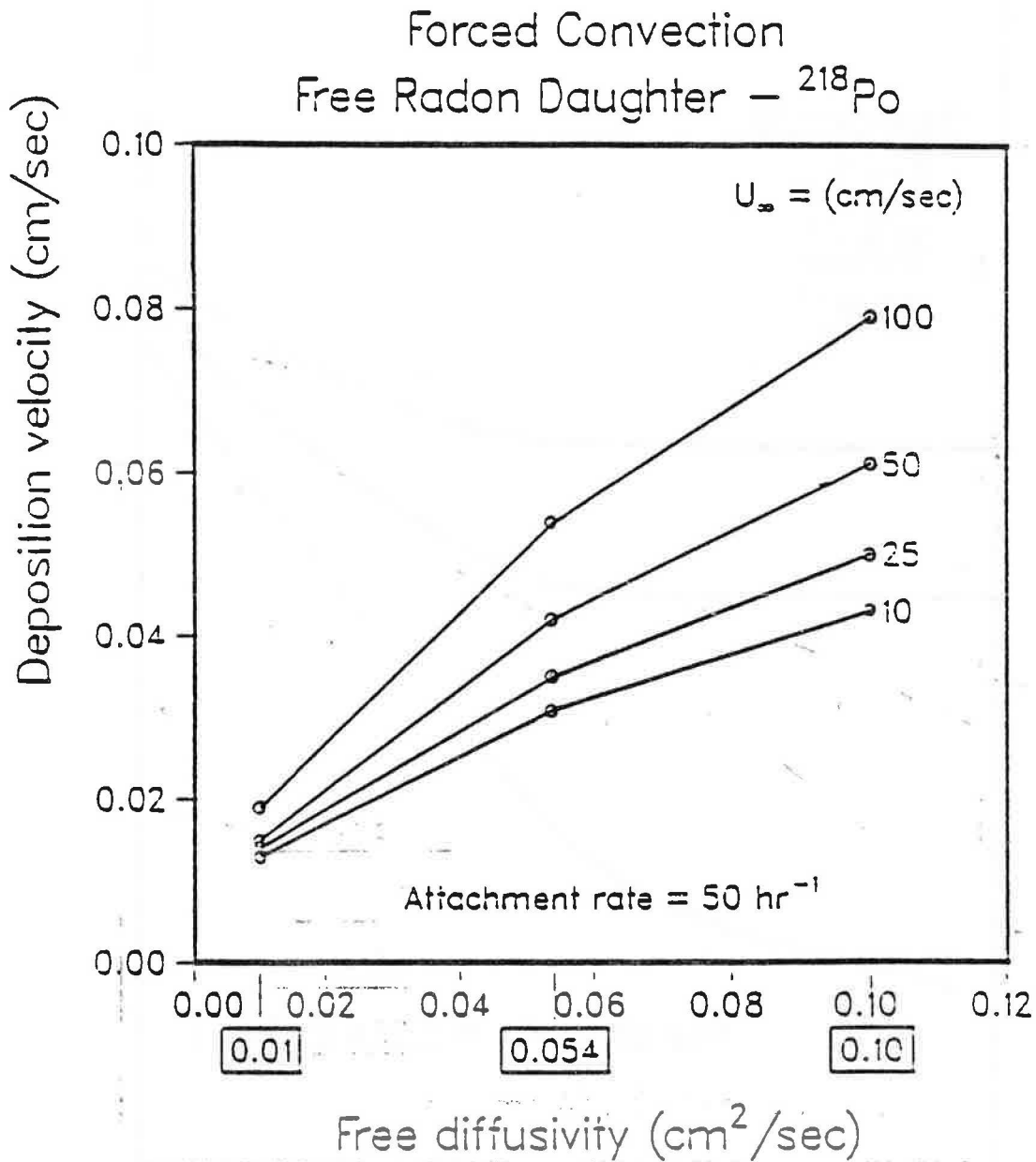


Figure 3. Variation of free deposition velocity with free diffusivity, for ^{218}Po . Deposition velocity is averaged across the 3 m long plate. The sensitivity of deposition velocity to diffusivity increases for higher air velocities.

Forced Convection
Free Radon Daughter — ^{218}Po

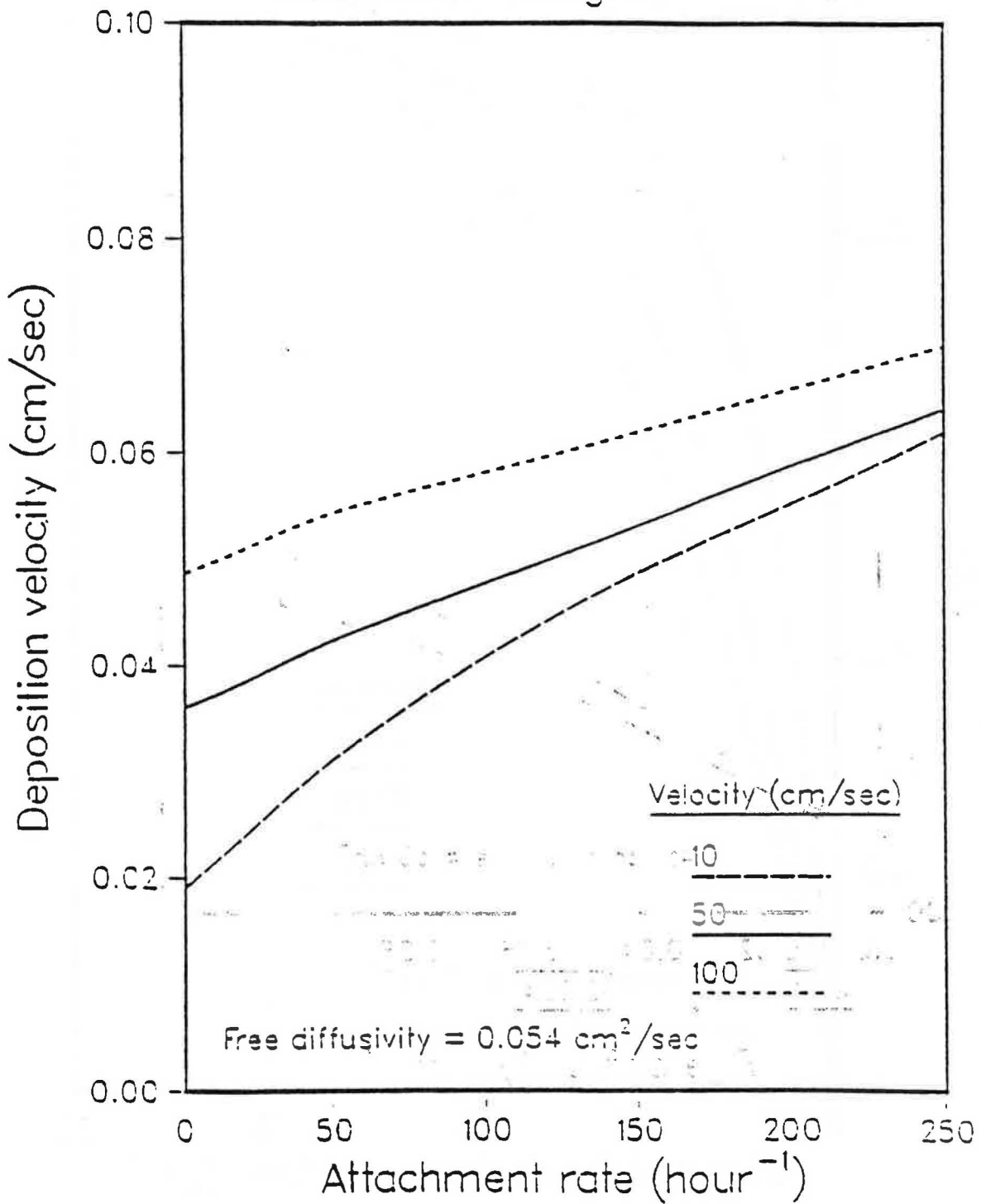


Figure 4. Variation of free deposition velocity with attachment rate, for ^{218}Po . Deposition velocity is averaged across the 3 m long plate. The sensitivity of deposition velocity to attachment rate decreases for higher air velocities.

One-Dimensional Diffusion Free Radon Daughters

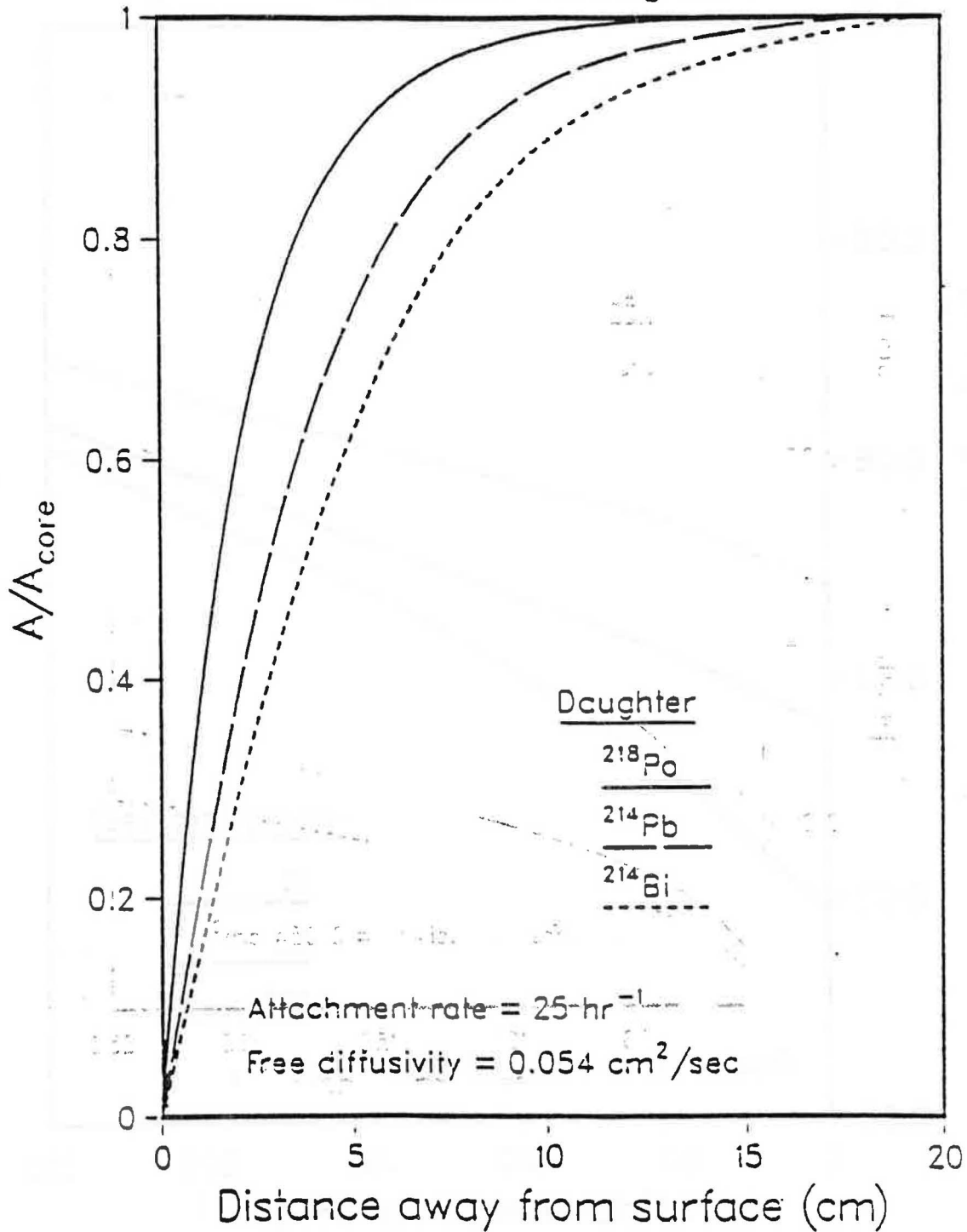


Figure 5. Free activity concentration profiles for the first three radon decay products. Concentrations are normalized by the core concentration outside of the boundary layer. Because of the non-uniform source, each subsequent decay product has a thicker normalized boundary layer and a broader gradient at the surface.

One-Dimensional Diffusion Free Radon Daughters

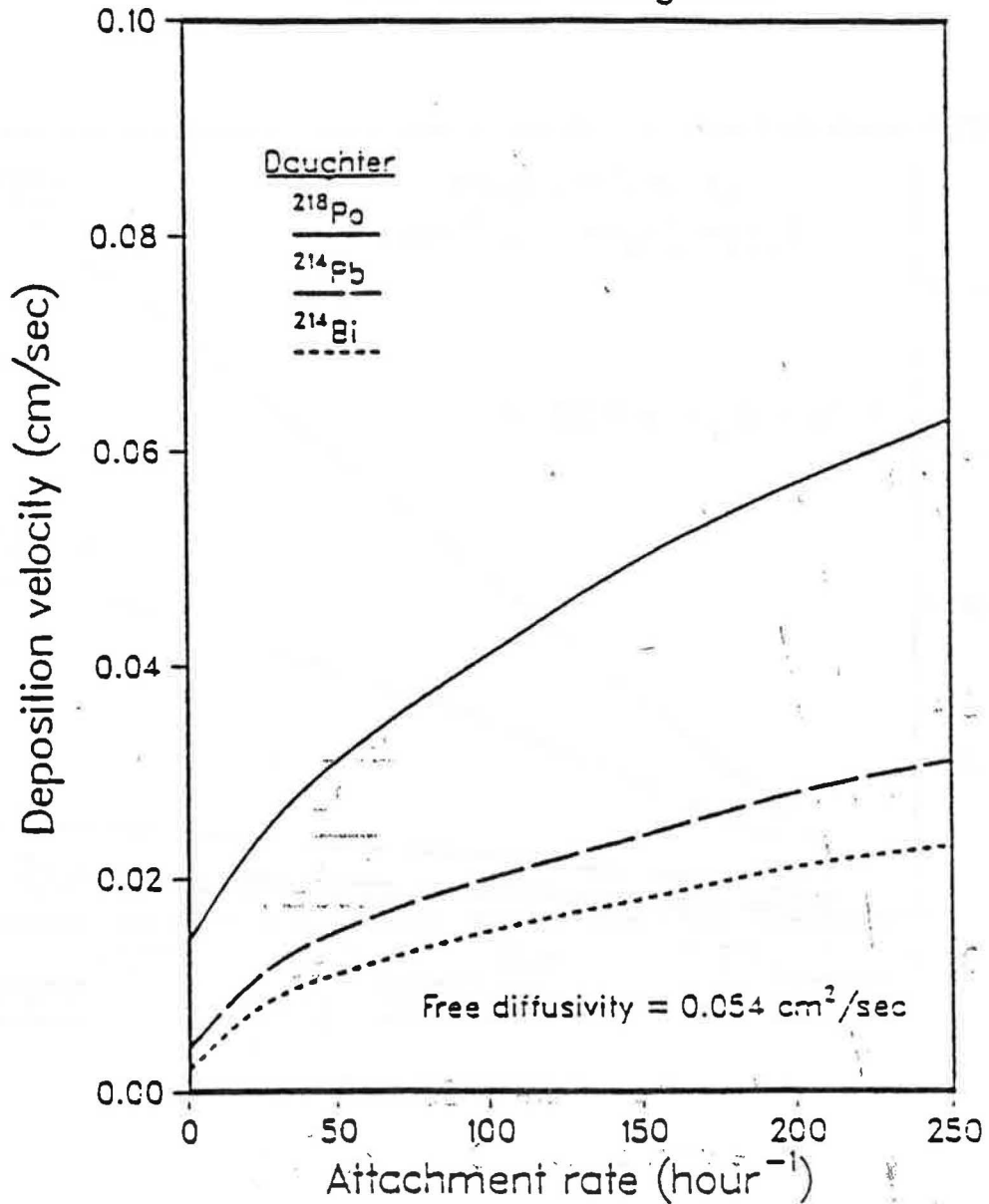


Figure 6. Variation of free deposition velocity with attachment rate, for the first three radon decay products, for one-dimensional diffusion. The broadening normalized concentration profile seen in Figure 5 results in a decrease in deposition velocity for daughters further down the decay chain.

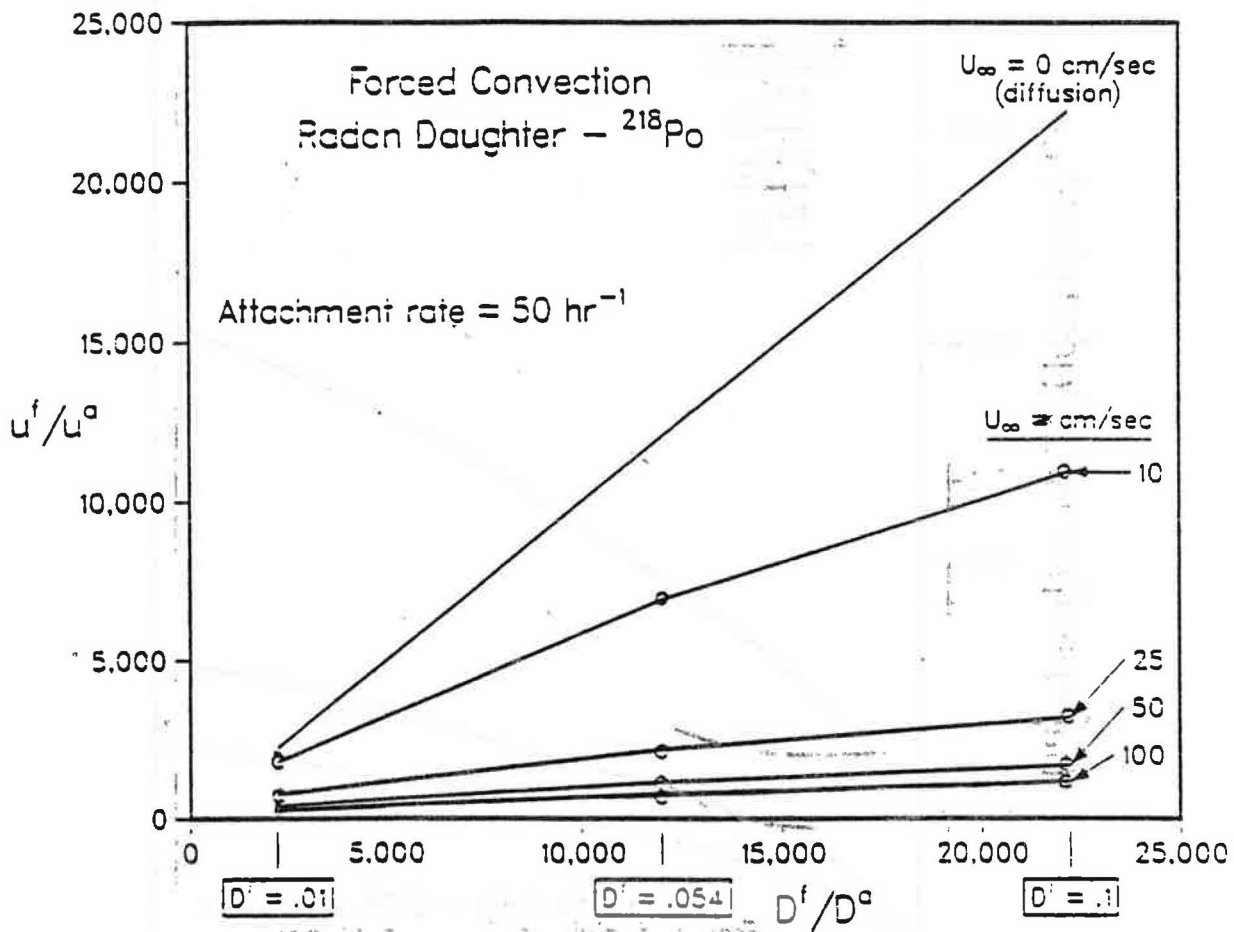


Figure 7. Relationship between the ratios of free-to-attached deposition velocity, and free-to-attached diffusivity, for ^{218}Po . Researchers often use this relationship to calculate one of the unknown parameters in terms of assumed or measured values of the others. The sensitivity of (u^f/u^a) to (D^f/D^a) decreases with higher air velocities.

Forced Convection
Attached Radon Daughter - ^{218}Po

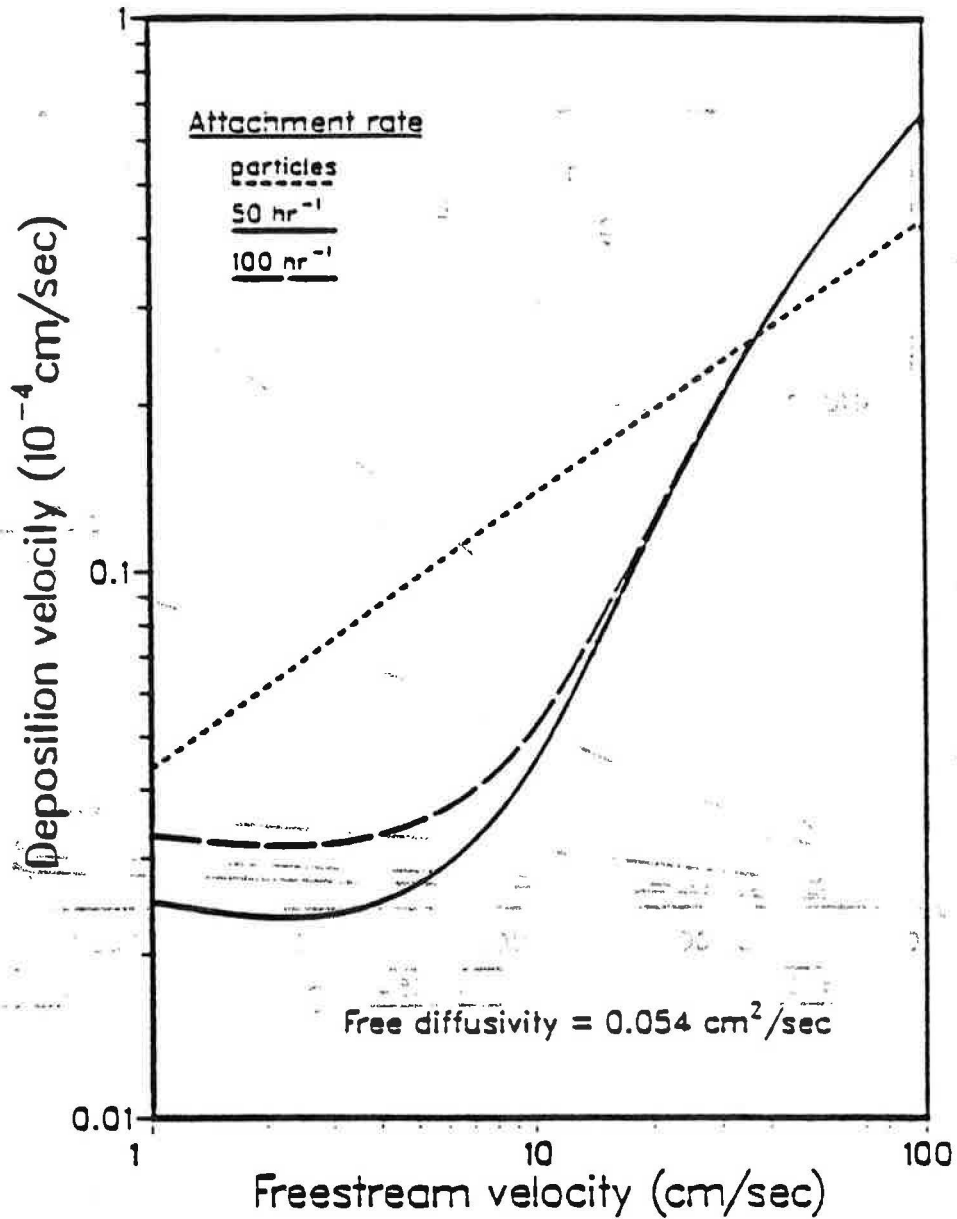


Figure 8. Variation of deposition velocity with air velocity, for attached ^{218}Po and also for particles. (It is assumed that the diffusivities are equal, i.e., $D^a = D_p$). Deposition velocities are averaged across the 3 m plate length. For low air velocity, deposition velocity is much higher for particles than for attached ^{218}Po .

