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G.E. Schiller
(Ph.D. Thesis)

July 1984

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A THEORETICAL CONVECTIVE-TRANSPORT MODEL
OF INDOOR RADON DECAY PRODUCTS

by
Gail Ellen Schiller

A Dissertation submitted in partial fulfillment
of the requirements for the degree of
Doctor of Philosophy
(Mechanical Engineering)
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April 1989

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A Theoretical Convective-Transport Model
of Indoor Radon Decay Products

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Gail Ellen Schiller
To my parents -

for giving me roots,
and wings.
A Theoretical Convective-Transport Model
of Indoor Radon Decay Products

Gail Ellen Schiller

ABSTRACT

A numerical model is developed which is capable of simulating the generation, convective and diffusive transport, and removal mechanisms of radon and radon decay products (daughters). The model is based on fundamental mass-transport equations. Specifically, separate differential equations describe the spatially-dependent concentrations of both unattached (free) daughters and daughters attached to airborne aerosols; the equations are coupled through source and sink terms which include the radioactive decay rate, attachment rate, and recoil factor. Surface deposition of airborne daughters by molecular diffusion is accounted for through boundary conditions.

The mass-transport model is employed to examine the influence of key parameters on free and attached deposition rates of the first three radon daughters, with a particular emphasis on the first free daughter, $\text{^{218}Po}$. These parameters - free daughter diffusivity, aerosol particle concentration, and convective air motion - provide a partial description of the physical and environmental conditions which are likely to have the greatest influence on deposition rates. The effects of air motion on the transport and deposition of radon daughters are examined for laminar, buoyancy-driven enclosure flows and for both free and forced convection boundary layer flows. The theoretical results indicate that
the deposition velocity decreases for subsequent daughters in the decay chain. It was also found that molecular diffusion in stagnant or laminar flow could not account for the deposition rates observed in experiments representative of realistic flow conditions.

The results of the parametric sensitivity studies are used to examine the adequacy of parametric models of deposition, interpret experimental measurements, and make recommendations regarding further experimental studies to investigate the dependence of deposition on environmental conditions. By treating a very complex process in a fundamental manner, the mass-transport model represents an important step in providing a realistic basis for understanding radon daughter behavior, and also provides a theoretical framework for quantitatively treating a variety of indoor pollutants for which spatial distributions and surface interactions are particularly important.
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<td>A</td>
<td>activity concentration (pCi/m³)</td>
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<tr>
<td>A&lt;sub&gt;avg&lt;/sub&gt;</td>
<td>average activity concentration</td>
</tr>
<tr>
<td>A&lt;sub&gt;c&lt;/sub&gt; or A&lt;sub&gt;core&lt;/sub&gt;</td>
<td>freestream or core activity concentration</td>
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<td>A&lt;sub&gt;s&lt;/sub&gt;</td>
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</tr>
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<td>b</td>
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<td>C</td>
<td>mole concentration (moles/m³)</td>
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<td>d&lt;sup&gt;a&lt;/sup&gt;, d&lt;sup&gt;f&lt;/sup&gt;</td>
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<td>free fraction</td>
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<td>f&lt;sup&gt;*&lt;/sup&gt;</td>
<td>dimensionless velocity vector = (\vec{u}/U)</td>
</tr>
<tr>
<td>F</td>
<td>air cleaning device filtration rate (hr⁻¹)</td>
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<tr>
<td>g</td>
<td>gravitational acceleration (9.81 m/sec²)</td>
</tr>
<tr>
<td>Gr</td>
<td>Grashof number = (g\beta TL³/v²), dimensionless</td>
</tr>
<tr>
<td>j</td>
<td>activity deposition flux (pCi/m² hr)</td>
</tr>
<tr>
<td>J</td>
<td>total rate of deposition (pCi/hr)</td>
</tr>
<tr>
<td>L</td>
<td>representative surface length (cm)</td>
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<tr>
<td>m</td>
<td>power coefficient</td>
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<tr>
<td>n</td>
<td>power coefficient, or direction normal to surface</td>
</tr>
<tr>
<td>N</td>
<td>particle concentration (particles/cm³)</td>
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<tr>
<td>p</td>
<td>power coefficient</td>
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<tr>
<td>Pe</td>
<td>Peclet number = (LU/D), dimensionless</td>
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<td>q&lt;sup&gt;a&lt;/sup&gt;, q&lt;sup&gt;f&lt;/sup&gt;</td>
<td>deposition rate (hr⁻¹)</td>
</tr>
<tr>
<td>r</td>
<td>recoil probability</td>
</tr>
<tr>
<td>(\hat{R})</td>
<td>dimensionless time step = (L/U\Delta t_o)</td>
</tr>
<tr>
<td>Ra</td>
<td>Rayleigh number = (Pr \cdot Gr), dimensionless</td>
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Re  \(\text{Reynolds number} = \frac{UL}{\nu}, \text{dimensionless}\)

S  \(\text{surface area (m}^2\)\)

\(S_1, S_2\)  \(\text{dimensionless source terms}\)

\(S_d\)  \(\text{stopping distance (cm)}\)

Sc  \(\text{Schmidt number} = \frac{\nu}{D}, \text{dimensionless}\)

t  \(\text{time (sec)}\)

\(\Delta t\)  \(\text{time step (sec)}\)

T  \(\text{temperature (°C)}\)

\(u^a, u^f, u^p\)  \(\text{deposition velocity (cm/sec)}\)

u  \(\text{velocity component (cm/sec)}\)

\(\vec{u}\)  \(\text{velocity vector}\)

\(u_*\)  \(\text{friction velocity (cm/sec)}\)

\(U, U_\infty\)  \(\text{characteristic or freestream velocity (cm/sec)}\)

v  \(\text{velocity component (cm/sec)}\)

\(\nu\)  \(\text{ventilation rate, air changes per hour (hr}^{-1}\)\)

V  \(\text{volume (m}^3\)\)

\(V_{th}\)  \(\text{thermophoretic deposition velocity (cm/sec)}\)

\(V_{ts}\)  \(\text{terminal setting velocity (cm/sec)}\)

x  \(\text{spatial coordinate}\)

X  \(\text{attachment rate (hr}^{-1}\)\)

y  \(\text{spatial coordinate}\)
Greek characters

α decay type, or thermal diffusivity (cm²/sec)

β decay type, or
attachment coefficient (hr⁻¹ (particles/cm³)⁻¹), or
coefficient of thermal expansion (°C⁻¹)

λ radioactive decay constant (hr⁻¹)

ν kinematic viscosity (cm²/sec)

θ dimensionless activity concentration (= A/A₀)

τ dimensionless time (t/Δt₀), or relaxation time (sec)

δ boundary layer thickness (cm)

∇ gradient operator

Superscripts

a attached daughter

f free daughter

p particle

Subscripts

i = 0, 1, 2, 3 radon and first three daughters
ACKNOWLEDGEMENTS

I would like to express my appreciation to the many generous people who helped me during this work. Sincere thanks are extended to Tony Nero and the Ventilation and Indoor Air Quality Group at Lawrence Berkeley Laboratory for their financial support, patience, and advice throughout the course of this research. In particular, I would like to thank Rich Sextro for his helpful discussions and suggestions, Bill Nazaroff for his invaluable literary critique, and Ashok Gadgil for sharing with me his convective model of air movement in enclosures. I am especially grateful to Ken Revzan for continually sharing with me his time, genuine interest, and expertise, and Buck Koonce for always doing all he could to make things easier for me. Credit must also be given to Gayle Milligan for her work-processor counselling, and to John Flambard for the final production graphics.

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In closing, I would like to express my most sincere gratitude and deepest appreciation to my family of friends. Their endless supply of love, encouragement, and understanding has always been a source of strength and support for me.
1.1 Objective

The objective of this work was to develop a physical model which uses a fundamental approach for examining the transport and removal mechanisms of radon daughters in enclosed spaces. The model has been used to study the deposition of radon daughters onto walls and other surfaces, an important removal mechanism at the low to moderate particle concentrations typically found indoors [Jac72, Po78a, Of84a]. Deposition rates vary substantially and, in the absence of a realistic theoretical model, inexplicably.

A theoretical model can provide detailed information about a variety of simulated conditions quickly and inexpensively. The predictions can be used to help interpret experimental results as well as formulate new experiments, and to determine the influence of building design and control techniques on indoor air quality. Foregoing the question of expense, a model's usefulness is only limited by the adequacy with which it mathematically represents realistic conditions.

The limitations of existing indoor radon models preclude examining many questions concerning the behavior and interactions of indoor radon and its daughters. These models are based on a macroscopic mass-balance, which requires the assumption of uniform concentrations within an enclosure. They do not account for the transport of pollutants or for spatially-variable pollutant sources and sink. They are only able to characterize average room concentrations in terms of estimated values of parameters which describe overall source and removal rates. In particular, a mass-balance model treats deposition as a volumetric
removal rate (e.g., an overall deposition rate) and requires prior knowledge of its value. This is a parameterization of the deposition process, which is actually a surface phenomenon and is strongly affected by environmental conditions.

In contrast, the model described here begins with the laws of physics which describe the motion of air and the transport of reactive pollutants, introduces appropriate boundary conditions to represent surface deposition, and then calculates the concentration distribution throughout the entire region of interest. With this approach, the "near-wall" behavior can be examined in greater detail than has previously been possible. Knowing the spatial concentration distribution, a local mass-transfer coefficient (deposition velocity) can be calculated, and the overall deposition rate for the space can be determined. This new approach enables one to examine how the deposition rate itself is influenced by the variable conditions found in rooms.

The model developed in this work treats transport and removal directly, rather than by gross parameterization. It is, therefore, an important step in providing a realistic basis for understanding radon daughter behavior and its dependence on physical and environmental conditions.

1.2 Scope

A two-dimensional numerical model, based on fundamental mass-transport equations, has been developed to simulate the behavior of indoor radon daughters. This mass-transport model has been employed to examine the effects of diffusivity, aerosol particle concentration, and convective air motion on the mechanism of diffusive deposition.
The major portion of this work focuses on the behavior of the first radon daughter in stagnant conditions and in laminar boundary layer and enclosure flows. The discussion places a particular emphasis on the deposition rates of daughters which are unattached to aerosol particles ("free" daughters). Concentration distributions of the second and third radon daughters were also simulated and this work briefly discusses the differences found in deposition rates for these decay products.

The complexity of the conditions for which the mass-transport model can be applied, and the corresponding questions which the model is currently able to address, are limited in part by the ability to simulate the air flows in those conditions. The air motion in buildings is extremely complex and varied and difficult to accurately measure or theoretically calculate in detail. Even the state-of-the-art models are only able to simulate relatively simplified single-zone flows. Although laminar flow is a simplification of realistic turbulent conditions, it is an important first step in attempting to characterize the effects of air motion on radon daughter transport and deposition. By treating a very complex process in a fundamental, but simplified manner, the mass-transport model provides the basis from which a more comprehensive and realistic model can be developed.

Chapter 2 constitutes a general introduction to the behavior of radon daughters indoors. Their interactions with airborne particles and room surfaces are summarized, and a brief description of existing indoor radon models and their limitations is presented.

Chapter 3 describes the physical basis, mathematical formulation, and numerical solution of the mass-transport model developed in this work.
Chapter 4 briefly summarizes the physical assumptions underlying the simulations which have been done to date. A general discussion is also presented concerning the characteristics of the enclosure and boundary layer flows that were investigated.

Chapter 5 is devoted to radon daughter deposition, which is the primary focus of this work. Brief descriptions of various deposition processes are given, followed by more detailed theory of deposition by molecular diffusion and how it is applied to the radon daughters. Physical interpretations of the measures of deposition, and how they are obtained through the mass-transport model, are also presented.

Chapter 6 is quite lengthy and involves detailed discussions of the parameters investigated in the parametric sensitivity studies, and their influence on deposition. Specifically, the parameters investigated include free daughter diffusivity, particle concentration, and air motion.

Chapter 7 relates the predictions of the theoretical model to experimental work. An critical review of existing experimental studies is first presented. The predictions are validated by comparing them to experimental results obtained in flow conditions which were most similar to those in the model simulations. The findings of the parametric studies are then used to interpret the results of a wide range of experimental measurements, assess the validity of some common assumptions used by experimental researchers, and made recommendations concerning future experimental studies.

The conclusions in Chapter 8 give an overall summary of the contributions of this work and the potential of the model in terms of recommendations for further study.
2.1 The Indoor Radon Problem

There has been increasing interest in recent years in the occurrence of elevated radon concentrations indoors [Ro83]. It has gradually become recognized that approximately half of the public's natural radiation exposure is due to radon daughter inhalation, and that this exposure predominantly occurs indoors [Ne83].

Radon-222 ($^{222}\text{Rn}$), a noble gas, is formed by the radioactive decay of $^{226}\text{Ra}$, a naturally occurring element in soil. Consequently, radon is also produced in earth-based building materials and groundwater. Being chemically inert, it migrates easily and can enter a building by a variety of pathways, which are illustrated in Figure 1. It is believed that pressure-driven transport from soil adjacent to the building structure is the predominant source of indoor radon in the U.S., Canada, and some European countries [Na84]. The building acts as a trap for the radon if the outside air available for dilution is relatively small, as in the case of low infiltration. As a result, indoor radon concentrations are often an order of magnitude greater, and can be much higher, than outdoor concentrations.

The radon decay chain is shown in Figure 2. There are four short-lived decay products (daughters) of radon: polonium-218 ($^{218}\text{Po}$), lead-214 ($^{214}\text{Pb}$), bismuth-214 ($^{214}\text{Bi}$), and polonium-214 ($^{214}\text{Po}$). ($^{214}\text{Po}$ has such a short half-life that for practical purposes it is considered to always be in equilibrium with $^{214}\text{Bi}$). For biological considerations, the decay chain effectively ends with the 5th daughter, lead-210 ($^{210}\text{Pb}$)
Figure 1. Primary pathways for radon entry in buildings. Radon, a naturally-occurring radioactive gas, originates in earth and earth-based materials. It can be transported into buildings via pressure-driven flow through penetrations in the substructure, by molecular diffusion from and through building materials, and via domestic water supplies.
Figure 2. The radon and radon daughter decay chain, with the corresponding half-lives and principal type of radiation associated with each. The shaded isotopes are those of primary biological concern due to inhalation and subsequent alpha decay. The shaded alpha decays are also those used to measure radon progeny concentrations.
which has a half-life of 22 years. Table 1 lists the physical properties of the first five radon daughters.

The short-lived decay products of radon are chemically active and pose a significant health risk. When inhaled, they can deposit in the lung and subsequently irradiate the surrounding tissue. Lung cancer is the most important health hazard, even though some of the inhaled decay products may pass into the bloodstream [Ta66].

2.2 Behavior of Radon Daughters

Indoor radon concentrations are usually moderately uniform within a single room, although they commonly vary among rooms, particularly on different floors. The behavior of indoor radon decay products is extremely complex, involving interactions with airborne particles and room surfaces. Figure 3 is a generalized schematic diagram which illustrates the various interactions and transformation processes of the radon daughters.

The first decay product, $^{218}$Po, initially exists either as a free ion, or as a small molecular cluster. In either form it is commonly referred to as a "free" daughter. A free daughter may become attached to an airborne particle, determining the size distribution of the radon decay products in the space. Experimental measurements show that the decay products follow a bimodal lognormal size distribution; the smaller mode comprising the free daughters, and the larger mode the attached [Ge78]. A typical size distribution of indoor radon daughters is shown in Figure 4.

For an attached daughter that undergoes alpha decay, i.e., a polonium (Po) isotope, the newly formed product can again be a free ion due to the high recoil energy of the decay. The free and attached
### TABLE 1

Properties of radon and its first five decay products.

<table>
<thead>
<tr>
<th>Isotopic designation</th>
<th>Principal type of radiation</th>
<th>Half-life</th>
<th>Decay rate (hr$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Name</td>
<td>Symbol</td>
<td>Radiation</td>
<td></td>
</tr>
<tr>
<td>radon-222</td>
<td>$^{222}$Rn</td>
<td>α</td>
<td>3.82 day</td>
</tr>
<tr>
<td>polonium-218</td>
<td>$^{218}$Po</td>
<td>α</td>
<td>3.05 min</td>
</tr>
<tr>
<td>lead-214</td>
<td>$^{214}$Po</td>
<td>β</td>
<td>26.8 min</td>
</tr>
<tr>
<td>bismuth-214</td>
<td>$^{214}$Bi</td>
<td>β</td>
<td>19.7 min</td>
</tr>
<tr>
<td>polonium-214</td>
<td>$^{214}$Po</td>
<td>α</td>
<td>0.00016 sec</td>
</tr>
<tr>
<td>lead-210</td>
<td>$^{210}$Po</td>
<td>β</td>
<td>22 yr</td>
</tr>
</tbody>
</table>
Radon Decay and Removal Processes

Other Removal Processes:

- Ventilation (v)
- Control Device (F)

Radon
Decay ($\lambda$)
Free Daughter
Attachment (X)
Attached Daughter
Particle

Recoil ($^{218}$Po only)
($r \lambda$)
Deposition ($q^f$)
Deposition ($q^a$)

Wall or Other Macro Surface

Figure 3. Schematic diagram of various decay and removal processes (and their associated rates) affecting concentrations of radon and radon progeny. The radioactive decay pathways for radon progeny are not explicitly noted in this diagram.
Figure 4. Histograms of typical indoor radon daughter size distributions, where the small mode represents the unattached ("free") radon daughters. The measurements were obtained by diffusion batteries and were confirmed by independent measurements with wire screens [Ge78].
fractions of each decay product depend on the relative magnitudes of the
decay and attachment transformation characteristics and the removal
rates in the space. The attachment and recoil processes are described
in more detail in later sections of this work.

The daughter-particle interaction and resulting size distribution
has biological implications by affecting both the extent to which the
radioactive daughters will pass through various parts of the human
respiratory system and the likelihood of deposition onto sensitive
tissues. With regard to determining the adverse health effects due to
daughter inhalation, the relative importance of the free and attached
daughters is not completely understood. However, it has been found
that, being smaller, the free daughters have a higher probability of
being deposited in the trachea-bronchial regions of the lung. Due to
the lack of a protective mucous layer, clearance mechanisms are slow
there and longer residence time in the lung results in a higher dose to
the bronchi, where the majority of lung cancers are first seen [Jam81].

The size distribution also influences the transport and removal of
the pollutants within the room. In particular, the diffusivity D (a
measure of mobility) is strongly dependent on particle diameter, with
higher values of D corresponding to smaller, more mobile particles. A
natural removal mechanism for radon daughters is deposition onto walls
and other surfaces. Although the mechanisms are not well understood,
this rate of removal can be significant for the free daughters. It is
comparatively small for the attached daughters due to their relative
immobility. Thus, deposition of the free daughters effectively
determines the total deposition rate for a particular decay product.
2.3 Existing Indoor Radon Models

2.3.1 Formulation

The most commonly used indoor pollutant models are based on a macroscopic mass-balance assuming uniform concentrations within the enclosure [Jac72, Po78a]. These are often referred to as "uniformly-mixed models" or "box models". The steady-state mass conservation laws, as applied in these models, give rise to simple algebraic equations which have no spatial or temporal dependence and can easily be solved.

For modeling radon daughter concentrations, the free and attached modes of each decay product are treated separately. Neglecting the spatial distribution, the steady-state mass-balance equation for each concentration simply equates the source terms to the removal terms. Each term is expressed by the product of the appropriate concentration and a corresponding transformation rate constant.

A flow chart of the partitioning used in the uniformly-mixed model is shown in Figure 5. Applying the mass conservation law to the free and attached daughters in terms of mole concentration, \( C_i^f \) and \( C_i^a \), the steady-state mass balance equations can be expressed as a general formulation for daughter i.

\[
\lambda_{i-1} C_{i-1}^f + r_{i-1} \lambda_{i-1} C_{i-1}^a = (\lambda_i + q^f + X) C_i^f \quad (1a)
\]

\[
(1-r_{i-1}) \lambda_{i-1} C_{i-1}^a + X C_i^f = (\lambda_i + q^a) C_i^a \quad (1b)
\]
Flow chart of partitioning used in uniformly-mixed model

Figure 5. Flow chart of the partitioning used in the uniformly-mixed model. Free and attached modes of each decay product are treated separately. The partitions represent the free and attached molar concentrations of each decay product, and are coupled through the transformation processes (radioactive decay, attachment, and recoil) which are described by corresponding transformation rate constants. Both free and attached daughters are removed by surface deposition. (Note that removal by ventilation and air cleaning devices is neglected in this diagram.)
The transformation rates in these equations are radioactive decay rate $\lambda$, attachment rate $X$, recoil probability $r$, and free and attached deposition rate constants $q_f$ and $q_a$. (It should be noted that the flow chart in Figure 5 and the above equations neglect removal by ventilation or air cleaning devices). The steady-state solutions to these equations are given in Appendix A. There are many papers which present detailed discussions of the uniformly-mixed model [Br83, Jac72, Kn83, Pa78a, Wi81]. The intent here is to discuss the parameters relevant to the mass-transport model described in this work.

2.3.2 Description of Parameters

Values of the radioactive decay rate constants, $\lambda$, are well known and are independent of the age of the atom or environmental conditions such as temperature, pressure, concentration, or chemical combination. Table 1 lists values of the decay constants for the short-lived radon daughters. The values of the other rate constants depend on various parameters and have varying degrees of uncertainty.

Radon daughter attachment is described by an overall attachment rate, $X$, which is a function of aerosol concentration $N$ and size distribution. Attachment is a diffusion process, controlled by the influences of electrostatic attraction and gas kinetics [Po78b]. The attachment rate constant, $X$, is a parameter used to represent this process. Its value is obtained by assuming a linear relationship between $X$ and $N$, with a constant coefficient of proportionality (the attachment coefficient, $\beta$), based on mean particle diameter. The mass-transport model treats attachment in his same manner, and therefore does not contribute anything new to understanding the actual attachment
process, but only its implications in terms of concentration distributions and deposition rates.

The probability of recoil is described by a recoil factor, $r$, which depends on the momentum transfer to the daughter molecule upon radioactive decay of the parent. During a decay process, the decay energy is distributed among the reaction products, with the lighter particles receiving most of the energy such that momentum is conserved [Co83]. The recoil probability will be determined by the relative mass of the emitted radioactive particle compared to the emitted radioactive particle, and the energy of the emitted particle. Alpha particles are much heavier and have significantly higher decay energy than beta particles. Thus, the nucleus will recoil with a much higher energy following an alpha decay, compared to a beta decay. As a result, the recoil probability is relatively high for alpha decay, and is negligible for low energy beta decay [Me76].

To calculate the probability of recoil, Mercer postulates that the recoiling atom will escape into the air only if it is unable to dissipate all its energy before it can emerge from the particle. Assuming that the radon daughter is on the surface of a spherical particle, and examining the total range of angles at which it can be projected, Mercer calculates a theoretical value of $r = 0.83$ (based on a lognormal particle size distribution with a geometric mean diameter of 0.2 $\mu$m and a recoil range of 0.14 $\mu$m, corresponding to a particle density of unit specific gravity) [Me76]. The mass-transport model treats the recoil process in the same way and presently assumes a constant recoil probability of $r = 0.83$ for alpha decay.
The uniformly-mixed model accounts for deposition on surfaces through an overall deposition rate constant, $q$, which is given different values for the free and attached daughters ($q_f$ and $q_a$) due to differences in diffusivity. Users of the model have always assumed that the values for $q_f$ and $q_a$ are independent of other parameters and are identical for all decay products, although there has generally been little agreement regarding the actual values of these removal rates.

2.3.3 Limitations of Existing Models

The uniformly-mixed model is a good tool for simple parametric studies, but does not examine the behavior of indoor radon daughters in a fundamental manner. The model is only able to characterize the average concentrations in a room, and does not consider the actual mechanisms of the transport and removal processes. Although the assumption of uniform concentration in a single room may be reasonably acceptable for most purposes, the applicability of the uniformly-mixed model is limited by the inability to track the movement of pollutants from their sources to other areas within the enclosure or to permit spatial- or time-dependent sources.

Using the uniformly-mixed model, predictions of indoor pollutant concentrations are highly dependent upon the choice of values for the rate constants. The model requires prior knowledge of all values of exchange rates and assumes these values to be uniform in space and constant over time. It does not have the ability to simulate the transfer process itself and therefore determine the influence of parameters on the values of the rate constants. The uncertainty in these required values presents an important limitation. The greatest uncertainty lies in the estimation of the deposition rate constants.
The mechanisms of deposition depend on the mobility of the decay product and a driving potential, both of which are characteristic of the particular mechanism. Diffusive deposition, for example, depends on both the diffusion coefficient of the decay product and also the concentration gradient near the surface. This profile is determined by the environmental conditions and, consequently, one would expect that the deposition rate constant would depend not on the diffusion coefficient alone, but also on indoor conditions such as particle concentration and air flow patterns [Br83], which are generally difficult to characterize in enclosed spaces. The concentration distribution of each daughter depends on the distribution of the parent concentration; consequently, one might also expect that deposition rates would vary for the different decay products.

In contrast to the uniformly-mixed model, the mass-transport model does not parameterize deposition but treats it in a fundamental manner based on physical principles. 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 daughter concentrations. This approach is an important step as it now enables one to examine the influence of indoor conditions on estimates of the deposition rate constants.
3.1 Mass Transport Equations

The mass-transport model is based on laws of physics which govern the behavior of indoor radon daughters in terms of their generation, convective and diffusive transport, and removal mechanisms. The velocity fields of a particular flow are assumed to be independent of the pollutant concentrations, and are obtained separately. Fundamental mass-transport equations, in two-dimensional form, separately describe the free and attached concentrations of each radon daughter. The equations are coupled through source and removal terms which include the radioactive decay rate, attachment rate, and recoil factor. Deposition is accounted for through boundary conditions. The solution of the mathematical formulation of the model is based on an approximating numerical technique.

The general mass-transport equation for any component, i, of a mixture is a differential equation derived by applying the law of conservation of mass of component i to an infinitesimal volume fixed in space. A transport law (Pick's first law of diffusion) is introduced so that the equation is written in terms of a single dependent variable (i.e., concentration) \([\text{Ka80, Sp63, Bir60}]\). The component can be carried through this volume element by convective transport and also by diffusion. Both modes of transport depend on factors which are a function of time and space.

Within the volume element, component i may also be created and destroyed by a homogeneous transformation process. The term "homogeneous" refers to a volumetric reaction, i.e., one which takes place throughout
the region of interest, and the net rate of production due to such a transformation appears in the differential equation as a source term. For this model of radon daughters, radioactive decay, attachment, and recoil are all represented as homogeneous transformation processes. Consequently, the net production term in the differential equations is equal to the source terms (parent atom decay, recoil for the free mode, attachment for the attached mode) minus the removal terms (daughter decay, attachment for the free mode, recoil for the attached mode).

A "heterogeneous" transformation is a surface reaction, i.e., one which occurs at a restricted region, and is represented as a boundary condition which is required to solve the differential equation. Boundary conditions can be expressed in terms of a specified concentration or flux at a surface, or as a mass flux which is written in terms of a mass-transfer coefficient or surface reaction rate [Bir60]. Radon daughter deposition is an example of a heterogeneous transformation.

For many pollutants it is often appropriate to speak in terms of mole concentration (C). For radon daughters, since exposure is due to radioactive decay, concentrations are commonly given in terms of activity concentration \( A = \lambda C \). For notational convenience, superscripts \( f \) and \( a \) are used to designate the free and attached daughters, respectively. Radon and the first three decay products, \( ^{218}\text{Po} \), \( ^{214}\text{Pb} \), and \( ^{214}\text{Bi} \), are represented by subscripts 0, 1, 2, and 3, respectively. Using vector notation, and writing in terms of activity concentration, the two-dimensional transport equations for decay product \( i \) (\( i=1,2,3 \))
are presented below in Equations (2a) and (2b).

\[
\frac{\partial A_i^f}{\partial t} + \nabla \cdot (\nabla A_i^f) = \lambda_i A_{i-1}^f + r_{i-1} \lambda_i A_{i-1}^a - (\lambda_i + X) A_i^f \quad (2a)
\]

\[
\frac{\partial A_i^a}{\partial t} + \nabla \cdot (\nabla A_i^a) = (1 - r_{i-1}) \lambda_i A_{i-1}^a + X A_i^f - \lambda_i A_i^a \quad (2b)
\]

The terms on the left side these equations represent, respectively, the time rate of change of activity concentration of component i, convective transport, and diffusive transport. The right side of each equation represents the source and removal terms. These equations are referred to as the "governing equations" for daughter concentration. The solution of these equations requires specification of initial conditions and boundary conditions.

Initial conditions of these recursion expressions (i.e., for radon, \(i=0\)) are \(A_0^f = A_0\), and \(A_0^a = 0\). In general, the radon concentration \(A_0\), which serves as the initial source distribution for all the daughters, can be any specified function of time or space.

Although the attachment process is parameterized in these equations by the attachment rate, \(X\), the model still presents an advantage over the uniformly-mixed model in that attachment does not necessarily have to occur uniformly over the entire space. The value of \(X\) in the equations can be specified as any generalized function of time or space, representative of variability in particle size or concentration.

By assuming that pollutant concentrations are not high enough to affect the air viscosity and velocity profiles, the pollutant mass-transport equations are effectively decoupled from the conservation of
momentum equations which are used to solve for the velocity fields. The velocity profiles can then be calculated independently and used as input to the mass-transport equations. The validity of this assumption is determined by evaluating the relative partial pressures of the air and the pollutant. The results of these calculations indicate that, even at concentrations much higher than those seen indoors, the assumption is an accurate one.

It has also been assumed that the pollutant concentrations are small enough such that the diffusion coefficients are not affected by the presence of other components. The partial pressure calculations previously mentioned also serve to validate this assumption.

For convenience, the governing equations can be expressed in terms of dimensionless variables. These dimensionless variables are defined by dividing the variables in equations (2a) and (2b) by constant reference parameters which are chosen to be characteristic of the particular problem (i.e., radon concentration $A_0$, surface length $L$, representative velocity $U$, time step $\Delta t_0$). The governing equations, and the initial and boundary conditions, are then formulated in terms of these non-dimensional variables. After rearranging, the resulting form of the equations contain important dimensionless combinations of the characteristic parameters. The non-dimensional form of the governing equations can be written as

\[
\frac{1}{R} \frac{\partial \theta^f_i}{\partial \tau} + \frac{1}{R} \frac{\partial \theta^f_i}{\partial r} - \frac{1}{\text{Re Sc}} \frac{\partial^2 \theta^f_i}{\partial r^2} = \hat{R} (S_1^f + S_2^f \partial \theta^f_i) \quad (3a)
\]

\[
\frac{1}{R} \frac{\partial \theta^a_i}{\partial \tau} + \frac{1}{R} \frac{\partial \theta^a_i}{\partial r} - \frac{1}{\text{Re Sc}} \frac{\partial^2 \theta^a_i}{\partial r^2} = \hat{R} (S_1^a + S_2^a \partial \theta^a_i) \quad (3b)
\]
The dimensionless groups in equations (3) have physical significance which are descriptive of various aspects of the flow. For the mass transport model, these groups include the following: the Schmidt number \((\text{Sc} = \nu/D)\), which is a measure of the relative potentials of momentum and mass diffusion; and the Reynolds number \((\text{Re} = LU/\nu)\), which is the ratio of the inertial to frictional effects in the flow. These two groups appear multiplied together, and their combination is called the Peclet number \((\text{Pe} = \text{Sc} \text{Re} = LU/D)\), which is the ratio of the potential strengths of convection and diffusion. In these expressions, \(\nu\) is the kinematic viscosity of the fluid (air).

3.2 Numerical Discretization Concepts

The coupled mass-transport equations are non-linear, partial differential equations which cannot be solved analytically. The Patankar-Spalding Differencing Technique [Pa80] provides the basis for their numerical solution; this is an approximating numerical technique which involves the conversion of each equation into discrete form ("discretization") and a matrix solution of the resulting algebraic equations. An important characteristic of this method is that it is based on physical considerations, rather than just on mathematical manipulations.

A finite number of non-overlapping subregions (control volumes), each represented by a unique grid point, divides the region of interest. The time period of interest is also divided into a finite number of steps. The discretization method replaces a continuous distribution of the dependent variable (activity concentration, \(A\)) with discrete values at each grid point and for each time step. The values for \(A\) at the grid
points are connected through the discretization equation. An iterative solution marches through the time steps to produce the time-dependent concentration distribution. The iterations are repeated until steady-state is reached (determined when the differences in concentrations between iterations is less than 0.1%). The following sections discuss these concepts in more detail.

3.3 Numerical Solution Techniques

3.3.1 Grid

Because of the approximating nature of a numerical solution, the grid scale must be sufficiently fine to obtain an accurate solution. To minimize computing costs, a non-uniform grid is used; a fine grid spacing is used to get high resolution in the regions near the wall, where most of the changes are occurring, while a coarser grid is used in the core region of the room, where the dependent variable changes rather slowly. The non-uniform grid used in the enclosure model is illustrated in Figures 6 and 7, respectively drawn for the entire enclosure and also expanded for one corner to illustrate the relative spacing.

3.3.2 Discretization Equation

The discretization equation is derived by integrating the governing differential equation over the control volume surrounding the grid point, and over the time step. The integration replaces the continuous derivatives by "finite differences", relating the value of A at the grid point to values at the neighboring grid points. For a region represented by M nodes, the differential equation is replaced by a set of M algebraic equations which can be solved by one of a variety of matrix solution techniques.
Figure 6. Non-uniform grid used for enclosure to define nodal points in finite-difference numerical scheme.

Figure 7. Non-uniform grid, expanded for corner of enclosure to illustrate the relative spacing near the walls.
The integration process involves assuming a set of piecewise profiles which describe how \( A \) varies between the grid points. There are many discretization formulations, distinguished by the choice of the assumed profile. The simplest formulation is the "central difference" scheme, which results from assuming a linear profile between the nodes. However, if convective transport is strong relative to diffusive transport, this formulation can lead to physically unrealistic results. The "upwind" scheme solves this difficulty by assuming, for the discretization of the convection terms, that a constant value of \( A \) exists between the nodes, equal to the value at the upstream node. This is only accurate, however, if convection is relatively strong. Alternative formulations, such as the "hybrid" scheme, are written such that they attempt to account for the direction and relative magnitude of convection at each node. Examples of the mathematical formulation using each of these schemes is given in Appendix B, and a discussion of the advantages or limitations of these various schemes is given elsewhere [Pa80]. The formulation of the mass-transport model described in this work uses the hybrid scheme.

When discretizing the spatial derivatives and the source terms, a profile must also be specified to describe how values of \( A \) vary during each time step. The model uses an "implicit" scheme; at the beginning of each time step, the value at each node suddenly changes to its new value and remains at that value during the entire interval of the time step. Using this method, the new value at each node is linked to the unknown new values at the neighboring nodes. A solution, therefore, requires that the whole set of equations for all the nodes be solved simultaneously. Although the "explicit" scheme permits direct
calculation of new values at each node, by linking them to the known old values at the neighboring nodes, the stability of this scheme is affected by the choice of grid spacing and time step interval. For problems requiring a large number of nodes for an accurate solution, the implicit scheme is preferred [Pa80]. The derivation of the discretization equation, assuming constant physical properties, and the final form of the discretization equation and coefficients is given in Appendix B.

3.3.3 Matrix Solution

Once the discretization equations are formulated, the set forms a matrix of the coefficients which can be solved by any suitable method. In the discretization equations, each node is linked only to its neighboring nodes. Consequently, the resulting matrix is very sparse (i.e., a large number of elements are zero) and iterative methods of solution can be used which are relatively simple and fast.

In this work, a line-by-line iterative solution technique was used which is referred to as the alternating-direction implicit (ADI) method [Pa80]. For a two-dimensional grid, this approach begins by splitting the time step into two parts. The first sweep of this iteration is made in one direction (e.g., the x-direction) and the discretization equations are solved for all the grid points along one line at a time (e.g., parallel to the y-axis). Only the values for the nodes on the particular line are considered unknown, while the values at all the other nodes are obtained from the last iteration. The second sweep would then be made in the opposite direction (e.g., the y-direction).

Using this method, the set of equations for the grid points along a single line form a matrix identical to a set of one-dimensional equations. Each equation involves only three unknowns (i.e., the grid
point and its two neighbors along the particular line), and the set forms a tri-diagonal matrix in which all the nonzero coefficients are aligned along the three main diagonals. The algorithms for inverting a tri-diagonal matrix are a simplified form of the standard Gaussian-elimination method, and are commonly referred to as the Tri-Diagonal Matrix Algorithm (TDMA) [Pa80].
4.1 Physical Assumptions

The mass transport model simulates the two-dimensional transport of radon daughters in both the free and attached modes. It is assumed that the radon concentration is uniform throughout the space. Radon decay is the only source for the first free daughter. Removal by ventilation or control devices is not considered.

Physical parameters in the equations include the transformation characteristics (decay, attachment, and recoil), and the diffusion coefficient. In simulations, it is assumed that the values of all these parameters are constant with respect to space and time. The values of decay rate and recoil probability were discussed in an earlier section. In actuality, the attachment rate will decrease to zero across a thin region near the walls due to particle deposition. This effect was accounted for in some test simulations and was found to make essentially no difference in predictions of radon daughter concentrations and deposition rates.

4.2.0 Flow Characterization

Understanding the airborne (convective) transport of pollutants requires detailed information about the air flow patterns in the space. Air movement in a building can be generated in a number of ways: infiltration, natural ventilation, mechanical ventilation, or buoyancy-driven natural convection. The air flow patterns are typically due to a combination of several influences, and the characterization of these flows is difficult.
Initial simulations using the mass-transport model have only looked at laminar conditions of buoyancy-driven enclosure flows, and free and forced convection boundary layer flows. As mentioned previously, assumptions have been made which decouple the Navier-Stokes equations and the mass-transport equations. The velocity fields can then be determined independently of the concentration distributions. For the complex recirculating flows in enclosures, the mass-transport model uses velocity data generated from a separate model of air movement in buildings. For the simple one-directional boundary layer flows, the model includes calculations of reasonable approximations of the velocity profiles. These are obtained through integral methods of solution of the boundary layer form of the Navier-Stokes equations.

The following sections provide more detailed discussions of the enclosure and boundary layer flows which were investigated.

4.2.1 Buoyancy-Driven Enclosure Flows

Buoyancy-induced fluid motion, commonly referred to as "natural convection", is the result of gravitational body forces acting on the fluid. These forces are due to density differences arising from temperature gradients in the space.

The motion of the fluid is described by the Navier-Stokes equations. These are non-linear partial differential equations which represent the laws of conservation of mass, momentum, and energy applied to a fluid element. The momentum and energy equations are coupled through the body force term; consequently, the velocity field depends on the temperature distribution. The Navier-Stokes equations are presented in Appendix C.
For input to the mass-transport model, data representing the velocity field in an enclosure are obtained from a convection model (developed by Gadgil at Lawrence Berkeley Laboratory), which is based on a numerical discretization of the coupled Navier-Stokes equations. The convection model simulates both natural and forced convection, for any combination of obstacles, openings, and velocity or heat sources and sinks in a rectangular shaped enclosure [Ga80]. The dimensionless parameters which characterize natural convection flow in an enclosure include: the aspect ratio (height/width), which describes enclosure geometry; the Prandtl number ($Pr = \nu / \alpha$), which characterizes fluid properties and is a measure of the relative potentials of momentum and thermal diffusion; the Grashof number ($Gr = g \beta \Delta T L^3/\nu^2$), which characterizes the flow and is a measure of the ratio of buoyancy to viscous forces; and the Rayleigh number ($Ra = Pr Gr$), which is a measure of the relative strengths of convective and conductive heat transfer.

(It should be noted that, in the expression for $Gr$, $\beta$ represents the coefficient of thermal expansion and should not be confused with the $\beta$ discussed previously which is used to represent the attachment coefficient).

As previously indicated, there are a number of influences which can contribute to the air flow patterns in buildings. There will always be some degree of natural convection due to temperature differences within the space, although the relative strengths of natural and forced convective flow will depend on factors such as the magnitude of the temperature differences (due to heat distribution and solar gain) and the source of the forced air flow.
Using the mass-transport model, initial simulations investigated a simple recirculating enclosure flow. Specifically, velocity fields were calculated for a two-dimensional square enclosure with boundary conditions corresponding to isothermal hot and cold walls and adiabatic floor and ceiling. A schematic drawing of streamlines in this flow is shown in Figure 8. For a given wall length and constant fluid properties, the flow is characterized by a specified Grashof number, which is proportional to the temperature difference between the walls.

4.2.2 Boundary Layer Flows

In addition to enclosure flows, pollutant transport was also simulated for simpler boundary layer flows. The velocity in a boundary-layer flow is predominantly in one direction without recirculation. The flow at one point is independent of the downstream behavior, and convection always dominates diffusion in this streamwise direction.

Boundary layer theory can also be applied to the mass-transport equations. When changes in concentration occur essentially only over a narrow layer near the surface (the boundary layer), which is thin relative to the dimensions of the surface, dimensional analysis shows that diffusion in the streamwise direction can be neglected.

A significant consequence of the boundary layer approximation is that it changes the character of the transport equation from an elliptic to a parabolic form. An elliptic equation is a boundary-value problem and can only be solved by specifying the boundary conditions on a complete contour enclosing the region. A parabolic equation, on the other hand, is a mixed initial- and boundary-value problem. Conditions need only be specified at one x-position, where x represents the streamwise direction. Computer solutions are then relatively easier.
Figure 8. Schematic diagram of streamlines, illustrating the velocity field in a simplified buoyancy-driven flow. Velocities are highest along the isothermal walls, as indicated by the streamlines being closer together.
because $x$ is now a marching variable; the profile can be initialized at $x=0$ and the solution then "marches" down the plate in the direction of the flow [Pa80]. The boundary layer forms of the Navier-Stokes equations are presented in Appendix C.

A boundary layer flow analysis is advantageous compared to a full enclosure flow. The solution at a particular point is independent of the behavior downstream; consequently, the iterative procedure used for recirculating flows is not necessary and storage requirements and computing costs are substantially less. The grid only needs to be extended out to the edge of the boundary layer, so one can get more detailed information near the wall by incorporating a much finer grid in this region than is possible for an entire enclosure. Overall, it is faster and less expensive to use a boundary layer analysis, particularly for the many simulations required for parametric sensitivity studies.

For many enclosure flows, specific regions of the flow can often be represented by simpler boundary layer flows. A good example of this is the simple enclosure flow that was investigated in these simulations. As discussed in a later section, the flow along the walls can be adequately represented by a boundary layer free convection flow along an isothermal single surface within quiescent surroundings, unbounded by other surfaces. The flow along the floor and ceiling can be fairly well represented by a boundary layer forced convection flow along a horizontal flat plate with an imposed freestream velocity. Schematic diagrams of velocity profiles in free and forced convection boundary layer flows are shown in Figures 9 and 10, respectively. The mathematical descriptions of these velocity profiles are presented in Appendix C.
Figure 9. Representative velocity profile, $u(y)$, of a free convection boundary layer flow along a hot, isothermal vertical surface within cooler, quiescent surroundings, unbounded by other surfaces.

Figure 10. Representative velocity profile, $u(y)$, of a forced convection boundary layer flow along a horizontal flat plate with an imposed freestream velocity, $U$. 

Schematic Diagram of a Free Convection Boundary Layer Flow

Schematic Diagram of a Forced Convection Boundary Layer Flow
5.1 Introductory Concepts

The current study employed the mass transport model primarily for examining the mechanisms by which airborne daughters are deposited on surfaces. (For convenience, the term "particle" will refer to either a free daughter, or a daughter-bearing particle (attached daughter)).

When a particle collides with a surface, it "sticks", or deposits, on that surface. An assumption used in the model is that the wall is a perfect sink; particles adhere perfectly to the wall without returning to a state of suspension. In this way, the model is essentially examining the "near-wall" behavior (i.e., diffusive and convective transport towards or away from the surface), while ignoring the "on-wall" behavior (i.e., detachment and reentrainment, or "bouncing").

The interaction between the particle and the wall is often described in terms of an accommodation coefficient. Perfect adherence at the wall is represented by an accommodation coefficient equal to one. The process of detachment, and the resulting value of the accommodation coefficient, may be strongly influenced by wall roughness. The assumption of negligible detachment probability from a surface seems to be reasonable, however. The recoil distance for alpha decay is approximately .015 cm [Br83], which is much less than the typical boundary layer thickness near the wall. Thus, a recoiling nucleus which detaches from the wall is likely to be redeposited.

Assuming that particles touching the wall are removed from the air implies that the airborne pollutant (activity) concentration at the surface ($A_g$) can be taken as zero [Fu64], and that there exists a thin
wall region over which the concentration increases to the freestream, or core, value \(A_c\). The thickness of the wall region depends on the relative magnitudes of the homogeneous source and removal terms (radioactive decay and attachment), the diffusion coefficient, and also the intensity of convection, which contributes to the transport of pollutants either towards or away from the wall. The resulting concentration gradient in this wall layer causes a continuous diffusion of pollutants towards the surface \([Hi82]\).

Beginning with a fundamental approach for calculating the rate of deposition along the surfaces, a mass transfer coefficient can be calculated which does not directly depend on the enclosure geometry and is representative of the surface deposition process. From this information, it is desirable to calculate for the enclosure a value for the overall deposition rate constant (as used in the uniformly-mixed model), which is essentially a measure of the average probability that particles will reach known boundaries \([Fu64]\). An overall deposition rate constant would allow one to calculate the total rate of deposition for the enclosure in terms of the average concentration in the space. The discussion in this chapter provides a basis for connecting experimental data, normally interpreted using the uniformly-mixed model, to results from the mass-transport model.

5.2 Deposition Processes

Deposition of particles can be due to the influence of many processes. As examples, these can include molecular diffusion, turbulent (eddy) diffusion, gravitational settling, inertial impaction, electostatic attraction (electrophoresis), and thermophoresis. This
section presents brief descriptions of these mechanisms and, when applicable, estimates of their significance for the particle sizes typically found indoors.

Molecular diffusion is the net transport of particles in a concentration gradient due to random Brownian motion. Diffusion increases in magnitude for smaller particle diameter and, excluding convection, it is the primary transport mechanism for particles less than 0.1 μm in size [Fu64]. The present formulation of the mass-transport model accounts for deposition by molecular diffusion, and this process is discussed in much more detail in Section 5.3.

In contrast to laminar flow, where the smooth streamlines can usually be described mathematically, turbulent motion involves the random fluctuating movement of large eddies, or "chunks", of fluid. These eddies result in mass being transported at rates much faster than those which can be accounted for by the relatively slow process of molecular diffusion. Consequently, deposition rates in a turbulent flow can be expected to be much higher than those in laminar conditions. Some of the difficulties in theoretically predicting turbulent deposition rates include: determining the functional form of the eddy viscosity, which is descriptive of the momentum transport in the various regimes of a turbulent flow; determining the extent to which particles follow the turbulent fluctuations, expressed by a relationship between the eddy diffusivity and eddy viscosity; and determining the rate of re-entrainment of the deposited particles, which is likely to occur when the turbulent flow velocity is high. Sehmel presents a critical review of models of turbulent particle deposition, as well as the results of his own experimental and theoretical studies [Se70, Se71]. The process
of turbulent deposition is extremely complex and has not been accounted for in the simulations described here. This will be an important next step for further applications of the mass-transport model.

Particles will deposit on upward-facing surfaces due to gravitational settling. The magnitude of this deposition process can be described by the particle's terminal settling velocity, $V_{ts}$, which is reached quickly and is the condition where the drag force of the air on the particle is exactly equal and opposite to the force of gravity. The value of $V_{ts}$ increases rapidly with particle size, and is negligible for the particle diameters associated with the free and attached radon daughters. This can be seen in Table 2, which lists values of $V_{ts}$ for a representative range of particle diameter.

An example of inertial impaction is the situation where a surface causes an airstream to turn, and particles with sufficient inertia are unable to follow the streamlines and will impact on the surface. It is extremely difficult to calculate the rate of inertial deposition, but its potential can be described by the relaxation time, $\tau$, which is a property of the particle and is proportional to the square of its diameter [Hi82]. The value of $\tau$ characterizes the time required for a particle to adjust its velocity to a new condition of forces, and can be used to determine the maximum distance a particle with an initial velocity $V_0$. This distance is often referred to as the "stopping distance", $S_d$, or "inertial range". Representative values of $\tau$ and $S_d$ (assuming $V_0 = 1$ cm/sec) for typical indoor particle sizes are given in Table 2. These values are very small, indicating that particles of this size cannot be thrown very far and deposition by inertial impaction can be considered to be negligible.
Most particles carry some electric charge and will experience an electrostatic force in the vicinity of charged surfaces. This force depends on the magnitudes of the particle charge, and the strength of the electric field in the space around the charged surface. A quantitative calculation of the magnitude of this deposition process is difficult and requires detailed information of the particle charge distribution and the charging of the walls. A general discussion of electrostatic forces on particles is given by Hinds [Hi82], and a simplified approach for examining electrophoretic deposition in enclosures is presented by van de Vate [Vat80]. The mass-transport model does not presently account for electrostatic forces, and further investigations are needed to determine the significance of this influence.

Thermophoretic deposition is caused by a thermal force on the particle which is a result of strong temperature gradients in the air. There is a greater momentum transfer from the gas molecules on the hot side of the particle compared to the cold side, resulting in a net force in the direction of cooler temperature. The mechanism is complicated, particularly for particles of diameter greater than 0.1 μm because a temperature gradient is established in the particle itself [Hi82]. Table 2 lists representative values of the thermophoretic velocity, \( V_{th} \), which is proportional to the temperature gradient, for a unit temperature gradient (1°C/cm). This temperature gradient is at least one or two orders of magnitude higher than those which might exist in typical conditions inside buildings. The values in Table 2 are very small, indicating that thermophoretic deposition is probably not significant for the conditions typically found indoors (except, possibly, for localized regions of
TABLE 2

Representative values of parameters characterizing various deposition mechanisms.

Characteristic parameters of deposition mechanisms

<table>
<thead>
<tr>
<th>Particle diameter (μm)</th>
<th>GRAVITATIONAL</th>
<th>INERTIAL IMPACTION</th>
<th>THERMOPHORETIC</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Terminal settling velocity</td>
<td>Relaxation time (sec)</td>
<td>Stopping distance (cm)</td>
</tr>
<tr>
<td>.01</td>
<td>$7.0 \times 10^{-6}$</td>
<td>$6.8 \times 10^{-9}$</td>
<td>$6.8 \times 10^{-9}$</td>
</tr>
<tr>
<td>.1</td>
<td>$8.8 \times 10^{-5}$</td>
<td>$8.8 \times 10^{-8}$</td>
<td>$8.8 \times 10^{-8}$</td>
</tr>
<tr>
<td>1.0</td>
<td>$3.5 \times 10^{-3}$</td>
<td>$3.6 \times 10^{-6}$</td>
<td>$3.6 \times 10^{-6}$</td>
</tr>
</tbody>
</table>

1 based on pressure $P = 1$ atm.
2 based on initial velocity $V_0 = 1$ cm/sec.
3 based on temperature $T = 20^\circ C$ and unit temperature gradient $= 1^\circ C/cm$. 
extreme uneven heating). An examination of thermophoretic deposition in enclosures is given by van de Vate [Vat80]. The mass-transport model does not account for this deposition mechanism.

In all flow conditions, there is a thin, relatively stagnant layer near the wall in which molecular diffusion predominates. The driving force for deposition by molecular diffusion is the concentration gradient across this wall layer. This is the deposition mechanism which the current simulations of the mass-transport model have investigated. The theory of diffusive deposition, and its application to radon daughters, is discussed in detail in the following sections.

5.3.1 Theory

Both convective and diffusive transport contribute to the motion of a pollutant. However, at a solid (non-porous) surface, the velocity components are zero and so the deposition flux is simply equal to the rate of diffusion at that surface.

Assuming that the diffusion of a particular pollutant is independent of temperature and pressure gradients, and concentration gradients of other components in the mixture, Fick's first law of diffusion can be applied at a surface to obtain the activity deposition flux, \( j \) (e.g., units of \( \text{pCi m}^{-2} \text{ hr}^{-1} \)). This law is analogous to Newton's law of viscosity and Fourier's law of heat conduction, and states that the rate of diffusion at any point in space is proportional to the concentration gradient of the diffusing substance at that point. Writing in terms of activity concentration, Fick's law can be applied at
the surface to obtain the activity deposition flux, \( j \)

\[
j = D \left. \frac{dA}{dn} \right|_{\text{surface}} \tag{5}
\]

The diffusivity, \( D \), is the coefficient of proportionality, and \( n \) is the distance measured normal to the wall. Diffusive transport is in the direction opposite the concentration gradient, and equation (5) is written with the convention that deposition flux \( j \) is positive towards the wall when \( n \) is measured positive away from the wall.

Deposition is a heterogeneous transformation and can be described in terms of a characteristic concentration driving potential and a mass transfer coefficient, or an effective surface reaction rate. Similar to the convective coefficient used in heat transfer theory, a mass transfer coefficient, \( u \), can be defined by

\[
j = u (A_C - A_s) = u A_C \tag{6}
\]

where \( (A_C - A_s) \), the concentration difference between the core and the surface, represents the driving potential, and \( A_s = 0 \) for deposition. Combining equations (5) and (6), the mass transfer coefficient is expressed as

\[
u = \frac{j}{A_C} = \frac{D}{A_C} \left. \frac{dA}{dn} \right|_{\text{surface}} \tag{7}
\]

The mass transfer coefficient has units of length/time and is often called the "deposition velocity" (commonly expressed in units of cm/sec). It is defined as the flux density per unit core concentration, and can be considered the effective velocity with which particles migrate to a surface [Hi82]. Equation (7) represents the local
deposition velocity at a point along a surface. One can also calculate the average deposition velocity, \( \langle u \rangle \), for an enclosure by using the average flux, \( \langle j \rangle \). Both average quantities, \( \langle j \rangle \) and \( \langle u \rangle \), can be calculated by integrating the expressions for \( j \) and \( u \), respectively, over all surfaces in the space, then dividing by the total room surface area, \( S \)

\[
\langle j \rangle = \frac{1}{S} \int j \cdot dS \tag{8}
\]

\[
\langle u \rangle = \frac{1}{S} \int u \cdot dS = \frac{\langle j \rangle}{A_c} \tag{9}
\]

The average deposition velocity, \( \langle u \rangle \), is useful because it can now be related to the deposition rate constant, \( q \), used in the uniform-mixing model. The deposition rate constant is essentially a volumetric reaction rate. In this way, the uniform-mixing model is expressing deposition as a homogeneous transformation. As used in the uniform-mixing model, the total rate of activity deposition, \( J \) (e.g., units of pCi/hr), in an enclosure can be written as

\[
J = q \cdot A_{\text{avg}} \cdot V \tag{10}
\]

where \( V \) is the total room volume and \( A_{\text{avg}} \) is the average activity concentration in the room.
If the steady-state mass transport equation is integrated over the entire space, the total rate of activity deposition can also be expressed by

\[ J = \int_{\text{all surfaces}} D \frac{\partial A}{\partial n} \cdot dS = \langle j \rangle \cdot S = \langle u \rangle \cdot A_c \cdot S \] (11)

Comparing equations (10) and (11), and noting that, for a large enclosure, \( A_{\text{avg}} \) is approximately equal to \( A_c \), the overall deposition rate constant can be defined by

\[ q = \langle u \rangle \cdot \frac{S}{V} \] (12)

This relationship, and the assumption that \( A_{\text{avg}} = A_c \), implies that room conditions are well-stirred and that all the surfaces are equally effective for deposition.

5.3.2 Deposition of Radon Daughters

The deposition velocities, or rates, of the free and attached radon daughters have to be determined independently, due to their differences in diffusivity and concentration profiles. Experimental measurements, on the other hand, are often in terms of total (free plus attached) daughter concentrations and removal rates. The individual (free and attached) quantities are connected to the total deposition rate through the free fraction, and the relationship between them is straightforward. Similar to previous notation, quantities which relate to the free or attached daughters are specified by superscripts \( f \) and \( a \), respectively. Total quantities for each daughter have no superscript.
For each daughter $i$, the free fraction $f_i$ is defined as the ratio of free to total daughter activity concentration:

$$f_i = \frac{A_i^f}{(A_i^f + A_i^a)} = \frac{A_i^f}{A_i}$$  \hspace{1cm} (13)

The free and attached daughter activity concentrations can then be expressed by

$$A_i^f = f_i A_i \quad A_i^a = (1-f_i) A_i$$  \hspace{1cm} (14)

and the free and attached rates of activity deposition can be written as

$$J_i^f = q_i^f A_i^f \cdot v \quad J_i^a = q_i^a A_i^a \cdot v$$  \hspace{1cm} (15)

where $A$ in equations (13), (14), and (15) represents average activity concentration. Combining equations (11), (14), and (15), the following relationship can be written for the total deposition rate constant for daughter $i$:

$$q_i = f_i q_i^f + (1-f_i) q_i^a$$  \hspace{1cm} (16)

An effective "total deposition velocity" can be mathematically defined in a similar way and written as

$$u_i = f_i u_i^f + (1-f_i) u_i^a$$  \hspace{1cm} (17)

However, this quantity is not very useful in that it does not have a clear physical interpretation, since the free and attached daughters clearly migrate to the surface with different "effective" velocities due to the differences in mobility.
In summary, the steps for examining radon daughter deposition using the mass-transport enclosure model involve:

1. solving for the spatial free and attached activity concentration distributions in the space, $A^f$ and $A^a$;
2. calculating local free and attached activity flux, $j^f$ and $j^a$, along each surface using Fick's law of diffusion, equation (5);
3. integrating the local flux over all surfaces to determine the free and attached rates of deposition in the space, $J^f$ and $J^a$;
4. calculating free and attached average deposition velocities, $u^f$ and $u^a$, and overall deposition rate constants, $q^f$ and $q^a$, using equations (11) and (12), respectively, and finally;
5. calculating a total deposition rate, $q$, using equation (16).

The mass-transport model represents a fundamental approach for obtaining values of deposition velocities and overall deposition rate constants for a specified set of environmental conditions. Depending on the type of measurements taken in experimental work, the mass-balance equations of the uniformly-mixed model are generally used to calculate values of the deposition rate constants to fit the measured data. The equations and concepts presented in this chapter provide the basis for comparing experimental results to predictions from the mass-transport model.
6.1 Introduction

The mass-transport model was used to perform parametric sensitivity studies, examining the parameters which initially seemed likely to vary the most in the published experiments, or have the greatest influence on concentrations and deposition rates.

Experimental measurements of radon daughter concentrations, and the subsequent attempts to calculate deposition rates from these measurements, vary widely for different researchers. A comparison of some of these experimental results is presented later in Chapter 7. The variations among these results may be due to several factors: personal, statistical, or systematic errors encountered in the measurement techniques; variations in the environmental conditions under which the experiments were conducted; or the assumptions that needed to be made to calculate deposition rates from the actual measurements [Ge80]. When reporting the results of their experiments, researchers have often attempted to evaluate the estimated range of error of their measurements, and have presented at least a brief discussion of the assumptions and calculations behind their values of free and attached deposition rate constants. However, to determine how much the variations in experimentally determined values of deposition rates might be due to experimental conditions, a better physical understanding is needed regarding how those conditions might influence pollutant behavior. Here a theoretical framework for addressing these questions is useful.
The sections in this chapter first discuss the parameters and flow conditions which were investigated in the parametric sensitivity studies, indicating the motivation behind their choice and the range of values which were examined. A summary of these parameters and flows is given in Table 3. A description of the velocity field in the enclosure flow, and a comparison of the predicted deposition velocities in the enclosure and representative boundary layer flows, is then presented. The following discussions concern the effects of the parameters on deposition and the variability among the daughters. The major findings, and some of their implications, are summarized in the final section.

6.2.0 Key Parameters

6.2.1 Diffusivity

It is clear that the value of the diffusion coefficient has an important influence on deposition rates. Because of its strong dependence on diameter, the value of the free daughter diffusion coefficient, $D^f$, depends on the daughter's physical and chemical nature. Many researchers have investigated the influence of environmental conditions on the diffusion coefficients of radon daughters, with a particular emphasis on the more mobile free daughters [Bi71, Bu81, Ch56, Po69, Po68, Raa68, Rag79, Th70].

Reported measurements have shown conflicting trends regarding the effect of increasing relative humidity on free daughter diffusivity [Bu81, Raa68, Rag79, Th70]. However, it has generally been found that free daughter diffusivity increases with higher ventilation rates, corresponding to shorter residence times [Bu81, Rag79]. This is consistent with other measurements that indicate that older atoms have
TABLE 3

Summary of parameter values and flow conditions investigated in simulations.

PARAMETRIC SENSITIVITY STUDIES

- FREE DAUGHTER DIFFUSIVITY
  
  0.01 - 0.1 cm$^2$/sec

- ATTACHMENT RATE
  
  0.0 - 250.0 hour$^{-1}$

- FLOWS
  
  One-Dimensional Diffusion
  Enclosure Diffusion
  Forced Convection Boundary Layer Flow
  Free Convection Boundary Layer Flow
  Buoyancy-driven Recirculating Enclosure Flow
more chance to grow and coalesce, resulting in larger effective
diameters and smaller diffusion coefficients [Bi71, Bu81, Raa68, Th70].

Raghunath's measurements show that the diffusivity of each daughter
is smaller than that of the parent atom, and this trend was seen for
various combinations of ventilation rates and humidity [Raa79]. A
smaller free diffusivity implies a larger effective diameter of the
molecular cluster. This might also be explained by the notion that
daughters further down the decay chain are effectively older (i.e.,
$^{214}$Po has a much longer half-life than $^{218}$Po and coalesces into a larger
cluster. It may not be likely that it will break apart upon beta decay,
implies that $^{214}$Bi will initially exist as a cluster and continue to
grow). Since the diffusion coefficient of the daughters varies, it seems
likely that the deposition velocity would also vary between daughters.

Researchers have measured a wide range of values for free daughter
diffusivity. Busigin gives a good overview of much of this 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 [Bu81].
The low value, 0.005 cm$^2$/sec, has been observed for the third daughter,
$^{214}$Bi, in humid air [Raa79], while the high value, 0.1 cm$^2$/sec, has been
observed for neutralized radioactive primary ions [Fo69, Ro68]. Busigin
also suggested that free daughters actually exist in many forms, and the
high values of deposition rates estimated from diffusion tube studies
may be explained by observing that only the most mobile species in these
experiments may have been measured [Bu81].

The average, or commonly accepted, value of free daughter
diffusivity is 0.054 cm$^2$/sec [Br83, Bu81, Ch56, Kn83]. As an attempt to
determine how much the variations in experimental measurements may
simply be due to differences in the physical state of the daughters, the sensitivity studies described here examined values of $D^E$ which vary over an order of magnitude, $0.01 - 0.1$ cm$^2$/sec, encompassing the range found by the majority of researchers and centering around the commonly used value. Examining the magnitudes of the resulting deposition rates will help determine whether the experimentally determined deposition rates can be accounted for by molecular diffusion.

The diffusivity of the relatively immobile attached daughters can effectively be determined by the activity-weighted mean particle diameter. Because of the higher probability of attachment to larger particles, there is a shift in the size distribution of the particles and attached daughters. The average particle diameter found in typical indoor environments is about $0.1 \, \mu m$ [Of84a], with a corresponding activity-weighted average of $0.15-0.2 \, \mu m$ [Kn83, Me76, Po83]. A commonly used value of attached daughter diffusivity is $4.5 \times 10^{-6}$ cm$^2$/sec [Kn83], and this is the value used in the sensitivity studies.

6.2.2 Particle Concentration

Researchers have not yet recognized the notion that particle concentration can affect the individual free and attached deposition rates. Consequently, this effect has not been accounted for by users of the uniformly-mixed model, or investigated through experimental measurement.

The uniformly-mixed model serves as a simple, but useful, parametric tool to illustrate the effect of particle concentration (expressed as an attachment rate, $X$) on overall radon daughter concentrations. For a specified value of $X$, and assumed values of the free and attached daughter deposition rates, $q^f$ and $q^a$, the free
fraction and total deposition rate for each daughter can be calculated. These calculations are given in Appendix A. However, because the uniformly-mixed model is not able to determine the influence of environmental conditions on the values of \( q^f \) and \( q^a \), users of this model have simply assumed that these values were constant, independent of environmental conditions, and identical for all daughters.

Some researchers have accompanied the radon daughter measurements with simultaneous measurements of particle concentration, and used this simple parametric approach to better understand the particle-daughter interaction and to compare mass-balance model predictions with experimental observations [Ge83, Kn83, Of84a]. Researchers who use this approach have generally believed that the effect of particle concentration, or attachment rate, on the total deposition rate constant, \( q \) for each daughter arise entirely from its direct effect on the free daughter fraction. This can be understood mathematically which implies that, in using equation (16) to calculate \( q_i \) for daughter \( i \), it is assumed that the values for \( q^f \) and \( q^a \) are identical for all daughters and independent of particle concentration. It does not appear that researchers have considered that the attachment rate also influences the values of the individual free and attached deposition velocities, \( u^f \) and \( u^a \), or rate constants, \( q^f \) and \( q^a \). Although this concept may not be intuitively obvious, it does have a physical basis which will be discussed in a later section in connection with the findings of this study. The significance of this influence will also be examined.

To examine the effect of particle concentration on the individual deposition velocities \( u^f \) and \( u^a \), \( X \) was varied over a range which corresponds to the 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, or a corresponding attachment coefficient of 4.3 x 10⁻³ hr⁻¹/(particles/cm³), this range of X = 0-250 hr⁻¹ corresponds to a particle concentration of up to 60,000 particles/cm³. This extends beyond the range typically found indoors, excepting the case of heavy smoking.

6.2.3 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. Although the characteristic flow conditions have varied considerably among experiments (ranging from stagnant to turbulent conditions), except for a few there have been no measurements of typical air velocities in the space or across any of the surfaces. Because of the lack of air flow measurements and the inability of the uniformly-mixed model to account for airborne transport, the effects of air motion on radon daughter deposition have not been characterized.

For similar "known" environmental conditions (i.e., particle concentration, ventilation rate, humidity, etc.), researchers still find wide variation in their measurements and calculations of deposition rates. Although it would be helpful to have better quantitative information concerning typical air velocities during the time of the experiment, it is still useful to have a qualitative understanding of the extent to which these differences might be accounted for by the variations in overall experimental flow conditions.
Perhaps the major contribution of the mass-transport model 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 the flows in real buildings or in the experiments, and to simulate a flow which is representative of those particular conditions. Given these limitations, however, the mass-transport model still provides a useful theoretical framework to investigate the relative effects of air motion on the predicted concentration distributions and deposition rates.

To examine the effects of air motion on deposition, several flow conditions were examined. Free daughter diffusivity and attachment rate were varied in each flow, over the appropriate ranges which were discussed in the two previous sections. As noted previously, Table 3 summarizes the various conditions which were examined.

Analytic calculations were first done using a simple film model (one-dimensional diffusion). This approach is useful in that, (for those comfortable with mathematics), the resulting expressions provide an informative means for conceptualizing the functional dependence of deposition in stagnant conditions. The condition of pure diffusion is also 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. As mentioned previously, the simpler boundary layer flows might adequately represent portions of the
enclosure flow, and this question will be addressed in a later section. 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 parametrics at consideraly less expense, and the results of the parametric studies provide a clearer picture of the observable patterns in terms of the near-wall behavior.

The magnitude of the flows was indirectly set by giving values to the representative temperature difference (for the free convection flows), or freestream velocity (for the forced convection boundary layer flow). For the free convection flows, a specified temperature difference determines the velocity field. The value corresponds to the temperature difference between the walls (for the enclosure flow) or between the wall and the surroundings (for the boundary layer flow). To compare the two flows, the wall-to-surrounding temperature difference in the boundary layer flow corresponds to half the wall-to-wall temperature difference in the enclosure flow. The temperature differences investigated varied by an order of magnitude, and the values correspond to varying the Grashof number of a 3 x 3 meter enclosure by an order of magnitude. For enclosure Grashof numbers equal to 3x10⁹ and 3x10¹⁰, the corresponding wall-to-wall temperature differences in the enclosure are 0.846 and 8.460 degrees Celsius. To compare the forced convection boundary layer flow to the enclosure flow, the imposed free stream velocity corresponds to the nominal horizontal air velocities along the floor and ceiling. For the two values of enclosure Grashof number given above, these velocities are approximately 0.3 and 0.6 cm/sec, respectively.
In addition to using the boundary layer flows to represent the recirculating enclosure flow, concentrations in a laminar forced convection boundary layer flow were simulated for free stream velocity ranging from 1 - 100 cm/sec, to examine the relative effects of a wide range of air velocity.

Needless to say, simulations using the various combinations of parameters and flow conditions generated an enormous amount of information. While the results of all the simulations were examined and analyzed, it is not necessary (nor effective) to present all of them here. The following discussions and graphs provide examples which appropriately illustrate the important findings of the parametric sensitivity studies. In particular, the final sections concern the effects of air motion and the variability among the daughters. These seem to be the most interesting findings, and probably have the greatest significance with regard to interpreting experimental results and leading to recommendations for further experimental studies.

6.3.0 Discussion of Findings

6.3.1 Enclosure Flow Conditions

As previously described, the recirculating enclosure flow corresponds to a square two-dimensional enclosure with isothermal hot and cold walls, and adiabatic floor and ceiling. For the enclosure, the overall velocity flow field can best be visualized by a plot of the streamlines, as shown in Figure 11 for Grashof = \(3 \times 10^9\). The streamlines are an informative pictorial representation of both the direction and magnitude of a fluid flow. The velocity vectors are always tangent to a streamline (i.e., no fluid crosses the streamlines), therefore the
Figure 11. Streamlines for the simulated buoyancy-driven enclosure flow for Grashof number $= 3 \times 10^9$. For a two-dimensional, 3 x 3 m square enclosure, this corresponds to a temperature difference of $T = 0.846 \, ^\circ C$ between the hot and cold walls. Floor and ceiling are adiabatic. Velocities are relatively high along the walls, and the boundary layer is correspondingly thin. Vortices form in the downstream corners of each wall flow, resulting in the streamlines being deflected away from the adiabatic surfaces.
volume rate of flow between any two streamlines is constant. This implies that the velocities are highest in the regions where the streamlines are closest together.

An immediate observation is that the velocities along the isothermal walls are relatively high compared to the stratified flow in the central core, indicating that a thin boundary layer exists along these walls. Vortices form in the downstream corners of each wall flow, resulting in the streamlines being deflected away from the adiabatic surfaces.

Representative magnitudes of the velocity components at various points in the room for this flow can be seen in Figures 12 and 13, which show the horizontal and vertical velocity profiles across selected sections. The convention used in these graphs is that a positive velocity corresponds to vertical flow upward and horizontal flow to the right. The profiles also illustrate the "swirling" and deflection of the flow in the regions of the corner eddies. In general, it is observed that velocities parallel to the surfaces are highest along the walls, ranging from about 1 to 7 cm/sec, and are slightly less than 0.5 cm/sec along most of the floor and ceiling. Recalling the previous discussion of the boundary layer concept, a comparison of the relative magnitudes of the streamwise and cross-stream velocity components along each surface (i.e., parallel and normal to the surface, respectively) indicated that the flow along the central regions of each surface, particularly the walls, can accurately be characterized as a boundary layer flow. These observations will now be discussed in more detail.

Figure 12 shows the vertical velocity profiles at selected points along the hot wall. The peak streamwise velocity of the developing flow
Figure 12. Vertical velocity profiles near the hot wall in the Grashof number $= 3 \times 10^9$ enclosure flow. Profiles are plotted across selected horizontal sections along the wall. The profiles illustrate the acceleration of the flow as it develops further along the wall, and the subsequent deceleration as the influence of the ceiling is felt.
increases from about 1 cm/sec near the floor or ceiling to a maximum of about 7 cm/sec midway along the wall (at 150 cm), while the boundary layer thickness correspondingly increases from about 2 cm to 6 cm. Past the midway point, the boundary layer continues to grow only slightly, but the flow is retarded and the velocities within the boundary layer begin to decrease again as the influence of the horizontal bounding surface downstream (the ceiling) is felt.

Because this work is concerned with the question of pollutant deposition, it is also of interest to examine the velocity component normal to the surfaces, since this is the component which will enhance or diminish the transport by diffusion by carrying the pollutants either towards or away from the wall, respectively. As mentioned previously, a comparison of the normal (cross-stream) and streamwise velocity components is also necessary to examine the boundary layer characteristics of the wall flow. It was found that, along the central region of the wall, the cross-stream velocity components are less than 0.1 cm/sec, and in the direction towards the hot wall (positive). This is at least an order of magnitude below the values of streamwise velocity, indicating that the wall floor is, indeed, characteristic of a boundary layer flow.

Figure 13 shows the horizontal velocity profiles along the floor. When the strong cold wall flow first turns the corner, horizontal velocities are initially high. Past the corner, the flow stabilizes and the velocity profiles continuing downstream are fairly consistent. For both the floor and ceiling, maximum velocities in this region are about 0.4-0.45 cm/sec, occurring at a distance of about 4-5 cm away from the horizontal surface. The velocities then decrease to a freestream
Figure 13. Horizontal velocity profiles above the floor in the Grashof number $= 3 \times 10^9$ enclosure flow. Profiles are plotted across selected vertical sections along the floor. The profiles are fairly consistent across the floor, with peak velocities which are low relative to those along the walls.
value of about 0.3 cm/sec. As suggested by the uniform profiles along the surface, the cross-stream velocity components normal to the floor are negligible.

6.3.2 Representation of simplified flows

As mentioned earlier, if results from the simpler boundary layer flows can adequately represent those from the enclosure flows, parametric studies using the boundary layer flows will be less expensive and the results will be easier to analyze.

Figures 14 and 15 show representative velocity profiles for the free and forced convection boundary layer flows, respectively. Comparing these to Figures 12 and 13, which show the velocity distributions along the hot wall and floor in the enclosure flow, illustrates the similarity between these simplified flows and the corresponding regions of the recirculating enclosure flow. Even more important than comparing the velocity fields, however, is a comparison of the predicted concentration profiles and values of deposition velocities. It is likely that the boundary layer treatments give generally less representative results for the profiles than for the transfer rates [Bir60].

Figure 16 is a plot of $u_f$ vs. $X$, as calculated from the enclosure model for Grashof number $= 3 \times 10^9$, and the representative free and forced convection boundary layer flows. As discussed in section 6.2.3, these representative boundary layer flows correspond to a temperature difference of 0.423 °C and a free stream velocity of 0.3 cm/sec, respectively. The Figure illustrates that there is excellent agreement between the predictions from these flows. This comparison essentially validates the findings of the parametric studies for the boundary layer flows as being representative of the enclosure flows. It also may
Figure 14. Representative vertical velocity profiles in a free convection boundary layer flow along a hot vertical wall. The profiles of this simplified flow are very similar to those along a hot wall in a recirculating enclosure flow.
Forced Convection
Horizontal Velocity Component

Figure 15. Representative horizontal velocity profiles in a forced convection boundary layer flow along a flat plate. Although slightly different, the profiles of this simplified flow resemble those along the floor in a recirculating enclosure flow.
Figure 16. Variation of free deposition velocity with attachment rate, for Po-218. Predictions from the Grashof number = $3 \times 10^9$ enclosure flow, and representative free and forced convection boundary layer flows are compared. The excellent agreement among these predictions serves to partially validate the findings of the parametric studies for the boundary layer flows as being representative of the enclosure flow.
justify further use of the simpler boundary layer flows for future parametric studies.

6.3.3 One-dimensional diffusion model

The simplest mass-transport model is a one-dimensional diffusion model, commonly known as the film model, in which the "film theory" assumes that the species diffuses through a stagnant film near the surface. The thickness of this film is defined such that beyond its outer edge the fluid is well-mixed and concentration gradients are negligible [Bir60]. The mathematical formulation of this model is essentially an equation describing one-dimensional diffusion of a chemically reactive species, and can be solved exactly. (It should be noted that the mathematical concept of a "boundary layer" does not apply in a one-dimensional model, thus the term "film thickness" or "wall region" is more appropriate for this case).

Applying this model to the free and attached radon daughters results in analytic expressions for the concentration distributions and mass-transfer coefficients, or deposition velocities, $u^f$ and $u^a$. Although the model may not physically represent room conditions, the correlations are informative and quite useful in terms of providing a simple physical picture. Much of the insight which can be gained from studying these results can also be applied to more complex systems; consequently, the mathematical formulations serve as a good starting point for illustrating some of the physical concepts which govern the behavior of indoor radon daughters.

The solutions to the one-dimensional diffusion equations for the three radon daughters are presented in Appendix D. The resulting expressions for deposition velocity in stagnant conditions illustrate
the complexities of the dependence of $u^f$ and $u^a$ on the various parameters and the potential differences between daughters. These differences will be discussed in later sections. The final expressions for $u^f$ and $u^a$ for the first daughter, $^{218}\text{Po}$, are presented here for reference and discussion:

$$u^f = \frac{D_f^f}{d_f^f} \quad u^a = \frac{P^a}{d_f^f + d^a}$$

(18)

where $d_f^f$ and $d^a$ are the free and attached daughter "diffusion lengths" defined as follows:

$$d_f^f = \left[ \frac{D_f^f}{(\lambda + X)} \right]^{1/2} \quad d^a = \left[ \frac{P^a}{\lambda} \right]^{1/2}$$

(19)

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 can then be expressed as:

$$u^f = \frac{D_f^f}{b_f^f} \quad u^a = \frac{P^a}{b^a}$$

(20)

Combining equations (18) and (20), one can write:

$$b_f^f = d_f^f \quad b^a = d_f^f + d^a$$

(21)

These expressions are simple, yet they are indicative of important physical concepts which cannot be illustrated from the mass-balance model or existing experimental measurements. The thickness of the wall
region for the free daughter concentration distribution, $b^f$, is obviously related to the free daughter diffusion length, $d^f$. The attached daughter concentration distribution is strongly determined by that of its source, the free daughter. Consequently, the attached daughter concentration profile, and the corresponding wall region thickness, $b^a$, will actually depend on both the free and attached diffusion lengths, $d^f$ and $d^a$. One implication of this result is that $u^a$ depends not only on attached daughter diffusivity, $D^a$, but also on the physical nature of the free daughter, as described by $D^f$.

This same concept can be applied to all the daughters. The concentration profiles are determined not only from the respective diffusion coefficients, but are strongly dependent on the source distribution of each decay product. Since each decay product is generated from different source distributions, it seems natural to assume that there will also be variations between the deposition velocities of each daughter.

Substituting equations (19) into (18), and noting that $d^f \gg d^a$, $u^f$ and $u^a$ can be written as

\[ u^f = (D^f (\lambda + X))^{1/2} \quad (22a) \]

\[ u^a = D_a \left[ \frac{(\lambda + X)}{D^f} \right]^{1/2} = \left[ \frac{D^a}{D^f} \right]^{1/2} \left[ (D^a (\lambda + X)) \right]^{1/2} \quad (22b) \]

Equations (22) express the functional dependence of $u^f$ and $u^a$ on $D^f$ and $D^a$, $\lambda$, and $X$, for $^{218}$Po in stagnant conditions. The following
sections will refer to these relationships, which will be used as a basis of reference for examining how air motion affects the various sensitivities.

6.3.4 Effect of free daughter diffusivity on deposition

The results of the parametric sensitivity studies were examined to determine the influence of $D^f$ on the predicted values of $u^f$ and $u^a$, and also the relationship between the ratios $(u^f/u^a)$ and $(D^f/D^a)$.

As expressed in equation (7), the deposition velocity is proportional to the diffusion coefficient $D$ and the concentration gradient at the surface, and is inversely proportional to the core concentration. Assuming a linear concentration profile at the surface, equation (20) expresses the deposition velocity in terms of an effective boundary layer thickness, $b$, which is descriptive of the shape of the concentration profile within the wall region.

The profile of the attached daughters depends on both the diffusivity $D^a$ and the distributions of its sources (i.e., the attached parent atom and also the free daughter through the attachment process). Consequently, it is expected that $b^a$ (and $u^a$) will also depend on the value of $D^f$. It was indicated in the previous section that the profiles of the attached daughters in stagnant conditions are dominated by the source distributions. Comparing equations (20) and (22) shows that

$$b^a \sim b^f \sim (D^f)^{1/2}$$

When there is a convective flow contributing to pollutant transport, it is expected that $D^f$ will have a relatively smaller influence on the thickness of both the free and attached boundary layers.
But \( b^f \) and \( b^a \) may not be similarly influenced by the increasing air motion and the relationship \( b^a \sim b^f \) will then no longer be valid. Compared to the free daughters, diffusive transport of the attached daughters is insignificant. Consequently, it is likely that the air motion will have a relatively larger influence on the transport of the attached daughters. Mathematically, this implies that

\[
b^f \propto (D^f)^m \quad b^a \propto (D^f)^n \quad (24)
\]

\[
u^f \propto (D^f)^{1-m} \quad u^a \propto (D^f)^{-n} \quad (25)
\]

where \( 0 < n < m < 1 \).

These physical considerations can be supported by examining some of the simulation results. The first point is illustrated in Figures 17 and 18, in which the concentration profiles of free and attached \(^{218}\text{Po}\), respectively, are plotted for two values of \( D^f \) (0.01 and 0.1 cm\(^2\)/sec) and for the cases of one-dimensional diffusion and forced convection with a freestream velocity \( (U) \) of 10 cm/sec. The profiles are plotted in terms of non-dimensional free and attached concentrations which have been normalized by the respective values of core concentration. In both graphs, comparing the curves for the two values of \( D^f \) illustrates that the sensitivity of both \( b^f \) and \( b^a \) to \( D^f \) is greater for diffusion than for convective flow.

The differences between \( b^f \) and \( b^a \) are illustrated by comparing the two figures. For stagnant conditions, the free and attached profiles are identical and both are strongly influenced by \( D^f \). The higher air velocity diminishes the sensitivity of \( b^a \) to changes in \( D^f \) much more strongly than that of \( b^f \) (i.e., \( n \ll m \) in equation (24)), thereby magnifying the differences between them.
Figure 17. Free daughter activity concentration profiles, for Po-218, illustrating the relative influence of free daughter diffusivity and air motion. The strong sensitivity of the concentration profiles to diffusivity is diminished only slightly when there is a convective flow contributing to pollutant transport. The sensitivity to air motion is also diminished for higher values of diffusivity.
Figure 18. Attached daughter activity concentration profiles, for Po-218, illustrating the relative influence of free daughter diffusivity and air motion. The free daughter serves as the source distribution of the attached daughter. As a result, the attached Po-218 profiles can be strongly influenced by free daughter diffusivity, particularly for stagnant conditions.
After investigating the effects of the concentration profiles, Figures 19 through 21 now serve to illustrate the effects of air motion on the sensitivity of the deposition velocities to changes in $D^f$. As indicated by physical considerations, and expressed by equation (25), it is expected that $u^f$ will become more sensitive and $u^a$ will become less sensitive to changes in $D^f$ as air motion increases. The relative magnitudes of both these effects are shown in Figures 19 and 20 in which $u^f$ and $u^a$ are respectively plotted vs. $D^f$, for various values of air velocity. In Figure 19, the sensitivity of $u^f$ to $D^f$ increases slightly over the range of air velocity investigated. Researchers have measured values of free daughter diffusivity which vary 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.

As shown in Figure 20, $u^a$ decreases slightly with higher values of $D^f$, but becomes increasingly less sensitive to changes in $D^f$ for higher air speeds. The curves indicate that at an air velocity of 30 cm/sec, $u^a$ is nearly independent of the value of $D^f$ (i.e., $n = 0$). For stagnant conditions, $u^a$ will decrease a maximum of three-fold for an order of magnitude increase in $D^f$.

Equations (20) can be used to express the ratio of $u^f$ to $u^a$ by

$$\frac{u^f}{u^a} = \frac{D^f}{D^a} \ast \frac{b^a}{b^f}$$

(26)

As noted previously, the values of $b^f$ and $b^a$ for each daughter are descriptive of the shapes of the respective free and attached concentration profiles within the wall region. These profiles are
Figure 19. Variation of free deposition velocity with free daughter diffusivity, for Po-218. This figure serves to illustrate the effects of higher air velocities on increasing the sensitivity of free deposition velocity to changes in free diffusivity.
Figure 20. Variation of attached deposition velocity with free daughter diffusivity, for Po-218. This figure serves to illustrate that attached deposition velocity decreases slightly with higher values of free diffusivity, but becomes increasingly less sensitive to free diffusivity for higher air velocities.
influenced not only by the diffusion coefficients but also the respective source distribution, relative strengths of the homogeneous source and removal terms, and the rate of convection which contributes to the transport of pollutants either towards or away from the wall. These influences can be mathematically described by expressing equation (24) in the following form

\[
\frac{u_f^e}{u_a^a} \sim \left( \frac{D_f^e}{D_a^a} \right)^p
\]

(27)

where \(0 < p < 1\) and its actual value is determined by the environmental conditions.

The significance of this relationship is indicated by the fact that researchers have often used this expression to calculate one of the unknown parameters in terms of assumed or measured values of the others. For example, if \(u^a\) is determined from particle measurements, values of the diffusivities are assumed and \(u^e\) is then calculated from this relationship [Jac72]. On the other hand, if assumptions have been made to extract values of both \(u^e\) and \(u^a\) from the measurements, the relationship is used to determine the effective \(D^e\) (or free daughter diameter) which would be necessary to account for these values [Kn83].

Although an accurate fundamental basis does not exist for analytically evaluating the value of \(p\), researchers have made various assumptions and used values of \(p = 1/2\) or \(p = 2/3\) in their calculations. It appears that these numbers may have come from two simplified cases which can be solved analytically. The first case corresponds to a reactive component with a uniform source in stagnant conditions, and the relationship between deposition velocity and diffusivity is \(u \propto D^{1/2}\), as
shown in equation (22a) for the first free daughter. If the relationship is assumed to be valid for both the free and attached daughters, then the value of $p$ in equation (27) would be equal to $1/2$. The second case corresponds to a non-reactive component (i.e., particles) in a forced convection flow, and the relationship is $u \propto D^{2/3}$. If the free and attached daughters are simply treated as particles with the respective values of diffusivity, then the value of $p$ in equation (27) would be equal to $2/3$.

Neither of these cases is perfectly accurate. Previous discussions pointed out that the attached daughter is generated from a non-uniform source, and its profile is therefore also determined by the value of $D_f$. Combining the expressions in equations (22) suggests that $p$ will have a value of 1, and not $1/2$, corresponding to the case of diffusion in stagnant conditions. When there is a convective flow contributing to pollutant transport, it is expected that the values of diffusivity will have a relatively smaller influence on the thickness of the concentration boundary layer, and also that $u^3$ will be less influenced by values of $D_f$. This implies that the coefficient $p$ will range from a maximum corresponding to stagnant conditions, to a minimum corresponding to conditions of higher velocities. It is not clear, however, that an analysis based on particle deposition (i.e., the second case), can determine the appropriate value of $p$ corresponding to conditions of faster air flow.

The results of the parametric studies will be examined to determine the extent of this influence. Figure 21 illustrates the relationship between the ratios $(u_f/u^3)$ and $(D_f/D^3)$, in a forced convection flow with freestream velocity $U$ ranging from 0 - 300 cm/sec. The greatest
Figure 21. Relationship between the ratios of free-to-attached deposition velocity and free-to-attached diffusivity, for Po-218. Researchers often use this dependence to calculate one of the unknown parameters in terms of assumed or measured values of the others. The relationship is linear for stagnant conditions, while the sensitivity is decreased with higher air velocities.
sensitivity is for stagnant conditions (maximum $p = 1$), and the decreased sensitivity with higher air speeds can be mathematically described by smaller values of $p$ in equation (27). At the highest velocity shown, 300 cm/sec, the curve can be represented by the minimum value of $p = 0.7$.

6.3.5 Effect of attachment rate on deposition

The main focus of the parametric sensitivity studies has been on the variability of the free daughter deposition velocity, $u_f$. For higher particle concentration, the increased attachment rate results in a smaller fraction of the daughter concentration existing as free daughters. The total deposition rate of a daughter is a function of both the free and attached deposition rates, and the free fraction, as indicated in equation (16). However, because the deposition rate of the free daughters is approximately two orders of magnitude greater than that of the attached, 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/cm$^3$, or an attachment rate $X$=130 hr$^{-1}$).

As noted previously, the attachment rate $X$ was used to represent particle concentration in the parametric sensitivity studies. For the simple case of one-dimensional diffusion, it was shown in equations (22) that for $^{218}$Po,

$$u_f \sim u^a \propto (\lambda + X)^n$$

(28)

where $n = 0.5$. Since $u_f$ and $u^a$ are proportional to $(\lambda + X)^n$, rather than simply $X^n$, increasing $X$ by an order of magnitude from 25–250 hr$^{-1}$
will result in $u^f$ and $u^a$ each increasing by a factor of 2.6, for $^{218}\text{Po}$ in stagnant conditions. This factor increase is slightly smaller compared to that when $D^f$ is increased by an order of magnitude.

The relationship between deposition velocity and attachment rate may not be as intuitive to many people as that between deposition velocity and diffusivity. Deposition depends on the concentration profile near the surface. As homogeneous removal rates, both the attachment rate and radioactive decay rate have a similar influence on the free daughter concentration distribution. As the removal rate increases, the growth of the wall region and broadening of the concentration profile is reduced. The region becomes thinner and the concentration gradient at the surface correspondingly becomes steeper. This is seen as an increase in the deposition velocity. Although attachment is a source term for the attached daughter, as mentioned previously the thickness $b^a$ is predominantly determined by the free daughter concentration profile. Consequently, as attachment rate $X$ increases, both $u^f$ and $u^a$ will also increase. (However, since $u^a$ is proportional to $D^a$, which is very small, the effect of increasing $X$ on the value of $u^a$ will be slight).

This influence is illustrated in Figure 22, which shows the non-dimensional concentration profiles, $A^f/A^f_{\text{core}}$ for free $^{218}\text{Po}$ in a stagnant enclosure (transport by diffusion only). The profiles are plotted for a single value of $D^f$ (0.054 cm$^2$/sec) and a range of values of attachment rate ($X = 0$-$200$ hr$^{-1}$). This representation clearly illustrates that as $X$ increases, the boundary layer thickness decreases slightly and the gradient at the surface steepens.
Figure 22. Free daughter activity concentration profiles, for Po-218, in a stagnant enclosure for various values of attachment rate (corresponding to a varying particle concentration). As attachment rate increases, the boundary layer thickness decreases slightly which the concentration gradient at the surface correspondingly steepens.
When the relative strength of the air motion increases, the attachment rate \( X \) will have a smaller influence on the thickness of the boundary layer. This influence is shown in Figure 23, in which non-dimensional profiles of \( \frac{A_f}{A_{\text{core}}} \) are plotted for \(^{218}\text{Po}\) in a forced convective flow, for two values of \( X \) (0 and 50 hr\(^{-1}\)) and two values of freestream velocity \( U_0 \) (10 and 50 cm/sec). For the higher value of air velocity, there is essentially no difference between the profiles for the two values of \( X \).

This influence of \( X \) on the profiles implies that \( u_f \) will increase for higher values of \( X \), although the sensitivity is lessened for higher air motion (expressed mathematically by \( n < 0.5 \) in equation (26)). The effect on \( u_f \) is illustrated in Figure 24, in which \( u_f \) increases by a factor of \( \sim 2.5 \) over the range of \( X \) for stagnant conditions, and the increasingly flatter curves indicate that \( u_f \) becomes less sensitive to \( X \) for higher values of air velocity.

6.3.6 Effect of air motion on deposition

A forced convection boundary layer flow was used to demonstrate the effects of a wide range of air velocity on daughter concentration profiles and deposition velocities. The effects of air motion on the concentration profiles are illustrated in Figures 25 and 26, in which the profiles for free and attached \(^{218}\text{Po}\) are plotted at different air speeds and also for diffusion only. Similar to previous figures, the concentrations have been non-dimensionalized by the respective free stream, or core, concentrations. A comparison of the plots shows that, for stagnant conditions, the non-dimensional profiles are identical. As air speed increases, the boundary layer thickness for both \( A_f \) and \( A_a \) is reduced, although the effect is much stronger for the attached daughter.
Forced Convection

Free Radon Daughter - $^{218}$Po

Figure 23. Free daughter activity concentration profiles, for Po-218, illustrating the relative influence of attachment rate and air motion. The concentration profiles are insensitive to changes in attachment rate when air velocities are high.
Figure 24. Variation of free deposition velocity with attachment rate, for Po-218, for various values of air velocity. As attachment rate increases, the concentration profile at the wall steepens, which is seen as an increase in the deposition velocity. Deposition becomes slightly less sensitive to changes in attachment rate for higher air velocities.
Figure 25. Free daughter activity concentration profiles, for Po-218, for various values of air velocity. As convective air motion increases, the concentration profiles near the surface gradually steepen as the boundary layer thickness is decreased.
Forced Convection
Attached Radon Daughter - $^{218}$Po

Velocity (cm/sec)

- 0.0 (diffusion)
- 10.0
- 50.0
- 100.0

Attachment rate = 50 hr$^{-1}$
Free diffusivity = 0.1 cm$^2$/sec

Figure 26. Attached daughter activity concentration profiles, for Po-218, for various values of air velocity. Although free and attached profiles are identical for stagnant conditions, the attached daughter concentration profiles are much more strongly influenced by increasing convective air motion.
Figures 27 and 28 are plots of $u^f$ and $u^a$ vs. freestream velocity $U$, for various values of attachment rate $X$. The values of $U$ vary up to 100 cm/sec, which is much higher than typical air velocities seen indoors but represents the maximum influence of air motion for laminar flow. The graphs indicate that both $u^f$ and $u^a$ become less sensitive to changes in air speed at higher values of $X$, corresponding to higher particle concentration. As indicated previously, the attached daughters are influenced by air motion to a greater degree than the free. When $U$ is varied over an order of magnitude from 1-10 cm/sec, there are negligible changes in $u^f$ while $u^a$ increases up to two-fold at the higher values of $X$. Even when $U$ is increased from 10-100 cm/sec, $u^f$ increases only by a maximum factor of 2, while $u^a$ increases by a factor of 10-15.

6.3.7 Variations among daughters

An assumption which has always been made by researchers, for all environmental conditions, is that the values of $u^f$ and $u^a$ are identical for all the daughters. The rationale behind this assumption has been that the values of $D^f$ and $D^a$ are identical for all the daughters. It has been seen, however, that deposition depends not only on the diffusion coefficient but also on the concentration gradient at the wall, and this profile is influenced by various factors. If all the daughters were generated from a uniform source, the differences in the free or attached profiles of the daughters would only be due to differences in the radioactive decay rate constants, and this influence might be negligible. But the concentration distribution of each daughter serves as the source distribution for the subsequent daughter, suggesting that the deposition velocities would actually vary between daughters.
Figure 27. Variation of free deposition velocity with air velocity, for Po-218, for various values of attachment rate. The magnitude of free deposition velocity is not strongly sensitive to changes in air motion, particularly over the low range of air velocities. The sensitivity is even smaller for higher values of attachment rate.
Forced Convection
Attached Radon Daughter $-^{218}$Po

Attachment rate
$50 \text{ hr}^{-1}$
$100 \text{ hr}^{-1}$

Deposition velocity ($10^{-4} \text{ cm/sec}$)

Free diffusivity $= 0.054 \text{ cm}^2/\text{sec}$

Freestream velocity (cm/sec)

Figure 28. Variation of attached deposition velocity with air velocity, for $^{218}$Po, for various values of attachment rate. Compared to the effect on free daughter deposition velocity, convective air motion has a much stronger influence on attached deposition velocity, particularly over the high range of air velocities.
This effect is mathematically described in the analytic solutions to the one-dimensional diffusion equations presented in Appendix D. Figure 29 shows the concentration profiles of the free daughters, non-dimensionalized by the core concentration, to get a better idea of the relative thickness of the boundary layer for each daughter. Because of the non-uniform source, each subsequent daughter has a thicker boundary layer and a broader gradient at the surface, indicating that deposition velocity decreases for each daughter further down the decay chain. This is shown in Figure 30 where $u^f$ vs. $X$ is plotted, for the three daughters. Specifying the three daughters by subscripts 1, 2, and 3, this figure shows that, across the range of $X$, $u_2^f$ is only $1/4 - 1/2$ the value of $u_1^f$, and $u_3^f$ is only $1/5 - 2/5$ the value of $u_1^f$.

6.4 Summary

The mass-transport model was used to examine the influence of key parameters on free and attached daughter deposition velocities, with a particular emphasis on the first free daughter, $^{218}$Po. These parameters, free daughter diffusivity, particle concentration, and air flow, provide a partial description of the physical and environmental conditions which are likely to have the greatest influence on deposition rates. The parametric sensitivity studies varied free daughter diffusivity ($D_{f}^f$) and particle concentration (represented by an attachment rate, $X$) over a broad range, encompassing the values representative of indoor conditions. The air flows that typically exist in real buildings are complex and difficult to accurately characterize through either measurements or simulations. The parametric studies looked at various laminar flows. Although laminar flow is admittedly a simplification of realistic conditions, these initial efforts represent
Figure 29. Free daughter activity concentration profiles, for the first three radon daughters, for one-dimensional diffusion. Because each subsequent daughter is generated from the previous non-uniform source, the boundary layer is thicker and there is a broader concentration gradient at the surface.
One-Dimensional Diffusion
Free Radon Daughters

![Graph showing variation of free deposition velocity with attachment rate for the first three radon daughters, for one-dimensional diffusion. The graph illustrates how the free diffusivity affects the deposition velocity.](image)

**Figure 30.** Variation of free deposition velocity with attachment rate, for the first three radon daughters, for one-dimensional diffusion. Because of the non-uniform source from which each subsequent daughter is generated, the broadening concentration profile is seen as a decrease in deposition velocity for daughters further down the decay chain.
an important and necessary first step in examining the effects of air motion of radon daughter deposition.

In addition to investigating deposition in stagnant conditions, a recirculating buoyancy-driven enclosure flow was also simulated. It was found that, in regard to the near-wall velocity profiles and the predicted values of deposition velocities, the enclosure flow could be adequately represented by simple free and forced convection boundary layer flows.

As noted, the parametric sensitivity studies focused primarily on the free daughter deposition velocity, \( u^f \), of the first daughter, \(^{218}\text{Po}\). With regard to the sensitivity of \( u^f \) to the parameters investigated, it was found that \( u^f \) increases with increasing values of both \( D^f \) and \( X \), although it is more strongly influenced by changes in \( D^f \). It was also found that \( u^f \) becomes more sensitive to \( D^f \), and less sensitive to \( X \), for higher air speeds. For the range of laminar air velocity investigated (0-100 cm/sec), \( u^f \) increases by a factor of 3.2-4.2, and by a factor of 1.4-2.6, when \( D^f \) and \( X \) are increased by an order of magnitude, respectively. The effect of air motion on the sensitivity of \( u^f \) to \( X \) suggests that, for higher air speeds representative of turbulent conditions, it is likely that the influence of \( X \) will be relatively insignificant and \( u^f \) can be considered to be independent of particle concentration.

While \( u^f \) increases for faster air motion, the relative influence of convective transport is reduced for higher values of \( X \). It was found that \( u^f \) increased by a minimum factor of 1.1 (corresponding to \( X = 250 \) hr\(^{-1}\)) when air velocity increased from 0-100 cm/sec. One interesting implication of this result is that, for conditions of high particle
concentration, the free daughter concentration profile is predominantly influenced by the high rate of removal by attachment, and $u^f$ is essentially independent of air velocity. It was also found that $u^f$ is not strongly affected by laminar air flow, and only increased by a maximum factor of 3.5 (corresponding to $X = 0 \text{ hr}^{-1}$) when air flow varied from stagnant conditions to a velocity of 100 cm/sec.

Summarizing the actual values of $u^f$ predicted by the model, $u^f$ varied from 0.014 - 0.054 cm/sec (an overall factor increase of 3.9), for the commonly accepted value of $D^f = 0.054 \text{ cm}^2/\text{sec}$, and for the full range of $X$ and air velocity. Accounting also for the full range of $D^f$, it was found that $u^f$ varied from 0.006 - 0.079 cm/sec (an overall factor increase of 13.1). These results indicate that the value of $D^f$ has the strongest influence on free daughter deposition rates for laminar conditions. In Chapter 7, the actual magnitudes of the predicted and experimentally determined values for $u^f$ will be compared to assess whether deposition by molecular diffusion can account for the measured deposition rates.

Although the discussion in this Chapter focused on the first free daughter, the influence of environmental conditions on the attached daughter deposition velocity, $u^a$, was also briefly examined. It was found that, for stagnant conditions, the attached daughter concentration profile was strongly influenced by its source distribution (i.e., the free daughter). However, this dependence rapidly diminished with increasing air motion. This was attributed to the fact that the air motion had a stronger influence on the transport of the attached daughters, compared to that of the free. It was observed that $u^a$ increased slightly for decreasing values of $D^f$ and increasing values of
X, in stagnant or low velocity conditions, and became relatively independent of these parameters as air velocity increased. In contrast to the effect on $u^f$, $u^a$ was strongly influenced by increasing air motion, and this influence was also reduced for higher values of $X$. When air velocity increased from 0–100 cm/sec, $u^a$ increased by a factor of 13–34, depending on the value of $X$. This result suggests that wide variations in experimentally observed values of $u^a$ can easily be accounted for by differences in flow conditions.

In addition to investigating the effects of environmental conditions on deposition velocities of the first daughter, an important contribution of the mass-transport model is its capability to examine the variations between daughters. It was found that $u^f$ decreases for subsequent daughters in the decay chain, and the range of values predicted for $u^f$ for the first daughter were 2–4 times greater than the corresponding values for the second daughter, and 2.5–5 times greater than those for the third daughter. These differences were 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 daughters in the decay chain.

The findings discussed in this Chapter clearly illustrate one of themany advantages of the mass-transport model over existing uniformly-mixed models. Specifically, the mass-transport model is able to examine the dependence of radon daughter deposition on environmental conditions and therefore provides a more realistic basis for understanding radon daughter behavior. These initial parametric sensitivity studies have demonstrated the relative influence of free daughter diffusivity,
particle concentration, and air motion of free and attached deposition velocities, and how the sensitivity of $u^f$ and $u^a$ to each of these parameters can be affected by the magnitudes of the others. The implications of these findings, with respect to interpreting experimental measurements and helping to formulate new experiments, will be discussed in the following chapter.
Assessment of Existing Experimental Studies

7.1.0 General Overview

As mentioned previously, there are a number of factors which contribute to the wide variability among researchers' experimentally determined values of deposition rates. An accurate comparison of these results requires a thorough examination of the various experimental techniques; however, such a critical review has not been provided by authors who have previously presented a comparison of these values.

Before discussing the implications of the model predictions in regard to experimental findings, it is useful to first present a brief overview of the methods of some existing experimental work. In particular, this overview will focus on the range of experimental conditions, types of measurements, and the assumptions and calculations which are commonly used to obtain estimations of the deposition rates. Table 4 summarizes a comparison of the conditions and results from the experiments which this work investigated in detail.

7.1.1 Experimental conditions

Measurements have been taken in occupied buildings, experimental rooms, small chambers, wind tunnels, and diffusion tubes. Although conditions in the small chambers, wind tunnels, and diffusion tubes are easiest to control and quantify, these are probably least representative of "realistic" conditions in buildings. In particular, the wind tunnel studies have predominantly been used to examine outdoor aerosol deposition and the air speeds investigated are often much higher than the magnitudes likely to be seen in indoor spaces.
**TABLE 4**

Summary of test conditions and experimental results from various researchers.

<table>
<thead>
<tr>
<th>Researcher</th>
<th>Test Space</th>
<th>S/V (m⁻¹)</th>
<th>Flow Conditions</th>
<th>Deposition velocity (cm/sec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Vanmarke [Va84]</td>
<td>chamber</td>
<td>6.2</td>
<td>Minimal air flow, stagnant</td>
<td>.02 - .07</td>
</tr>
<tr>
<td>Rudnick [Ru83]</td>
<td>experimental room</td>
<td>1.4</td>
<td>Stagnant; Mixing fans, turbulent</td>
<td>.025 - .05</td>
</tr>
<tr>
<td>George, Knutson [Ge83, Kn83]</td>
<td>chamber</td>
<td>4.7</td>
<td>Minimal air flow, stagnant</td>
<td>.033 - .066 .0005 - .001</td>
</tr>
<tr>
<td>Porstendorfer [Po83]</td>
<td>wind tunnel</td>
<td>——</td>
<td>Umin ~ 10 cm/sec (u* ~ 1 cm/sec)</td>
<td>.06 - .17 .00056 - .0017</td>
</tr>
<tr>
<td>Scott [Sc88]</td>
<td>occupied buildings</td>
<td>——</td>
<td>Free convective flow</td>
<td>.14  .005</td>
</tr>
<tr>
<td>Offermann Sextro, et al. [Of84]</td>
<td>experimental room</td>
<td>1.9</td>
<td>oscillating fans, turbulent</td>
<td>22  .0023</td>
</tr>
</tbody>
</table>
Reports of experimental work typically give only qualitative descriptions of the air motion in the test spaces; these have included relatively stagnant conditions [Ge83, Kn83, Ru83, Van84], natural convective flow [Sc83], laminar and turbulent controlled flows (wind tunnels and tube flows) [Cl73, Po83, Se71], and conditions in which mixing fans were used and the flow was most likely turbulent [Of84a, Ru83]. In general, flow conditions in the experiments have not been well characterized. Although volumetric flow rates (ventilation rates) are often known, there have only been a few cases where air velocities were actually measured. These have either been in the form of typical air speeds at representative points in an enclosure flow [Sc83], or detailed measurements of the velocity profiles in the wind tunnel experiments [Se71].

7.1.2 Measurements

Although most of the experiments of interest look at radon daughter deposition, some investigate only particle deposition for particles in a given size range. The results of these experiments are useful only if the particle size range includes values representative of the free or attached daughter size distribution. Even if the diameters are identical, however, it has not been clearly established that particle deposition is an accurate representation of radon daughter deposition.

Almost all the radon daughter experiments include measurements of radon concentration and ventilation rates in the space. Airborne radon daughter measurements are generally in terms of total (free plus attached) activity concentrations. There have rarely been direct measurements of the free fraction. Total surface activity concentrations are also sometimes measured, and a few of these
experiments have examined the variations along different surfaces in the room. Equilibrium activity concentrations are either given for the individual daughters or simply in terms of the total working level. Transient measurements of the individual airborne daughter concentration have also been taken.

For some experiments, particle concentration has been measured. Because the attachment rate has a significant effect on the free fraction and deposition rates, experiments which include particle concentration measurements produce more informative results. With this information, fewer assumptions are involved in the calculations which are made from the radon daughter measurements in these experiments.

7.1.3 Assumptions

The intent of this section is to briefly summarize some of the assumptions most frequently used by experimental researchers, give some initial assessment of whether they might be reasonable, and indicate the capability of the mass-transport model to address their validity. A later section will use the findings of the parametric sensitivity studies to examine these assumptions in more detail.

Researchers have commonly assumed that the values for free and attached deposition velocity (or deposition rate) are constant and identical for all daughters, and the variation of total deposition rate is consequently only due to changes in the free fraction. The validity of this assumption has been the main focus in the discussion of the findings of this work. The results presented in Chapter 6 clearly indicate that $u^f$ and $u^a$ are both influenced by environmental conditions and that they also vary for the different daughters.
An even greater simplification was made by Rudnick, who only measured total working level [Ru83]. Because neither individual daughter activity or particle concentration was measured, it was necessary to assume that the total deposition rate, q, was identical for all daughters. From other experimental measurements, and even if one assumes constant values of $q^f$ and $q^a$ and uses the simple mass-balance equations from the uniformly-mixed model, it is known that the free fraction varies significantly between daughters. This implies that q also varies significantly, and that this assumption made by Rudnick is not valid. Results from these experiments should be viewed with caution.

In the experiments in which the removal rate of particles was determined [Of84a, Po83], it was typically assumed that the attached daughter deposition velocity is equivalent to the particle deposition velocity, $u^p$. The value of $u^a$, however, depends on both $D^a$ and the gradient of the profile of $A^a$ near the wall. Both of these may differ from the corresponding values for the particles, and the implications of these differences, in terms of a comparison between $u^a$ and $u^p$, will be examined in a later section.

7.1.5 Calculations

All the radon daughter experiments examined here measured only total (free plus attached) daughter activity. Based on these measurements, the calculations give values for the corresponding total deposition rate, $q_i$, or velocity, $u_i$, for each daughter.

There seem to be three predominant methods that are used for determining total q or u. These can, in general, be distinguished by the type of measurements which were taken in the experiments. In the first method, applicable when surface activity measurements are taken,
the effective total deposition velocity can be directly calculated from the surface and airborne concentrations of each daughter [Ru83, Sc83]. Total deposition rate can then be estimated in terms of the surface-to-volume ratio of the room, S/V. (Note that a deposition rate is not defined for the wind tunnel studies and only a deposition velocity has meaning for this case).

The second and third methods are used with only airborne daughter measurements. When equilibrium concentrations are measured, the second method uses steady-state mass balance equations to calculate an overall removal rate for each daughter [Kn83]. For transient airborne measurements, the third method uses the time-dependent form of the mass balance equations and the overall removal rate is determined from the rate at which the concentration decreases over time [Of84a]. For both these cases, total deposition rate for each daughter, \( q_i \), can be calculated from the total removal rate and known values of the daughter radioactive decay constant, ventilation rate, and (if applicable) air cleaning device removal rate. A total effective deposition velocity is then estimated in terms of the surface-to-volume ratio of the room.

To estimate the individual free and attached deposition rates (\( q^f \) and \( q^a \)) separately, researchers use various methods which involve combinations of the previously discussed assumptions. The mass-balance equations are typically used to find values of \( q^f \) and \( q^a \) to fit the measured data. When measurements have been made for all three daughters, or under various experimental conditions, it is assumed that \( q^f \) and \( q^a \) are constant, and the values are obtained by an optimization technique [Kn83] or a simple average [Of84a]. This can be done using equations for all three daughters [Kn83] or for only the first daughter [Of84a].
The details of these and other calculations used by researchers can be found in the references for a particular experiment.

7.1.5 Discussion

Although the value of deposition velocity is a better measurement of deposition as a surface transformation process, it is the overall deposition rates for a space and their effect on average room concentrations that are of interest in terms of evaluating exposures. As outlined in previous sections, either the deposition velocity or the deposition rate constant is directly calculated from the measurements, while the other can be obtained in terms of the estimated value of S/V through the use of equation (12). The value of the parameter obtained in this way will have the greater degree of uncertainty due to possible limitations of the assumptions involved in equation (12), i.e., well-mixed conditions and all surfaces being equally effective for deposition. An additional factor is that S/V could possibly be higher than the measured value, due to increased deposition surface by furniture, etc. [Po83]. This influence is probably not significant, however, since a room must be cluttered with surprisingly enormous amounts of furniture to achieve values of S/V higher than 3.0 [Of84b].

The calculational method which obtains deposition velocity directly from measurements of surface activity is the only way to determine variations along the surfaces in a room. Airborne concentrations are also measured in this method to represent the driving potential for deposition. Although the deposition velocity calculated with this method will be a better description of the deposition process, it should be recognized that its value will strongly depend on the location and type of surface on which the measurements were taken. If there is a
complex non-uniform flow in the room, it is important to assess how representative the measurement location might have been of the average deposition flux in the space (i.e., are all surfaces equally effective for deposition?). Because of this influence, and possible uncertainties in the value of $S/V$, the quoted value of deposition rate constants in these surface-measurement experiments should be viewed with caution and it is important to examine the method of measurement in the particular experiment.

In contrast, the experiments which only measure airborne concentrations calculate deposition rate constants from a mass balance. These experiments will produce a better measurement of the overall removal rates for the space. However, overall removal rates are very geometry-dependent, and the calculation of the corresponding average deposition velocities is preferable for comparing the "effectiveness" of the deposition process to other conditions. Using the same arguments as above, the estimated values of the effective deposition velocity in these experiments may involve varying degrees of uncertainty.

For both methods, the measured airborne concentrations are assumed to represent the average concentrations in the room. The accuracy of the calculated deposition rates will depend on the validity of this assumption. If the radon distribution was not uniform throughout the space, or there were large non-uniformities in the flow conditions, a single airborne measurement may not be adequate to represent average conditions.
7.2 Comparison of Results

7.2.1 Model Validation

Experimental flow conditions have varied considerably. To validate the model as a prediction tool, it is necessary to compare the predictions to measurements from experiments in which the conditions were most similar to the simplified flows simulated in the model. These include stagnant conditions or laminar flows.

Figure 31 shows a summary of the values of $u^f$ and $u^\alpha$ obtained by various researchers, and the range of values predicted by the mass-transport model from the parametric studies. Experiments which can best be used to validate the model include those by George and Knutson which were taken in a small 1.9 m$^3$ chamber with minimal air flow [Ge83], by Vanmarcke in a 1 m$^3$ chamber with stagnant conditions [Van84], and by Rudnick in a 78 m$^3$ experimental room without mixing fans [Ru83]. Because the geometry and value of $S/V$ in these experiments varied considerably (1.9, 6.0, and 4.7 m$^{-1}$ for the three experiments, respectively), the comparison needs to be made in terms of deposition velocity rather than the geometry-dependent deposition rate. An examination of Table 4 and Figure 31 shows that, for these three experiments, there is excellent agreement between the experimental values and the mass-transport predicted values for $u^f$. (As indicated previously, the reliability of Rudnick's results may be questionable).

Of these experiments, only those of George and Knutson provided a value of $u^\alpha$. They estimated $u^\alpha$-0.0005-0.001 cm/sec for tests where the geometric mean particle diameter varied in the range 0.06-0.15 μm. Values of $u^\alpha$ predicted from the model were at least an order of magnitude lower than this, ranging from ~0.00004-0.00007 cm/sec. This
Free Daughter Deposition
Experimental measurements

Deposition velocity (cm/sec)

0 0.05 0.10 0.15 0.20 0.25

Model

Vanmarke (chamber) [Va84]

Rudnick (experimental room) [Ru83]

George/Knutson (chamber) [Ge83, Kn83]

Porstendorfer (wind tunnel) [Po84]

Scott (building) [Sc83]

Offerman (experimental room) [Of84]

Deposition rate* (hour\(^{-1}\))

*Deposition rate calculated from deposition velocity, assuming ratio \(S/V = 2\) m\(^{-1}\)

Figure 31. Free daughter deposition velocity, as measured by various experimental researchers. Measurements are compared to the range of values predicted by the mass-transport model from the parametric studies. A proper comparison must take note of the various conditions under which the experiments were carried out.
is most likely due to the fact that only deposition by molecular diffusion was accounted for in the simulation model. Compared to the free daughters, the attached daughters are much more sensitive to convective, gravitational, and electrostatic effects due to their larger size and inertia. Although the inertial and gravitational settling effects are not large due to the small size of the attached daughters, these deposition mechanisms may not be negligible compared to the small values of diffusive deposition velocity which have been calculated, indicating that these effects may need to be accounted for. Further studies of deposition due to electrostatic forces may also account for these differences.

While the uncertainty in the predicted values of \( u^a \) is high, except for very high particle concentrations the total deposition rate is almost entirely influenced by \( u^f \). Consequently, it is not expected that these low values of \( u^a \) will contribute to large uncertainties in predictions of overall room concentrations.

7.2.2 Overall Comparison

There are many experiments which have been conducted under conditions which the mass-transport model has not yet simulated. However, the physical understandings obtained through the parametric studies can still be helpful. The differences among the experimental results and the model predictions will be examined to try to evaluate whether these differences can be accounted for by variations in the environmental conditions.

Results of the parametric studies indicated that \( u^f \) increases only by a maximum factor of ~3.5 when air velocity increases over a wide range (0-100 cm/sec) in laminar forced convection conditions. The
maximum value of $u_f$ in these conditions was approximately 0.08 cm/sec. This implies that experimental values of $u_f$ greater than $\sim 0.1$ cm/sec cannot be accounted for by molecular diffusion alone, and can most likely be attributed to deposition mechanisms which have not yet been simulated, such as electrostatic attraction of turbulence.

In contrast, $u_a$ can increase by a factor of 20-30 for air velocity increasing by two orders of magnitude. Although the magnitudes of the values for $u_a$ obtained from the simulations are largely underestimated, the relative effects of air velocity are based on physical principles and are still valid under realistic conditions. Consequently, even if particle size and methods of measurement and calculations were similar, it is likely that wide variations of $u_a$ will be seen among the experimental work due to the differences in flow conditions.

The range of experimentally determined values of $u_f$ and $u_a$ can be compared in terms of the variations among the test spaces and flow conditions under which the measurements were made. Tests by Rudnick [Ru83] and Offerman/Sextro [Of84a] were both done in experimental rooms of slightly different sizes (volumes 78 and 36 m$^3$, and S/V ratios 1.4 and 1.9 m$^{-1}$, respectively). A comparison of the range of Rudnick's values of $u_f$ with (0.2-0.7 cm/sec) and without (.025-.05 cm/sec) mixing fans showed that $u_f$ increased by a factor between 4-30. In comparison, tests by Offerman/Sextro showed no difference between removal rates when measurements were taken with fans operating at different speeds or turned off. This invariability of $u_f$ with fan operation could indicate that electrostatic attraction (which is independent of flow conditions) may have been the dominant deposition mechanism during these experiments, or that the air flow was still turbulent even when the fans
were not operating. This turbulent flow might have been generated by infiltration, natural convection driven by radiant heat gain through the windows, or the operation of the small fans in the equipment in the room [Of84b].

The values of $u_f$ from Offerman/Sextro's experiments and those at the low end of the range of Rudnick's results are in good agreement. Offerman/Sextro estimate values of $u_f \approx 0.22$ cm/sec, which is ~4 times higher than the average simulation predictions and the experimental values of $u_f$ for stagnant or laminar flow. They also estimate a value of $u_\infty \approx 0.0023$ cm/sec for particle size distribution with a geometric mean diameter of 0.15 μm. This value is ~3 times higher than Knutson's value of $u_\infty \approx 0.00075$ cm/sec for stagnant conditions. The model predictions indicate that variations in air flow can easily account for this relative discrepancy.

Scott made surface activity measurements in occupied buildings along various surfaces [Sc83]. The air flow patterns were visualized using smoke and air velocities were measured, indicating a natural convection air flow pattern in the room with downward air speeds along the cool walls of ~4-12 cm/sec, and typical air velocities over the floor and table tops of ~0.5-1.5 cm/sec. Although these measurements indicate that flow conditions might have been similar to the simulated buoyancy-driven enclosure flow, it is likely that there was some small degree of turbulence in the experimental flow. The disturbances which exist in an occupied space, compared to the simulated flow, are likely to result in transition to turbulence at the typical enclosure Grashof number which might produce these velocities. This would account for the average value obtained by Scott of $u_f \approx 0.14$ cm/sec, which is ~3 times
higher than the average $u_f$ for stagnant or laminar conditions. The fact that Scott's value of $u_f$ is about midway between the values for laminar and fully turbulent conditions may indicate the relative degree of turbulence in the natural convection (buoyancy-driven) vs. forced convection (mixing fans) conditions. Scott obtained his values of $u_a$ by assuming that it was equal to the deposition velocity of RaC ($^{214}$Bi), which was assumed to be all attached. His value of $u_a \approx 0.005 \text{ cm/sec}$ is considerably higher than the other experimentally determined values.

Porstendorfer estimated values of $u_f$ and $u_a$ from experimental wind tunnel measurements of free daughter and particle deposition in turbulent flow [Po83]. Assuming an average air velocity of $\sim 10 \text{ cm/sec}$ in turbulent flow with a friction velocity of $\sim 1 \text{ cm/sec}$, and average particle diameter of $0.15 \mu \text{m}$, he derives values of $u_f$ and $u_a$ implying that they are appropriate for poorly ventilated rooms with air exchange rates less than $0.3 \text{ hr}^{-1}$. Porstendorfer then assumes that $u_f$ and $u_a$ will increase by a factor of 3 for higher friction velocities corresponding to ventilation rates between 1-2 hr$^{-1}$ and gives the resulting range of $u_f \approx 0.056-0.17 \text{ cm/sec}$ and $u_a \approx 0.00056-0.0017 \text{ cm/sec}$. (It should be noted that no theoretical basis was given for assuming that deposition velocities increase 3-fold with an average 5-fold increase in ventilation rate). The low end of the range of $u_f$ compares well with the simulation predictions, and towards the higher end compares with Scott's average value of $u_f$. Knutson's value of $u_a$ is also comparable with Porstendorfer's range.

Jacobi's values of $u_f \approx 0.3-0.6 \text{ cm/sec}$ come from measurements of $^{218}$Po in cylindrical tubes with air velocities in the range 3-20 cm/sec [Jac72]. His values of $u_a \approx 0.01 \text{ cm/sec}$ is extrapolated from
Chamberlain's wind tunnel work, assuming air velocities of ~10 cm/sec for particle diameter in the range of 0.05-0.2 μm. The flow pattern in tubes is very different from that in wind tunnels or enclosures, and so it is difficult to compare values of $u^f$ from tube experiments to any others. It is unclear why Jacobi's estimation of $u^a$ is so much higher than those other experimentally determined values.

Sehmel also took wind tunnel measurements of particle deposition in turbulent flow, and then developed correlations with which he extended his results with theoretical prediction [Se71]. As his interest was not particularly concerned with indoor radon daughter deposition, the range of air velocities investigated was much higher than those representative of indoor conditions. The lowest air velocity he examined was 1 mph (~45 cm/sec) in a turbulent flow with a friction velocity of 2.3 cm/sec. For representative free and attached daughter diameters of 0.001 and 0.1 μm, respectively, this produces values of $u^f$~0.1 cm/sec and $u^a$~0.0003 cm/sec. It's interesting to note that Sehmel's representative value for $u^a$ in a turbulent flow is less than the values estimated by Forstendorfer for wind tunnel studies of lower air speeds and by George/Knutson for stagnant conditions, for a similar average diameter. The reason for this inconsistency is not clear.

7.3 Validity of Assumptions

The mass-transport model has the capability of addressing many of the various assumptions made by experimental researchers. The initial use of the model focused primarily on the most commonly made assumption concerning the invariability of the individual deposition velocities.

The discussions in Chapter 6 described the dependencies of $u^f$ and $u^a$ on the physical parameters investigated, and also variations between
daughters. Although these sensitivities exist, it was indicated that they decrease with increasing particle concentration and air motion. This suggests that, in general, the assumption of constant values of $u^f$ and $u^a$ will be most appropriate for conditions of higher particle concentration and air speeds, and are likely to be best for turbulent conditions. The magnitudes of these constant values will, of course, still be influenced by the conditions themselves.

The acceptability of assuming constant values of $u^f$ and $u^a$ (or $q^f$ and $q^a$) depends on how sensitive predicted room concentrations are to the magnitude and variations in these values. For a general picture of this sensitivity, Figure 32 is a plot of total working level vs. a range of values of $q^f$, calculated using the mass balance equations of the uniformly-mixed model, for chosen values of $X$ and assuming $q^f/q^a = 100$. For the range of $u^f$ predicted for stagnant or laminar conditions, working level is not significantly affected by these low values, except for very low particle concentration. When deposition velocity varies across the range of experimentally observed values, however, there is a much more pronounced effect on the average working level in the space.

This figure illustrated the uncertainties which might result from the various choices for a constant value of $u^f$. A better indication of the validity of the invariability assumption would be an examination of how the predicted value of total working level changes if one assumes constant values of $u^f$ compared to using values which vary for each daughter and also depend on environmental conditions such as particle concentration. This will be done using values calculated for the case of one-dimensional diffusion, which represents the condition for which the greatest variability might be expected. Figure 33 shows total
Figure 32. Variation of total working level over a range of values of free daughter deposition velocity (or deposition rate). Deposition has a pronounced effect when varied across the range of experimentally observed values. The effect does not appear to be significant for the range of deposition velocity predicted by the model for stagnant or laminar conditions.
Figure 33. Variation of total working level over a range of values of attachment rate, calculated using constant values of free and attached deposition velocity and also using the predicted values which vary for each daughter and also depend on the attachment rate itself. For stagnant or laminar conditions, the variability of deposition rates has little influence on overall working level, except for very low particle concentration.
working level vs. attachment rate \( X \), using the variable, or dependent, values of \( q_f \) and \( q_a \) and also using constant values corresponding to those which were calculated for the first daughter at \( X=50 \text{ hr}^{-1} \). Although the dependent values of \( q_f \) and \( q_a \) varied by several factors, the figure indicates that this variability had little influence on overall predicted room concentrations compared to using constant values, except for very low particle concentration. This is not surprising, however, since the total deposition rate in stagnant conditions is relatively small and so even a three-fold increase will not make much difference. Although the higher values of \( q_f \) and \( q_a \) which result from higher air speeds will have a greater influence on predicted values of total working level, the variability itself will diminish. These results indicate that the assumption of invariability of \( q_f \) and \( q_a \) may be fairly valid, as long as the constant values chosen for these exchange rates are appropriate for the environmental conditions.

For most experiments in which particle concentration was measured, the assumption has been made \( u_a \) is equivalent to particle deposition velocity, \( u_P \). For deposition by molecular diffusion this may not be valid because the deposition rate depends on both the diffusion coefficient and the concentration gradient at the surface.

Looking first at the influence of the diffusion coefficient, it is possible that the effective attached daughter diffusivity \( D_a \) may be slightly less than that for the particles, \( D_P \). This is because the daughters are more likely to attach to larger particles, resulting in a slight shift between the particle size distribution and the attached daughter size distribution [Ge78]. The average diameter of the attached daughters is therefore slightly larger than the average particle
Figure 33. Variation of total working level over a range of values of attachment rate, calculated using constant values of free and attached deposition velocity and also using the predicted values which vary for each daughter and also depend on the attachment rate itself. For stagnant or laminar conditions, the variability of deposition rates has little influence on overall working level, except for very low particle concentration.
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diameter, consequently $D_a < D_P$. It is not clear how large this shift is, and the resulting difference between $D_a$ and $D_P$, but it is likely that its effect on the difference between $u_a$ and $u_P$ is not significant.

As noted previously, the differences between the attached daughter and particle concentration gradients near the surface stem from the fact that the attached daughter concentration distribution, $A_a$, depends on both the particle concentration, $N$, and the free daughter concentration, $A_f$ (the source for the attached daughters). It was demonstrated in an earlier section that, particularly for lower air velocities, the attached daughter profile is dominated by that of the free daughter. Because of the larger value of $D_f$ compared to $D_a$ or $D_P$, the boundary layer thickness of $A_f$ and $A_a$ is larger than that of $N$, and the gradient at the surface is not as steep. This results in $u_a < u_P$. For higher particle concentrations, or higher values of attachment rate $X$, the free fraction is reduced and has a smaller influence on the attached daughter profile. Therefore, $u_a$ increases with increasing values of $X$ and approaches the value of $u_P$. For higher air velocities, the attached daughter profile is also less influenced by $A_f$ because of higher inertial effects. This also results in $u_a$ approaching the value of $u_P$ for faster air motion.

These effects are illustrated in Figure 34, in which $u_a$ vs. air velocity is plotted for various values of $X$, along with a curve for $u_P$, both calculated using the same diffusivity. It can be seen in this figure that, at low attachment rate and air velocity, $u_P$ is as much as four times greater than $u_a$, while the values become nearly equivalent at the higher values of velocity. The equivalence of $u_a$ and $u_P$ at higher attachment rate is also illustrated in this figure. (It is not yet...
Forced Convection
Attached Radon Daughter $-^{218}\text{Po}$

Attachment rate
- particles
- $50 \text{ hr}^{-1}$
- $100 \text{ hr}^{-1}$

Deposition velocity ($10^{-4} \text{ cm/sec}$)

Free diffusivity = $0.054 \text{ cm}^2/\text{sec}$

Freestream velocity (cm/sec)

Figure 34. Variation of deposition velocity with air velocity, for attached Po-218 and also for particles (same effective diameter). For low air velocity, deposition velocity is much higher for particles than for attached daughters.
clear why \( u_D \) becomes less than \( u_a \) at the highest values of velocity). Since total deposition rate is almost entirely determined by free daughter deposition at the lower particle concentrations, even a factor of four difference between \( u_a \) and the assumed value of \( u_a = u_D \) in this region is not likely to have a significant influence on predicted values of room concentration. The value of \( u_a \) begins to have an influence at the higher particle concentrations, and for these conditions the assumption \( u_a = u_D \) is quite valid. The assumption also appears to be accurate enough for flow conditions with moderate air velocities, and is likely to be most accurate for turbulent conditions.

7.4 Recommendations

The wide range of experimentally determined values of deposition velocity, together with the findings of the mass-transport model, suggests that deposition is strongly dependent on environmental conditions. The uniformly-mixed model can be used to calculate effective deposition rates from concentration measurements, but is not in itself able to explain the differences between values obtained from various experiments. Experimental methods (i.e., measurements, assumptions, and calculations) commonly used by researchers have not enabled them to account for the variability of the individual free and attached deposition rates. Consequently, many of the trends observed in the mass-transport model predictions have not been able to be validated by experimental measurements. The findings of the parametric sensitivity studies, however, indicate what parameters are most likely to account for the variability in observed deposition velocities. Based on these findings, several recommendations can be made regarding how experiments might be improved to investigate the sensitivity of free and
attached deposition and increase our understanding of the behavior of indoor radon daughters.

As mentioned in previous sections, very few researchers have measured air speeds in the experimental space. To determine to what extent variations in experimental measurements might be 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, and/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, for conditions which are representative of those typically found in buildings.

In regard to the actual radon daughter measurements, a great deal of information could be gained (and the necessity for making certain assumptions reduced) if free daughter concentrations were directly measured. Needless to say, it is also important to measure the daughter concentrations separately, rather than measuring only total working level, in addition to simultaneously measuring particle concentration.

Measuring daughter concentrations at various locations in the experimental space would also provide useful information. For example, measurements at several locations in the core would indicate the validity of the well-mixed hypothesis. This will be particularly important in conditions of minimal air circulation, non-uniform flows, or variable source distribution. Airborne measurements might also be taken at several locations close to the surfaces to determine the boundary layer thickness and concentration profile within the wall region. This will be particularly informative if air speed measurements
are also taken simultaneously. If surface activity is measured in a chamber or room, the measurements should be made on different surfaces to investigate the possible variations. These measurements will also be most useful if the variation in air flow along the surfaces is known.

Given the improved measurements, the assumptions and calculations which have commonly been used also need to be modified. There is no need to assume that $u_f$ and $u_a$ are constant in order to fit the equations of the uniformly-mixed model to the measurements. If measurements have been made of the free and attached activity concentrations of each daughter, then the uniformly-mixed mass balance equations can be used to calculate the values of $u_f$ and $u_a$ for each daughter, separately. By carrying out this procedure for each experiment conducted under various conditions, the variation of $u_f$ and $u_a$, for each daughter, can be determined.

Following these recommendations, the additional information generated from these measurements can be compared to the patterns predicted by the mass-transport model. If the experimental flow conditions can adequately be simulated, the measurements will serve to further validate the model predictions. The model can then be used to interpret experimental results and investigate the behavior of indoor radon daughters in a variety of other conditions.
8.1 Overview

The complex behavior of indoor radon daughters is influenced by physical and environmental conditions such as the size distribution of the decay products, airborne particle concentration, and air flow patterns in the space. A mass-transport model was developed which is able to account for the variability in source distributions and transport mechanisms, and their influence on the concentration distributions and deposition rates of the radon daughters. The basis of this model is the mass conservation and transport laws governing the movement of reactive components in specified flow conditions. The model offers many advantages over existing indoor radon models, and also provides theoretical insight to experimental results. The applicability of the model is primarily limited by the ability to simulate flow conditions representative of realistic conditions.

As a particular application of the model, parametric sensitivity studies were performed which examined the effects of free daughter diffusivity, aerosol particle concentration, and convective air motion on the free and attached deposition rates of the first three radon daughters, in boundary layer and enclosure configurations. Researchers have commonly assumed that these deposition rates are constant, independent of environmental conditions and identical for all the daughters. The results of these studies provided a basis for evaluating the validity of this, and other, assumptions which are often made. Based on the findings, recommendations were made concerning further
experimental studies to investigate the dependence of deposition on environmental conditions.

8.2 Recommendations for Further Study

The current state of the mass-transport model represents an important first step in attempting to characterize the effects of air motion on radon daughter transport and deposition. The model provides a theoretical framework for further studies concerning the general behavior and interactions of indoor radon and its daughters.

The parametric sensitivity studies described in this work generated a great deal more information than has presently been discussed here. Further analysis of the results will first focus on the effect of air motion on the variability of deposition velocities among the daughters. The predicted deposition velocities in the enclosure flows will also be examined in more detail. This examination will pay particular attention to the variations that were observed along the surfaces, especially in the corners where eddies formed. In addition to the effects of the magnitude of the air motion, it is also useful to look at the type of flow. The characteristics of free and forced convection flows are quite different, and it was observed that the sensitivity of deposition velocity in these two flows also differed. These patterns will be investigated further in future work.

It was found that molecular diffusion in stagnant or laminar flow could not account for the deposition rates observed in experiments representative of realistic flow conditions. Consequently, an important next step will be to simulate other deposition mechanisms and flow patterns. Simplified approaches will first be taken for examining electrostatic and turbulent deposition. Enclosure flows generated by
infiltration and ventilations will also be investigated. For these cases, in particular, the model will be expanded to three-dimensions.

The mass-transport model is based on fundamental laws of physics, and the approach can easily be applied to other pollutants simply by appropriately changing the homogeneous transformation rates and boundary conditions. As an example of the utility of this approach for other pollutants, the model can be used to investigate the transport and distribution of a pollutant within a space for a specified source distribution. For instance, a point source might represent combustion products from a stove, heater, or cigarette, and a line or area source might represent the emanation of pollutants from building materials.

Perhaps even more important, the fundamental basis of this model enables it to be used for quantitatively examining the behavior of pollutants for which surface interactions are important. One example of such a pollutant is formaldehyde, for which the walls can constitute both a source and a sink. Another example is NO₂, which arises from combustion but can be removed by homogeneous interactions, by surface

By treating a very complex process in a fundamental manner, the mass-transport model represents a notable contribution towards understanding the behavior of indoor radon daughters, and provides the basis from which a more comprehensive and realistic model of indoor air quality can be developed.
REFERENCES


[Ch56] Chamberlain, A.C., and E.D. Dyson, "The dose to the trachea and bronchi from the decay products of radon and thoron", Br. J. Radiol., 29, 1956.


[Po78a] Porstendorfer, J., A. Wicks, and A. Schraub, "The influence of exhalation, ventilation and deposition processes upon the concentration of radon (Rn-222), thoron (Rn-220) and their decay products in room air", Health Physics, Vol.34 (May), 1978.


[Vat80] van de Vate, Joh. F., "Investigations into the dynamics of aerosols in enclosures as used for air pollution studies", Research Report, Netherlands Energy Research Foundation, 1980.


APPENDIX A

Uniformly-mixed Model

Equilibrium concentrations

\[ A_i^f = \frac{\lambda_i \ A_i^{f-1} + r_{i-1} \lambda_i \ A_i^{a-1}}{(\lambda_i + q^f + X)} \]

\[ A_i^a = \frac{(1 - r_{i-1}) \lambda_i \ A_i^{a-1} + X \ A_i^{f}}{(\lambda_i + q^a)} \]

where:

- \( A_i \) = activity concentration for decay product \( i \), \( i = 1, 2, 3 \)
- \( A_i^{f} = A_{\text{radon}} \)
- \( A_i^{a} = 0 \)
- \( r_2 = r_3 = 0 \)

Free Fractions

\[ f_1 = \frac{\lambda_1 + q^a}{\lambda_1 + q^a + X} \]

\[ f_2 = \frac{\lambda_2 + q^a - \lambda_2(1-r_1)(1-f_1)(\frac{A_1}{A_2})}{\lambda_2 + q^a + X} \]

\[ f_3 = \frac{\lambda_3 + q^a - \lambda_3(1-f_2)(\frac{A_2}{A_3})}{\lambda_3 + q^a + X} \]
APPENDIX B

Discretization Equations

General formulation (assuming constant properties)

continuity:

\[
\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} = 0
\]

mass-transport:

\[
\frac{\partial \theta}{\partial t} + \left[ \frac{\partial (u \theta)}{\partial x} + \frac{\partial (v \theta)}{\partial y} \right] - D \left[ \frac{\partial^2 \theta}{\partial x^2} + \frac{\partial^2 \theta}{\partial y^2} \right] = (S_1 + S_2 \theta)
\]

where the quantities in the first set of brackets represent the convection terms, those in the second set of brackets represent the diffusion terms, and the quantities on the right hand side of the equation represent the source terms.

Isolate a single node, P, with neighboring nodes, control volume interfaces, and total (convection plus diffusion) fluxes defined as follows:

- uppercase letters denote nodal points of grid.
- lowercase letters denote control volume interfaces.
- \( J \) = total mass flux across interface.
- \( \Delta x, \Delta y \) = control volume faces.
- \( \delta \) = distance between nodes.

The differential equations will be integrated over the control volume, resulting in an algebraic equation which relates the value of \( \theta_P \) to the values at the neighboring nodes, \( \theta_E, \theta_W, \theta_N, \theta_S \). This algebraic equation (referred to as the "discretization equation") will take the following form:

\[
a_P \theta_P = a_E \theta_E + a_W \theta_W + a_N \theta_N + a_S \theta_S + b_P
\]

where the a's and b are coefficients. All values of \( \theta \) are those at the end of the timestep \( \Delta t \), except for \( \theta_P \) in the time derivative term which is the "old" known value at the beginning of the timestep.
Integrating over the control volume yields:

(continuity)

\[(u_e - u_w)\Delta y + (v_n - v_s)\Delta x = 0\]

(mass-transport, combined with continuity)

\[\frac{\Delta x \Delta y}{\Delta t} (\theta_p - \theta_p^2) + (J_e - u_e \Delta y \theta_p) - (J_w - u_w \Delta y \theta_p) + (J_n - v_n \Delta x \theta_p) - (J_s - v_s \Delta x \theta_p) = \Delta x \Delta y (S_{1p} + S_{2p} \theta_p)\]

where the flux terms are:

\[J_e = [u_e \theta_e - D \frac{\partial \theta}{\partial x} \mid_e] \Delta y\]

\[J_w = [u_w \theta_w - D \frac{\partial \theta}{\partial x} \mid_w] \Delta y\]

\[J_n = [v_n \theta_n - D \frac{\partial \theta}{\partial y} \mid_n] \Delta x\]

\[J_s = [v_s \theta_s - D \frac{\partial \theta}{\partial y} \mid_s] \Delta x\]

and the discretization coefficients are related by:

\[(J_e - u_e \Delta y \theta_p) = a_E (\theta_p - \theta_E)\]

\[(J_w - u_w \Delta y \theta_p) = a_W (\theta_W - \theta_p)\]

\[(J_n - v_n \Delta x \theta_p) = a_N (\theta_p - \theta_N)\]

\[(J_s - v_s \Delta x \theta_p) = a_S (\theta_S - \theta_p)\]

\[a_p = \left( \frac{1}{\Delta t} - S_{2p} \right) \Delta x \Delta y + a_E + a_W + a_N + a_S\]

\[b_p = \left( \frac{\theta_p^2}{\Delta t} - S_{1p} \right) \Delta x \Delta y\]

\[a_E, a_W, a_N, a_S\] are determined by the discretization formulation.
Discretization Formulations

To complete the formulation and find expressions for the coefficients $a_E$, $a_W$, $a_N$, $a_S$, one needs to make assumptions regarding the profile of $\theta$ between the nodes. The “hybrid” scheme used in this work is a combination of two simpler formulations - the “central difference” and “upwind” schemes.

Central Difference Formulation

The central difference formulation assumes that convective and diffusive transport between nodes are of comparable magnitude. The interface value is taken to be the average of the nodal values, and a linear profile is used to calculate the gradient at the interface. Using interface “e” as an example:

$$\theta_e = \frac{1}{2}(\theta_E + \theta_P); \quad \frac{\partial \theta}{\partial x} \bigg|_e = \frac{\theta_E - \theta_P}{\delta_e}$$

$$(J_e - u_e \Delta y \theta_P) = \left( \frac{D}{\delta_e} - \frac{u_e}{2} \right) \Delta y (\theta_P - \theta_E)$$

$$a_E = \left( \frac{D}{\delta_e} - \frac{u_e}{2} \right) \Delta y = \left( 1 - \frac{P_e}{2} \right) \frac{D \Delta y}{\delta_e}$$

where

$$P_e = \frac{u_e \delta_e}{D} \equiv \text{grid Peclet number, evaluated at interface “e”}.$$  

Upwind Formulation

The upwind formulation assumes that convective transport is dominant and the interface value is taken to be equal to the upstream nodal value. Diffusion is neglected, so the gradient at the interface is zero. Using interface “e” again as an example:

for $u_e > 0$:

$$\theta_e = \theta_P; \quad \frac{\partial \theta}{\partial x} \bigg|_e = 0$$

$$(J_e - u_e \Delta y \theta_P) = 0$$

$$a_E = 0$$
for $u_e < 0$:

$$\theta_e = \theta_E; \quad \frac{\partial \theta}{\partial x} |_e = 0
$$

$$(J_e - u_e \Delta y \theta_P) = -u_e \Delta y (\theta_P - \theta_E)
$$

$$a_E = -u_e \Delta y = -P_e \frac{D \Delta y}{\delta_e}
$$

**Hybrid Formulation**

The hybrid formulation is a combination of the central difference and upwind formulations. It accounts for the relative magnitudes of convection and diffusion at each interface. Using interface "e" as an example:

$$\frac{\delta_e}{D \Delta y} a_E = \begin{cases} 
-P_e & \text{if } P_e < -2 \\
(1 - u_e P_e) & \text{if } -2 < P_e < 2 \\
0 & \text{if } P_e > 2
\end{cases}
$$

Another way of writing this is:

$$a_E = \frac{D \Delta y}{\delta_e} \left[ [-P_e, (1 - \frac{P_e}{2}), 0] \right] = \Delta y \left[ [-u_e, (\frac{D}{\delta_e} - \frac{u_e}{2}), 0] \right]
$$

where the symbol $[[]]$ denotes the largest of the quantities contained within it.

Applying a similar procedure to all the interfaces of the control volume surrounding node $P$:

$$a_W = \Delta y \left[ [u_w (\frac{D}{\delta_w} + \frac{u_w}{2}), 0] \right]
$$

$$a_N = \Delta x \left[ [-v_n (\frac{D}{\delta_n} - \frac{v_n}{2}), 0] \right]
$$

$$a_S = \Delta x \left[ [v_s (\frac{D}{\delta_s} + \frac{v_s}{2}), 0] \right]
$$

$$a_P = \left( \frac{1}{\Delta t} - S_{\theta_P} \right) \Delta x \Delta y + a_E + a_W + a_N + a_S
$$

$$b_P = \left( \frac{\theta_P^0}{\Delta t} - S_{\theta_P} \right) \Delta x \Delta y
$$

$$a_P \theta_P = a_E \theta_E + a_W \theta_W + a_N \theta_N + a_S \theta_S + b_P$$
Non-Dimensional Form

The mass-transport equation can be non-dimensionalized using characteristic parameters \( U, L, \) and \( \Delta t \). The equation can then be written as:

\[
\hat{R} \frac{\partial \theta}{\partial \tau} + \vec{\nabla} \cdot (\vec{f} \theta) - \frac{1}{P_{ch}} \nabla^2 \theta = \hat{R} (\hat{S}_1 + \hat{S}_2 \theta)
\]

where all quantities are dimensionless and the following are defined:

\[
\hat{R} = \frac{L}{U \Delta t}, \text{ dimensionless characteristic time step;}
\]

\[
P_{ch} = \frac{UL}{D}, \text{ characteristic Peclet number.}
\]

The discretization coefficients then take the following form (where the quantities \( u, v, \Delta x, \Delta y, \) and \( \delta \) are all non-dimensional):

\[
a_E = \frac{1}{P_{ch}} \frac{\Delta y}{\delta} \left[ [-P_e,(1 - \frac{P_e}{2}),0] \right]
\]

\[
a_W = \frac{1}{P_{ch}} \frac{\Delta y}{\delta_w} \left[ [P_w,(1 + \frac{P_w}{2}),0] \right]
\]

\[
a_N = \frac{1}{P_{ch}} \frac{\Delta x}{\delta_n} \left[ [-P_n,(1 - \frac{P_n}{2}),0] \right]
\]

\[
a_S = \frac{1}{P_{ch}} \frac{\Delta x}{\delta_s} \left[ [P_s,(1 + \frac{P_s}{2}),0] \right]
\]

\[
a_P = \hat{R} \left( \frac{1}{\Delta r} - \hat{S}_{2p} \right) \Delta x \Delta y + a_E + a_W + a_N + a_S
\]

\[
b_P = \hat{R} \left( \frac{\theta_P}{\Delta r} - \hat{S}_{1p} \right) \Delta x \Delta y
\]

\[
a_P \theta_P = a_E \theta_E + a_W \theta_W + a_N \theta_N + a_S \theta_S + b_P
\]

where the grid Peclet numbers are now defined as:

\[
P_e = u_e \delta_e P_{ch} \ (u_e, \delta_e \text{ non-dimensional})
\]
APPENDIX C

Navier-Stokes Equations; Velocity Profiles

Navier-Stokes Equations, Two-Dimensional Form

Assuming constant physical properties, buoyancy-drive flow, and gravity in negative-x direction.

Continuity:

$$\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} = 0$$

Momentum:

$$\frac{\partial u}{\partial t} + [u \frac{\partial u}{\partial x} + v \frac{\partial u}{\partial y}] = \nu \left[ \frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2} \right] + g \beta \Delta T$$

$$\frac{\partial v}{\partial t} + [u \frac{\partial v}{\partial x} + v \frac{\partial v}{\partial y}] = \nu \left[ \frac{\partial^2 v}{\partial x^2} + \frac{\partial^2 v}{\partial y^2} \right]$$

Energy:

$$\frac{\partial T}{\partial t} + [u \frac{\partial T}{\partial x} + v \frac{\partial T}{\partial y}] = \alpha \left[ \frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} \right]$$
Boundary Layer Form of Navier-Stokes Equations

assuming \( x = \) streamwise direction, \( y = \) cross-stream direction, and neglecting streamwise diffusion \( \left( \frac{\partial^2}{\partial x^2} \right) \)

momentum:

\[
\frac{\partial u}{\partial t} + (u \frac{\partial u}{\partial x} + v \frac{\partial u}{\partial y}) = \nu \frac{\partial^2 u}{\partial y^2} + g \beta \Delta T
\]

\[
\frac{\partial v}{\partial t} + (u \frac{\partial v}{\partial x} + v \frac{\partial v}{\partial y}) = \nu \frac{\partial^2 v}{\partial y^2}
\]

energy:

\[
\frac{\partial T}{\partial t} + (u \frac{\partial T}{\partial x} + v \frac{\partial T}{\partial y}) = \alpha \frac{\partial^2 T}{\partial y^2}
\]

Forced Convection Boundary Layer Flow

Imposed freestream velocity; \( U = \) constant

Velocity profile:

\[
\frac{u(y)}{U} = \frac{3}{2} \frac{y}{\delta} - \frac{1}{2} \frac{y^3}{\delta}
\]

Boundary layer thickness:

\[
\delta(x) = \left( \frac{280}{13} \frac{\nu x}{U} \right)^{1/2}
\]
Free Convection Boundary Layer Flow

Imposed wall-freestream temperature difference,
\[ \Delta T = T_{\text{hot}} - T_{\text{cold}} = \text{constant}. \]

Velocity profile:
\[
\frac{u(y)}{u_x} = \frac{y}{\delta} \left[ 1 - \frac{y}{\delta} \right]^2
\]

\[ u_x = 5.17 \nu \left[ \frac{20}{21} + \frac{\nu}{\alpha} \right]^{-1/2} \left[ \frac{g \beta \Delta T}{\nu^2} \right]^{1/2} x^{1/2} \]

\[ u_{\text{max}} = \frac{4}{27} u_x \text{ at } y = \frac{\delta}{3} \]

Boundary layer thickness:
\[ \delta(x) = 3.93 \left[ \frac{20}{21} + \frac{\nu}{\alpha} \right]^{1/4} \left[ \frac{g \beta \Delta T}{\nu^2} \right]^{-1/4} x^{1/4} \]
APPENDIX D

One-Dimensional Diffusion Equations

*General Formulation* (for decay product $i = 1, 2, 3$)

Governing equations:

\[
D_f \frac{d^2 A_i}{dy^2} = (\lambda_i + X) A_i - \lambda_i A_i - r_{i-1} \lambda_i A_{i-1}^a
\]

\[
D_a \frac{d^2 A_i^a}{dy^2} = \lambda_i A_i^a - X A_i' - (1-r_{i-1}) \lambda_i A_{i-1}^a
\]

where:

\[
A_i' = A_o = A_{radon} \quad A_i^a = 0
\]

\[
r_1 = r \quad r_2 = r_3 = 0
\]

Boundary conditions:

at $y = 0$; $A_i = 0$

at $y \to \infty$ \quad $\frac{dA_i}{dy} = \frac{d^2 A_i}{dy^2} = 0$ \quad $A_i = A_{i,\infty}$ \quad finite

Deposition velocity:

\[
u_i = \frac{D}{A_{i,\infty}} \frac{dA_i}{dy} \bigg|_{y=0}
\]

Define non-dimensional concentration:

\[
\theta_i = \frac{A_i}{A_o}
\]

Define diffusion lengths:

\[
d_i' = \left[ \frac{D_f}{\lambda_i + X} \right]^{1/2}
\]

\[
d_i^a = \left[ \frac{D_a}{\lambda_i} \right]^{1/2}
\]
First Free Decay Product

Non-dimensional governing equation:
\[
\left( d_1 \right)^2 \frac{\partial^2 \theta_1}{\partial y^2} = \theta_1 - \frac{\lambda_1}{\lambda_1 + X}
\]

Freestream concentration \( y_{\to \infty} \):
\[
\theta_1 \left( y_{\to \infty} \right) = \frac{\lambda_1}{\lambda_1 + X}
\]

Solution:
\[
\theta_1 \left( y \right) = \theta_1 \left( y_{\to \infty} \right) \left[ 1 - \exp \left( - \frac{y}{d_1} \right) \right]
\]

Deposition velocity:
\[
u_1 = \frac{D_1}{d_1} = \left[ D \left( \lambda_1 + X \right) \right]^{1/2}
\]

First Attached Decay Product

Non-dimensional governing equation:
\[
\left( d_1 \right)^2 \frac{\partial^2 \theta_1}{\partial y^2} = \theta_1 - \frac{X \theta_1}{\lambda_1}
\]

Freestream concentration \( y_{\to \infty} \):
\[
\theta_1 \left( y_{\to \infty} \right) = \frac{X}{\lambda_1} \theta_1 = \frac{X}{\lambda_1 + X}
\]

Solution:
\[
\theta_1 \left( y \right) = \theta_1 \left( y_{\to \infty} \right) \left[ 1 - \frac{\left( d_1 \right)^2 \exp \left( - \frac{y}{d_1} \right) - \left( d_1 \right)^2 \exp \left( - \frac{y}{d_1} \right) \left( d_1 \right)^2}{\left( d_1 \right)^2 - \left( d_1 \right)^2} \right]
\]
Approximate solution:

(assuming $D^f \gg D^a$, $d^f \gg d^a$)

$$\theta_1^a(y) = \frac{X}{\lambda_1} \theta_1^f(y) = \theta_1^a \left[1 - \exp\left(-\frac{y}{d^f}\right)\right]$$

Deposition velocity:

$$u_1^a = \frac{D^a}{d^f + d_1^a} \approx \frac{D^a}{d^f} = \left[\frac{D^a}{D^f}\right]^{1/2} \left[D^a(\lambda_1 + X)\right]^{1/2}$$

Second Free Decay Product

Non-dimensional governing equation:

$$(d_2^f)^2 \frac{d^2 \theta_2^f}{dy^2} = \theta_2^f - \frac{\lambda_2}{\lambda_2 + X} [\theta_1^f + r \theta_1^a]$$

Freestream concentration ($y \to \infty$):

$$\theta_2^f(\infty) = \frac{\lambda_2}{\lambda_2 + X} [\theta_1^f \theta_1^a] = \frac{\lambda_2(\lambda_1 + rX)}{(\lambda_2 + X)(\lambda_1 + X)}$$

Solution:

$$\theta_2^f(y) = \theta_2^f(\infty) \left[1 - \exp\left(-\frac{y}{d_2^f}\right)\right] + \frac{(d_2^f)^2 [\exp\left(-\frac{y}{d_2^f}\right) - \exp\left(-\frac{y}{d_2^f}\right)]}{(d_2^f)^2 - (d_2^f)^2}$$

Deposition velocity:

$$u_2^a = \frac{D^f}{d_2^f + d^f} = \frac{|D^f|^{1/2}}{(\lambda_2 + X)^{-1/2} + (\lambda_1 + X)^{-1/2}}$$

Second Attached Decay Product

Non-dimensional governing equation:

$$(d_3^2)^2 \frac{d^2 \theta_2^a}{dy^2} = \theta_2^a - \frac{X}{\lambda_2} \theta_2^f - (1 - r) \theta_1^a$$
Freestream concentration \((y \to \infty)\):

\[
\theta_{2,\infty} = \frac{X}{\lambda_2} \theta_{2,\infty} + (1 - r) \theta_{1,\infty} = \frac{X(\lambda_1 + rX)}{(\lambda_2 + X)(\lambda_1 + X)} + \frac{X(1 - r)}{\lambda_1 + X}
\]

Approximate solution:

(assuming \(D_f \gg D_a, d_f \gg d_a\))

\[
\theta_{2,e}(y) = \frac{X}{\lambda_2} \theta_{2,e}(y) + (1 - r) \theta_{1,e}(y)
\]

(see previous expressions for \(\theta_{2,e}(y)\) and \(\theta_{1,e}(y)\))

Deposition velocity:

\[
u_2 = \frac{D_a}{\theta_{2,e}} \left[ \frac{X}{\lambda_2} \theta_{2,e} u_2 + (1 - r) \theta_{1,e} u_1 \right]
\]

(see previous expressions)

**Third Free Decay Product**

Non-dimensional governing equation:

\[
(d_f^2)^2 \frac{d^2 \phi}{dy^2} = \theta_f - \frac{\lambda_3}{\lambda_3 + X} \theta_f
\]

Freestream concentration \((y \to \infty)\):

\[
\theta_{3,\infty} = \frac{\lambda_3}{\lambda_3 + X} \theta_{2,\infty} = \frac{\lambda_3 \lambda_2 (\lambda_1 + rX)}{(\lambda_3 + X)(\lambda_2 + X)(\lambda_1 + X)}
\]

Solution:

\[
\theta_f(y) = \theta_{3,\infty} \left[ \left(1 - \exp(-y/d_f^2)\right) + \frac{(d_f^2)^4 \left[ \exp(-y/d_f^2) - \exp(-y/d_f^2) \right]}{[(d_f^2)^2 - (d_f^2)^2] \cdot [(d_f^2)^2 - (d_f^2)^2]} \right]
\]

\[
+ \frac{(d_f^2)^4 \left[ \exp(-y/d_f^2) - \exp(-y/d_f^2) \right]}{[(d_f^2)^2 - (d_f^2)^2] \cdot [(d_f^2)^2 - (d_f^2)^2]} \right]
\]
Deposition velocity:

\[
\mathbf{u}_s^a = D^f \left\{ \frac{1}{d^f} + \frac{(d^f)}{[(d^f)^2 - (d_s^f)^2] \cdot [(d^f)^2 - (d_s^f)^2]} \left[ \frac{1}{d^f} - \frac{1}{d_s^f} \right] \right. \\
+ \left. \frac{(d^f)}{[(d^f)^2 - (d_s^f)^2] \cdot [(d^f)^2 - (d_s^f)^2]} \left[ \frac{1}{d^f} - \frac{1}{d_s^f} \right] \right\}
\]

**Third Attached Decay Product**

Non-dimensional governing equation:

\[
(d_{a_s^f})^2 \frac{d^2 \theta_{s^a}}{dy^2} = \theta_{s^a} - \frac{X \theta_{s^f}}{\lambda_3} - \theta_{s^a}^2
\]

Freestream concentration \((y \to \infty)\):

\[
\theta_{s_{3\infty}}^a = \frac{X}{\lambda_3} \theta_{s^f}^a + \theta_{s_{2\infty}}^a
\]

\[
= \frac{X \lambda_2 (\lambda_1 + rX)}{(\lambda_3 + X)(\lambda_2 + X)(\lambda_1 + X)} + \frac{X (\lambda_1 + rX)}{(\lambda_2 + X)(\lambda_1 + X)} + \frac{X (1 - r)}{(\lambda_1 + X)}
\]

Approximate solution:

(assuming \(D^f \gg D^a, d^f \gg d^a\))

\[
\theta_{s^a}(y) = \frac{X}{\lambda_3} \theta_{s^f}(y) + \theta_{s^a}(y)
\]

(see previous expressions for \(\theta_{s^f}(y)\) and \(\theta_{s^a}(y)\))

Deposition velocity:

\[
u_3^a = \frac{D^a}{\theta_{s_{3\infty}}^a} \left[ \frac{X}{\lambda_3} \theta_{s^f}^a u_s^f + \theta_{s_{2\infty}}^a u_2^a \right]
\]