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Effects of Vegetation on Radon Transport Processes in Soil: The Origins and Pathways of ^{222}Rn Entering into Basement Structures

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MASTER

EFFECTS OF VEGETATION ON RADON TRANSPORT PROCESSES IN SOIL: THE ORIGINS AND PATHWAYS OF ^{222}Rn ENTERING INTO BASEMENT STRUCTURES

The entry rate of ^{222}Rn into a basement structure was measured continuously. These measurements demonstrated that radon entry did not vanish even when the structure was slightly pressurized. This persistent entry has been determined to be dominated by diffusion through the floor and walls and a combination of diffusion and convection through the floor-wall joint. The highest indoor radon concentrations occurred during calm periods when the pressure differentials between the inside and outside of the structure were small. The objectives of this work were to identify the origins of the radon and investigate the entry pathways. The radon could originate either in the concrete or in the soil surrounding the structure. Entry pathways into the basement were through the concrete floor and walls as well as through the floor-wall joint. The contributions of the origins and entry pathways were determined by continuously measuring the radon entry rate into the basement, using a trace gas system, and the flux density through portions of the floor and walls. Radon entry through the floor-wall joint could be controlled using a baseboard barrier system. Results indicated that, during calm conditions with wind speeds less than 1 m s^{-1} , 25 % of the radon enters through the floor-wall joint and 75 % enters through the

concrete. About 30 % of the radon originated in the concrete floor and walls. A method for in-situ determination of the diffusion length and emanation fraction of radon in concrete was developed. For the concrete used in the structure, the average diffusion length and emanation fraction were 27 ± 4 cm and 0.19 ± 0.02 respectively.

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TABLE OF CONTENTS

ABSTRACT	iii
ACKNOWLEDGEMENTS	v
TABLE OF CONTENTS	iv
LIST OF TABLES	viii
LIST OF FIGURES	ix
CHAPTER I. INTRODUCTION	.1
 CHAPTER II. REVIEW OF LITERATURE	4
Health Risks Associated with ^{222}Rn	4
^{222}Rn Dynamics	10
Investigations of Transport Mechanisms	15
Origins and Entry Pathways of Indoor ^{222}Rn	16
^{222}Rn in Concrete	25
 CHAPTER III. MATERIALS AND METHODS	30
Experimental Facility	30
Data Acquisition System	40
Meteorological Parameters	40
Measurement of Pressure Differentials	42
Detection of ^{222}Rn	42

Ventilation Rate Measurement	44
^{222}Rn Entry Rate	46
Measurement of ^{222}Rn Flux Density	47
Partitioning of ^{222}Rn Entry	52
In-situ Measurement of F and D_e	56
 CHAPTER IV. RESULTS AND DISCUSSION	 60
Permeability and Soil Gas Radon	60
Pressure Differentials	63
Ventilation and ^{222}Rn Entry	68
^{222}Rn Flux Density	76
Radon Flux From Surfaces	82
Partitioning of Radon Origins and Entry Pathways	93
Radon Generation and Diffusion in Concrete	100
 CHAPTER V. CONCLUSIONS	 111
 REFERENCES	 114
 APPENDIX A. Derivation of Equation for Flux From a Slab	 120
APPENDIX B. Comparison of Linear and Non-linear Least Squares Fitting Techniques.	127

LIST OF TABLES

Table 1.	Short-lived decay products of ^{222}Rn and their half-lives, primary emission types and energies.	2
Table 2.	Lung cancer risk factors from exposure to ^{222}Rn	9
Table 3.	Contribution of various sources and transport mechanisms to the radon entry into a structure.	21
Table 4.	Properties of backfill soil surrounding the basement structures. . . .	34
Table 5.	Properties of the concrete used in the basement floor and walls. . .	34
Table 6.	Sperman rank correlation coefficients and 95 % confidence intervals for 1.0 m below ground flux as a function of wind speed, indoor-soil pressure, indoor temperature, barometric pressure and the first time derivative of barometric pressure.	82
Table 7.	Measured ^{222}Rn flux densities and entry rates.	95
Table 8.	Relative contributions of the origins and entry pathways to the global radon entry rate.	98
Table 9.	Radon entry results scaled to a 6.3 m x 16 m x 2.3 m basement with a ventilation rate of 0.2 h ⁻¹	100
Table 10.	Estimated emanation fraction (F), bulk diffusion coefficient (D _e) and diffusion length (l _c) for ^{222}Rn in concrete.	101

LIST OF FIGURES

Fig. 1.	Schematic diagram of the possible origins and entry pathways of radon entry into a structure.	19
Fig. 2.	Relative location of the basement structures.	32
Fig. 3.	Elevation view of one of the basement structures.	37
Fig. 4.	Diagram of baseboard barrier system used to control radon entry through the floor wall joint.	39
Fig. 5.	Schematic diagram of the flux measurement system.	49
Fig. 6.	Relative location of accumulator plate measurements.	55
Fig. 7.	Geometry used in the solution of the transport equation (eqn. 8). . .	57
Fig. 8.	Air permeability of the backfill soil surrounding the structures. . . .	62
Fig. 9.	Box plot of the soil gas ^{222}Rn concentrations at 30, 100 and 180 cm below the soil surface.	65
Fig. 10.	Wind speed, indoor-outdoor pressure and indoor-soil pressure at 30 and 100 cm below the soil surface.	67
Fig. 11.	Indoor and outdoor air temperature and soil temperature at 5, 30, 100 and 180 cm below the soil surface.	70
Fig. 12.	Wind speed at 1.0 m above ground, global radon entry rate, indoor radon concentration and ventilation rate.	72

Fig. 13. Box plot of the persistent radon entry rate with the floor-wall joint closed and open.	75
Fig. 14. Box plot of the net count rate from the floor accumulator plate radon detector with and without the delay line in place.	78
Fig. 15. Smoothed ^{222}Rn flux, barometric pressure, indoor-soil pressure and global radon entry.	81
Fig. 16. Box plot of the ^{222}Rn flux density from the wall and floor.	84
Fig. 17. Radon flux density and subslab radon concentration for a 14 d period.	87
Fig. 18. Theoretical steady-state concentration profile for radon in a 10 cm thick concrete slab.	89
Fig. 19. Theoretical radon flux from 10 and 15 cm thick concrete slabs as a function of the subslab radon concentration.	92
Fig. 20. Measured radon flux density from the floor slab as a function of the subslab radon concentration.	97
Fig. 21. Theoretical relationship between: concrete slab thickness, emanation fraction and diffusion length.	103
Fig. 22. Relationship between the calculated diffusion length and emanation fraction for slab thicknesses of 4, 8, 10, 12 and 15 cm. . . .	106
Fig. 23. Theoretical relationship between: the thickness of a concrete slab, porosity and the calculated diffusion length.	108

Fig. 24. Relationship between the calculated diffusion length and concrete porosity for 4, 8, 10, 12 and 15 cm thick slabs. 110

Fig. B-1. Ratio of the coefficients β and γ as a function of diffusion length. . .133

I. Introduction

Within the past decade indoor radon has been identified as the largest single source of radiation exposure to the general population. This exposure is primarily from the isotope ^{222}Rn . Based on studies of underground miners, ^{222}Rn and its decay products are known to cause lung cancer in humans. Exposure to radon concentrations typically found in homes in the United States are estimated to account for 0.5% to 5% of the total number of lung cancer deaths which occur each year (NCRP 1988a). Because of this, programs have been initiated in several countries which have the goal of reducing indoor radon concentrations. To effectively perform this task it is necessary to understand the properties of radon and how it enters dwellings. This includes understanding the transport mechanisms, where the radon originates and the pathways by which it enters the structure.

Of the 26 isotopes of radon only 2 occur naturally in great enough abundance to be of concern to human health. These are ^{220}Rn , which is often referred to as thoron, and ^{222}Rn . Of these thoron has been typically ignored in studies of lung cancer induction and radon transport. Historically it has been assumed that the risk of lung cancer from exposure to thoron and its decay products is much less than the exposure to the same concentration of ^{222}Rn (Schery 1989).

The isotope ^{222}Rn , hereafter referred to as radon, is generated in the decay of ^{238}U and its immediate precursor is ^{226}Ra . The average ^{226}Ra concentration of soil in the United States is $40 \pm 10 \text{ Bq kg}^{-1}$ ($1.0 \pm 0.3 \text{ pCi g}^{-1}$) with a measured range of 9 to 155 Bq kg^{-1} (Myrick et al. 1983). Since the radionuclides in the decay chain prior to radon are chemically reactive they are unable to travel any great distance. Radon, however, is an inert gas and with its 3.82 d half-life is able to be transported away from its place of origin. Following the decay of radon there is a series of 4 radionuclides which have half-lives ranging from $164 \mu\text{s}$ to 29 min (table 1). These are often referred to as the short-lived radon progeny.

Table 1. Short-lived decay products of ^{222}Rn and their half-lives, primary emission types and energies.

	Half-life	Decay mode	Energy (MeV)
^{218}Po	3.05 min	α	6.0
^{214}Pb	26.8 min	β	0.65, 0.71, 0.98
		γ	0.295, 0.352
^{214}Bi	19.7 min	β	1.0, 1.5, 3.3
		γ	0.609, 1.120, 1.764
^{214}Po	$164 \mu\text{s}$	α	7.7

Before radon can be a risk to human health it must be transported from its place of origin in a porous medium to the atmosphere. Since the diffusion length and recoil range for radon atoms in soil grains is approximately 70 nm (Tanner 1980) the atoms, in order to be available for transport, must be released into the pore spaces of the material. Generation of radon in a porous medium is a

function of the properties of the radon isotope and the material. The important isotope properties are the concentration of the precursor (^{226}Ra and ^{224}Ra) and the half-life of the isotope of radon. Properties of the material which are important are its bulk density, moisture content and porosity. Another factor which must be taken into account is the emanation fraction. This is the fraction of radium decays which release radon into the pore spaces. This is a function of the grain structure, pore size and moisture content. It is also dependent on the location of the radium in grain and the energy of the decay. The emanation of radon in a porous medium is discussed in detail by Tanner (1980).

Two of the more important radon transport mechanisms are convection and diffusion. Convection, frequently referred to as pressure-driven flow, is the transport of a material due to a pressure differential. Pressure fields around a structure can be caused by meteorological parameters such as wind, changes in barometric pressure, temperature differentials and precipitation. Occupants can also influence the pressure fields around a structure through the use of appliances, fireplaces, furnaces and the opening and closing of windows and doors (Nazaroff et al. 1988). Diffusion is the transport of a molecular species which results from a concentration gradient and occurs in the absence of, or even against, a pressure differential. The diffusive flux density is the rate at which atoms cross a surface area and has units of atoms, or activity, per unit time per unit area. The flux density is proportional to the concentration gradient with the constant of proportionality being the diffusion coefficient. The mathematical expression of this is known as Fick's law.

Radon can enter a structure through the floor, walls or through openings which are in direct "communication" with the soil without an intervening material other than air. Radon entering through the floor and walls can originate either in the soil or in the building materials. Transport can also occur through openings in the building shell such as cracks, utility entry points and joints in the floor and wall. In both cases the transport mechanism can be either diffusion, convection or a combination of both. The use of radon laden water, natural gas and the infiltration of outdoor air are also sources of indoor radon entry. The objectives of this study were to partition the radon entry according its origins (soil and concrete) and entry pathways (through the floor, walls and floor-wall joint). An in-situ determination of the diffusion coefficient and emanation fraction of ^{222}Rn in concrete was also made.

The difficulty in understanding the origins, transport mechanisms and entry of radon in houses is their multi-compartment nature as well as confounding factor introduced by the activities of the occupants. Therefore, it is desirable to have a system which, while being simpler than a full size house, still maintains similarities of construction techniques and materials. The structures used in this investigation consisted of a single room with no partitions and were constructed using materials and techniques typical for the Colorado front range area. The radon entry was partitioned into radon originating in the soil and in the concrete. The contributions of radon entering through the floor, walls and floor-wall joint to the total entry was also determined. This was achieved by continuously

measuring the indoor radon concentration, radon entry rate and the flux density through the floor and walls. Additionally, by measuring the flux from the floor slab while changing the subslab radon concentration an in-situ determination of the diffusion and emanation coefficients for ^{222}Rn in concrete was made. The coefficients were estimated by performing a non-linear, least-squares fit to the steady-state solution to Fick's law for 1-dimensional diffusion through a concrete slab.

II. Review of Literature

Health Risks Associated with ^{222}Rn

The primary health concern created by exposure to radon and its short-lived progeny is the induction of lung cancer. This was first observed in studies of underground miners (NRC 1988). Risk to other organs, from inhaled radon progeny, has been estimated to be at least 2 orders of magnitude less than the risk to the lungs (James 1988). The risk is not from radon itself but from its short-lived, alpha emitting, decay products. While this is true for all radon isotopes, studies analyzing the risk have focused on ^{222}Rn and its short-lived progeny: ^{218}Po , ^{214}Pb , ^{214}Bi and ^{214}Po . The tissue at risk is not the entire lung but the undifferentiated cells in the bronchial epithelium (James 1988). Since the dose to the bronchial epithelium is delivered by alpha particles, produced by decay of the progeny which plate out in the lungs, the concept of the potential alpha energy concentration (PAEC) has been developed. The PAEC is the amount of alpha kinetic energy which would potentially be released in the complete decay of the short-lived radon progeny. This is expressed in units of energy per unit volume (J m^{-3}) or the working level (WL). One working level is equivalent to a PAEC of $2.08 \times 10^{-5} \text{ J m}^{-3}$. The PAEC is not directly related to the dose to the tissues at risk. This is because the dose delivered to the undifferentiated cells in the bronchial epithelium, and not the entire lung, is of

importance for the induction of lung cancer (NCRP 1984b). The deposition of the progeny in the lungs will affect the dose to the bronchial epithelium. Some factors which determine the location that the progeny are deposited are the particle size, respiratory rate and unattached fraction (NCRP 1984b).

When deposited in the lungs the progeny may be removed before decaying, be absorbed into the blood stream or decay. Removal rates and mechanisms are dependent on the location in the lung. In the alveolar region removal is primarily from absorption into the blood stream. The mean residency time of particles in this region is 700 min (Jacobi 1964). In the bronchi the primary removal mechanism is mucociliary clearance. The velocity of the mucus layer which lines the bronchi is approximately 5 mm min^{-1} (Hui et al. 1990). Generally, this process is not rapid enough to remove the ^{222}Rn progeny from the bronchi and trachea prior to decaying (Jacobi 1964).

Those alpha emitting radon progeny which decay in the tracheo-bronchial region must be located where the alpha particle can cause damage to the nuclei of the undifferentiated cells in the bronchial epithelium. Although the dose to the basal cells is often considered to be of the greatest importance doses received by all of the epithelial cells except for the goblet and ciliated cells can lead to cancer (James 1988). The median thickness of the epithelial layer is $15 \mu\text{m}$ with a range of 5 to $30 \mu\text{m}$ (Gastineau 1972). The progeny are deposited on the mucus layer which is approximately $7 \mu\text{m}$ thick. This places the nuclei of the sensitive cells within the ranges of the ^{218}Po and ^{214}Po alpha particles in tissue (48 and $71 \mu\text{m}$ respectively).

The other naturally occurring radon isotopes (^{219}Rn and ^{220}Rn) have typically been ignored as causes of lung cancer. Radon-219 is not of concern because of the low concentrations of its progenitor, ^{235}U , found in the earth's crust. The reason indoor thoron has usually been discounted is that concentrations have been assumed to be low and the dose to the tracheo-bronchial epithelium is a factor of 3 less, per unit of inhaled alpha energy, than from ^{222}Rn (NRC 1988). This assumption may not always be true. Based on measurements in 17 U.S. homes Schery (1989) estimates that the average thoron concentration is between 5 and 30 Bq m^{-3} which is comparable to the US median ^{222}Rn concentration of 25 Bq m^{-3} (USEPA 1992). However, he has also found that the potential alpha energy concentration (PAEC) of thoron progeny is approximately half that of ^{222}Rn progeny. It is estimated that the dose to the lung, per unit exposure, due to thoron progeny is around 20% that from ^{222}Rn progeny (Schery 1989, James 1988). Little epidemiological data exists on lung cancer incidence due to exposure to thoron.

Most risk estimates are based on the analysis of lung cancer incidence in underground miners. The populations investigated include: uranium miners on the Colorado plateau and in New Mexico, Czechoslovakia, Ontario and Saskatchewan, iron miners in Sweden and fluorospar miners in Newfoundland (USEPA 1992a). Table 2 contains a summary of the risk estimates from these analyses. The estimated cancer incidence rates all fall between 10 and 100 cases per year per 10^6 people exposed. At the current United States population of

2.5 x 10⁸ (USDC 1992) this would relate to 2500 to 25 000 additional lung cancers annually. This estimate is based on the total population being exposed to the median indoor radon concentration in U.S. houses of 25 Bq m⁻³ (USEPA 1992b) for 24 h d⁻¹ with the radon progeny at 50% equilibrium. These estimates are averaged over the total population without making any corrections for age, sex, exposure time or smoking status. Some studies estimate the risk to sub-populations by; smoking status (USEPA 1992a, NRC 1988, ICRP 1987), sex (NRC 1988, ICRP 1987) and age and duration of exposure (NRC 1988, ICRP 1987).

Table 2. Lung cancer risk factors from various studies. Risk factors are averaged over the entire population with no differentiation for age, sex, exposure time or smoking status. Cancer incidence is based on an exposure rate of 0.2 WLM y⁻¹.

Source	Risk Factor (WLM ⁻¹)	Lung Cancer Incidence Rate (Cases 10 ⁻⁶ y ⁻¹)
NCRP 1984a	2 x 10 ⁻⁴	40
NCRP 1984b	1 - 4 x 10 ⁻⁴	20 - 80
NRC 1988	3.5 x 10 ⁻⁴	70
UNSCEAR 1977	2 - 4.5 x 10 ⁻⁴	40 - 90
ICRP 1987	0.6 - 1.8 x 10 ⁻⁴	12 - 36
USEPA 1992b	1.12 - 4.80 x 10 ⁻⁴	22 - 96
Nero 1983	0.5 - 5 x 10 ⁻⁴	10 - 100
All Causes ^a (NCRP 1984a)		1900

^a Annual lung cancer incidence from all causes including radon.

The authors of the studies admit that uncertainties are introduced in trying to estimate the risk of lung cancer to people chronically exposed to low concentrations in homes based on data from the high exposure conditions

encountered in mines. This is because of differences in the atmosphere (eg. particulate size distributions and presence of other carcinogens), respiratory rates, and radon-radon progeny equilibrium ratios. Also all of the miner data is for males of working age. Even with these differences the difference between the lung dose per unit exposure in a mine atmosphere and a home is expected to be less than a factor of 2 (NRC 1988). In order to better estimate the risk due to exposures in homes a series of epidemiological and animal studies are underway (Neuberger 1992; Borak and Johnson 1988; NRC 1988). The use of case-control and cohort studies of the risk from long term domestic radon exposures are difficult due to the uncertainty in historical exposure information (Mahaffey et al. 1993). Attempts to avoid this problem through the use of ecological studies has led to uncertain results because of they use population averages rather than characteristics of the individual. This is sometimes referred to as the "ecological fallacy" (Stidley and Samet 1992).

²²²Rn Dynamics

The radon concentration in the pore spaces of a medium can be described by using a mass balance model. Radon is released into the pore volume by the decay of ²²⁶Ra in the pore walls and the subsequent emanation into the pore space. The loss of radon from a given pore volume is through: diffusion and convective transport through the connected pores and from radioactive decay. The decay of ²²²Rn and is controlled by its decay constant ($\lambda = 2.1 \times 10^{-6} \text{ s}^{-1}$). For

materials transported in the gas phase the effects of gravity are small and generally ignored. Each of the input terms will be discussed in detail.

The generation of radon in a porous medium is a function of its ^{226}Ra concentration, bulk density, emanation fraction, porosity and the decay constant of ^{222}Rn . Mathematically this is expressed by:

$$\phi = \frac{A_{Ra} \rho_b F \lambda}{\epsilon} \quad (1)$$

where;

ϕ = ^{222}Rn activity generation rate into the pore volume ($\text{Bq m}^{-3} \text{ s}^{-1}$).

A_{Ra} = ^{226}Ra concentration (Bq kg^{-1}).

ρ_b = bulk density of the medium (kg m^{-3}).

F = ^{222}Rn emanation coefficient.

λ = decay constant for ^{222}Rn ($2.1 \times 10^{-6} \text{ s}^{-1}$).

ϵ = porosity of the medium.

The emanation coefficient is the ratio of ^{226}Ra decays which result in ^{222}Rn being released into the pore space. This is a function of the location of the radium in the grain, the grain size and the moisture content and temperature of the medium (Tanner 1980, Stranden et al. 1984). The emanation coefficient has been found to increase up to a maximum as the moisture content is increased. If the moisture content continued to increase the emanation coefficient rapidly decreases (Stranden 1984). In the same study F was found to increase as the temperature increased.

Transport by diffusion is the net migration of atoms from a region of high concentration to one of lower concentration. This process is independent of any pressure differentials as long as they do not disturb the concentration gradient. The diffusive flux of radon atoms across a surface area is directly proportional to the concentration gradient. The constant of proportionality is the diffusion coefficient. This relationship is known as Fick's law and can be expressed by:

$$J_d = -D_e \cdot \nabla C_{Rn} \quad (2)$$

where;

J_d = diffusive flux density (Bq m⁻² h⁻¹).

D_e = effective diffusion coefficient (m² h⁻¹).

∇C_{Rn} = radon concentration gradient in the pore volume.

There is a certain amount of confusion in the regarding the definition of the diffusion coefficient. Collé (1981) defines D_e as the effective diffusion coefficient relating the concentration gradient to the flux across the bulk, or geometric, surface area of the material. The flux across the pore area can be calculated by using the interstitial diffusion coefficient, D_e^* . The interstitial diffusion coefficient is related to the bulk diffusion coefficient by the porosity of the material (eqn. 3).

$$D_e = \epsilon D_e^* \quad (3)$$

An alternative definition is the use of D_e as the pore, or interstitial, diffusion coefficient (Nazaroff et al. 1988). Confusion in the exact definition being used

can result in errors in the diffusion coefficient of an order of magnitude or more. The definition used by Coll will be used here. The diffusion coefficient is sometimes expressed in terms of a relaxation or diffusion length. This is the linear distance in which the steady-state concentration profile decreases by e^{-1} (37 %). The distance actually traveled by the atoms will be greater than the diffusion length since the pore spaces are generally not connected in straight lines. In the notation used here, the diffusion length, l_c , is:

$$l_c = \sqrt{\frac{D_e}{\epsilon \lambda}} \quad (4)$$

Convection, frequently referred to as pressure-driven flow, is the transport of atoms along a pressure gradient. The atoms are carried along with the fluid (air or water) as it moves through the medium. Although water can transport radon into structures, this discussion will focus on air as the transporting fluid. The mathematical expression which describes the velocity is known as Darcy's law and is written as:

$$\mathbf{v} = -\frac{\mathbf{K}}{\xi} \cdot \nabla P \quad (5)$$

where:

$\mathbf{v}$ = superficial velocity vector across area A ($\text{m}^3 \text{s}^{-1} \text{m}^{-2}$).

$\mathbf{K}$ = 3 x 3 permeability matrix (m^2).

ξ = viscosity of air ($\text{kg m}^{-1} \text{s}^{-1}$).

∇P = pressure gradient (Pa m^{-1}).

The area, A , in the superficial velocity is a geometrical area which is greater than the area of the pore openings. The permeability matrix is a measure of the ease with which air flows through the medium (Davis 1969). For an isotropic medium the matrix can be expressed as a single value, k . The convective flux vector, $\mathbf{J}_c$, across the geometric area of an isotropic medium can be expressed by:

$$\mathbf{J}_c = C_{Rn} \mathbf{v} \quad (6)$$

Based on conservation of mass, the above terms can be combined to give a general partial differential equation for the time rate of change of the pore space radon concentration in a medium with negligible water content (eqn. 7).

$$\frac{\partial C_{Rn}}{\partial t} = -\frac{1}{\epsilon} \nabla \cdot \mathbf{J}_d - \frac{1}{\epsilon} \nabla \cdot \mathbf{J}_c - \lambda C_{Rn} + \phi \quad (7)$$

This relation has been expanded to include semi-saturated media by taking into account the fraction of the pore volume occupied by water, the emanation of radon into the water and the transfer of radon between the water and air (Rogers and Nielson 1993; Nero et al. 1988).

Although finite element models have been used to arrive at numerical solutions to the transport equation (eqn. 7) (Schery et al. 1988a, 1988b; Tsang and Narasimhan 1992; Holford et al. 1993), it is common to make simplifying

assumptions in order to allow an analytical solution. Common assumptions are: D_e and K are isotropic and homogenous, the media is free of water and transport occurs in 1-dimension. Applying these, eq. 7 reduces to:

$$\frac{\partial C_{Rn}}{\partial t} = \frac{D_e}{\epsilon} \frac{\partial^2 C_{Rn}}{\partial x^2} - \frac{1}{\epsilon} \frac{\partial (v C_{Rn})}{\partial x} - \lambda C_{Rn} + \phi \quad (8)$$

This can be further simplified by evaluating for steady state conditions and for diffusive or convective transport only.

Investigations of Transport Mechanisms

As indicated above the 2 primary mechanisms for radon transport are convective flow and diffusion. Studies of these transport mechanisms have led to a variety of conclusions. In a review of available data for subslab radon concentrations in houses, air exchange rates and fluxes from building materials Bruno (1983) concluded that flow through cracks makes a much greater contribution to the radon entry than does diffusion. Eaton and Scott (1984) state that the daily variations in the indoor radon concentration indicate that the entry rate is highly variable and, consequently not controlled by diffusion.

Measurements of radon concentrations and ventilation rates in 100 houses in the U.S. and Canada indicated that in some houses there was a correlation between the radon source and the ventilation rate (Nazaroff et al. 1981). From this it was concluded that the dominant radon entry mechanism was soil gas influx rather

than diffusion. The National Council on Radiation Protection and Measurements concludes that diffusion cannot account for the observed radon entry into a structure (NCRP 1989).

Other investigators have concluded that diffusion can be an important contributor to indoor radon. In a series of flux measurements, carried out in 3 houses in Grand Junction, Colorado, Kendrick and Langner (1991) found that 80 % of the entry through cracks in the structure boundary was due to diffusion. Holub et al. (1985) concluded that diffusion was the dominant transport mechanism in an energy efficient house in Denver, Colorado. Ward et al. (1993) continuously measured the ^{222}Rn concentration, ventilation rate and ^{222}Rn entry rate in 2 basement structures. They concluded that, for periods with winds less than 1 m s^{-1} , diffusion was the dominant transport mechanism and could account for greater than 80 % of the long term entry. In a theoretical analysis Tanner (1990) concluded that diffusion would be dominant in materials with permeabilities less than $7.6 \times 10^{-12} \text{ m}^2$. A study conducted in a 4.9 m^3 test structure by Harley and Terilli (1986) indicated that 60 % of the radon entry was due to diffusion.

Origins and Entry Pathways of Indoor ^{222}Rn

Indoor ^{222}Rn can originate in any material which contains ^{226}Ra and where a pathway exists for the radon to be transported into a structure. The materials of interest here are the soil surrounding the structure and the concrete used in construction. Radon can enter a structure through the floor and walls or through

cracks and joints in the building shell. Other sources include infiltrating outdoor air and the use of radon laden water and natural gas (fig. 1). The relative contributions of the origins of the radon to the total entry or concentration have been studied theoretically, in laboratory measurements and in studies in houses.

Only a limited number of studies have partitioned the radon entry into a structure by the origins and transport mechanisms of the radon. In his review Bruno (1983) partitioned the entry rate based on a steady-state analysis of a "typical house". The United Nations Scientific Committee on the Effects of Atomic Radiation has issued a report which estimates the fraction of radon entering through the building materials and along with water, infiltrating outdoor air and natural gas (UNSCEAR 1982). While they do not separate the radon originating in the soil from that which originated in the building materials they state that, in some houses, soil may be the dominant source. The International Commission on Radiological Protection (ICRP 1987) and Nero et al. (1987) estimated the contribution the building materials, soil gas, tap water and outdoor air make to the indoor concentration. These calculations were based on a theoretical analysis of average fluxes from building materials and soil. Based on measurements in 14 houses located in the pacific northwest, Turk et al. (1990) estimated the contributions soil gas, building materials and tap water made to the indoor concentration during the winter months. Kahn et al. (1983) estimated the contributions of soil gas entry, building materials and outdoor to the indoor concentration based on flux measurements in 3 houses. Table 3 contains a

Fig. 1. Schematic diagram of the possible origins and entry pathways of radon entry into a structure.

summary of these studies. Quantitative comparisons of the results are difficult because of differences in the units and assumptions made for the theoretical calculations. However, it is clear that soil gas is considered to be the most important contributor to the indoor radon concentration. In order to help compare the results it is helpful to understand the assumptions and general conclusions of each of the studies.

Bruno (1983) used a slab-on-grade structure with 150 m² floor and 2.5 m ceiling. An additional 150 m² of masonry was used in the walls. The ventilation rate of the structure was 1 h⁻¹. Contributions of the building materials to the indoor concentration were based on a flux density from the masonry of 10 Bq m² h⁻¹. He concluded that: 1) in order to reduce the radon entry, cracks and other openings in the slab should be sealed and 2) the use of diffusion coatings is pointless since diffusion is a minor contributor to the total entry rate. However, he does admit that the building materials and diffusion may become important contributors in houses with low ventilation rates (0.1 - 0.2 h⁻¹).

In a review of available data UNSCEAR (1982) evaluated the contributions of radon entering through the building shell, infiltrating outdoor air, natural gas and LP gas to the entry rate into a structure. The structure under consideration had a volume of 200 m³ and a surface area of 350 m². The ventilation rate was 1 h⁻¹. Of the radon entering through the building shell there was no differentiation between radon which originated in the soil as opposed to

Table 3. Contribution of various sources and transport mechanisms to the radon entry into a structure.

	Soil Gas, Diffusion	Soil Gas, Transport	Building Materials	Tap Water	Outdoor Air
Bruno 1983 (Bq s ⁻¹)	0.1 - 0.2	0 - 5.6	0.01 - 1.1	0 - 2.2	0
UNSCEAR 1982 (Bq s ⁻¹)		0.7 ^a		0.08	0.1
ICRP 1987 (Bq m ⁻³)		2 - 60 ^b	3 - 30	≤ 0.1	3 - 7
Nero 1980 (Bq m ⁻³)		50 ^b	2	0.4	10
Turk et al. 1990 (Bq m ⁻³)		350 - 5000 ^b	7 - 96	< 4 - 11	0
Kahn et al. 1983 (Bq m ⁻³)		7 - 40 ^b	1 - 2		4 - 8

^a Combination of diffusion, convection and emanation.

^b Both diffusive and convective flow.

that originating in the building materials. It is stated that soil can be a significant source particularly when there are cracks exist in the structure boundary. Water became important only when the concentration in the water was greater than 10 kBq m^{-3} . The contributions from natural and LP gas were highly variable but were considered to be insignificant when compared to the other sources.

The International Commission on Radiological Protection estimated that building materials will account for approximately 30 % of the indoor radon concentration (ICRP 1987). Their conclusions are based on typical ranges of volume-specific entry rates found in the literature. The volume-specific entry rate is the rate of increase of the radon concentration and has units of $\text{Bq m}^{-3} \text{ h}^{-1}$. The ventilation rate used for the calculations was 0.7 h^{-1} .

In an analysis of average emanation rates from concretes and soil, Nero et al. (1990) conclude that for 1 or 2 story houses that the soil is the primary source of indoor radon. Infiltrating outdoor air is the second most important contributor. Other sources, such as natural gas are insignificant when compared to soil gas and outdoor air. The hypothetical structure used in these calculations was a single story wood frame construction with a concrete slab-on-grade foundation. The slab was 20 cm thick, the floor area 100 m^2 and the ceiling 2.4 m. The ventilation rate was assumed to be between 0.2 and 0.8 h^{-1} . In an earlier analysis Nero and Nazaroff (1984) concluded that the extent to which transport mechanisms account for the actual entry rate will depend on the specific characteristics of the building and that, for smaller buildings, soil gas entry is the most important contributor.

Measurements carried out in 14 houses located in the Pacific Northwest concluded that soil gas entry was the most important contributor to the indoor radon concentration (Turk et al. 1990). The flow rate of soil gas into the structure was calculated based on the: soil gas radon concentration at infinite depth, average indoor concentration during the winter months, measured ventilation rate and structure volume. The ventilation rate was measured at least once in each house using a tracer gas technique. Measurements of flux density were used to calculate the contribution from the building materials.

The contribution of the building materials to the indoor radon concentration in 3 houses investigated was investigated by Kahn et al. (1983). The flux from the building surfaces was measured using 10 cm charcoal canisters. The contribution to the indoor concentration was calculated using an assumed ventilation rate of 1.1 h^{-1} . Since the flux measured over cracks and seams was much greater than that from the intact slab it was concluded that most of the soil gas entry was through such openings. For surfaces with ^{226}Ra concentrations near 40 Bq kg^{-1} the contribution from the building materials was approximately a factor of 10 less than the contributions from soil gas or outdoor air.

Other authors have made qualitative determinations about the contributions of the origins and entry pathways to indoor radon. In a laboratory experiment Ingersoll (1983) measured the emanation of radon from concrete cylinders placed in sealed cans for 1 to 3 d. The average exhalation rate was used to calculate the contribution of concrete to the indoor concentration in a 170 m^2

wood frame structure with a thick concrete slab foundation and a 2.4 m ceiling. With an assumed ventilation rate between $0.5 - 1 \text{ h}^{-1}$ the concrete would contribute 4 to 8 Bq m^{-3} to the indoor concentration. From this he concludes that building materials are not normally an important contributor to indoor radon concentrations in U.S. houses.

Harley and Terilli (1986) evaluated diffusion and convection as sources of indoor radon. Measurements of the radon flux density from a 20 cm thick concrete block wall were carried out. The contribution of the radon generated in the concrete to the total flux was calculated using measured values of the ^{226}Ra concentration, emanation fraction and porosity and the diffusion coefficient of radon in concrete. It was calculated that approximately 70 % of the flux was from radon generated in the concrete.

A series of measurements was carried out in 20 homes with high indoor radon concentrations at Elliot Lake, Canada (DSMA 1978). The approach was to measure the flux density from the walls and floor and the soil gas concentrations. Theoretical calculations for diffusion through and emanation from the concrete were carried out using diffusion coefficients and porosities measured by Culot et al. in Grand Junction, Colorado (1976). They concluded that in order for diffusion through the concrete to be the transport mechanism the radon concentration in the soil gas would have to be between 3 and 30 MBq m^{-3} . For the concrete to be the source the emanating ^{226}Ra concentration would have to be 300 to 370 Bq kg^{-1} . Since these conditions were not met it was concluded

that diffusion or emanation from the concrete could not be significant sources of indoor radon concentrations at Elliot Lake.

The actual contributions that the soil and building materials make to the radon entry rate or indoor concentration is highly variable. The flux density from different building materials has been observed to vary over 2 orders of magnitude (Stranden 1988). This is caused by the variation of the ^{226}Ra content of the material (Stranden 1983, Ulbak et al. 1984), its density, porosity and emanation fraction. These factors can be further affected by the moisture content in the material, temperature (Stranden et al. 1984) and the presence of micro-cracks (Holub and Brady 1981). Turk et al. (1990b) state that in order to properly assess the importance of a transport mechanism that it is necessary to make measurements of diffusion and emanation from the building materials as well as convection and soil gas entry. In general making a-priori judgements of the transport mechanism, origin or entry pathway of radon based on other "similar" situations is not appropriate.

^{222}Rn in Concrete

An array of methods have been used in order to determine the emanation coefficient and diffusive properties of radon in concrete. As early as 1955, Gabrysh studied the effects of heating on radon emanation from concrete samples. Since that time the majority of the investigations have been carried out on small concrete blocks, or cylinders, in a laboratory setting. Only a limited

amount of information is available on measurements carried out on an intact wall or slab.

The most common laboratory method involves sealing the sample in a container and allowing the radon to grow into equilibrium with the radium. The canister is then counted on a γ -spectroscopy system and the radon activity determined (A_c). A second measurement of the radon activity is made with the canister unsealed (A_o). The emanation coefficient, F , is then calculated using:

$$F = 1 - \frac{A_o}{A_c} \quad (9)$$

Another common method is to measure the radon which is released into the air volume surrounding the sample in a sealed container (Jonassen 1983).

Both methods were used by Ingersoll (1983) to determine the emanation coefficient for 106 concrete samples obtained from 8 U.S. states. The measured values ranged from 13 to 25 %. Rogers and Nielson (1992) used the second method to determine the emanation coefficient for concrete samples from Florida. Values ranged from 0.019 to 0.078. Using a similar procedure, Stranden et al. (1984) measured the emanation of radon from concrete samples as a function of moisture content and temperature. Emanation coefficients for 4 concrete samples from Poland were between 0.008 and 0.085 (Peńsko et al. 1980).

Laboratory measurement of the diffusion coefficient is generally conducted by applying a known radon concentration to one side of a sample and measuring

the concentration on the opposite side. This can be done either after equilibrium has been reached or as a function of time. The appropriate solution to the transport equation (eqn. 7), assuming that diffusion is the only transport mechanism, is then used to determine the diffusion coefficient.

Zapalac (1983) observed the time-dependent ingrowth of radon on one side of a thin cylinder with a fixed concentration on the other side. He assumed that steady state was reached rapidly and the sample was very thin compared to the diffusion length. The sample itself was ignored as a source of radon. In measurements of 2 different samples the diffusion lengths were 13 and 17 cm which correspond to effective diffusion coefficients of $3.5 \times 10^{-4} \text{ cm}^2 \text{ s}^{-1}$ and $6.1 \times 10^{-4} \text{ cm}^2 \text{ s}^{-1}$ respectively.

In a similar study Folkerts et al. (1984) measured the diffusion coefficient for samples of "aerated" and "heavy" concrete under steady-state conditions. The effective diffusion coefficient ranged from $1.3 \times 10^{-2} \text{ cm}^2 \text{ s}^{-1}$ ($l_c = 50 \text{ cm}$), for the aerated sample, to $7 \times 10^{-3} \text{ cm}^2 \text{ s}^{-1}$ for the heavy concrete ($l_c = 2 \text{ cm}$). As in the previous study the sample was not considered to be a source of radon.

Measurements of Florida concretes conducted by Rogers and Nielson (1992) resulted in diffusion coefficients ranging from $1.8 \times 10^{-4} \text{ cm}^2 \text{ s}^{-1}$ to $4.6 \times 10^{-3} \text{ cm}^2 \text{ s}^{-1}$. It is not clear which definition of diffusion coefficient was used. The method used was described only as the Florida Radon Research Program protocol.

A unique approach was used by Jonassen and McLaughlin (1980). The measured the diffusion coefficient by measuring the steady state radon concentration profile in a wall. This was done by drilling a hole partway into the wall and sealing a sample tube in place leaving only a small airspace around the end of the tube. Air samples were then collected and the radon concentration determined. The pore diffusion coefficient was calculated based on the steady-state solution to eqn. 7 for known, fixed radon concentrations on both sides of the wall and with the concrete being a source of radon. The estimated pore diffusion coefficient was $3.4 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$. Based on a porosity of 0.45 the bulk diffusion coefficient is $1.5 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$ and the diffusion length 4 cm. They admit that the results may be highly uncertain since the method of collecting the air samples was crude.

An in-situ method of determining the diffusion coefficient was developed by Culot et al. (1976). They measured the radon flux from a wall in a basement surrounded by U mill tailings. The flux was measured by sealing a metal container to the wall and observing the buildup of radon. The radon concentration in the mill tailings adjacent to the wall were monitored periodically. A water manometer was used to insure that a pressure difference did not occur across the wall. The steady-state transport equation was solved for known concentrations on either side of the wall. The concrete was assumed to not significantly contribute to the flux. The resulting diffusion length was $2.5 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$ ($l_e = 10 \text{ cm}$).

As part of a study carried out in an energy efficient house in Denver, Colorado, Holub et al. (1985) estimated the diffusion coefficient for the structure as a whole. A general steady-state solution for the emanation of radon into an enclosed space was used for this purpose. The primary assumptions for the solution were that the walls were considered to be infinitely thick, radon was generated in the concrete and the outdoor air did not significantly contribute to the radon entry. The resulting diffusion length was 43 cm. These results were disputed by Nazaroff (1988) and a re-analysis resulted in a diffusion length between 80 and 110 cm (Holub and Borak 1988).

The diffusion lengths, of radon in concrete, measured in the above studies ranged from 2 to 110 cm. This compares to a range of values of 17 to 40 cm given in a review by Nero and Nazaroff (1984). The large variation in values indicates the need for further study in order to properly assess diffusion as a radon transport mechanism. In order to evaluate the various experimental methods and their results there needs to be use of a common definition of the diffusion coefficient as well as documentation of the assumptions and boundary conditions applied to the transport equations.

III. Materials and Methods

Experimental Facility

Experiments investigating the origins of available radon and pathways of radon entry were carried out in 2 specially constructed basement structures. These structures are located on the Colorado State University campus in Fort Collins, Colorado and are part of a continuing investigation of how deep rooted vegetation affect radon transport into basements. They were constructed using building materials and methods typical to the Colorado front range area. The area surrounding the basements is flat with no large obstructions such as trees or other buildings. They are separated from each other by approximately 18 m and are surrounded by a backfill area extending 2.5 m from the walls (fig. 2). During this investigation the backfill area was kept free of all vegetation.

Construction began with the excavation of two cavities measuring at least 8.7 m x 8.7 m x 4 m. This allowed for a minimum of 2 m of backfill material extending from the walls and floor. The extent of the backfill was selected to be greater than the approximate 1 m diffusion length for ^{222}Rn in soil. The backfill was carried out alternately into each cavity in 15 cm lifts using the soil which was originally excavated. This was done in order to reduce the probability of having a

Fig. 2. Relative location of the basement structures. The backfill material extends 2.5 m from the walls of the structures.

location with a high ^{226}Ra concentration, or "hot spot", occurring near one of the structures and not the other. It also served the purpose of homogenizing the soil; eliminating any cracks, large stones or roots which might influence the transport of radon into the basements. The moisture content and packing density of the lifts were measured periodically by a soils testing firm with a nuclear moisture/density gauge. During the backfill process 6 soil samples were collected at random and analyzed for ^{226}Ra content, porosity, emanation fraction, and the fraction sand, silt and clay. The results of these analyses are listed in table 4.

When the when the cavities had been filled to 2 m below ground level the footers for the basement were poured. Next the walls were poured as a single unit. The area inside the footers was filled with a 15 cm thick gravel layer. The mean diameter of the gravel was approximately 2 cm. A perforated PVC pipe was installed for use at a later date as part of a subslab ventilation system. Finally, a 0.2 mm (0.008") thick vapor barrier was installed over the gravel layer and the floor poured. This resulted in a basement structure with no wall-to-wall joints and a single continuous floor-wall joint. The inside dimensions of the basements are 3.35 m x 3.35 m x 2.3 m. The walls and floor are 15 and 10 cm thick respectively. Samples of the concrete used in the floor, walls and footers were also analyzed for ^{226}Ra content, bulk density and porosity and the results are summarized in table 5.

Once the structure was completed the backfill continued as described above.

Table 4. Properties of backfill soil surrounding the basement structures. Soil is classified as a sandy clay loam.

^{226}Ra Concentration	$52 \pm 4 \text{ Bq kg}^{-1}$
^{222}Rn Emanation Fraction (Dry)	0.10 ± 0.02
Grain Density	$2660 \pm 60 \text{ kg m}^{-3}$
Porosity	0.39 ± 0.03
Bulk Density	1600 kg m^{-3}
Composition:	
Sand	49 %
Silt	27 %
Clay	24 %

Table 5. Properties of the concrete used in the basement floor and walls.

	Walls	Floor
Thickness (m)	0.15	0.10
^{226}Ra Concentration (Bq kg^{-1})	62 ± 4	62 ± 4
Porosity	0.13 ± 0.02	0.13 ± 0.02
Bulk Density (kg m^{-3})	2100	2100

During the backfill process plastic tubes for the measurement of indoor-soil pressure differentials, soil gas ^{222}Rn concentrations and air permeability were placed. The tubes were 6 mm OD PVC and were terminated in the soil by an 90° elbow which was oriented downward. This was done to inhibit water entry into the tubes. The tubes were passed into the building through one of 9 conduits

in each wall or 3 in the floor. After all tubes had been installed the conduits were sealed with a silicon caulking compound. When not in use the tubes were capped to prevent soil gas entry into the structure. Figure 3 is an elevation view of the completed basement. The roof peak is approximately 1 m above ground level. The walls extend 2 m below the soil surface and a 30 cm section is above ground. Circles in the backfill area represent locations where pressure differentials, soil gas radon concentrations and permeability were measured. There are a total of 191 locations around each basement. The vertical lines in the backfill area represent the location of access ports used for the measurement of soil moisture with a neutron moisture probe.

In one basement a baseboard barrier system has been installed to control radon entry through the floor-wall joint (fig. 4). The barrier was sealed in place using a combination of rope caulk and a polyurethane sealant. The system was periodically tested for leaks by injecting sulphur hexafluoride (SF_6) gas behind the barrier and "sniffing" using an infrared spectrophotometer. Any leaks were resealed using the polyurethane sealant and the system retested. This was repeated until no further leaks were found. A series of 5 cm diameter ports were installed in the baseboard barrier. There were 2 ports located on along each wall. When the ports were capped the floor-wall joint was sealed. This prevented gas from the subslab air space and soil from entering the structure through the joint. The barrier could be "removed" by uncapping the ports.

Fig. 3. Elevation view of one of the basement structures. Circles represent locations of pressure tubes. The vertical lines in the backfill region represent access ports used to measure soil moisture with a neutron moisture probe.

Fig. 4. Diagram of baseboard barrier system used to control radon entry through the floor-wall joint. When the caps were in place the floor-wall joint was sealed. There were 2 caps located along each wall.

Data Acquisition System

Continuous data were collected using a computer based acquisition system. Transducers with voltage signals were connected directly to a 32 channel datalogger which had an 8 bit analog-to-digital converter and a RS-232 serial interface. One datalogger was located in each structure. Since the dataloggers did not have a pulse counting option, transducers with digital outputs (eg. radon detectors and precipitation gauge) were connected to 12 bit digital-to-analog (D/A) converters. The D/A outputs were then connected to the datalogger. The dataloggers could be programmed to integrate selected input signals over the data collection interval. At the end of each measurement interval the computer would collect the data from the dataloggers, store it in an ASCII file on the hard drive and reset the integrators and D/A cards for the next interval. Files were periodically transferred from the hard drive to a floppy disk for further analysis. At this time any changes in sensor configurations would be made, a new datafile started and length of the collection interval selected. The interval could range from 1 to 10^4 min per interval. For all data presented here, the interval length was 15 min. The system could also be controlled and data transferred from a remote terminal without entering either of the structures.

Meteorological Parameters

Two meteorological stations were located on the site. One station, at 1 m above ground level, was located between the basements and a 3 m station was

located in the southwest corner of the site (fig. 2). The height of the 1 m station was selected as being most representative of the winds which would influence the structures. A tipping-bucket precipitation gauge¹ was also located on site. The precipitation gauge had a resolution of 0.25 mm and was heated during periods when the outdoor temperature was less than 7°C to measure precipitation due to snowfall. Wind speed, wind direction, barometric pressure and outdoor temperature were measured at the 1 m station. The wind speed and direction were also measured at the 3.0 m station. Wind speed was measured using a 3 cup anemometer connected to a DC generator². The threshold of the anemometer was 0.4 m s⁻¹ with a range of 0 to 45 m s⁻¹. A wind vane, with a 0.34 m s⁻¹ threshold was used to measure the wind direction³. The angular resolution of the wind vane (0.5°) was controlled by the data collection software. Since large variations in the wind speed and direction can occur during the 15 min between readings they were averaged over the collection interval. The outdoor temperature was measured with a resistance temperature device (RTD). An absolute pressure transducer with a range of 0 to 1293 mmHg was used to measure barometric pressure⁴. The outdoor temperature and barometric pressure

¹ Model 6028-A. Qualimetrics, Inc. 3213 Orange Grove Ave., Sacramento, CA 95660.

² Model 2011. Qualimetrics, Inc. 3213 Orange Grove Ave., Sacramento, CA 95660.

³ Model 2005. Qualimetrics, Inc. 3213 Orange Grove Ave., Sacramento, CA 95660.

⁴ Model 280E. Setra Systems, Inc., 45 Nagog Park, Acton, MA 01720.

readings were taken once an interval since they are unlikely to change greatly during the 15 min period between readings.

Measurement of Pressure Differentials

Differential pressure transducers were used to measure the pressure change between the inside of the structures and the outdoor air and soil. The inside of the structure was used as a reference for all pressure measurements. A negative pressure indicated that the pressure inside the basement was lower than the exterior point. Therefore, the net airflow would be into the structure. The point used to measure the indoor-outdoor pressure differential was located approximately 2 m from the structure and 10 cm above ground level. It was protected from the wind by a metal enclosure. The soil pressure locations were at the ends of the 6 mm OD tubes placed during construction (fig. 3). The transducers had a range of ± 25 Pa and a lower limit of detection of 0.2 Pa⁵. Because changes in pressure differentials can occur over short periods of time the readings were averaged over the 15 min collection interval. Periodic transducer calibrations were conducted using an inclined plane manometer.

Detection of ²²²Rn

Continuous measurements of indoor, outdoor and subslab concentrations were carried out using a flow-through scintillation flask which was fabricated from

⁵ Model 264. Setra Systems, Inc., 45 Nagog Park, Acton, MA 01720.

a commercially available flask counter (Ward and Borak 1991). The continuous detectors had a volume of 1 l and were operated at a flow rate of 0.2 l min^{-1} . The D/A converter cards used with the indoor and subslab continuous detectors could accumulate a total of 2^{11} and 2^{15} counts, in an interval, respectively. If this number of counts was exceeded then the counter would reset and begin accumulating again at 0 V. The D/A card used with the outdoor radon detector could accumulate the same total number of counts as those used with the indoor detectors. The outdoor concentration was measured at 1 m above ground level. Continuous measurements of the subslab radon concentration were collected through the perforated PVC pipe placed in the subslab gravel layer during construction. After passing through the detector the subslab air was vented outside the basement.

Instantaneous, or grab samples, of radon in the soil gas as well as the indoor and subslab radon concentrations were taken on a weekly basis using 100 cm^3 scintillation flasks. The scintillation flasks were calibrated with an National Institute of Standards and Technology (NIST) traceable ^{226}Ra solution (Monette 1990). Also, the performance of the systems were evaluated annually in a blind intercomparison conducted by the Environmental Measurements Laboratory in New York. All samples were collected by pulling a known volume of air with a syringe and injecting it into a scintillation flask. The flasks were allowed to "age" for a minimum of 3 h and counted on a photomultiplier tube. Grab samples of indoor and subslab air were used to verify that the continuous detectors were operating properly.

Ventilation Rate Measurement

The ventilation rate is the total rate of air exchange in the building from all sources. Nero (1988) divides the ventilation into 3 components: infiltration through cracks and holes, mechanical ventilation caused by fans and exchange through windows and doors. Since these structures have no windows and data was collected with the door closed, the only components of ventilation are infiltration and mechanical ventilation. Mechanical ventilation occurred when a small blower fan was used to pressurize the structure. Continuous measurement of the total ventilation rate was made using the trace gas sulfur hexafluoride (SF_6) and a continuous sampling spectrophotometer⁶. Sulfur hexafluoride was selected since it does not naturally occur in the environment, is non-toxic and is easily detectable. The SF_6 was released into the room at a well regulated rate and the concentration in the center of the room measured with the spectrophotometer. A first order differential equation (eq. 10) was used to describe the concentration in the room at any given time.

$$\frac{dC_s(t)}{dt} = \left(\frac{R_s}{V_{eff}} \right) - \lambda_v(t) C_s(t) \quad (10)$$

where:

$C_s(t)$ = concentration of SF_6 in the structure (PPM).

⁶ Miran-1A. Foxboro Analytical, P.O. Box 5449, South Norwalk, CT. 06856.

R_s = SF₆ flow rate (cm³ h⁻¹).

V_{eff} = effective volume of the structure = 23 m³.

$\lambda_v(t)$ = ventilation rate of the structure (h⁻¹).

The effective volume of the structure is slightly smaller than the physical volume due to space occupied by the instruments and furnishings. Measurement of the effective volume was accomplished using a tracer gas dilution technique (Ward 1992).

In order for eq. 10 to be valid there must be rapid mixing of the trace gas within the room. This assumption was tested using the SF₆ concentration as a function of time data used to determine the effective volume. After the release of trace gas the concentration in the room reached a steady state concentration within 10 min. Since this is less than the 15 min interval between readings the assumption of rapid mixing was not violated.

Assuming the only source of SF₆ is the regulated release in the room and the only loss through ventilation then eq. 10 can be solved for the ventilation rate (eq. 11). This can be calculated since F and V_{eff} are known and the SF₆ concentration is measured every 15 min. The first time derivative of the concentration was calculated numerically using a 5 point moving average filter (Stummel and Hainer 1980).

$$\lambda_v(t) = \frac{\left(\frac{R_s}{V_{eff}} - \frac{dC_s(t)}{dt} \right)}{C_s(t)} \quad (11)$$

²²²Rn Entry Rate

Determination of the total rate of radon entry was possible since the ventilation rate and indoor ²²²Rn concentrations were measured continuously. The first order differential equation used to describe the indoor radon inventory as a function of time was:

$$\frac{dQ(t)}{dt} = R(t) - Q(t) \lambda_v \quad (12)$$

where:

$Q(t)$ = ²²²Rn inventory in the structure (Bq).

$R(t)$ = Total rate of ²²²Rn input into the structure from all sources (Bq h⁻¹).

Loss due to radioactive decay can be ignored since it is much smaller than the typical ventilation rates of 0.03 h⁻¹. Based on a diffusion coefficient for radon in plywood of 10⁻⁹ m² s⁻¹ and a ceiling thickness of 13 mm the diffusion of radon through the ceiling is at least 10 times less than the loss of radon through ventilation. The inventory of radon in the structure can be calculated from the indoor concentration ($C_{Rn}(t)$) using:

$$C_{Rn}(t) = \frac{Q(t)}{V_{eff}} \quad (13)$$

The effective volume (V_{eff}) in eq. 13 is the same as that used in the determination

of the ventilation rate (eq. 11). Solving eq. 12 for the total entry rate and substituting eq. 13 gives:

$$R(t) = V_{eff} \left(\frac{dC_{Rn}(t)}{dt} + \lambda_v C_{Rn}(t) \right) \quad (14)$$

$R(t)$ is the total ^{222}Rn input rate from all sources including soil, building materials and infiltrating outdoor air. The contribution of the radon in outdoor air to the total entry rate can be removed using the measured outdoor radon concentration ($C_o(t)$) and the ventilation rate. The corrected entry rate ($R_c(t)$), henceforth referred to as the global entry rate, was calculated using eq. 15. All entry rates measured by the trace gas system presented in this work represent the global radon entry rate from all sources other than outdoor air.

$$R_c(t) = R(t) - V_{eff} \lambda_v C_o(t). \quad (15)$$

Measurement of ^{222}Rn Flux Density

The radon flux density through the floor and walls of the basement was measured using an accumulator plate system (fig. 5). The plates consisted of 13 mm thick plexiglass sheets which had a 6 mm deep cavity milled in one side. In order to measure the flux density the plate was sealed to the surface using a combination of rope caulk and a silicon sealant. Room air was pulled through the plate at approximately 0.1 l min^{-1} and into a flow-through scintillation flask of the

Fig. 5. Schematic diagram of flux measurement system. The arrows represent the direction of airflow.

same type used to measure the indoor radon concentration. The air then passed through a mass flowmeter and exhausted back into the room. The flowmeter was calibrated at ambient temperature and pressure (21°C and 630 mmHg respectively) using a volumetric flow standard⁷. By knowing the radon concentrations at the inlet and outlet of the plate, the flow rate and the plate area the flux density from the surface can be calculated using:

$$J = \frac{(C_2 - C_1) \times \dot{v}}{A} \quad (16)$$

where:

J = radon flux density (Bq m⁻² h⁻¹).

C_2 = radon concentration at plate outlet (Bq m⁻³).

C_1 = radon concentration at plate inlet (Bq m⁻³).

$\dot{v}$ = air flow rate through plate (m³ h⁻¹).

A = plate surface area (m²).

The plates were constructed in 4 sizes: 0.059 m² (140 mm x 420 mm), 0.0621 m² (140 mm x 440 mm), 0.12 m² (290 mm x 420 mm) and 0.13 m² (290 mm x 440 mm). The 2 small plates were used primarily to measure the flux from the walls and the large plates were used to make measurements on the floor. The flow rate and plate volumes were selected so that the mean residency time of the air in the plates (8 min for the largest plate) would be less than the 15 min

⁷ Gillian Instrument Corp. 35 Fairfield Place, W. Caldwell, NJ 07006-6206.

collection interval while still allowing for a measurable difference in the radon concentrations between the plate inlet and outlet. This made it possible to measure any temporal changes in the flux density. When calculating the flux it was not necessary to insert a time delay between the radon concentrations since the mean residency time of the air in the largest plate and the transit time from the plate to the detector was less than the data collection interval. A differential pressure transducer was used to measure the pressure differential between the inside the cavity and the room. The flow rate was adjusted until the pressure inside the cavity was within ± 0.1 Pa of the room pressure.

The minimum detectable flux (MDF) is a function of the radon concentration at the inlet (C_1), the plate size and the flow rate. Treating the inlet concentration as the "background" of the system the MDF was calculated for the probability of committing a type 1 and type 2 error of 0.05 (NCRP 1978). The inlet radon concentration was assumed to be 1100 Bq m^{-3} which is a typical radon concentration encountered in the basements. For this case the MDF for the small and large plates are 12 and $6 \text{ Bq m}^{-2} \text{ h}^{-1}$ respectively. The MDF can be decreased by decreasing inlet radon concentration.

This system has several advantages over other methods of measuring the radon flux from a surface. The primary one is that, since room air is used to flush the plate, the radon concentration under the plate is very similar to the concentration seen by the rest of the wall. This eliminates the possibility of increased diffusion due to a lower concentration, such as would be experienced by

adsorption onto activated charcoal or flushing with low radon air. Also, the problems of back diffusion are much smaller than encountered with a closed container. Another advantage with using room air to flush the plate is the seal with the surface is not critical as long as any airflow out of the plate passes through the outlet radon detector. Finally, the plate volume, air flow rate and continuous radon detector make it possible to detect short term changes in the flux. Since the residency time in the plates is less than the length of the 15 min reading interval it would be possible to measure temporal variations with periods greater than 30 min. These changes would result in changes in the global radon entry rate. Changes in the radon entry rate have been shown to be caused by winds which occur over periods of minutes to hours (Ward et al. 1993). A measurement method which integrates the flux over several hours would not be able to detect these wind induced fluctuations. The automated nature of the data collection system also makes it possible to measure the fluxes over extended periods without disturbing the system by entering the structure.

Partitioning of ^{222}Rn Entry

Diffusion has been found to be the dominant mode of radon transport into these structures during periods with low average wind speeds (Ward et al. 1993). During these periods it can account for over 80 % of the long term global entry rate and results in the greatest indoor radon concentrations. For these reasons the relative contributions of the origins of the entering radon and the entry

pathways to the diffusive global radon entry rate were determined. Possible origins are the soil surrounding the basement and the concrete floor and walls. Radon can enter the structure through the floor, walls and floor-wall joint. Entry through the floor and walls was measured using the accumulator plate system. The baseboard barrier system was used to determine the amount of radon entering through the floor-wall joint.

Flux measurements were taken at 7 locations on the wall and 2 locations on the floor (fig. 6). One of the locations on the wall was above ground level. This plate measured the radon entering through the wall which originated in the wall concrete. The flux from the below ground portions of the wall were measured at 30, 100 and 180 cm, each at 2 different locations. These measured the radon entering through the wall which originated in the soil as well as in the concrete. Similarly, the plates in 2 different locations on the floor measured the radon entering through the floor which originated in the subslab airspace as well as in the floor concrete. The contributions to the global entry were calculated by multiplying the flux density by the area associated with that portion of the structure. For example, the flux density through the above ground portion of the wall would be multiplied by the surface area of this section of the wall (4 m^2). This procedure assumes that the flux measured at one location is representative of the flux through the rest of the surface.

The baseboard barrier system was used to determine the amount of radon entering through the floor-wall joint. The global entry rate was measured with

Fig. 6. Relative location of accumulator plate measurements in the basement structure. Measurements of the flux from the below ground portion of the wall were conducted on the east and south walls. Radon flux from the floor was measured in the northeast and northwest corners of the basement.

and without allowing radon entry through the floor-wall joint. The change in the global entry rate was used to estimate the portion of the radon entering through the floor-wall joint.

In-situ Measurement of F and D_e

An in-situ method of determining the emanation and diffusion coefficients for ^{222}Rn in concrete was developed. The general approach was to continuously measure the flux from a surface while changing the concentration on the other side. A non-linear, least squares, fit to the steady-state solution to Fick's law for the 1-dimensional flux from a slab with known concentrations on both sides using the measured flux and radon concentrations. The emanation and diffusion coefficients were treated as unknowns.

Fig. 7 shows the geometry used to reach the solution to the flux equation. The boundary conditions were:

$$C_c(Z=0) = C_s \quad (17)$$

and

$$C_c(Z=L) = C_I \quad (18)$$

where C_c , C_I and C_s are the radon concentrations in the concrete, inside and outside the structure respectively (fig. 7). Using the above boundary conditions and solving eq. 8 for the 1-dimensional diffusive flux under steady-state conditions

Fig. 7. Geometry used in the solution of eq. 8 for diffusion from a concrete slab.

gives:

$$J(L) = l_c \epsilon \phi_c \left(\frac{\cosh\left(\frac{L}{l_c}\right) - 1}{\sinh\left(\frac{L}{l_c}\right)} \right) - \frac{l_c \epsilon \lambda C_l \cosh\left(\frac{L}{l_c}\right)}{\sinh\left(\frac{L}{l_c}\right)} + \frac{l_c \epsilon \lambda C_s}{\sinh\left(\frac{L}{l_c}\right)} \quad (19)$$

This derivation assumes that the concrete was a source of radon and that steady-state was "rapidly" reached. Although the relaxation time for the diffusion of radon in a 10 cm thick concrete slab is approximately 11 h the steady-state solution to Fick's law was used for the curve fit. This violates the assumption that steady-state conditions were achieved within each measurement interval.

However, the subslab concentration (C_s) changed from one equilibrium condition to another within reached equilibrium within 4 d and data was collected for a period of a minimum of 5 d at each concentration. Therefore, sufficient data was collected under equilibrium conditions that the use of a time-dependent solution was not necessary. A detailed derivation of eq. 19 is given in appendix A.

The fitted parameters were the emanation fraction and the diffusion length. They were treated as unknowns because of the large variation in the values found in the literature and the difficulty in measuring them accurately using laboratory techniques. Other parameters, such as ^{226}Ra concentration, porosity, slab thickness and bulk density, can be easily and accurately measured using conventional techniques.

The problem with current methods of determining D_e and F are that they typically use small samples of concrete. Frequently these samples are prepared in the laboratory with careful attention given to the removal of air bubbles and curing of the sample. Also, laboratory samples have not been subjected to the temperature and moisture conditions experienced in a house. These conditions can lead to the formation of micro-cracks which will influence the diffusive and emanation properties of the concrete (Samaha and Hover 1992; Holub and Brady 1981). Results from core samples removed from existing buildings are also questionable due to the introduction of additional cracking in the coring process.

The most common method for determining the ^{226}Ra concentration of a material involves sealing the sample in a radon tight container, allowing the ^{222}Rn to grow into equilibrium with the ^{226}Ra and then counting the gamma rays from ^{214}Pb and ^{214}Bi (Stranden 1988). The mixing performed in the making the concrete should result in a uniform ^{226}Ra concentration within a batch of concrete. Therefore, several small samples of the concrete should be representative of the entire batch. Total porosity and bulk density of materials can be determined using methods described in Klute (1986). The measurements will represent the concrete in the structure as long as the samples are prepared from the same batch of concrete and in the same manner as used in constructing the structure. Factors which can influence the porosity and density of the concrete are: tamping to remove air bubbles, temperature, moisture content and curing conditions (Rogers and Neilson 1989; Samaha and Hover 1992).

IV. Results and Discussion

Comparisons of the 2 basements with respect to: indoor radon concentrations, ventilation rate, pressure differentials, temperature and radon entry rate were conducted by Ward (1992). He found that while they are not identical they have similar responses to changes in wind speed and direction. Also, the same techniques and materials were used in the construction of the basements. Therefore, conclusions based on measurements carried out in one structure can be applied to the other. The following discussion will focus primarily on measurements conducted in the west basement between December 1992 and April 1993.

Permeability and Soil Gas Radon

Convective flow of soil gas is governed by the permeability of the medium and the pressure gradient. The permeability of the soil surrounding the basements has been measured on a weekly basis for approximately 2 years. Fig. 8 is a box plot representation of the air permeability of the soil in the backfill area surrounding the 2 basements. The variability in the readings are caused by changes in the soil moisture content and local inhomogeneities, such as cracks, in the backfill (Schumann et al. 1992). The median air permeabilities for the soil

Fig. 8. Air permeability of the backfill soil surrounding the structures. The median of all data is $8 \times 10^{-12} \text{ m}^2$. Dotted lines represent the average value. The solid horizontal represent the median. The limits of the box indicate 25 % below and above the median. The vertical lines extend to the 10th and 90th quantiles. Symbols represent extreme values. Test refers to a reference soil with fixed permeability used to check the operation of the permeability measuring device.

surrounding the east and west basements are $1.3 \times 10^{-11} \text{ m}^2$ and $2.1 \times 10^{-12} \text{ m}^2$ respectively. However, it should be noted that the measured permeabilities range over 4 orders of magnitude. The medians for the soil around the structures fall within the 25th and 75th quantiles for all data values. The median of all measurements is $8 \times 10^{-12} \text{ m}^2$.

Soil gas radon concentrations were measured on a weekly basis. Fig. 9 is a box plot of the concentrations measured at 3 depths near the west basement during the period between Dec. 1992 and Apr. 1993. The concentration increases with increasing depth below ground. Temporal variations in the soil gas radon concentration are caused by changes in the soil moisture content (Monette 1991). Correlation of changes in the soil gas radon concentration with changes in barometric pressure and wind speed were not possible. This is because of the samples were collected once a week. Based on the Nyquist sampling theorem (Ramirez 1985) only frequency components with periods greater than 2 weeks will be observable. Changes in wind speeds typically take place on the order of minutes to hours while barometric pressure changes occur over a periods of hours to several days.

Pressure Differentials

Pressure differentials between the inside and outside of the structure are caused by winds, temperature differentials and changes in barometric pressure. Fig. 10 shows the wind speed at 1.0 m above ground, indoor-outdoor, and indoor-soil pressure differentials for a 2 week period in march, 1993. A quadratic

Fig. 9. Box plot of the soil gas ^{222}Rn concentrations at 30, 100 and 180 cm below the soil surface. Samples were taken from a point 30 cm from the wall between December 1992 and April 1993. Boxes enclose data between the 25th and 75th percentiles. The median is represented by the horizontal line. Data between the 10th and 90th percentiles fall within the vertical lines. Symbols represent extreme values.

Fig. 10. Wind speed (A), indoor-outdoor pressure (B) and indoor-soil pressure at 30 cm (C) and 100 cm (D) below the soil surface. Soil pressures were measured 30 cm from the south wall.

relationship has been found between the wind speed and the pressure differentials at a given location in the soil (Ward et al. 1993). This relationship is modified by the wind direction. The effect of wind direction on the indoor-soil pressure differentials decreases at greater depths and distances from the walls.

Temperature differences between the inside and outside of the basements did not effect the pressure differentials. This is because most of the structures lie below ground level where the soil temperature is relatively constant (fig. 11).

Barometric pressure also did not have an observable effect on the pressure differentials. This is because the relaxation time for pressure changes to propagate through 1 m of soil (permeability = 10^{-11} m²) is approximately 2.4 s (Collé 1980). Also, the time required for changes in barometric pressure to propagate into the structure through leakage sites is instantaneous. Therefore, any pressure differentials created by changes in barometric pressure will reach steady-state within a single measurement interval.

Ventilation and ²²²Rn Entry

Since pressure differentials are driven by winds it would be expected that the entry rate would increase with increasing wind speeds. Fig. 12 A and B are, respectively, plots of the wind speed at 1.0 m above ground level and the global radon entry rate, determined with the trace gas system, for a 2 week period. It is clear that as the wind increases the entry rate also increases. The reason that a given wind speed does not always result in the same entry rate is the directional

Fig. 11. (A) Indoor and outdoor temperature and (B) soil temperatures at 5, 30, 100 and 180 cm below the soil surface for a 2 week period in May.

Fig. 12. Wind speed at 1.0 m above ground (A), global radon entry rate (B), indoor radon concentration (C) and ventilation rate (D) for a 14 d period. Data was collected every 15 min. Radon entry through the floor wall joint was not obstructed during this period.

dependance of the pressure differentials and the locations of the entry sites. It is important to note that even though the entry rate increases during periods of high winds the indoor radon concentration generally decreases (fig. 12 C). This is because there is the competing process of increased ventilation (fig. 12 D) removing the radon at a greater rate than it can be replaced by the increased entry. However, no definite correlation between the radon entry rate and indoor concentration has been found (Ward et al. 1993). This is because the ventilation rate of a structure is not constant. Therefore, for these structures, it is improper to make judgements about the importance of a transport mechanism or entry pathway based solely on measurements of concentration.

When the wind speeds, and therefore the pressure differentials, approach zero a non-zero component of the global radon entry rate remains. This entry persists even when the structure is slightly overpressurized indicating that diffusion is an important transport mechanism. It is during calm periods, when this entry is dominant that the indoor ^{222}Rn concentration reaches its greatest values (fig. 12 B and C). This persistent entry, over an extended period, can account for over 80 % of the total entry (Ward et al. 1993). Since this entry results in the highest indoor concentrations the origins of the entering radon and the entry pathways were investigated in greater detail.

Fig. 13 is a box plot of the persistent entry components with the floor-wall joint open and closed. When the radon entry through the floor-wall joint was

2 of 2

Fig. 13. Box plot of the persistent radon entry rate with the floor-wall joint closed and open. During the closed condition radon entry through the joint was prohibited. The persistent entry is due to diffusion and remains even when the structure is slightly overpressurized.

blocked the median persistent entry rate decreased by approximately 20 %. This indicates that at least 80 % of the radon entry is from radon passing through, or emanating from, the concrete floors and walls.

²²²Rn Flux Density

Initial measurements carried out on the above ground portion of the wall indicated that the flux from the surface was greater than expected. At the same time a side-by-side comparison of 2 continuous flux measurement systems was being conducted on a portion of the floor. One system was developed at Colorado State University (CSU) and is described above. The other was developed by Kendrick and Langner of the U.S. Dept. of Energy, Grand Junction Projects Office, Radon Laboratory (Kendrick and Langner 1991). The flux measured by the Grand Junction system was approximately 55 % less than that measured by the CSU system. Further investigation revealed that this was caused by ²²⁰Rn emanating from the concrete surface. A delay line was installed between the plate outlet and the radon detector on the CSU system (fig. 5). The line was long enough to allow the thoron to decay before entering the detector. Fig. 14 is a box plot of the net detector count rates with and without the delay line in place. After the removal of the thoron the fluxes measured by the systems were indistinguishable (Kendrick et al. 1993). The Grand Junction system was insensitive to ²²⁰Rn because the accumulator consisted of a large, cylindrical container with a ventilation rate of 0.5 h⁻¹. This allowed the thoron to decay before entering the radon detector. All subsequent measurements were carried out with the delay line in place.

Fig. 14. Box plot of the net count rate from the radon detector connected to the floor accumulator plate with and without the delay line in place. The delay line was of sufficient length to allow the thoron to decay prior to entering the detector.

The flux density at all locations showed some temporal variation. Fig. 15 is a plot of the wall flux at 1 m below the soil surface, barometric pressure, indoor-soil pressure differential and global ^{222}Rn entry rate over a 2 week period. The wall flux was smoothed using a 15 point, linear, moving average filter to remove the variations which are due to counting statistics. Spearman rank correlation coefficients were used to determine the ability of wind speed, indoor-soil pressure differentials, indoor temperature, barometric pressure and the first derivative of barometric pressure with respect to time to account for the variations in the flux. The correlation coefficients were calculated for each parameter as a single predictor of the flux. A rank correlation was used to avoid problems created by differences in the ranges of the data (Kleinbaum et al. 1988). This procedure results in a measure of the degree of correlation between the 2 variables. However, since the ranks and not the actual values are used, the procedure cannot be used to predict the flux based on the independent variable (e.g. wind speed, pressure differential, barometric pressure, etc.). All of the coefficients were statistically significant with the wind speed having the strongest correlation (table 6). The values of the coefficients were slightly different for other time periods and locations but in all cases the order of strongest to weakest correlation remained the same. It should be noted that the variations in the flux are insufficient to account for the changes in the global entry rate caused by convective flow. For example, the highest observed entry rate shown in fig 15 C is 2.7 kBq h^{-1} . If this entry were to occur uniformly through all below ground

Fig. 15. Smoothed ^{222}Rn flux (A), barometric pressure (B), indoor-soil pressure (C) and global radon entry rate (D) for 14 days. Data was collected every 15 min. The flux was measured on the wall, 1.0 m below ground. Smoothing was carried out using a 15 point, linear, moving average filter. The soil pressure was measured 1.0 m below ground and 30 cm from the wall.

surfaces of the basement the change in the flux would be approximately 50 Bq m² h⁻¹. The observed change in flux was less than 10 Bq m² h⁻¹. Since changes in the global entry rate are created by pressure differentials this indicates that pressure driven flow through intact concrete may be a small contributor to the dynamic portion of the global radon entry. The convective flow of gas into the basement must take place through other portions of the structure boundary such as the floor-wall joint.

Table 6. Sperman rank correlation coefficients, and their 95 % confidence intervals, for 1.0 m wall flux as a function of wind speed, indoor-soil pressure, indoor temperature, barometric pressure and the time rate of change of barometric pressure (dB/dt).

Parameter	r _s	95% Confidence Interval
Wind Speed	0.26	0.21 - 0.31
Indoor-Soil Pressure	-0.13	-0.18 - (-0.08)
dB/dt	0.13	0.08 - 0.18
Barometric Pressure	0.11	0.05 - 0.16
Indoor Temperature	0.08	0.03 - 0.14

Radon Flux From Surfaces

Fig. 16 is a box plot of the ²²²Rn flux densities measured on the walls and floor. The fluxes measured on the below ground portions of the wall (30, 100 and 180 cm) were compared using the Kruskal-Wallis test. The medians are significantly different from each other (B = 89, p < 0.0005). However, the range

Fig. 16. Box plot of the ^{222}Rn flux density from the wall and floor. The wall flux was measured: above ground level (0) and at 30, 100 and 180 cm below ground. Measurements of the floor flux were conducted with (closed) and without (open) the baseboard barrier in place. The horizontal line within the box represents the median and the box encloses 25 % of the data greater than and less than the median. The vertical lines extend to the 10th and 90th percentiles. Symbols represent data outside this range.

of the means is small enough (21 to 24 Bq m² h⁻¹) that the use of the average of all values (22 ± 5 Bq m² h⁻¹) to represent the entire below ground portion of the wall does not affect the final result. The change in the floor flux with the floor-wall joint open and closed was found to be caused by a change in the subslab radon concentration (fig. 17).

In order to explain these results a theoretical analysis was made. Assuming that diffusion was the only transport mechanism the radon concentration in, and flux through, a concrete slab system were calculated. The diffusive flux of radon through a porous medium is described mathematically by Fick's law (eq. 2). Since the concrete contains ²²⁶Ra and, therefore, is a source of ²²²Rn the flux is not purely a function of the radon concentrations on either side of the slab. Fig. 18 is a plot of the computed radon concentration profile in a concrete slab system. For this case the slab was 10 cm thick and the diffusion length in concrete assumed to be 20 cm. The generation rate was calculated using eq. 1 for concrete used in the construction of the basements (table 5) and assuming an emanation fraction of 0.1. This results in a generation rate of 1.5 kBq m⁻³ h⁻¹. The indoor (C_i) and subslab (C_s) concentrations were 1.5 and 7.4 kBq m⁻³ respectively.

The radon flux into the room is proportional to the slope of the concentration at the boundary (Z = 10 cm). Note that the concentration gradient at the boundary due to radon originating in the concrete (dashed line) is greater than that due to radon originating outside the concrete (dotted line). This means

Fig. 17. Radon flux density (A) and subslab radon concentration (B) for a 14 d period. The floor-wall joint was closed on day 4.

Fig. 18. Theoretical steady-state concentration profile for radon in a concrete slab system. The radon generation rate in the concrete was $1.5 \text{ kBq m}^{-3} \text{ h}^{-1}$ and the concentration outside the concrete was $7.4 \text{ kBq m}^{-3} (C_s)$ and $1.5 \text{ kBq m}^{-3} (C_l)$.

that the actual flux would be greater than would be expected based only on the radon concentrations on either side of the concrete. This might lead to an overestimate of the importance of convective flow. Also, the diffusion length of radon in concrete would be overestimated based only on measurements of the flux and concentration on either side of the slab and ignoring radon originating in the concrete.

The diffusive flux of radon into the structure is proportional to the concentration gradient at 10 cm. The calculated flux as a function of the subslab concentration (C_s) is shown in Fig. 19. For this case the indoor concentration (C_i) was assumed to remain constant at 1.5 kBq m^{-3} . When C_i is greater than C_s the flux from radon originating outside the concrete will be into the concrete slab. This is referred to as back diffusion. However, since the flux from radon originating in the concrete is independent of the concentration on either side of the slab and is greater than the back diffusion the total flux will be into the room. As the thickness of the slab increases the flux due to radon originating in the concrete increases and the dependence on the concentration on the opposing side decreases. This is illustrated in fig. 19. The slope of the flux as a function of C_s decreases as the thickness of the slab is increased from 10 to 15 cm. This is a partial explanation why the measured fluxes below ground are similar. Another possibility is the possibility of a region of fairly uniform radon concentration immediately near the exterior surface of the basement.

Fig. 19. Theoretical radon flux from 10 and 15 cm thick concrete slabs as a function of the subsurface radon concentration (C_s). The flux due to radon originating inside the concrete is independent of the radon concentration outside the slab. The radon generation rate was $1.5 \text{ kBq m}^{-3} \text{ h}^{-1}$ and the concentration inside the structure was assumed to be a constant 1.5 kBq m^{-3} .

Partitioning of Radon Origins and Entry Pathways

Since the greatest indoor ^{222}Rn concentrations result from the persistent radon entry rate it was partitioned according to the origin of the radon and its entry pathway. First it was necessary to determine if the radon entry rate determined from the measured flux densities could account for persistent radon entry rate. Once this had been accomplished the relative contributions of the entering radon that originated in the soil and concrete and entered through the floor, walls and floor-wall joint could be calculated.

The first step was to determine the entry rate due to radon passing through the floor-wall joint. This is not simply the change in the global entry rate with and without the floor-wall joint sealed since the flux through the floor changes as a function of the subslab radon concentration (fig. 17). The mean entry rate through the floor-wall joint was calculated using the following relations:

$$E_o = E_{WAG} + E_{WBG} + E_{Fo} + E_{FWJ} \quad (20)$$

and

$$E_c = E_{WAG} + E_{WBG} + E_{Fc} \quad (21)$$

where:

E_o = average global radon entry rate, measured using the trace gas system,
with the floor-wall joint open (Bq h^{-1}).

E_{WAG} = average radon entry rate through the concrete walls above ground level
(Bq h⁻¹).

E_{WBG} = average radon entry rate from radon entering through the concrete
walls below ground level (Bq h⁻¹).

E_{Fo} = average radon entry rate from radon entering through the concrete floor
with the floor-wall joint open (Bq h⁻¹).

E_{FWJ} = average radon entry rate from radon entering through the floor-wall joint
(Bq h⁻¹).

E_c = average global radon entry rate, measured using the trace gas system,
with the floor-wall joint closed (Bq h⁻¹).

E_{Fc} = average entry rate from radon entering through the concrete floor with
the floor-wall joint closed (Bq h⁻¹).

Subtracting eq. 20 from eq. 21 and solving for E_{FWJ} gives:

$$E_{FWJ} = E_o - E_c - E_{Fo} + E_{Fc} \quad (22)$$

Table 7 summarizes the flux densities and their contributions to the global radon entry rates. Based in the flux data, the global radon entry rates were 840 ± 30 Bq h⁻¹ (eqn. 20) with the floor-wall joint closed and 1000 ± 200 Bq h⁻¹ (eqn. 21) with the floor-wall joint open. These values are not significantly different from the mean of the global entry rate readings measured by the trace gas system ($Z = 1.26$, $p = 0.052$ and $Z = 0.64$, $p = 0.13$ respectively).

Table 7. ^{222}Rn flux densities and entry rates. Uncertainties represent 1 standard deviation of the readings. Global refers to measurements carried out with the trace gas system.

Location	Flux Density (Bq m ⁻² h ⁻¹)	Surface Area (m ²)	Entry Rate (Bq h ⁻¹)
Global (E _o) (Open) ^a			850 ± 120
Global (E _c) (Closed) ^b			660 ± 140
Wall Above Ground	7 ± 2	4	30 ± 10
Wall Below Ground	22 ± 1	27	620 ± 30
Floor (Open) ^a	11 ± 1	11	120 ± 10
Floor (Closed) ^b	18 ± 2	11	200 ± 20
Floor-Wall Joint			260 ± 180

^a Floor-wall joint open.

^b Floor-wall joint closed.

Assuming that diffusion is the only transport mechanism, a first-order approximation of the width of the floor-wall joint was made using eq. 23. It was assumed that the joint was filled with air ($D_{\text{air}} = 1 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$), the diffusion length of radon in the floor concrete was 0.3 m ($D_c = 2.5 \times 10^{-8} \text{ m}^2 \text{ s}^{-1}$) and the tortuosity of the joint was 1. The floor area is 11 m² and the area of the floor wall joint (A_{FWJ}) is 13.4 m times the width (w). The estimated width is 4.5 mm.

$$\frac{E_{\text{FWJ}}}{E_{\text{Fo}}} = \frac{D_{\text{air}} \frac{\Delta C}{\Delta Z} A_{\text{FWJ}}}{D_c \frac{\Delta C}{\Delta Z} A_F} \quad (23)$$

This is greater than seems reasonable since the floor was poured directly on top of the footers. This suggests that there maybe a flow of gas between the room and the subslab airspace. However, the pressure differentials between these 2 regions were not detectable (< 0.2 Pa). A possible explanation is that an air current loop from the room, through the subslab volume and returning to the room exists. This flow may be driven by uneven solar heating of the structure and the directional component of wind velocity.

If all of the entry through the floor-wall joint is due to convection then the airflow rate through the joint would be 0.67 l min^{-1} . This corresponds to a flow of 0.05 l min^{-1} per 1 m of the floor-wall joint. It has not been possible to measure this very small flow. However, qualitative measurements conducted using smoke tubes have shown that there is a flow into the joint along some sections, while the flow is out of the joint along other portions. This supports the existence of an air current loop through the room and subslab airspace.

The radon entry was further separated into the radon which originated in the concrete and that which originated in the soil. The contribution of radon originating in the wall concrete to the persistent radon entry was measured by the accumulator plate on the above ground portion of the wall. However, there was no such direct measurement available to determine how much, if any, of the radon entering through the floor originated in the concrete. To determine this the flux from the floor was plotted as a function of the subslab radon concentration (fig. 20). The intercept of a linear regression applied to this data

Fig. 20. Measured radon flux density from the floor at 2 locations as functions of the subslab radon concentration. The error bars represent 1 standard deviation of the readings. Uncertainties on the intercepts are 95 % confidence limits. The number of data points at each concentration range from 60 to 900.

represents the flux with zero subslab radon concentration. This is the best estimate of the flux due to radon originating in the floor concrete. The intercept ($6 \pm 0.6 \text{ Bq m}^{-2} \text{ h}^{-1}$) was the same for both floor locations and was not significantly different from the flux density measured from the above ground portion of the wall ($Z = 0.49$, $p = 0.16$). Table 8 shows the relative contributions of the origins and entry pathways to the global ^{222}Rn entry rate with the floor-wall joint open. It is important to note that 29% of the radon is generated in the concrete. Based on an average ventilation rate of 0.03 h^{-1} , the concrete alone would account for an indoor concentration of 430 Bq m^{-3} .

Table 8. Relative contributions of the origins and entry pathways to the global radon entry rate.

Entry pathway	Origin of radon (%)	
	Soil	Concrete
Floor-wall joint	25	0
Floor	6	7
Walls	40	22
Total	71	29

Because of the small size of the structures and their high surface-to-volume ratio (1.8:1) these results must be scaled in order to compare them with other studies. A hypothetical basement structure with a floor area of 100 m^2 (6.3 m x 16 m) and 2.3 m ceilings was used for these purposes. The surface-to-volume ratio for this basement is 0.9:1. Like the experimental structures the walls are 15 cm thick concrete with a 30 cm section above ground level. The floor slab is 10 cm thick and lies on a gravel layer. The ventilation rate used for these

calculations was 0.2 h^{-1} . The global entry rate was calculated using the fluxes measured in the experimental basement and scaled by the appropriate surface area. The contributions to the indoor concentration were calculated using:

$$C_{Rn} = \frac{E_o}{\lambda_v V} \quad (24)$$

The results of the calculations are contained in table 9. For this case the steady-state radon concentration would be 90 Bq m^{-3} (2.4 pCi l^{-1}) 33% of which originated in the concrete floor and walls. The median annual radon concentration measured in U.S. basements is 76 Bq m^{-3} (USEPA 1992). This indicates that, for dwellings with radon concentrations near the median, molecular diffusion and building materials are significant contributors to the radon entry and cannot be ignored.

Table 9. Radon entry results scaled to a $6.3 \text{ m} \times 16 \text{ m} \times 2.3 \text{ m}$ basement with a ventilation rate of 0.2 h^{-1} . The area associated with the floor-wall joint has units of length (m).

Origin and pathway	Flux ($\text{Bq m}^{-2} \text{ h}^{-1}$)	Area (m^2)	Entry Rate (Bq h^{-1})	C_{Rn} (Bq m^{-3})
Concrete	6.5	203	1300	30
Wall	15	90	1350	30
Soil gas				
Floor	5	100	500	10
Soil gas				
Floor-wall Joint	19	45	860	20
Total			4010	90

Radon Generation and Diffusion in Concrete

In order to characterize the generation and diffusion of ^{222}Rn in concrete it is necessary to know the ^{226}Ra content, density, porosity and emanation fraction of the concrete and the diffusion coefficient for radon in concrete. Of these the radium content, density and porosity are easily and accurately measured in the laboratory. Laboratory measurements of the emanation and diffusion coefficients are much more uncertain. This is because the concrete encountered in a basement setting has been subjected to environmental conditions (temperature and moisture) loading that is not typically applied to laboratory samples. Also, typically much greater care is taken in the preparation of laboratory samples. For these reasons, in-situ measurements of the emanation fraction and diffusion coefficient are the best method for characterizing the properties of the concrete.

Taking advantage of the fact that the flux from the floor changes with the subslab radon concentration an in-situ determination of the emanation fraction and diffusion coefficient was made at 2 locations on the floor. The measured flux density (J), indoor (C_I) and subslab (C_s) radon concentrations were fitted to eqn. 19 using a Marquardt-Levenberg algorithm. This method minimizes the sum of the square of the differences between the data and the equation. The fitted parameters were the diffusion length (l_c) and the emanation fraction (F). Equation 19 can also be written as a linear combination of the indoor and outdoor radon concentration. A linear regression can be used to estimate the

intercept and coefficients. The diffusion length and emanation fraction can then be expressed terms of the estimated parameters. This was investigated as an alternative to performing a non-linear regression. Both procedures properly determined the diffusion length and emanation fraction for an "ideal" data set. However, when there is uncertainty in the flux values (ie. from counting statistics) the linear regression results deviated from the true values. Details of this analysis is contained in appendix B.

The best estimates of the diffusion length and emanation fraction, determined using the non-linear regression, are contained in table 10. The fitted values for the emanation fraction and diffusion length are significantly different at the 2 locations. There are several possible reasons for this difference. One is the number of micro-cracks existing under the plates. A greater number of cracks would result in a larger diffusion coefficient. Another possibility is the thickness of the concrete slab.

Table 10. Emanation fraction (F), bulk diffusion coefficient (D_e) and diffusion length (l_c) for ^{222}Rn in concrete. Measurements were taken at 2 locations in the west structure. Uncertainties represent 95% confidence intervals.

	Northwest Corner	Northeast Corner
F	0.172 ± 0.007	0.209 ± 0.002
D_e ($\text{cm}^2 \text{ s}^{-1}$)	2.5×10^{-4}	1.5×10^{-4}
l_c (m)	0.302 ± 0.003	0.232 ± 0.001
N	1228	5766
R^2	0.66	0.57

Fig. 21 shows the results of a theoretical calculation of the dependence of the diffusion length on slab thickness and emanation fraction. Values were calculated by determining the diffusion length necessary to maintain a flux of $10 \text{ Bq m}^{-2} \text{ h}^{-1}$ for a given slab thickness and emanation fraction. The ^{226}Ra content, porosity and bulk density of the concrete were as given in table 5. The diffusion length changes rapidly for emanation fractions between 0.1 and 0.3 and with slab thicknesses between 4 and 10 cm. Fig. 22 shows this relationship for selected slab thicknesses in this range. This indicates that the use of an incorrect slab thickness or emanation fraction can result in errors in the estimated diffusion length of up to an order of magnitude.

Measurements of the flux density and radon concentrations have been used to determine the diffusion coefficient and porosity (Zapalac 1983). Fig. 23 shows the relationship between the slab thickness, porosity and diffusion length. The emanation fraction used in these calculations was 0.2. For porosities greater than 0.1 the porosity has little effect on the calculated diffusion length over a large range of concrete thicknesses. At a porosity of 0.1 the calculated diffusion length changes by approximately a factor of 2 between thicknesses of 4 and 15 cm (fig. 24). Therefore, uncertainties in the porosity will not have as great an effect on the estimated diffusion length as uncertainties in the emanation fraction. This supports the choice of using the curve fit procedure was used to estimate the emanation fraction, rather than the porosity.

Fig. 21. Theoretical relationship between: concrete slab thickness, emanation fraction and the calculated diffusion length of ^{222}Rn in concrete. The diffusion length was calculated using eq. 19. The properties of the concrete are listed in table 5. The radon concentrations C_1 and C_s were 1.1 and 4.0 kBq m^{-3} respectively and the flux was assumed to be $10 \text{ Bq m}^{-2} \text{ m}^{-1}$.

Fig. 22. Relationship between the calculated diffusion length and emanation fraction for slab thicknesses of 4, 8, 10, 12 and 15 cm. Note that the calculated diffusion length changes rapidly for emanation fractions between 0.1 and 0.3.

Fig. 23. Theoretical relationship between: the thickness of a concrete slab, porosity and the calculated diffusion length for radon in concrete. Diffusion lengths were calculated using the same method as for fig. 21 except that the emanation fraction was assumed to be 0.2.

Fig. 24. Relationship between the calculated diffusion length and concrete porosity for 4, 8, 10, 12 and 15 cm thick slabs. The calculated diffusion length is relatively insensitive to the porosity for porosities greater than 0.1.

V. Conclusions

Experiments conducted in 2 unique basement structures have been used to investigate the origins of indoor ^{222}Rn , the entry pathways and the diffusion of radon through concrete. The basements were constructed using techniques and materials typical to the Colorado front range. A computer controlled data acquisition system was used to continuously measure: indoor, outdoor and subslab radon concentrations, building ventilation, pressure differentials and meteorological parameters. Also, a flow-through accumulator plate system was used to continuously measure the radon flux density through portions of the floor and walls. Using the measured indoor radon concentration and ventilation rate it was possible to determine the global radon entry rate as a function of time. The global radon entry rate is the input rate of radon into the structure from all sources except outdoor air.

The global radon entry rate into the basement structures has persistent, non-zero component and a time varying component. The persistent radon entry does not vanish even when the structure is slightly over pressurized. This indicates that diffusion is an important transport mechanism. It is during calm periods, when the persistent entry is dominant, that result in the greatest indoor radon concentrations. For this reason this entry was partitioned according to the origins of the radon and the entry pathways. Possible origins of the radon are in

the concrete floor and walls and in the soil. Entry into the basements occurs through the concrete floor and walls and through the floor-wall joint. Radon entry through the floor-wall joint was controlled by a baseboard barrier system. Results indicated that only 25 % of the radon entry during calm conditions occurs through the floor-wall joint. Entry through the floor-wall joint was a combination of diffusion and an air current through the room and subslab airspace. It was not possible to separate the portions of the entry through the joint which were due to diffusion and convection since the actual width of the joint and the airflow were not known. It may be possible to measure the transfer of air from the subslab volume to the room through the use of a second trace gas injected into the subslab airspace or with very sensitive flowmeters connected to the baseboard barrier system. The remaining 75 % of the entry takes place through the concrete floor and walls. Of the radon which enters through the floor and walls, 40 % (30 % of the total entry) originated in the concrete. Scaling these results to a hypothetical basement with a 100 m² floor area and a ventilation rate of 0.2 h⁻¹ resulted in a steady state radon concentration, from radon diffusing through the floor, walls and floor-wall joint, of 90 Bq m⁻³. This is slightly greater than the U.S. median basement radon concentration (74 Bq m⁻³). Approximately 33 % of the radon entering into the hypothetical basement originated in the concrete floor and walls. This indicates that, for houses with basement radon concentrations near the U.S. average, the building materials and molecular diffusion cannot be ignored as sources of radon entry.

The accumulator plate system was also used to make in-situ measurements of the diffusion length and emanation fraction for radon in concrete. This method has the advantage of measuring a large area of concrete which has been exposed to temperature, moisture and loading conditions which would be experienced in a typical house. This is rarely true for samples used in laboratory measurements. The diffusion length and emanation fraction were estimated by fitting the measured floor flux density, subslab and indoor radon concentrations to the steady-state solution to Fick's law for diffusion from a concrete slab with known concentrations on either side. The average diffusion length and emanation fraction were 27 ± 4 cm and 0.19 ± 0.02 respectively. A theoretical analysis indicated that the diffusion length, estimated using this procedure, was strongly dependent on the thickness of the concrete slab and the emanation fraction. The porosity had little effect on the estimate.

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Appendix A

Derivation of Equation For Flux From a Slab

The solution for 1-dimensional diffusive transport of ^{222}Rn through a concrete slab was derived using the notation used by Coll et al. (1991). The defining equations are:

$$J = -D_e \frac{\partial C_c}{\partial Z} \quad (\text{A-1})$$

and

$$\frac{\partial C_c}{\partial t} = \frac{D_e}{\epsilon} \frac{\partial^2 C_c}{\partial Z^2} - \lambda C_c + \phi_c \quad (\text{A-2})$$

where:

D_e = Bulk diffusion coefficient ($\text{m}^2 \text{s}^{-1}$).

C_c = ^{222}Rn concentration in the concrete pore spaces (Bq m^3).

ϵ = porosity of concrete.

λ = decay constant for $^{222}\text{Rn} = 0.00756 \text{ h}^{-1}$.

ϕ_c = ^{222}Rn production rate in the concrete pore spaces ($\text{Bq m}^{-3} \text{ h}^{-1}$).

The bulk diffusion coefficient (D_e) relates to the flux density across the geometric area of the surface. Flux density across the pore area can be determined using the relation:

$$J_p = -D_e^* \frac{\partial C_e}{\partial Z} \quad (\text{A-3})$$

The pore diffusion coefficient (D_e^*) is related to the bulk diffusion coefficient by:

$$D_e = \epsilon D_e^* \quad (\text{A-4})$$

Production of ^{222}Rn in the pore spaces can be calculated based on the ^{226}Ra concentration (Ra), bulk density (ρ_b), emanation fraction (F), ^{222}Rn decay constant (λ) and the porosity (ϵ).

$$\phi_c = \frac{Ra \rho_b F \lambda}{\epsilon} \quad (\text{A-5})$$

Equations A-1 and A-2 were solved for a concrete slab, of thickness L, with known radon concentrations (C_i and C_s) on either side. Boundary conditions were:

$$C(Z = 0) = C_s \quad (\text{A-6})$$

and

$$C(Z = L) = C_I \quad (\text{A-7})$$

At steady state eq. A-2 becomes:

$$\frac{\partial C_c}{\partial t} = 0 = \frac{D_e}{\epsilon} \frac{\partial^2 C_c}{\partial Z^2} - \lambda C_c + \phi_c \quad (\text{A-8})$$

The general solution for the concentration in the concrete is:

$$C_c(Z) = A \sinh\left(\frac{Z}{l_c}\right) + B \sinh\left(\frac{L-Z}{l_c}\right) + \frac{\phi_c}{\lambda} \quad (\text{A-9})$$

The parameter l_c is the diffusion length of radon in concrete and is related to the bulk diffusion coefficient by :

$$D_e = l_c^2 \epsilon \lambda \quad (\text{A-10})$$

Applying the boundary conditions A-6 and A-7 gives:

$$A = \frac{C_I - \frac{\phi_c}{\lambda}}{\sinh\left(\frac{L}{l_c}\right)} \quad (\text{A-11})$$

and

$$B = \frac{C_s - \frac{\phi}{\lambda}}{\sinh\left(\frac{L}{l_c}\right)} \quad (\text{A-12})$$

Equation A-9 becomes:

$$C_c(z) = \frac{C_1 \sinh\left(\frac{z}{l_c}\right)}{\sinh\left(\frac{L}{l_c}\right)} + \frac{C_s \sinh\left(\frac{L-z}{l_c}\right)}{\sinh\left(\frac{L}{l_c}\right)} - \frac{\phi_c}{\lambda} \left[\frac{\sinh\left(\frac{L-z}{l_c}\right) + \sinh\left(\frac{z}{l_c}\right)}{\sinh\left(\frac{L}{l_c}\right)} - 1 \right] \quad (\text{A-13})$$

Taking the first derivative of eq. A-13 with respect to Z gives:

$$\frac{\partial C_c}{\partial z} = \frac{C_1 \cosh\left(\frac{z}{l_c}\right)}{l_c \sinh\left(\frac{L}{l_c}\right)} - \frac{C_s \cosh\left(\frac{L-z}{l_c}\right)}{l_c \sinh\left(\frac{L}{l_c}\right)} - \frac{\phi_c}{\lambda} \left[\frac{\cosh\left(\frac{z}{l_c}\right) - \cosh\left(\frac{L-z}{l_c}\right)}{l_c \sinh\left(\frac{L}{l_c}\right)} \right] \quad (\text{A-14})$$

Evaluating eq. A-14 at $Z=L$ and applying eq. A-1 gives the equation for the flux into the interior of the structure from the slab (eq. A-15).

$$J(L) = -D_s \left[\frac{\partial C_s}{\partial z} \right]_{z=L} = D_s \left[\frac{\phi}{\lambda} \left(\frac{\cosh\left(\frac{L}{l_c}\right) - 1}{l_c \sinh\left(\frac{L}{l_c}\right)} \right) - \frac{C_l \cosh\left(\frac{L}{l_c}\right)}{l_c \sinh\left(\frac{L}{l_c}\right)} + \frac{C_s}{l_c \sinh\left(\frac{L}{l_c}\right)} \right] \quad (\text{A-15})$$

Since $D_s = l_c^2 \epsilon \lambda$ (eq. A-10) the flux can be expressed as:

$$J(L) = l_c \epsilon \phi_c \left(\frac{\cosh\left(\frac{L}{l_c}\right) - 1}{\sinh\left(\frac{L}{l_c}\right)} \right) - \frac{l_c \epsilon \lambda C_l \cosh\left(\frac{L}{l_c}\right)}{\sinh\left(\frac{L}{l_c}\right)} + \frac{l_c \epsilon \lambda C_s}{\sinh\left(\frac{L}{l_c}\right)} \quad (\text{A-16})$$

Appendix B

Comparison of Linear and Non-linear Least Squares Fitting Techniques

The use of the non-linear regression to determine the emanation fraction and diffusion length of radon in concrete requires special curve fitting algorithms. These can be difficult to use and, since an iterative procedure is used, may require large amounts of computer time. However, the equation used in the curve fit can be written as a linear combination of the indoor and subslab radon concentrations. The coefficients of the model can easily be computed using the linear regression function found in many spreadsheet packages. The emanation fraction and the diffusion length can then be expressed as functions of the coefficients. Since this may provide a simple alternative to the use of a non-linear regression, the two methods were compared for the ability to accurately calculate the diffusion length and emanation fraction.

Assuming that diffusion is the only transport mechanism the equation for the 1-dimensional, steady-state flux from one side of a concrete slab is:

$$J(L) = l_c \epsilon \phi_c \left(\frac{\cosh\left(\frac{L}{l_c}\right) - 1}{\sinh\left(\frac{L}{l_c}\right)} \right) - \frac{l_c \epsilon \lambda C_i \cosh\left(\frac{L}{l_c}\right)}{\sinh\left(\frac{L}{l_c}\right)} + \frac{l_c \epsilon \lambda C_s}{\sinh\left(\frac{L}{l_c}\right)} \quad (\text{B-1})$$

where:

J = radon flux density ($\text{Ba m}^{-2} \text{ h}^{-1}$).

l_c = diffusion length of radon in concrete (m).

ϵ = porosity of concrete.

L = thickness of concrete slab (m).

λ = decay constant for radon = 0.00756 h^{-1} .

C_i = indoor radon concentration (Bq m^{-3}).

C_s = subslab radon concentration (Bq m^{-3}).

The production rate (ϕ_c) is:

$$\phi_c = \frac{Ra \rho_b F \lambda}{\epsilon} \quad (\text{B-2})$$

where:

Ra = ^{226}Ra concentration in concrete (Bq kg^{-1}).

ρ_b = bulk density of concrete (kg m^{-3}).

F = ^{222}Rn emanation fraction.

Equation B-1 can be re-written as:

$$J(L) = \alpha - \beta C_i + \gamma C_s \quad (\text{B-3})$$

where:

$$\alpha = l_c \epsilon \phi_c \left(\frac{\cosh\left(\frac{L}{l_c}\right) - 1}{\sinh\left(\frac{L}{l_c}\right)} \right) \quad (\text{B-4})$$

$$\beta = \frac{l_c \epsilon \lambda \cosh\left(\frac{L}{l_c}\right)}{\sinh\left(\frac{L}{l_c}\right)} \quad (\text{B-5})$$

$$\gamma = \frac{l_c \epsilon \lambda}{\sinh\left(\frac{L}{l_c}\right)} \quad (\text{B-6})$$

The coefficients α , β and γ can be determined by performing a linear regression on the flux as a function of the indoor and subslab radon concentrations.

The production rate of radon in the concrete (ϕ_c) and the diffusion length (l_c), written in terms of α , β and γ , are:

$$\phi_c = \frac{\alpha \lambda}{\beta - \gamma} \quad (\text{B-7})$$

and

$$l_c = \frac{L}{\cosh^{-1}\left(\frac{\beta}{\gamma}\right)} \quad (\text{B-8})$$

The emanation fraction is then:

$$F = \frac{\phi_c \epsilon}{Ra \rho_b \lambda} = \left(\frac{\alpha}{\beta - \gamma} \right) \frac{\epsilon}{Ra \rho_b} \quad (\text{B-9})$$

The linear and non-linear fitting techniques were compared by simulating the flux using eqn. B-1 and randomly selected indoor and subslab radon concentrations. The concrete slab was 8 cm thick. Two data sets were generated for the "ideal" case where the flux is exactly equal to that calculated by eqn B-1. For one set the diffusion length and emanation fraction were 10 cm and 0.02 respectively. The parameters for the other set were $l_c = 30$ cm and $F = 0.2$. To simulate the affect that counting statistics would a uniform random variation of ± 20 % and ± 50 % was applied to the flux data. Both regression procedures were performed on each of the data sets. Table B-1 contains the estimates of the diffusion length and emanation fraction based on the 2 techniques. For some simulations it was not possible to calculate the diffusion length because the ratio of β to γ was less than 1 where the hyperbolic cosine does not exist.

Table B-1. Results of linear and non-linear regression techniques.

		Linear	Non-Linear
$l_c = 0.1 \text{ m}; F = 0.02$	l_c	0.100	0.100
	F	0.020	0.020
Variation = 20%	l_c	0.111	0.100
	F	0.025	0.020
Variation = 50%	l_c	0.060	0.100
	F	0.009	0.020
$l_c = 0.3 \text{ m}; F = 0.20$	l_c	0.300	0.300
	F	0.200	0.200
Variation = 20%	l_c	0.164	0.300
	F	0.060	0.199
Variation = 50%	l_c	0.165	0.300
	F	0.060	0.195

In both cases either method was able to accurately estimate the diffusion length and emanation fraction for the ideal data. However, when the flux varied from the ideal case only the non-linear technique was still able to calculate the actual values. This may be due to the ratio of β to γ approaching unity as the diffusion length increases (fig. B-1). Uncertainties in β and γ , created by deviations from the ideal case, can result in a large uncertainty in the ratio. This means that the uncertainty in the estimate of the diffusion length and emanation fraction will also be large. In general, the linear regression method can be used to predict F and l_c for cases where the diffusion length is less than 10 cm where the ratio of β to γ is much greater than 1. The non-linear regression was able to accurately estimate the diffusion length and emanation fraction in all cases.

Fig. B-1. Ratio of linear regression coefficients β and γ as a function of the diffusion length of radon in concrete. The ratio approaches 1 as the diffusion length increases.

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