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Using $^{220}Rn/^{222}Rn$ ratios as indicators of soil gas transport

Hutter, Adam Richard, Ph.D.

City University of New York, 1994

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USING 220RN/222RN RATIOS AS INDICATORS OF SOIL GAS TRANSPORT

by

ADAM R. HUTTER

A dissertation submitted to the Graduate Faculty in Earth and Environmental Sciences in partial fulfillment of the requirements for the degree of Doctorate of Philosophy, The City University of New York

1994

This manuscript has been read and accepted for the Graduate Faculty in Earth and Environmental Sciences in satisfaction of the dissertation requirement for the degree of Doctor of Philosophy.

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Abstract

USING 220RN/222RN RATIOS AS INDICATORS OF SOIL GAS TRANSPORT

by

ADAM R. HUTTER

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$^{220}Rn/^{222}Rn$ ratios were used to address changing soil gas transport mode and migration distance as a cause and control of soil gas ^{222}Rn concentration variations. First, a method to accurately and precisely measure soil gas ^{220}Rn was developed. The lowest ^{220}Rn concentration that can be measured using this method while maintaining an uncertainty (1σ) of about 30% is approximately 500 Bq m^{-3} (13 pCi l^{-1}). The uncertainty can be decreased to about 20% if multiple samples are obtained in series and an arithmetic mean reported. At typical soil gas concentrations, the 1σ single-measurement error is approximately 20%. Bi-weekly ^{220}Rn, ^{222}Rn, and permeability measurements were obtained at sites in Chester and Matawan, NJ for a period of two years. At a third site, located in Cheesequake State Park, NJ, these measurements were obtained on a bi-weekly basis whenever possible. Soil gas ^{222}Rn concentrations at the Chester, NJ site were highest during September/October and up to 10X early-winter lows, variations too large to be explained by a diffusion-only equation. At the other two sites, no seasonal ^{222}Rn variations were observed. Spatial variations in ^{222}Rn and ^{220}Rn concentrations up to 10 times over distances on the order of 10 to 12 m were observed. Soil gas ^{220}Rn concentrations at the Chester, NJ site were highest during mid-summer, with lowest concentrations occurring during mid-winter, as described according to a diffusion-only equation. At the other two sites, no temporal trends in the ^{220}Rn soil gas concentration were observed. Permeability measurements, thought to be an indicator of soil parameters

controlling soil gas ^{222}Rn variations, showed no correlations with ^{222}Rn, ^{220}Rn, or $^{220}Rn/^{222}Rn$ ratios. The $^{220}Rn/^{222}Rn$ ratios at the Chester site showed a strong dependence on ^{222}Rn only, whereas at the Matawan site, the $^{220}Rn/^{222}Rn$ ratios were controlled by ^{220}Rn, indicating longer soil gas migration at the Chester site. The data indicate that changing transport mode may be responsible for unexpectedly large seasonal ^{222}Rn concentrations, thus providing information on transport distances of soil gas, a critical factor in many environmental concerns.

Acknowledgments

I have so many people to thank for helping me reach this milestone, it is obvious to me that the adage "no man is an island" is as close to being a truth as is possible, despite as much as I sometimes may seem to aspire to the contrary. I have had help both scientifically and personally from many people, and I am most appreciative of those that fit both categories. My thesis advisor, Dr. Anthony Prave, has helped in building and enjoying a true passion for science. More than any specific tidbit of information or bureaucratic guidance offered, it is this quality that I will carry forever and that I am most grateful to him for. Drs. John Wehmiller of the University of Delaware and Arthur W. Rose of The Pennsylvania State University, previous academic advisors, are also thanked for sparking and developing my interest in the geosciences in general, and geochemistry specifically, through their enthusiasm, knowledge, and teaching abilities. Without their considerable input to my academic progress, I certainly would not have pursued further advancement.

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"The fewer the words, the greater the prophet."

Anonymous

"Imagination is more important than knowledge."

Albert Einstein

"All men want, not something to do with, but something to do, or rather, something to be."

Henry David Thoreau, Walden

TABLE OF CONTENTS

ABSTRACT iii

ACKNOWLEDGMENTS v

LIST OF TABLES x

LIST OF FIGURES xii

Chapter 1 INTRODUCTION 1

Chapter 2 A SUMMARY OF ^{222}RN GENERATION, CONCENTRATION AND MIGRATION IN SOIL GAS 7

Chapter 3 METHODS, SITE DESCRIPTIONS AND QUALITY ASSURANCE 27
31 Soil Gas ^{222}Rn and ^{220}Rn 27
32 Field Procedures 40
33 Subsurface Characterization 42
34 Permeability 45
35 Site Descriptions 45
36 Quality Assurance 49

Chapter 4 DATA PRESENTATION 51
41 Site 1 Soil Gas ^{222}Rn and ^{220}Rn 51
42 Site 2 Soil Gas ^{222}Rn and ^{220}Rn 62
43 Site 3 Soil Gas ^{222}Rn and ^{220}Rn 70
44 ^{220}Rn/^{222}Rn Ratios 73

Chapter 5 DATA INTERPRETATION AND DISCUSSION 77
51 ^{222}Rn and ^{220}Rn 77
52 ^{220}Rn/^{222}Rn Ratios 84

Chapter 6 SUMMARY, CONCLUSIONS AND RECOMMENDATIONS 102
61 Summary of Investigation 102
62 Summary of Results 103
63 Conclusions 104
64 Recommendations 108

APPENDIX A ^{222}RN AND ^{220}RN CALCULATION PROGRAM 109

APPENDIX B DATA TABLES 121
Site 1 122

Site 2 131
Site 3 134

APPENDIX C QUALITY ASSURANCE DATA 135

APPENDIX D SAMPLE CALCULATION FOR 222RN AND 220RN 141

BIBLIOGRAPHY 143

LIST OF TABLES

Table 3.1 Results of ^{220}Rn method determination tests in the EML Radon, Thoron and Progeny Exposure Facility 37

Table 5.1 Range of $^{220}Rn/^{222}Rn$ ratio for Site 1 88

Table 5.2 Range of $^{220}Rn/^{222}Rn$ ratios for Sites 2 and 3 89

Table 5.3 ^{222}Rn correlation coefficients (r) between sampling locations (hole #) at Site 1 91

Table 5.4 ^{220}Rn correlation coefficients (r) between sampling locations (hole #) for Site 1 91

Table 5.5 $^{220}Rn/^{222}Rn$ ratio correlation coefficients (r) between sampling locations (hole #) for Site 1 91

Table 5.6 ^{222}Rn correlation coefficients (r) between sampling locations (hole #) at Site 2 92

Table 5.7 ^{220}Rn correlation coefficients (r) between sampling locations (Hole #) at Site 2 92

Table 5.8 $^{220}Rn/^{222}Rn$ ratio correlation coefficients (r) between sampling locations (Hole #) at Site 2 92

Table 5.9 Site 1 correlation coefficients (r) by sampling location (hole #) 95

Table 5.10 Site 2 correlation coefficients (r) by sampling location (hole #) 96

Table 5.11 Site 3 correlation coefficients (r) by sampling location (hole #) 97

Table B.1 Site 1 Data: Meteorological Data 122

Table B.2 Site 1 Data 123

Table B.3 Site 2 Data 131

Table B.4 Site 3 Data 134

Table C.1 222Rn duplicate measurements data 135

Table C.2 220Rn duplicate measurements data 138

Table C.3 Calibration data 140

LIST OF FIGURES

Figure 2.1 ^{238}U decay series 8

Figure 2.2 ^{232}Th decay series 9

Figure 2.3 ^{222}Rn concentration profile with depth calculated using equation (10) and (12) 20

Figure 2.4 ^{222}Rn concentration profile with depth calculated using equation (11) 21

Figure 3.1 Growth of ^{222}Rn and progeny activity after isolation 30

Figure 3.2 Histogram of measurement errors based on duplicate ^{222}Rn samples 33

Figure 3.3 Histogram of measurement errors based on duplicate ^{220}Rn samples 33

Figure 3.4 Cartoon of the components of an electrical resistivity measurement for the determination of subsurface profile 43

Figure 3.5 New Jersey map showing site locations 46

Figure 3.6 Schematic of Site 1 47

Figure 3.7 Schematic of Site 2 48

Figure 3.8 Schematic of Site 3 49

Figure 4.1 ^{222}Rn data from Site 1 52

Figure 4.2 ^{220}Rn data from Site 1 53

Figure 4.3 ^{222}Rn data from Hole 1, Site 1 54

Figure 4.4 ^{222}Rn data from Hole 2, Site 1 54

Figure 4.5 ^{222}Rn data from Hole 3a, Site 1 55

Figure 4.6 ^{222}Rn data from Hole 3b, Site 1 55

Figure 4.7 ^{222}Rn data from Hole 4a, Site 1 56

Figure 4.8 ^{222}Rn data from Hole 4b, Site 1 56

Figure 4.9 ^{222}Rn data from Hole 5, Site 1 57

Figure 4.10 ^{222}Rn data from Hole 6, Site 1 57

Figure 4.11 ^{220}Rn data from Hole 1, Site 1 58

Figure 4.12 ^{220}Rn data from Hole 2, Site 1 58

Figure 4.13 ^{220}Rn data from Hole 3a, Site 1 59

Figure 4.14 ^{220}Rn data from Hole 3b, Site 1 59

Figure 4.15 ^{220}Rn data from Hole 4a, Site 1 60

Figure 4.16 ^{220}Rn data from Hole 4b, Site 1 60

Figure 4.17 ^{220}Rn data from Hole 5, Site 1 61

Figure 4.18 ^{220}Rn data from Hole 6, Site 1 61

Figure 4.19 ^{222}Rn data from Site 2 63

Figure 4.20 ^{220}Rn data from Site 2 64

Figure 4.21 ^{222}Rn data from Hole 1, Site 2 65

Figure 4.22 ^{222}Rn data from Hole 2, Site 2 65

Figure 4.23 ^{222}Rn data from Hole 3, Site 2 66

Figure 4.24 ^{222}Rn data from Hole 4a, Site 2 66

Figure 4.25 ^{222}Rn data from Hole 4b, Site 2 67

Figure 4.26 ^{220}Rn data from Hole 1, Site 2 67

Figure 4.27 ^{220}Rn data from Hole 2, Site 2 68

Figure 4.28 ^{220}Rn data from Hole 3, Site 2 68

Figure 4.29 ^{220}Rn data from Hole 4a, Site 2 69

Figure 4.30 ^{220}Rn data from Hole 4b, Site 2 69

Figure 4.31 ^{222}Rn data from Site 3 71

Figure 4.32 ^{220}Rn data from Site 3 72

Figure 4.33 ^{220}Rn/^{222}Rn Ratio from Site 1 74

Figure 4.34 ^{220}Rn/^{222}Rn Ratio from Site 2 75

Figure 4.35 ^{220}Rn/^{222}Rn Ratio from Site 3 76

Figure 5.1 Histogram of ^{222}Rn data from Site 1 81

Figure 5.2 Histogram of ^{220}Rn data from Site 1 81

Figure 5.3 Histogram of ^{222}Rn data from Site 2 82

Figure 5.4 Histogram of ^{220}Rn data from Site 2 82

Figure 5.5 Histogram of ^{222}Rn data from Site 3 83

Figure 5.6 Histogram of ^{220}Rn data from Site 3 83

Figure 5.7 Scatterplot of paired ^{222}Rn and ^{220}Rn data from Site 2 86

Figure 5.8 Scatterplot of paired ^{222}Rn and ^{220}Rn data from Site 3 86

Figure 5.9 Scatterplot of paired ^{222}Rn and ^{220}Rn data from Site 1 87

Chapter 1

INTRODUCTION

Radon (Rn) moved from the realm of relative obscurity as a scientific entity to a household word in the mid 1980's following detection of very high levels of this inert radioactive gas in the basement of a house in eastern Pennsylvania. Initial public hysteria, due to the nature of the hazard, namely radiation, and thus, in turn, cancer, slowly evolved over several years into widespread public awareness. Through the last decade, an entire industry has developed for the measurement of indoor Rn and remediation of homes having elevated concentrations which present an unusually high health risk. It has been estimated that approximately 10% (or about 13,000 per year) of all lung cancer deaths in the United States are directly attributable to exposure to Rn and its decay products (Lubin and Boice, 1989). As often happens, science follows public demand, and research directed towards the Rn problem increased. There have been numerous projects examining a myriad of aspects pertaining to this issue, encompassing such diverse disciplines as biology, chemistry, physics and geology, to epidemiology, sociology and even law. However, most scientific studies have focused on the end result, namely, Rn measurements in homes and buildings. Relatively few studies by similarly few researchers have been concerned with the root of the problem, i.e., the source characteristics, which, in the case of Rn, is the soil gas (Nero and Nazaroff, 1984).

In an attempt to address this issue, a project was designed at the U.S. Department of Energy's Environmental Measurements Laboratory (EML) to study the long-term trends of soil gas ^{222}Rn relative to geologic setting, with particular emphasis on fracture traces (these are surface expressions in deeply weathered rock and soil of nearly vertical zones of fractured and faulted bedrock). As part of the initial plan of action, a pilot study was initiated at a site where no known geologic features existed in order to first establish

background levels and dynamics of ^{222}Rn in the soil gas. The site selected was EML's Regional Baseline Station in Chester, NJ operated by EML as part of its mission to study the background dynamics of the environment, i.e., pollutant sources and dispersal, natural radiation background and effects, etc., because it met the geological requirements and is relatively close to EML, thereby simplifying other logistical considerations. The initial data obtained, however, were far from what was expected, with widely variable ^{222}Rn concentrations both temporally and spatially. The results of the Chester, NJ site ^{222}Rn concentrations were so complex it became readily apparent that other, more fundamental questions needed to be addressed before a complicating variable such as a fracture trace affecting the soil gas ^{222}Rn concentrations could be integrated into the study. The problem addressed in this dissertation does just that, to design and focus an investigation to enhance our understanding of the factors influencing the temporal and spatial variations in soil gas ^{222}Rn.

Statement of Problem

The problem addressed in this dissertation is to design and focus an investigation to enhance our understanding of the factors, and their interactions, influencing the temporal and spatial variations in soil gas ^{222}Rn. The issues and hypotheses defined and tested in this study are (described in detail in the following sections):

(1) Do differing geologic settings have quantitative affects on ^{222}Rn concentrations?

(2) Can the relative importance of the numerous variables affecting soil gas transport (e.g., soil moisture, porosity, type, atmospheric conditions, emanation coefficients, etc.) be determined from multi-variate statistical analysis of the data?

(3) Is, as commonly thought, diffusion the dominant mechanism of soil gas transport or are other mechanisms operative thereby causing unexpected variations in ^{222}Rn concentrations?

(4) Can variations in the isotopic ratio $^{220}Rn/^{222}Rn$ over time be used to distinguish between steady-state diffusion-dominant migration of soil gas versus advection-dominant?

(5) Finally, can a predictive model for ^{222}Rn soil gas concentrations be developed from the measurement of an array of time dependent soil, atmospheric, and geologic variables?

The data obtained herein will form the foundation from which the continuing effort of the EML soil gas program, aimed at determining the long term trends of soil gas ^{222}Rn and ^{220}Rn and their prediction, will be based.

Discussion

The variations observed in the soil gas ^{222}Rn concentrations at the Chester, NJ site (hereafter referred to as Site 1) were puzzling without attempting to relate them to other parameters such as soil moisture, soil porosity, emanation coefficient of the soil, soil type, etc. The classic manner in which to proceed to study the importance of these parameters on the soil gas ^{222}Rn concentration would have been to measure all the possible influencing factors and statistically determine their levels of significance. However, the procurement of all the necessary equipment is seldom available to the extent the science dictates. The fact that this investigation was financially limited turned out to be serendipitous, as the lack of funds provided the incentive to develop a new way (described below) of trying to discover the causes and controls of the observed variations.

The half-life of ^{220}Rn (55 sec) is very short when compared to the 3.8 day half-life of ^{222}Rn, which in turn limits the transport distance in the environment, whether the milieu is soil air, the atmosphere, or indoor air. Isotopic ratios are commonly used to provide valuable information in many geochemical investigations (Holland, 1973; Veizer et al., 1980). A hypothesis of using $^{220}Rn/^{222}Rn$ ratios was developed to see if any useful information could be obtained from an analysis of long-term trends to help explain observed ^{222}Rn variations. The reason for using $^{220}Rn/^{222}Rn$ ratios is that, relative to

^{220}Rn, the likelihood exists that ^{222}Rn can be transported significant distances from its source (or its measurement site). If there was a significant flow, i.e., advection, of soil gas from depth (or perhaps laterally), then it is reasonable to assume that the ^{222}Rn concentration would increase reflecting the higher concentration of parent material in the source region, whereas the ^{220}Rn concentration would not, given its much shorter transport distance (currently thought to be approximately 1 cm, Nazaroff, 1992). Conversely, if advective processes were not present, i.e., only diffusive transport was operating, changes in soil gas ^{222}Rn concentration over time would likely reflect localized soil conditions, such as changing soil moisture and temperature (as suggested by Washington and Rose, 1990). If the above is true, then the $^{220}Rn/^{222}Rn$ ratio change over time would reflect the dominant soil gas transport mode, either diffusion or advection, rather than localized soil condition changes, given that those would affect similarly the ^{222}Rn and ^{220}Rn concentrations. Even though the prevailing opinion is that diffusion is the dominant mechanism of transport of soil gas, the variable ^{222}Rn concentrations observed at Site 1 indicated that the normal parameters associated with diffusional transport could not produce such variations and that a change in soil gas transport mode was a plausible explanation. In short, one of the hypotheses to be tested in this dissertation is if constant $^{220}Rn/^{222}Rn$ ratios over time indicate a steady-state diffusion-dominant migration of soil gas, or if changing values represent times when the dominant migration mode varies between diffusion and advection.

Thus, one of the main purposes of this investigation is to determine if $^{220}Rn/^{222}Rn$ ratios can give new information concerning the mode of soil gas transport. There is a dire need for a greater understanding of the transport mechanisms of soil gas (Narasimhan, et al., 1990), not only in relation to ^{222}Rn, but also to the migration of other pollutants (Dorr, 1990), many relevant to the U.S.D.O.E.'s past and present activities. However, in order to obtain a record of the $^{220}Rn/^{222}Rn$ soil gas ratios over time, a quick, accurate and precise method of measuring soil gas ^{220}Rn was developed for

this dissertation work. As is detailed herein, the development of a reliable ^{220}Rn soil gas measurement is an important contribution to the radon research community. Additionally, there are very few reported studies in which the long term trends of soil gas ^{222}Rn and ^{220}Rn have been investigated, so this study fills that void. Finally, in an attempt to investigate regional geologic factors and their possible influence on soil gas ^{222}Rn and ^{220}Rn, data were collected from two sites in very different geologic settings.

This dissertation is a product of trying to understand local baseline ^{222}Rn and ^{220}Rn concentrations as a precursor to future studies at the Chester, NJ site as well as other sites in the EML soil gas program. Consequently, certain constraints were placed on data collection for this dissertation. For example, it would have been scientifically prudent to collect soil samples from the bottom of all sampling holes in order to integrate the magnitude of the parent material concentration into the ^{222}Rn concentration analyses. However, some of these same holes are currently being used in new studies, so that destruction of the sampling holes for soil analysis can not be undertaken until these studies are completed. Fortunately, not having parent concentrations from all sites is not that problematic because the current study focuses on temporal variations and attempts to explain their causes. The possibility, and sometimes probability, of parent material inhomogeneity was taken into consideration when spatial variations were detailed, as noted in the discussions. Additionally, previous soil samples have been obtained for Site 1 and these data were used in the present analyses (note that inhomogeneity was considered).

There is one (at least) caveat in attempting to use $^{220}Rn/^{222}Rn$ ratios to discern soil gas transport mechanisms - the method is largely site dependent. Using $^{220}Rn/^{222}Rn$ ratios as a tool can only be substantiated if at least one measurement site indeed has advective and diffusive processes occurring. It may be the case that changing soil gas migration processes only occur given certain soil conditions, geologic terrains, climatic conditions, or some other site specific factor(s). It is conceivable that within the relatively

limited scope of this study the three sites selected may not adequately validate the generic usefulness of $^{220}Rn/^{222}Rn$ ratio analyses. The ^{222}Rn data variations at the Chester, NJ site imply that both migration modes may be occurring, but finding an additional site where soil gas advection was likely to occur was desired. In the midst of this dissertation project, a site was found that fulfilled all requisite considerations, but after almost a year of making logistical arrangements, the owners prohibited using their property for fear of future liability if high ^{222}Rn levels were found. However, as the soil gas program at EML continues, it is hoped that such a site will be obtained in order to continue studying these phenomena.

In summary, the goals of this study are to:

(1) Develop a reliable method for determining soil gas ^{220}Rn concentrations;

(2) Develop the first data of long-term trends of soil gas ^{220}Rn;

(3) Add to the understanding of long-term trends in soil gas ^{222}Rn;

(4) Study certain aspects of geological setting and soil dynamics to the ^{222}Rn and ^{220}Rn concentrations in the soil gas;

(5) To investigate the feasibility of using long-term trends of soil gas $^{220}Rn/^{222}Rn$ ratios as potential indicators of soil gas transport.

Towards these ends, the project was designed and carried out as follows:

(1) A technique developed herein used Bateman equations to determine accurately, precisely, and rapidly soil gas ^{220}Rn concentrations using scintillation cells and photomultiplier (PM) tube equipment;

(2) At three sites, on a bi-weekly frequency, soil gas samples were obtained from 1.0 m depth and analyzed for ^{220}Rn and ^{222}Rn;

(3) While taking soil gas samples, soil permeability was measured to determine the effects of soil moisture and porosity;

(4) The $^{220}Rn/^{222}Rn$ time series data were analyzed to determine its usefulness as an indicator of the dominant soil gas transport process, either diffusion or flow.

Chapter 2

A SUMMARY OF ^{222}RN GENERATION, CONCENTRATION AND MIGRATION IN SOIL GAS

The element Rn, discovered in 1900 by Dorn who named it radium emanation, is a gaseous product of the ^{238}U decay series, and has an atomic weight of 86 making it the heaviest inert gas. The solid progeny of radon were originally monikored RadiumA, RadiumB, etc., terminology now used by only the most senior researchers. There are 20 known isotopes of Rn, with ^{222}Rn, ^{220}Rn, and ^{219}Rn being alpha emitters. ^{222}Rn has a half-life of 3.825 days, and is colloquially referred to as radon; ^{220}Rn has a half-life of 54.5 seconds, and is commonly termed thoron; ^{219}Rn, a product of actinium decay, is known as actinon, with a half-life of 3.92 seconds, which is too short to be of major importance in most environmental concerns.

^{222}Rn is a product in the ^{238}U decay series, shown in Figure 2.1, the immediate parent of which is ^{226}Ra. ^{220}Rn is a product of the ^{232}Th series, shown in Figure 2.2. ^{222}Rn follows the universal radioactive decay law, which states that the decay rate is proportional to the number of atoms remaining at any time t (using the proportionality constant λ) (for more detailed discussions of the following summary, see such texts as Nazaroff and Nero, 1988; Hopke, 1987; Wilkening, 1990 and the references cited therein):

$$-\frac{dN}{dt} = \lambda N \qquad (2.1)$$

where:

t = time; N = number of atoms of a radioactive element;

λ = decay constant = $\ln 2/T_{1/2}$, $T_{1/2}$ = half life = 3.82 days

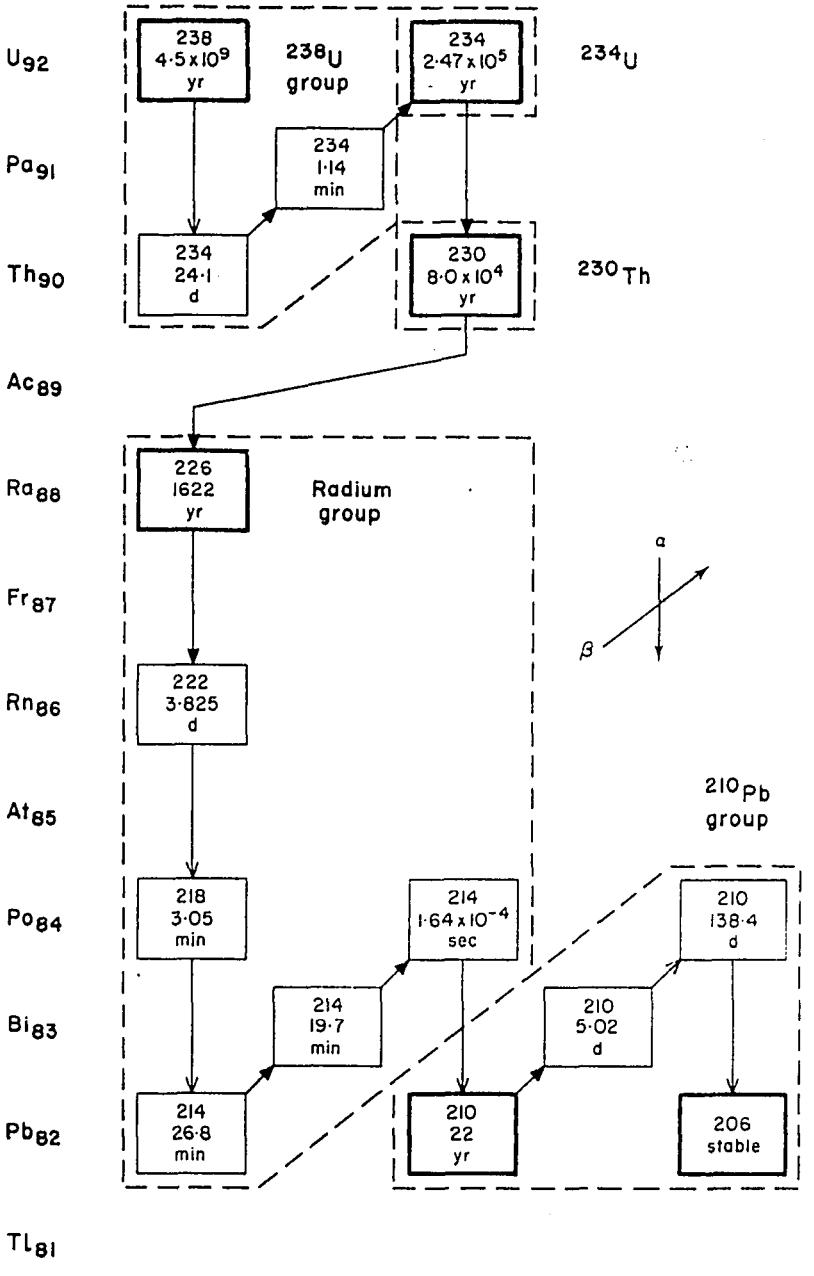

Figure 2.1 ^{238}U disintegration series. Half-lives and atomic weights are in small boxes. After Rose et al., 1979.

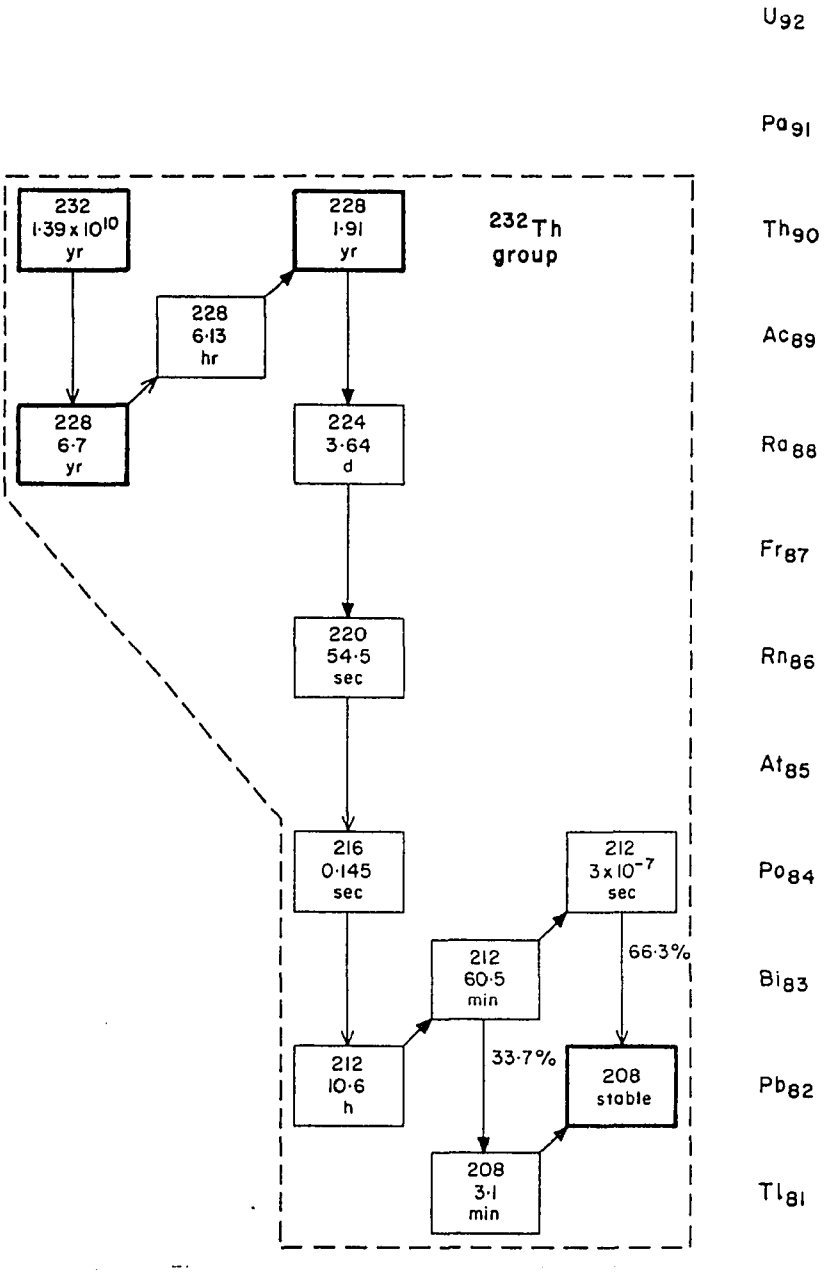

Figure 2.2 ^{232}Th disintegration series. Half-lives and atomic weights are in small boxes. After Rose et al., 1979.

Solving the first order differential equation 2.1 gives the number of radioactive atoms (N) that remain at any time t of an original number of atoms:

$$N = N^o e^{-\lambda t} \tag{2.2}$$

where: N^o = the initial number of atoms at t=0

In the ^{238}U decay series, the progenies are also radioactive so that the abundances change with time in a more complex manner than is shown in equation 2.1. To determine the activity of ^{222}Rn produced from the decay of radium the following equations must be combined:

$$\lambda_{Ra} N_{Ra} = - dN_{Ra}/dt = + dN_{Rn}/dt \ (^{222}Rn \text{ produced}) \tag{2.3}$$

$$\lambda_{Rn} N_{Rn} = - dN_{Rn}/dt \ (^{222}Rn \text{ Loss}) \tag{2.4}$$

where: N_{Rn} = number of ^{222}Rn atoms

N_{Ra} = number of radium atoms

λ_{Ra} = Radium decay constant

λ_{Rn} = ^{222}Rn decay constant = 2.1×10^{-6} sec^{-1}

to yield the net rate of change in ^{222}Rn with time:

$$dN_{Rn}/dt = [(\lambda_{Ra})(N_{Ra})] - [(\lambda_{Rn})(N_{Rn})] \tag{2.5}$$

The solution of the first order differential equation 2.5 gives the number of ^{222}Rn atoms at any time (assuming no change in N_{Ra} with time except by decay to ^{222}Rn):

$$N_{Rn} = [(\lambda_{Ra})/(\lambda_{Rn} - \lambda_{Ra})] N^o_{Ra}\{e^{-\lambda_{Ra}t} - e^{-\lambda_{Rn}t}\} + N^o_{Rn} e^{-\lambda_{Rn}t} \tag{2.6}$$

Because $\lambda_{Ra} << \lambda_{Rn}$ ($T_{1/2Ra} >> T_{1/2Rn}$), equation 2.6 simplifies to:

$$N_{Rn} = (\lambda_{Ra}/\lambda_{Rn})\, N^{o}_{Ra}\, [(e^{-\lambda_{Ra}t}) - (e^{-\lambda_{Rn}t})] + N^{o}_{Rn}\, e^{-\lambda_{Rn}t} \qquad (2.7)$$

If a radioactive nuclide is produced at the same rate as it decays, it is said to be in secular equilibrium. In the case of ^{222}Rn, if the rate of decay of ^{226}Ra equals the rate at which ^{222}Rn decays to ^{218}Po, secular equilibrium is established. Therefore, equation 2.7 can be further simplified, when t is large compared to the half-life of ^{222}Rn, to:

$$\lambda_{Ra}\, N_{Ra} = \lambda_{Rn}\, N_{Rn} \qquad (2.8)$$

One of most commonly used units to describe ^{222}Rn activity is the curie (Ci), defined as "the quantity of any radioactive nuclide that undergoes 3.7×10^{10} disintegrations per second" (Handbook of Chemistry and Physics, 1982-1983), which is equal to the decay rate for one gram of ^{226}Ra. ^{222}Rn concentrations in indoor air or water are commonly reported in units of picocuries per liter (pCi l^{-1}), where 1.0 pCi = 10^{-12} Ci, and in outdoor air in units of pCi m^{-3}. The International System of Units (S.I.) unit of radioactivity is the bequerel (Bq), equal to one disintegration per second, so that 1.0 Ci = 3.7×10^{10} Bq and 1.0 pCi l^{-1} = 37 Bq m^{-3}. The Bq is becoming more widely accepted due to the mandatory use of S.I. units in most journals, conferences, meetings, and symposia. However, there are fractions amongst workers in the ^{222}Rn industry seemingly unwilling to accept the S.I. units. The U.S. Environmental Protection Agency (EPA), the regulatory agency for indoor Rn under a memorandum of understanding with the U.S. Department of Energy, which focuses on basic research, continues to use pCi in all reports. The ^{222}Rn industries of indoor ^{222}Rn testing and mitigation will probably never switch to S.I. units, as the 4.0 pCi l^{-1} indoor "action level" has become quite

ingrained into the vocabulary. Consequently, in this dissertation, S.I. units will be used, kBq m^{-3} for soil gas concentrations, with pCi l^{-1} included in parentheses for clarity where appropriate.

In older literature, a commonly used unit, though of somewhat dubious value, is the working level (WL). The WL corresponds to any combination of ^{222}Rn progeny in one liter of air that will ultimately release 1.3 X 10^5 MeV of alpha energy during decay to ^{210}Pb. The WL is the sum of alpha energies from the short-lived progeny in equilibrium with 100 pCi l^{-1} ^{222}Rn. One WL is approximately equal to 200 pCi l^{-1}, assuming that 50% of the alpha energy is derived from ^{222}Rn itself. The Potential Alpha Energy Concentration (PAEC), with S.I. units of nJ m^{-3}, has replaced the WL as the unit of preference when discussing total ^{222}Rn progeny concentrations.

Also used in the literature are the units of the Working Level Month (WLM) and the Working Level Ratio (WLR), units carried over from the industrial hygiene and mining industries. Exposure is measured in WLM where the working month is taken to be 170 hours. Thus, exposure at one WL for 170 hours results in one WLM (Cliff, 1980). Clearly, the WLM has no direct use in explaining ^{222}Rn exposure in the home, since the exposure period per month varies depending on how much time an individual spends indoors. Nevertheless, the WLM has been used by researchers in reporting exposure in homes. Fortunately, the use of this unit is waning. The WLR is equal to the WL divided by the ^{222}Rn concentration in pCi l^{-1} multiplied by 100. The WLR is not a commonly used unit, though the ratio of PAEC/^{222}Rn activity provides a useful equilibrium ratio to characterize the ^{222}Rn series in air.

Several articles present a good overall description of the indoor ^{222}Rn problem (Nero, 1983; Akerblom et al., 1984; Gesell, 1983; Sachs et al., 1982). Additional thorough, yet not exhaustive, bibliographies of ^{222}Rn-related articles have been compiled (Lepman et al., 1981; Tanner, 1964, 1980; Bujdoso, 1991). Collections containing many articles with diverse perspectives are also available (Health Physics, special issue volume

45, 1983; The Natural Radiation Environment, Symposiums I, II, III, IV and V, 1964, 1972, 1980, 1988, 1992; Environment International, v. 8, no. 1-6, 1982; International Meeting on Radon-Radon Progeny Measurements, 1981).

The results of many studies concerning the ^{222}Rn concentrations in houses worldwide have been published. Swedish homes are monitored under a government ordered and funded plan. The ^{222}Rn concentration of every new house built must not exceed 70 Bq m^{-3} (1.89 pCi l^{-1}). Houses already in existence are regarded as "unsanitary" if the ^{222}Rn concentration exceeds 400 Bq m^{-3} (10.8 pCi l^{-1}), of which nearly 15% exceed that limit (Swedjemark, 1980; Hildingson, 1982). Other studies of ^{222}Rn in homes have been conducted in The Netherlands (Wolfs et al., 1984), Switzerland (Buchli and Burkart, 1989), Italy (Facchini et al., 1981), Great Britain (Cliff, 1980; Abu-Jarad and Fremlin, 1983), Hong Kong (Tso and Leung, 1991), Poland (Pensko et al., 1969), Norway (Stranden, 1980), Turkey (Koskal et al., 1993) and Canada (McGregor et al., 1980), among many others. Additionally, there have been many reports of the ^{222}Rn concentration in U.S. houses (George and Breslin, 1980; White et al., 1989; Doyle et al., 1984; Cohen and Shah, 1991; Nero et al., 1986; Moschandreas and Rector, 1982; Alter and Oswald, 1983), where the EPA has established a 4.0 pCi l^{-1} (148 Bq m^{-3}) "action level" guideline; houses with concentrations above this level should seek steps to mitigate the indoor ^{222}Rn.

In many of the European studies, low ventilation rates and building materials are commonly given as the major influencing factors in the ^{222}Rn accumulation (Schuler, et al., 1991; Fleischer et al., 1982; Akerblom and Wilson, 1981; Stranden et al., 1979). In the U.S. and Canada, the substrate, specifically the soil gas, is frequently given as the most common source (Nero and Nazaroff, 1984; Sachs et al., 1982; Li et al., 1992). In a relatively few isolated instances, the water supply has been found to be a major contributing source of the ^{222}Rn (Lawrence et al., 1992; Cross, 1985; Hess et al., 1980, 1982; Gesell and Prichard, 1980). Nevertheless, in most instances where elevated ^{222}Rn

concentrations in houses exist, the soil gas is most likely to be the major contributing source.

The first study to establish that ^{222}Rn and ^{220}Rn are contained in soil gas was performed by Elster and Geitel (1902). Subsequent investigations were made in the U.S. by Bumstead and Wheeler (1904), Sanderson (1911), Wright and Smith (1915) and Kovach (1945). Additionally, there were a few studies conducted in Europe and elsewhere (see Kovach, 1945, and Israel and Bjornsson, 1966). Most of these early investigations studied the source and magnitude of the natural radiation phenomena. Recent investigations of soil gas ^{222}Rn have shown temporal and spatial variations to be large and often not well understood (Rose et al., 1990).

A number of studies involve the methods and techniques of measuring ^{222}Rn. Solid state nuclear track detectors, such as Track-Etch detectors, integrate the short-term variations of ^{222}Rn using exposure times of up to three months to a year (Budnitz, 1974; Gingrich and Fisher, 1976). These solid state track detectors consist of a plastic film that is damaged by alpha particles during radioactive decay of ^{222}Rn and ^{220}Rn. The plastic strips are later acidically etched to determine the ^{222}Rn concentration from the track density (Kvasnicka, 1980). Field portable detectors, based on scintillation cells and photomultiplier tubes (Lucas, 1957), are convenient, accurate and quick; many measurements can be obtained in a day (Hesselbom, 1985; Dyck, 1969).

The first observations of anomalous ^{222}Rn concentrations over faults were made by Ambronn (1921). A few studies were conducted using ^{222}Rn in soil gas for prospecting of faults (Israel and Bjornsson, 1966) and other geological structures (Borchiellini et al., 1991). Additionally, ^{222}Rn has been used as a prospecting tool for uranium, as anomalous uranium concentrations in underlying strata may produce anomalous ^{222}Rn concentrations in the soil gas (Smith et al., 1976; Dyck, 1969; Gingrich and Fisher, 1976; Severne, 1978).

^{222}Rn has also proven to be very useful tool as an atmospheric tracer of air masses (Balkanski et al., 1992; Whittlestone, S. et al., 1992; Hutter, A. R. et al., 1991), and is used by climatologists to test and validate global transport models (GTM's; Jacob and Prather, 1990; Feichter and Crutzen, 1990; Brost and Chatfield, 1989).

The variability of ^{222}Rn in the soil gas has been studied in connection with tectonic activity as a potential predictive tool. ^{222}Rn monitoring in earthquake-prone regions was started in the U.S. in 1975, with the first reports published in by Mogro-Campero and Fleischer in 1977 and by King in 1978. ^{222}Rn has been used as a tool to predict the magnitude, location and time of earthquakes using methods described by King (1980), Mogro-Campero et al. (1980) and Fleischer and King (1985), and is summarized by Monnin and Seidel (1992).

There have been several soil gas ^{222}Rn studies attempting to identify temporal patterns. Three general trends have been identified:

(1) high summer and low winter ^{222}Rn (Rose et al., 1990)

(2) high winter/spring and low summer/fall (Klusman and Jaacks, 1987; Schumann et al., 1989)

(3) no annual pattern (Fleischer et al., 1980).

Clearly, understanding temporal patterns of soil gas ^{222}Rn needs to be better defined, in order to incorporate its dynamics in understanding indoor ^{222}Rn concentrations. The current study focuses on transport mechanisms and processes in the soil gas as a means of understanding temporal and spatial variations of soil gas ^{222}Rn.

Transport processes of ^{222}Rn in the near surface environment have been studied but definitive conclusions concerning the mode of ^{222}Rn transport differ (Tanner, 1964, 1980). Diffusion in soil air has generally been considered to be the dominant process of ^{222}Rn migration (Tanner, 1964; Schroeder et al., 1965). However, there have been suggestions that in soils with connected air-filled pores, bulk flow processes are locally significant (Fleisher and Mogro-Campero, 1979). The proposed microflow of ascending

"geogas" may act as a carrier gas for the ^{222}Rn atoms (Malmqvist and Kristiansson, 1984). The carrier gas originates either in an immense reservoir in the interior of the earth or as part of a cyclic process that involves an exchange of gases between the atmosphere and the ground (Kristiansson and Malmqvist, 1984). The stream of carrier gas moving upward as bubbles through water-filled cracks and fissures can transport ^{222}Rn atoms with a velocity much greater than diffusional transport (Kristiansson and Malmqvist, 1984).

If diffusion is the major mode of ^{222}Rn transport, ^{222}Rn migration distance in soils can be described by the diffusion length. The diffusion length (defined as the distance in which an e-fold decrease in ^{222}Rn concentration occurs) depends upon the diffusion coefficient for ^{222}Rn in the soil and on the ^{222}Rn decay constant (the pertinent diffusion equations are discussed later). The diffusion constant depends on soil parameters and atmospheric variables such as humidity, porosity, barometric pressure, wind, precipitation, etc. (Kvasnicka, 1980). For dry soil of normal porosity, the diffusion length is approximately one m, which translates into an upper limit of ^{222}Rn transport on the order of a few meters (Tanner, 1964). However, if ^{222}Rn migration is caused by a flow of soil air, ^{222}Rn transport distances can exceed 100 m (Malmqvist and Kristiansson, 1984). It is thought that ^{222}Rn entry into houses is largely controlled by advection processes induced by a temperature and pressure stack effect between the building and soil interface (Nazaroff, 1992).

Research directed towards understanding the significance of soil gas dynamics to the problem of ^{222}Rn accumulation in houses and buildings has not substantially advanced since the early 1980's. It would seem imprudent to neglect this aspect of the ^{222}Rn problem, for in order to understand the dynamics of indoor ^{222}Rn, the characteristics of its source must be well understood first. Understanding the controls on the variability of soil gas ^{222}Rn will increase the ability to predict areas likely to have elevated ^{222}Rn

concentrations in the soil gas and consequently which houses and buildings are likely to have elevated indoor ^{222}Rn concentrations.

To date, it is regarded that ^{222}Rn generation in the soil is largely dependent upon three factors:

(1) parent concentration in rocks and soil

(2) emanation coefficient, the fraction of ^{222}Rn generated that in soil that escapes the soil grain

(3) partitioning among solid (i.e., rock and soil), liquid (i.e., soil water content) and gas (i.e., soil gas).

The rate of ^{222}Rn migration through the soil is either by diffusion or advection. Diffusion is governed by Fick's Law, written as:

$$J_s^d = -D_e \nabla C_{Rn} \qquad (2.9)$$

where:

J_s^d = diffusive flux density of ^{222}Rn activity per unit of air-filled pore area of soil, Bq m^{-2} s^{-1}

∇ = gradient, m^{-1}

D_e = Diffusion coefficient, m^2 s^{-1}

C_{Rn} = ^{222}Rn concentration, Bq m^{-3}

Advection in the soil gas is described by Darcy's Law:

$$vD = -(\kappa/\mu)\nabla P, \qquad (2.10)$$

where:

vD = volumetric flow rate per unit cross-sectional area

κ = soil permeability

μ = dynamic viscosity of the fluid

∇P = pressure gradient

Diffusion is thought to be the dominant process by which ^{222}Rn enters the atmosphere from uncovered soil. However, advection is thought to be the dominant transport process of ^{222}Rn entering buildings (Nazaroff, 1992).

One of the most important physical characteristic to soil gas migration is soil permeability (Nazaroff, 1992). Permeabilities of soils range over six orders of magnitude, with diffusion being the dominant process at the low end and advection being dominant at the high end. The value of soil permeability that separates the two regimes is on the order of 10^{-11} m^2 (Nazaroff and Sextro, 1989). Soil permeability, in turn, is largely dependent upon grain size and soil moisture.

Models of ^{222}Rn concentration in the soil gas have been developed based on the above equations. Most studies have indicated that diffusion is the dominant transport process in the soil gas, and largely ignore the advection component.

The ^{222}Rn concentration (C_{Rn}) in the soil air of a thick soil layer below the depth affected by diffusion towards the surface follows the equation:

$$C_{Rn} = Q/\lambda_{Rn} = C_{Ra}E\rho/P \text{ (Nielson and Rogers, 1984)} \qquad (2.11)$$

where:

C_{Ra} = Radium activity, Bq kg^{-1}

Q = ^{222}Rn production rate, number of ^{222}Rn atoms that are free to migrate through the pore system per unit time per unit pore volume, Bq m^{-3} sec^{-1}

E = ^{222}Rn emanation coefficient

P = connected soil porosity

ρ = soil density, kg/cm^3

λ_{Rn} = ^{222}Rn decay constant, 2.1 X 10^{-6} sec^{-1}

Given typical values (C_{Ra} = 37 Bq kg^{-1}, E = 0.3, P = 30 %, ρ = 2.5 g/cm^3) the soil air ^{222}Rn concentration is calculated to be 92.5 kBq m^{-3} (2500 pCi l^{-1}).

However, soil gas ^{222}Rn concentrations decrease toward the surface as a result of diffusion from high ^{222}Rn concentrations in the deep soil to low ^{222}Rn concentrations in the atmosphere (Tanner, 1964). Examples of the ^{222}Rn concentration profile in soil are given below for simple diffusion-only transport. The soil gas ^{222}Rn concentration as a function of depth (Z) in a homogeneous soil under steady-state conditions is given by the equation (Colle et al., 1981):

$$C(Z) = [Q/\lambda]\{1 - e^{(Z/v)}\} \qquad (2.12)$$

where:

Z = soil depth, m

v = diffusion length, or lifetime diffusion distance, a measure of the distance in which there is an e-fold (1/e = 37%) decrease in ^{222}Rn concentration, m

$= (D/(P\lambda))^{1/2}$,

D = diffusion coefficient, m^2/sec.

$= D_o\rho\exp(-6F\rho - 6F^{14}\rho)$, $D_o = 1.1 \text{ X } 10^{-5} \text{ m}^2 \text{ s}^{-1}$ (Rogers and Nielson, 1991)

F = water saturation fraction

As discussed in the example above,

$[Q/\lambda] = C_{Rn} = C_{Ra}E\rho/P = 92.5 \text{ kBq m}^{-3}$

Figure 2.3 shows the ^{222}Rn concentration profile calculated using equation 2.12 (labeled A) with commonly used variables Z = 8.0 m, F = 0.5, P = 0.35, E = 0.3, C_{Ra} = 37 Bq kg^{-1}, and $\rho = 2.5$ g/cm^3.

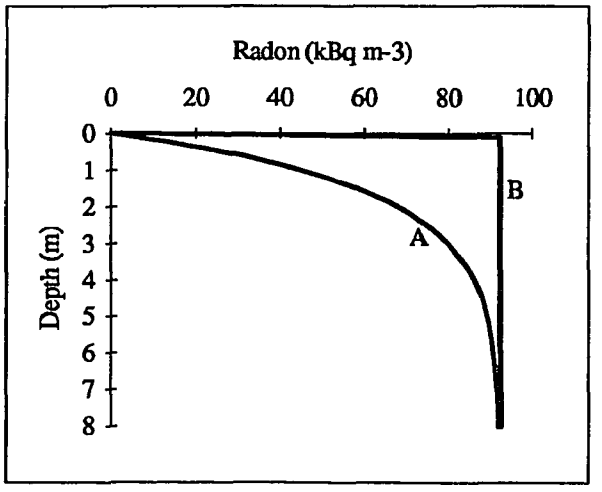

Figure 2.3 ^{222}Rn profile for 8m soil calculated from equation 2.12 labeled A. ^{222}Rn profile for 8m soil calculated using equation 2.14 labeled B for surface-saturated or ice-capped soil. See text for explanation.

To calculate the soil gas ^{222}Rn concentration profiles for soils of differing depths, equation 2.13 is used, given by (Colle et al., 1981):

$$C(Z) = (Q/\lambda)*[1-\{\sinh(Z/v) + \sinh(L-Z)/v\}/\sinh(L/v)] \qquad (2.13)$$

where:

L = thin soil layer thickness, m

Z = depth in soil, m

$Q/\lambda = 92.5$ kBq m^{-3}

Figure 2.4 shows the ^{222}Rn concentration profile for depths of 2.0 m, 4.0 m, 6.0 m and 8.0 m. Notice that at greater depths, the profile calculated using equation 2.13 approaches that used for a deep soil calculated using equation 2.12.

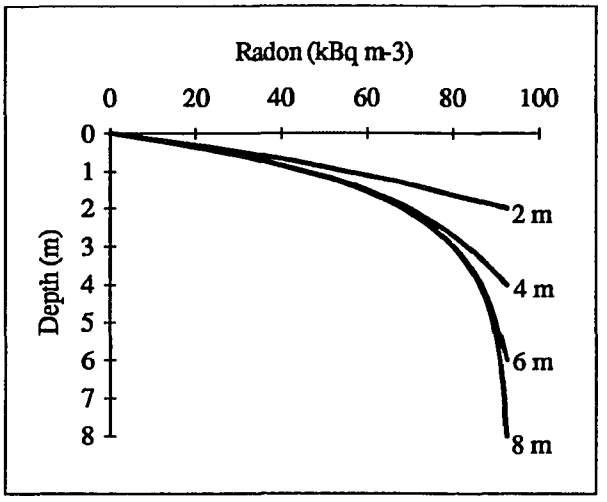

Figure 2.4 ^{222}Rn concentration profiles calculated using equation 2.13 for depths 2m, 4m, 6m and 8m as denoted in text.

The ^{222}Rn concentration in the soil gas (C(Z)) of a thick soil layer beneath a 10 cm water-saturated surface layer was calculated using equation 2.14 (Colle et al., 1981):

$$C(Z) = Q/\lambda[1 - \{e^{(Z/v_g)}\}/\{1 + (v_s/v_g)*(D_g/D_s)*\tanh(L/v_s)\}] \quad (2.14)$$

where:

v_g = diffusion length in the soil gas, m

v_s = diffusion length in the saturated soil layer, m

D_g = diffusion coefficient in soil gas, cm^2/sec

D_s = diffusion coefficient in saturated soil layer, cm^2/sec

L = depth of saturated soil layer, m

$[Q/\lambda] = C_{Ra}E\rho\lambda_{Rn}/P = 92.5$ kBq m^{-3}

The ^{222}Rn concentration profile calculated using equation 2.14 (using commonly-used variables v_s = .04 m, v_g = 2.2 m, $D_s = 1 \times 10^{-5}$, D_g = .03, L = .10 m) is shown in Figure 2.3 and labeled B. A saturated surface soil acts as a cap to the exhalation of soil gas, so that the ^{222}Rn concentration increases in the shallow soil layers to a value

approaching that in the deeper layers of the soil profile, if the surface soil remains saturated for a period longer than approximately two weeks.

The soil gas ^{222}Rn concentration described by simple diffusion can be modified by many factors, temperature, barometric pressure, precipitation, wind direction and speed, relative humidity, soil moisture, soil porosity and permeability to name a few. The magnitudes of these effects on the soil gas ^{222}Rn concentration are time dependent and each affect the ^{222}Rn concentration to different depths. Furthermore, these variables affect the ^{222}Rn concentration over time periods as short as a few minutes or hours, or as long as a year. Additionally, bedrock type and structure, as well as uranium and uranium progeny concentrations and distributions in the rock and soil can similarly affect ^{222}Rn concentrations.

The barometric pressure of the atmosphere tends to affect the ^{222}Rn concentration in the soil gas in a piston-like manner. When the barometric pressure is increasing, the ^{222}Rn along with the bulk soil gas is "pushed" downward in the soil profile, and escape to the atmosphere is decreased. On the other hand, when the barometric pressure is dropping, the ^{222}Rn is "pulled" from the soil pores into the atmosphere (Rudakov, 1985). Atmospheric pressure changes of one to two percent occurring over a period of one to two days results in ^{222}Rn flux changes from the soil to the atmosphere of 20 to 60 % (Clements and Wilkening, 1974). Furthermore, if the ^{222}Rn concentration being exhaled to the atmosphere is changing by 20 to 60%, the soil gas ^{222}Rn concentration (to some depth) must also change by an approximately equivalent factor.

Precipitation has at least two effects on the soil gas ^{222}Rn concentration. Water tends to form a thin layer around the soil grains, which may increase the emanation coefficient, thereby increasing the soil gas ^{222}Rn concentration (Tanner, 1964). The emanation coefficient of a soil grain surrounded by water is greater than if the grain is only surrounded by air because water can absorb recoiled alpha particles whereas a greater

number of alpha particles become embedded in an adjacent soil grain if only surrounded by air. Conversely, during periods of little precipitation, the soil dries out which increases soil pore space and allows ^{222}Rn to exhale and diffuse to the atmosphere more rapidly than from saturated soils (Tanner, 1964). Additionally, water tends to fill the soil pores at the surface and, because the diffusion coefficient in water is approximately three orders of magnitude less than in air (Tanner, 1964), decreases the diffusion rate. Wind direction and speed each affects the ^{222}Rn concentration in the soil gas down to a depth of one or two meters (Kraner et al., 1964).

Temperature can affect the ^{222}Rn concentration both diurnally and seasonally. Diurnal variations are associated with the competition between the changes in convective flow due to the temperature differences in the soil from day to night and the turbulent mixing in the atmosphere during the daytime leads to an increase in ^{222}Rn exhalation (Colle et al., 1981; Wilkening, et al., 1972). Additionally, the seasonal temperature variation affects the soil gas ^{222}Rn concentration (Okabe, 1956; Kraner et al., 1964; Megumi and Mamuro, 1973; Washington and Rose, 1990). During the winter, a frozen ground cover, occurring if the surface soil is saturated, caps the soil, thus prohibiting ^{222}Rn escape to the atmosphere and causing an increased ^{222}Rn concentration in comparison to other times.

A model incorporating temperature effects of the seasonal variations found in soil gas ^{222}Rn at a limited number of sites is given by Washington and Rose (1990) for depths below which diffusional transport to the surface is negligible, about 1.0 m:

$$C_{Rn} = 10^3 C_{Ra} E\rho_b / P[F(K_T - 1)+1] \qquad (2.15)$$

where:

C_{Rn} = ^{222}Rn concentration, Bq m^{-3}

C_{Ra} = parent concentration, Bq kg^{-1}

E = emanation coefficient

ρ_b = dry bulk density

F = water saturation fraction

K_T = partition coefficient of ^{222}Rn between unit volumes of air and water, C_{water}/C_{air}. (values vary from 0.525 at 0 °C to 0.226 at 25 °C; Battino, 1979).

P = volume fraction of total pore space (porosity)

which is equation 2.11 with a temperature dependence added into the model. Variations in the ^{222}Rn concentration of up to a factor of about four can be accounted for due to effects of F and K_T, with warm (25 °C) moist (F = 0.95) soils having a ^{222}Rn concentration of 78.3 kBq m^{-3} (2116 pCi l^{-1}; calculated with C_{Ra} = 30 Bq/kg, E = 0.2, and ρ = 1.5 g/cm^3) compared to a cold (0 °C) and dry (F = 0.05) soil having a ^{222}Rn concentration of 21.2 kBq m^{-3} (573 pCi l^{-1}).

Parameters affecting the soil gas ^{222}Rn and ^{220}Rn concentrations, such as the soil moisture, temperature, barometric pressure, etc. will affect the emanation of ^{220}Rn and ^{222}Rn in soil gas equally. Therefore, the ^{220}Rn to ^{222}Rn ratio taken over time should be nearly constant; i. e., as the ^{222}Rn concentration rises due to an increase in soil moisture increasing the emanation coefficient, so should the ^{220}Rn emanation increase and thusly the ^{220}Rn concentration. The net result would be that the ^{220}Rn to ^{222}Rn ratio remains constant.

From equation 2.15:

$$C_{222Rn} = C_{226Ra} E\rho_b/P[F(K_T - 1)+1] \qquad (2.16)$$

where:

C_{222Rn} = ^{222}Rn concentration, kBq m^{-3}

C_{226Ra} = ^{226}Ra concentration, kBq kg^{-1}

and written for ^{220}Rn:

$$C_{220Rn} = C_{224Ra} E\rho_b/P[F(K_T - 1)+1] \quad (2.17)$$

where:

C_{220Rn} = ^{220}Rn concentration, kBq m^{-3}

C_{224Ra} = ^{224}Ra concentration, kBq kg^{-1}

Taking the ratio of equation 2.17 to 2.16 to use $^{220}Rn/^{222}Rn$ ratios, the pertinent equation becomes simply:

$$C_{220Rn}/C_{222Rn} = \text{constant}, \quad (2.18)$$

since all parameters simply cancel out, or the ratios become a constant themselves, such as the parent concentrations and the emanation coefficients (Greeman, pers. comm., 1991). Additionally, other soil parameters possibly affecting the soil gas ^{222}Rn and ^{220}Rn concentrations, e.g., barometric pressure, permeability (reflected by porosity), etc. would also cancel in any equation including these parameters into the model calculation. The exact value of this constant will be site specific, of course, dependent upon local soil conditions.

If there was a soil gas advection from depth of high ^{222}Rn concentration, due to deposition of uranium, thorium, and/or radium in faults and fractures, the ^{222}Rn concentration in nearer surface soils would also be elevated but without an accompanying increase of ^{220}Rn. The short half-life of only 55 sec does not permit ^{220}Rn to travel more than a few cm's, even at high flow rates. If these advective conditions were present, the ^{220}Rn to ^{222}Rn ratio would be expected to be low. Conversely, even if the flow conditions were present and low ^{222}Rn concentration soil gas was being transported, the ^{220}Rn to ^{222}Rn ratio would still be elevated for the same reason. In either case, the

change from advective to diffusive migration process should be detected by a change in the $^{220}Rn/^{222}Rn$ ratios over time. On the other hand, if diffusion was the only mechanism of soil gas transport, the ^{220}Rn to ^{222}Rn ratio would be expected to be constant. Therefore, observing how the ^{220}Rn to ^{222}Rn ratio over time changes could give an indication of whether or not the transport type changed during the year. If only ^{222}Rn concentrations were measured, as has previously been the case in all soil gas studies, then all other parameters would have to be concurrently measured in order to ascertain the controls of ^{222}Rn concentration variations. The proposed method of using ^{220}Rn and ^{222}Rn in soil gas eliminates the need to measure all other aforementioned parameters.

Chapter 3

METHODS, SITE DESCRIPTIONS AND QUALITY ASSURANCE

3.1 Soil Gas ^{222}Rn and ^{220}Rn

This section was written and submitted for publication to the journal Health Physics. Figure, Table and equation numbers have been changed for consistency within this dissertation.

A Method for Determining Soil Gas ^{220}Rn (Thoron) Concentrations

Abstract

A technique to accurately, precisely and quickly measure soil gas ^{220}Rn was developed for use with commercially available grab-sample scintillation detectors. The method requires a one-minute-count as soon as the sample has been drawn into the scintillation cell, and a five- or ten-minute-count at least five minutes after the soil gas sample has been obtained. The ^{222}Rn concentration is determined from the second count, and used to subtract the counts due to ^{222}Rn and progeny during the first count. The remaining counts are due to ^{220}Rn and progeny. The overall uncertainty when using this method to measure typical soil gas ^{220}Rn and ^{222}Rn concentrations, i.e. greater than approximately 3 kBq m^{-3} (80 pCi l^{-1}) was determined to be 20.7% and 10.8% (90% confidence levels), respectively, determined from analyses of duplicate field measurements. The lowest ^{220}Rn concentrations that can be measured using this technique while maintaining an overall error no greater than about 30% is approximately 500 Bq m^{-3} (13 pCi l^{-1}). The ^{220}Rn measurement uncertainty at this concentration level decreases to approximately ± 20 to 25% if at least three measurements sampled in series are arithmetically averaged.

Introduction

The term radon generally refers to the specific ^{222}Rn isotope of the element radon (Rn). Much less is known and published concerning the isotope ^{220}Rn, generally referred to as thoron, owing to difficulties in measurement arising from its short half-life of about 56 seconds. It has been estimated, however, that 10-20% of the effective dose equivalent from all indoor ^{222}Rn and ^{220}Rn progeny are due to ^{220}Rn progeny (UNSCEAR, 1982). Recent evidence has shown soil gas to be a major source of indoor ^{220}Rn (Li et al., 1992). Understanding the source term spatial and temporal variability of ^{220}Rn is clearly an important consideration in this regard. Additionally, $^{220}Rn/^{222}Rn$ ratios are currently being used to assess variations of soil gas transport processes (Hutter, 1993).

The most common measurement techniques for ^{220}Rn require gross alpha counting solid progeny deposited on filters hours after sampling, and calculations extricating alpha counts due to ^{220}Rn from those due to ^{222}Rn and progeny. There are currently no known published methods for measuring ^{220}Rn directly using scintillation cells/PM tube instruments. For an investigation of soil gas transport mechanisms, rapid determination of ^{222}Rn and ^{220}Rn concentrations for a large number of soil gas samples were needed (Hutter and Feely, 1990). The technique described here allows a large number of samples to be obtained, and also allows the scintillation cell to be purged and reused, thereby limiting the number of cells needed.

Description of Method

The method developed entails a one-minute count immediately following the end of entry of the soil gas into the cell, followed by a five-minute-count sometime after most of the ^{220}Rn has decayed (about 4 or 5 half-lives, or about 5 minutes). The time required for ^{222}Rn and ^{220}Rn determination for each sample is approximately 15 minutes (including background check, pumping and flushing), and thus the effects of contamination by ^{222}Rn progeny plating out onto the zinc phosphor lining of the

scintillation flask are minimized, allowing the flask to be reused with a low background count even when a high ^{222}Rn concentration sample was previously obtained.

The sample collection can be either via pumping or into an evacuated cell. The pumping method is necessary for soil gas sampling, due to common sampling depths of 1 m, so that corrections are needed that account for the time the soil gas took to reach the scintillation cell from the end of the sampling hole, as well as the decays that occurred during this time. The time correction is calculated from the known flow rate and volume of sampling tubes. The correction for decays is calculated knowing this time correction and using standard decay equations. These corrections are easily incorporated into a computer program for calculation of concentrations from measured counts.

Determination of ^{222}Rn

By calculating the expected radioactive decay of ^{222}Rn and the radioactive growth of ^{222}Rn progeny in the presence of ^{220}Rn and its progeny, it was concluded that a five-minute count beginning at the end of five minutes and continuing until 10 minutes after the soil gas was introduced into the counting chamber was satisfactory to calculate the ^{222}Rn concentration. During this five-minute period, the counts due to ^{220}Rn are negligible, since more than five half-lives have elapsed.

The technique requires calculation of counts obtained back to counts per minute at time zero, i.e., as soon as sampling ended. In order to obtain the initial counts per minute due to ^{222}Rn (X_{222Rn}), an integration factor (β_{222Rn}) must be derived to take into account the counts due to ^{222}Rn daughter growth and ^{222}Rn decay during this time. The integration factor is calculated from integrating the following equation for the time period selected:

$$\beta_{222Rn} = (^{222}Rn + ^{218}Po + ^{214}Po)/^{222}Rn_o \qquad (3.1)$$

where: $^{222}Rn_0$ = initial ^{222}Rn activity at t = 0 (cpm), and ^{222}Rn, ^{218}Po, ^{214}Po are activities in counts per time period.

Figure 3.1 shows the curve of equation 3.1, the total ^{222}Rn and progeny alpha activity divided by initial ^{222}Rn alpha activity, versus time.

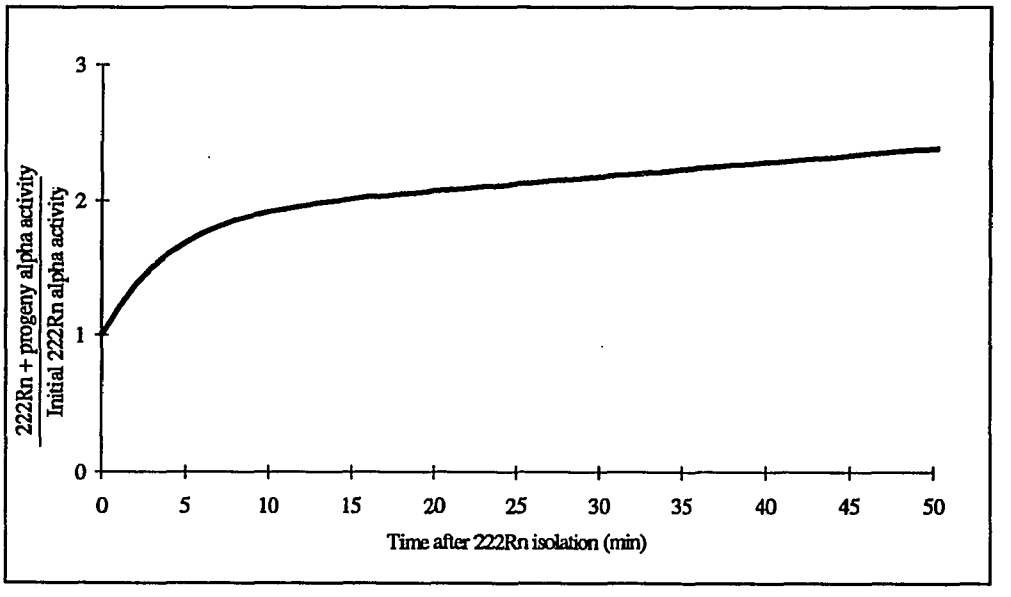

Figure 3.1 Growth of ^{222}Rn and progeny activity after isolation.

The integration factor, β_{222Rn}, has units of min^{-1}, i.e., counts in specified time period divided by initial counts per minute. The 5-to-10-minute integration factor was calculated to be 9.08 counts per 5-to-10 minutes divided by the initial ^{222}Rn activity in counts per minute. Equation 3.2 gives the initial counts per minute due to ^{222}Rn (X_{222Rn}) for any sample:

$$X_{222Rn} = \{[(N_S) - (N_b)]/\beta_{222Rn}\} \quad (3.2)$$

where: N_S = x-minute count, x = 5, 10, or longer minutes; N_b = x-minute background count, x = 5, 10, or longer minutes; β_{222Rn} = integration factor in units of min^{-1} (counts per time period divided by initial counts per minute).

A computer program was written to calculate the integration factor for any specified time periods so that any counting periods could be used if necessary.

The initial counts per minute due to ^{222}Rn (X_{222Rn}) are then multiplied by a calibration factor determined for the detector/scintillation cell apparatus to give ^{222}Rn concentration. The commercial ^{222}Rn detector (photomultiplier tube and scintillation cells) has the same counting efficiency for all the ^{222}Rn, ^{220}Rn and progeny alpha particles, regardless of energy.

Determination of ^{220}Rn

^{220}Rn concentrations can be calculated after the ^{222}Rn concentration has been determined. As mentioned earlier, a one minute count is obtained as soon as possible after sampling has ended, normally about 10 seconds. The initial counts per minute due to ^{222}Rn, previously calculated, are multiplied by an integration factor (calculated using the computer program integrating the curve in Figure 3.1 for the one-minute period) to give the counts in this one-minute-period due to ^{222}Rn and ^{222}Rn progeny. These counts are subtracted from the total counts measured during the one-minute-count obtained after the soil gas is introduced into the scintillation cell to give counts not due to ^{222}Rn, that is, the counts due to ^{220}Rn and ^{220}Rn progeny. The initial counts per minute due to ^{220}Rn (X_{220Rn}) are obtained by dividing the counts in the one-minute period due to ^{220}Rn and progeny by a second integration factor (β_{220Rn}) calculated from:

$$\beta_{220Rn} = (^{220}Rn + ^{216}Po \text{ activity}) / ^{220}Rn_o \qquad (3.3)$$

where: $^{220}Rn_o$ = initial ^{220}Rn activity in cpm, and ^{220}Rn and ^{216}Po are activities in counts per time period, by integrating for the time of the first count (1 min). The initial

counts per minute due to ^{220}Rn are then multiplied by a calibration factor determined separately for each scintillation cell to obtain the ^{220}Rn concentration in kBq m^{-3}. The apparatus used with this method was calibrated using the EML Radon, Thoron, and Progeny Exposure Facility for ^{222}Rn and a Pylon[1] source for ^{220}Rn.

Error Analysis

The overall uncertainty when using this method for determining soil gas ^{220}Rn and ^{222}Rn incorporates systematic errors introduced from counting statistics, and calibrations, as well as random errors from sampling procedures, each of which are evaluated below. The overall uncertainties in the ^{220}Rn and ^{222}Rn concentrations are estimated from analyses of duplicate field measurements, defined as measurements performed on the same hole on the same day approximately 10 - 15 minutes apart using a different scintillation cell. The time dependent variations of the ^{222}Rn and ^{220}Rn in soil gas over 10 or 15 minutes are assumed to be negligible. The uncertainties obtained in this manner are interpreted to be the total error associated with this technique because they incorporate the errors due to counting statistics and calibrations, as well as random errors arising from sampling procedures.

From previous studies (Hutter, 1993), 101 duplicate measurements for ^{222}Rn and 74 for ^{220}Rn were obtained. The determination of the overall uncertainty can be inferred from the histogram of the % differences from the mean of the duplicate measurements, shown in Figures 2 and 3, for ^{222}Rn and ^{220}Rn, respectively. The mean % difference is 0.64% for ^{222}Rn and 2.8 % for ^{220}Rn. Based on one standard deviation of this distribution, the overall uncertainty associated with the soil gas measurements is estimated to be 10.8% for ^{222}Rn and 20.7% for ^{220}Rn.

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[1]Pylon Electronic Development company. ltd.
147 Colonnade Rd.
Ottawa, Ontario
Canada K2E 7L9

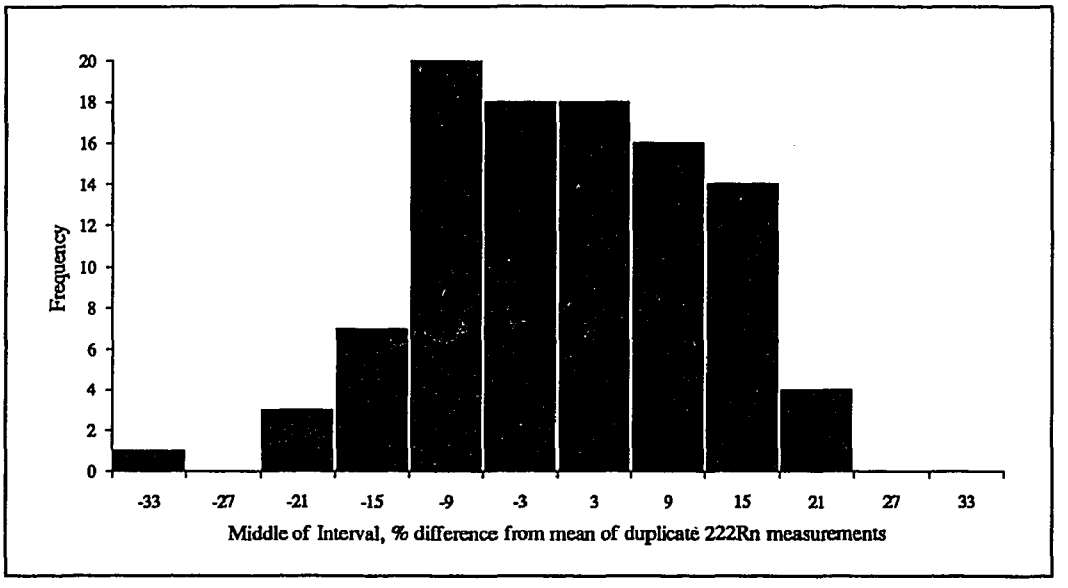

Figure 3.2 Histogram of % difference from mean of duplicate ^{222}Rn measurements. The duplicate ^{222}Rn measurements (n=101) ranged from 8.40 to 1308 kBq m^{-3} (227 to 35358 pCi l^{-1}), and averaged 457 kBq m-3 (12361 pCi l^{-1}) with a standard deviation of 374 kBq m^{-3} (10129 pCi l^{-1}).

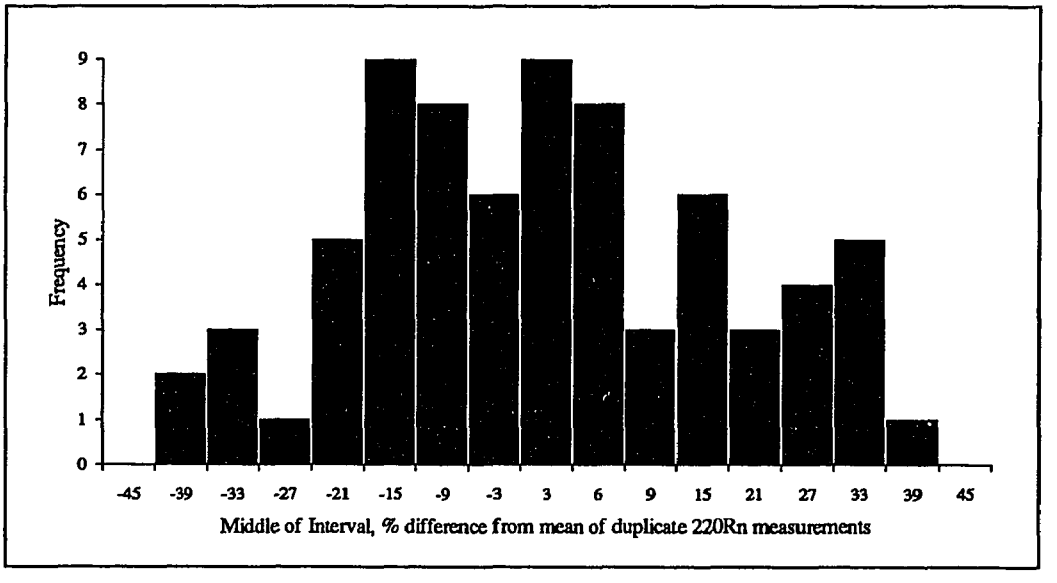

Figure 3.3 Histogram of % difference from mean of duplicate ^{220}Rn measurements. The duplicate ^{220}Rn measurements (n=74) ranged from 5.0 to 801 kBq m^{-3} (135 to 21657 pCi l^{-1}), and averaged 294 kBq m-3 (7955 pCi l^{-1}) with a standard deviation of 211 kBq m^{-3} (5713 pCi l^{-1}).

As with any radioactivity measurement, there are errors arising from counting statistics. The simple error for counting statistics is simply the square root of the number of counts divided by the number of counts, according to the equation (Friedlander et al., 1981):

$$s = (N^{1/2})/N = (N_S + N_b)^{1/2}/(N_S - N_b) \quad (3.4)$$

where: s = relative standard deviation; N = number of counts for the sample; N_S = counts attributed to the element of interest; N_b = background counts.

Using equation 3.4, the 1σ error introduced to a single count, as is used for ^{222}Rn determination, due to these counting statistics alone is between 14% and less than 1%, respectively, for normal counting rates between 50 and 5000 cpm. From analyses of multiple counts obtained from 45 soil gas samples ranging in ^{222}Rn concentration from 2 to 1219 kBq m^{-3} (55 to 32953 pCi l^{-1}), the counting statistical error (1σ) was empirically estimated to be 4.5% for ^{222}Rn. The error in the counts used to determine ^{220}Rn is propagated from the resulting errors in the two count periods, such that:

$$\varepsilon_{220Rn} = ((\varepsilon_1)^2 + (\varepsilon_2)^2)^{1/2} \quad (3.5)$$

where: ε_{220Rn} = error in counts due to ^{220}Rn; ε_1 = error in 1st count (used for ^{220}Rn measurement); ε_2 = error in 2nd count (used for ^{222}Rn measurement). At low soil gas ^{220}Rn and ^{222}Rn concentrations (on the order of 7.5 kBq m^{-3} (200 pCi l^{-1})), typically the first count (1 min) is approximately 50 cpm and the second count (5 min) approximately 200 cpm. At these levels the counting error ε_1 of 14% and ε_2 of 7% (1σ) result in a counting error ε_{220Rn} of approximately 16% (1σ). At higher soil gas ^{220}Rn and ^{222}Rn concentrations observed at a New Jersey site on the order of 200 kBq m^{-3}

(Hutter and Feely, 1990), where the count rates in both counting periods commonly exceed 2000 cpm, the ε_{220Rn} is estimated to be only 3% (1σ).

Although this method was developed for soil gas measurements, the technique can also be applied to indoor air measurements, albeit with higher uncertainties due to low count rates. Compared to soil gas levels, at relatively low ^{222}Rn and ^{220}Rn concentrations such as those found in buildings, e.g., 185 Bq m^{-3} (5 pCi l^{-1}), the count rate using similar apparatus may be only 8 - 10 counts per minute, so that the error in ^{220}Rn and ^{222}Rn measurements due to counting statistics alone at such low levels may be 33% (1σ) or more.

Uncertainties are also introduced during calibrations of the apparatus, which includes an error from counting statistics as well. The uncertainty introduced for both ^{222}Rn and ^{220}Rn from calibrations was estimated to be 5% (1σ).

The final source of uncertainty when using this technique to measure soil gas ^{220}Rn and ^{222}Rn are errors introduced during sampling. These random errors may be caused by leaks in the scintillation cells, leaks in any tubing used due to holes or loose connections, etc. These sampling errors can be estimated by using the standard rule for propagating independent errors, i.e., square root of the sum of the errors squared, as the errors introduced due to counting statistics, the errors from calibrations and overall uncertainty have all been previously estimated. Using error values for typical soil gas ^{220}Rn and ^{222}Rn concentrations, i.e., for ^{220}Rn: 20.7% overall error, 8% counting error and 5% uncertainty from calibrations; for ^{222}Rn: 10.8% overall error, 4.5% counting error and 5% for the uncertainty from calibrations, result in estimates of sampling errors of 8.5% for ^{222}Rn and 18.4% for ^{220}Rn.

Measuring low levels of ^{220}Rn in the presence of high concentrations of ^{222}Rn increases the error in the ^{220}Rn measurement. That is, if the counts in the first counting period become mostly counts due to ^{222}Rn and progeny, the error in the resulting net counts due to ^{220}Rn is larger than if the ^{222}Rn and ^{220}Rn concentrations were similar,

i.e., the error in the difference between two increasingly large numbers becomes larger. This situation is not expected to cause a problem in soil gas measurements, as ^{220}Rn and ^{222}Rn concentrations are normally of the same magnitude. However, for indoor measurements, ^{222}Rn and ^{220}Rn concentrations are often vastly different, and this method may only be useful in arriving at an order-of-magnitude ^{220}Rn level.

It is important to use as a basis of the total uncertainty data obtained in the environment that is to be sampled, if possible. In many reports of ^{222}Rn investigations, the reported errors are based on the calibration errors determined in a ^{222}Rn chamber, where conditions are close to ideal. Sampling soil gas, or even indoor air, can greatly increase random errors, and thus the total uncertainty in a measurement should be the error reported. The ill-treatment of reported errors associated with ^{222}Rn measurements leads to a questionable data base from which to further the science (Harley, 1993). Reporting an error based on the standard deviation of a large population of duplicate field measurements gives a much more meaningful description to the actual measurement value and the probability that the sample is valid and has a real basis from which to interpret and make conclusions. It is not common practice, however, because these numbers are normally much larger than errors obtained in a chamber where complicating measurement conditions are purposefully minimized.

Method Evaluation

The method described for measuring the ^{220}Rn concentration was compared with the two-filter technique for ^{220}Rn concentration determination using the EML Radon, Thoron and Progeny Exposure Facility. The two-filter technique for ^{220}Rn concentration determination requires several hours, as the ^{220}Rn progenies deposited on filters need to be continually counted for at least 2 - 3 hours after sampling. The results are summarized in Table 1 where the Nominal Concentration refers to ^{220}Rn concentration measured via the two-filter technique; # of Samples refer to the number of single measurements performed in parallel (using the method described in this paper) while the two-filter

measurement was in progress; Arithmetic Mean, Standard Deviation and % are results of all the ^{220}Rn measurements using the method described for each Nominal Concentration.

Table 3.1 Results of ^{220}Rn method tests in EML Radon, Thoron and Progeny Exposure Facility.

Nominal Concentration ($Bq\ m^{-3}$)	# Samples	Arithmetic Mean ($Bq\ m^{-3}$)	Standard Deviation (1σ) ($Bq\ m^{-3}$)	%
550	5	480	110	22.9
925	8	918	104	11.3
1300	4	1202	410	34.1
4800	4	4995	866	17.3

The method for calculating the ^{222}Rn concentration was also tested in the EML Radon, Thoron, and Progeny Exposure Facility. The ^{222}Rn concentration levels in the chamber ranged from 50 to 80 $pCi\ l^{-1}$, with the method described achieving an average error (1σ) of less than 7%, versus approximately 5% using longer 2- to 3- hour-wait-methods.

Due to the relatively small number of test measurements (n = 21), a conservative estimate of 500 $Bq\ m^{-3}$ (13 $pCi\ l^{-1}$) is the lowest ^{220}Rn concentration that can be measured using this technique while maintaining an overall uncertainty no greater than approximately 30%. At this concentration level, however, the uncertainty can be decreased to 20 to 25% if multiple samples are obtained in series and an arithmetic mean is obtained.

Conclusions

- It has been shown that, by using a special two-count protocol, a scintillation cell and photomultiplier tube can be used for rapid (15 minute) simultaneous determination of both ^{222}Rn and ^{220}Rn in soil gas.
- The overall uncertainties when using this method to measure typical soil gas ^{220}Rn and ^{222}Rn concentrations, i.e. greater than approximately 2000 Bq m^{-3} (54 pCi l^{-1}), were determined to be 20.7% and 10.8% (1σ), respectively, determined from analyses of duplicate field measurements.
- The lowest ^{220}Rn concentrations that can be measured using this technique while maintaining a reasonable uncertainty is approximately 500 Bq m^{-3} (13 pCi l^{-1}). At this concentration level, the overall uncertainty in the measurement is estimated to be ± 30% for single measurements, decreasing to approximately ± 20 - 25% if at least three measurements sampled in series are arithmetically averaged. Lower concentrations are able to be measured, however, with higher uncertainties.
- At the very low count rates encountered in indoor measurements, this method for ^{220}Rn determination is neither very precise nor accurate, as the uncertainty increases with decreasing count rates. If at least three samples are obtained for determination of indoor ^{220}Rn concentrations, the total uncertainty is expected to be at least 50%, assuming concentrations on the order of 150 Bq m^{-3}. Nevertheless, this method may be useful to obtain an order-of-magnitude indoor ^{220}Rn concentration.

References

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Hutter, A. R., $^{220}Rn/^{222}Rn$ (thoron/radon) as indicators of soil gas transport, Geological Society of America Abstracts with Programs, vol. 25, no. 6, October, 1993.

Li, Y., S. D. Schery and B. Turk, Soil as a source of indoor ^{220}Rn, Health Physics, vol. 62, no. 5, 1992.

United Nations Scientific Committee on the Effects of Atomic Radiation. Ionizing radiation: Sources and biological effects. New York: United Nations; 1982.

3.2 Field Procedures

It is widely thought that at about 1.0 m depth, atmospheric effects on the soil gas ^{222}Rn concentration become negligible. Therefore, for the purposes of this study, a 1.0 m sampling depth was chosen so these influencing parameters would not dramatically affect the soil gas ^{222}Rn and ^{220}Rn concentrations and hence would not have to be incorporated into the final analysis of understanding the variability in the data.

The detailed field procedures used for this method have been incorporated into a project QA Plan at EML and are presented here to clarify the nomenclature used throughout this report:

1. At each measurement site there are 4 to 6 sampling locations.
2. Each of the 4 to 6 sampling locations are linearly spaced about 3 m apart.
3. At each of the sampling locations at each measurement site, sampling tubes of 1.0 m length and 0.95 cm I.D. are permanently emplaced into the soil.
4. At two (or more, depending on local soil conditions) of the sampling locations at each measurement site, sampling tubes of length 0.25, 0.5, and 1.5 m and 0.95 cm I.D. are additionally permanently placed into the soil. These four sampling tubes at a sampling location are spaced 5 cm apart along a line trending perpendicular to the direction of the linear arrangement of the sampling locations.
5. Two tubes of the same length (usually 1.0 m, or, 1.5 m if the soil permits) are placed within 10 cm of each other at at least one sampling location at each measurement site.
6. At the measurement site, a 5-minute-background count for each scintillation cell to be used is obtained before the sample is drawn. The counts per minute (cpm) from the sample must turn out to be at least 3 times the background cpm. If this requirement is not met, a new sample must be obtained.

7. Calibrations of scintillation cells-^{222}Rn detector apparatus are performed on at least a semi-annual basis using the EML environmental ^{222}Rn chamber
8. On a bi-weekly basis, soil gas samples are obtained by drawing soil gas from the tubes using a portable battery-operated DuPont[2] (or similar type) pump, at a flow rate of 1 to 3 l min^{-1} for one minute.
9. The sample is drawn directly into scintillation cells that have been previously inserted into the EDA Instruments Inc.[3] RDA-200 ^{222}Rn detector.
10. The pump is turned off, the tubes to the scintillation cell are removed and the cap to the EDA RDA-200 ^{222}Rn detector is screwed on. A one-minute-count is then obtained. The time between the end of pumping and the start of counting must be less than 90 seconds (one-and-a-half half-lives of ^{220}Rn), and should be as small as possible. Normally, this time is about 10 seconds. This time between the end of pumping and the start of counting must be recorded to within $\pm$ 1 second.
11. The scintillation cell is then removed from the EDA RDA-200 and another sample obtained.
12. At least 5 minutes after the soil gas sample is drawn into the scintillation cell, and no longer than three or four days, a 5- or 10-minute-count is obtained using the EDA RDA-200. Normally, these counts are obtained

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[2]DuPont Model # 2500 Air Sampler
E.I. du Pont de Nemours & Co. (Inc.)
Wilmington, DE 19898

[3]EDA Model # RDA-200 Radon Detector
EDA Instruments Inc. (Scintrex)
4 Thorncliffe Dr.
Toronto, Ontario
Canada M4H 1H1

either on-site at the end of sampling, or within an hour or two at a convenient (warm and dry) site.

13. Second samplings of soil gas from previously drawn sampling tubes are obtained at a frequency of 5 to 10% of all samples obtained (i.e., duplicates). Additionally, at the same frequency, selected samples are counted a second time.
14. Concentrations are calculated using the program shown in Appendix A. This program has been verified to be accurate by comparisons with manual mathematical methods, and by checks with the known ^{222}Rn concentrations in the EML Environmental ^{222}Rn Chamber.

3.3 Subsurface Characterization

In order to better characterize each site, the soil depth and/or depth to bedrock was determined by an electrical resistivity method using a Bison Earth Resistivity Meter.[4] The technique is based on differing resistivities of different types of subsurface, e.g., soils having lower resistivities (higher conductivities due to ubiquitous water) than rocks (with higher resistivities, or lower conductivities, due to a lack of water). With this technique, it is possible to determine a subsurface profile (i.e., soil horizons, sand and gravel deposits, depth to bedrock, saturated zones, etc.) at a single spot in about one hour.

For the current study, two sets of electrodes were set out at distances from the point where the profile is determined at 15.2, 22.9, 30.5, 40.6, 66.0, 94.0, 134.6, 195.6, 281.9, 406.4, 584.2, 845.8, 1209.0, 1745.0, and 2512.1 cm in a straight line. Current is sent to an electrode pair of equal distance from the center point and the voltage drop is then measured across two different electrodes also of equal distance from the center point but closer than the current electrodes. The electrodes are positioned in a Wenner array, so

[4]Bison Earth Resistivity Meter Model # 2350B
Bison Instruments, Inc.
5708 W. 36th St.
Minneapolis, MN 55416-2595

that the distances among the four electrodes in use are the same, and that distance is defined as the A-spacing (Telford et al., 1976). A series of such measurements are obtained with approximately 12 different A-spacings. Figure 3.4 shows the basic configuration for one such measurement. The A-spacings for each measurement used in the current study were 30.4, 45.8, 61.0, 81.2, 132.0, 188.0, 269.2, 391.2, 563.8, 812.8, 1168.4, and 1691.6 cm.

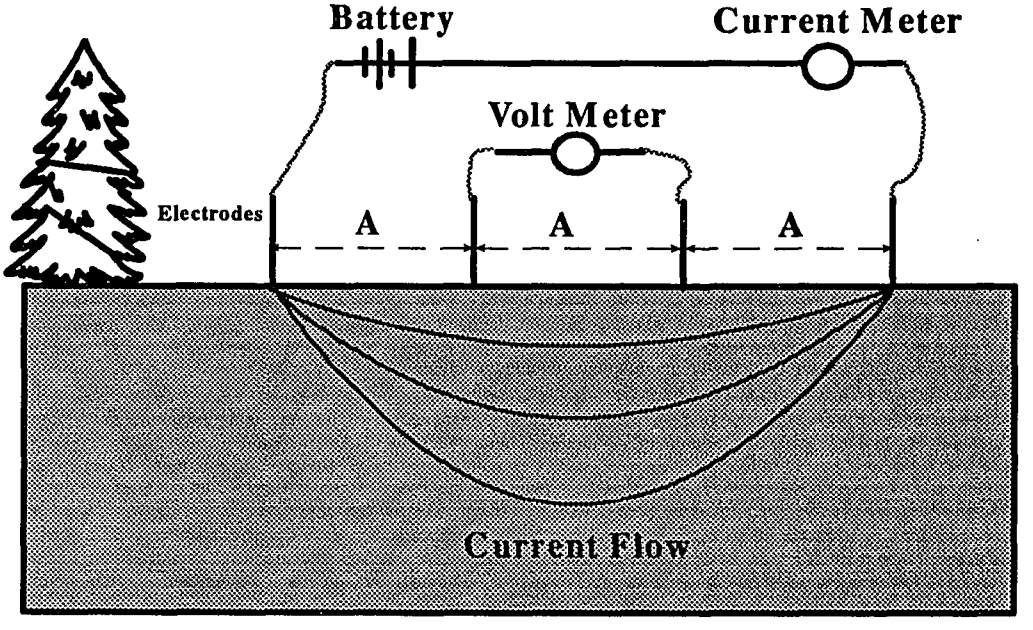

Figure 3.4 Cartoon showing the components of an electrical resistivity measurement for the determination of subsurface profile. A denotes the A-spacing between electrodes.

The maximum depth that can be profiled depends upon how far the array of electrodes, and thus larger A-spacings, are continued. The farther out the array of electrodes is continued, the deeper the modeling of the resistance measurements can be performed. The farthest set of electrodes used in this study were 2512.1 cm (about 82 feet) from the center point, translating into a maximum depth of determination of 20 meters.

Each set of current and voltage readings for a particular A-spacing is then converted to an apparent resistivity, according to the following equation:

$$\Omega = 2\pi A(V/I) \qquad (3.6)$$

where:

Ω = apparent resistivity, ohm-m

A = A-spacing, m

V = Voltage

I = Current

The series of apparent resistivities are plotted against the A-spacings on a log-log basis, and using commercially available computer-modeling software[5], the depth of each layer of differing resistivities is determined from the shape of the resulting curve. The modeling technique is based on a ridge-regression inversion algorithm, and an estimated error based on the least squares error between the data and the resulting curve is produced. Changing the input model parameters, i.e., number of layers and estimated resistivities, produces several possible scenarios. The "answer" is selected according to the smallest estimated error produced by the ridge-regression inversion algorithm.

In addition, soil samples have been collected from Site 1. The total ^{238}U and ^{232}Th series concentrations were determined using gamma spectral techniques developed at EML, and are 50.32 ± 0.74 and 92.50 ± 1.11 Bq kg^{-1}, respectively (Miller and dePlanque, 1978).

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[5]Resix
Interprex Limited
PO. Box 839
Golden, CO

3.4 Permeability

As previously discussed, soil permeability and ^{222}Rn diffusion coefficient are two of the key soil-related parameters characterizing soil gas ^{222}Rn transport. For investigations of ^{222}Rn transport, the ^{222}Rn diffusion coefficient can be theoretically determined from the soil water saturation fraction (Rogers and Nielson, 1991). Measuring the soil permeability can give vital information concerning major controlling variable parameters of soil gas transport, such as soil moisture and soil porosity.

The soil permeability of each hole sampled at each site was estimated using flow and pressure differential measurements at the time of sampling. Soil permeability (k) is estimated using the equation (Rogers and Nielson, 1991):

$$k = 5.2 \text{ X } 10^{-4} \, Q / \Delta P \qquad (3.7)$$

Where: k = soil permeability, (m^2); Q = flow rate, ($m^3 \, s^{-1}$); ΔP = suction pressure, (Pa)

3.5 Site Descriptions

Figure 3.5 shows the locations of the three New Jersey field sites.

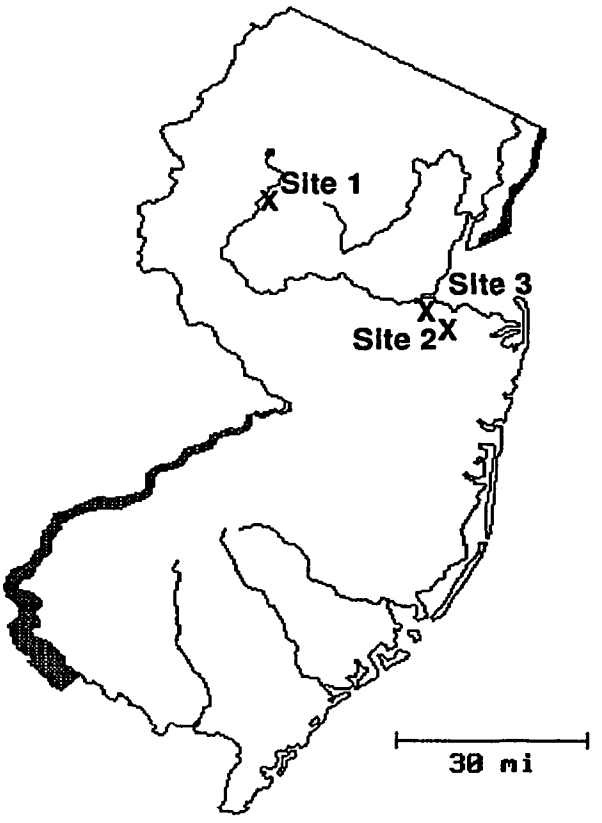

Figure 3.5 New Jersey map showing site locations demarked with an X. Site 1 is located in Chester, Site 2 in Matawan and Site 3 in Cheesequake State Park.

Site 1 is located on the edge of the Reading Prong in central northern New Jersey. The site is situated in an open grassy field on the side of a hill, and is underlain by granitic gneiss. Figure 3.6 is a schematic of Site 1.

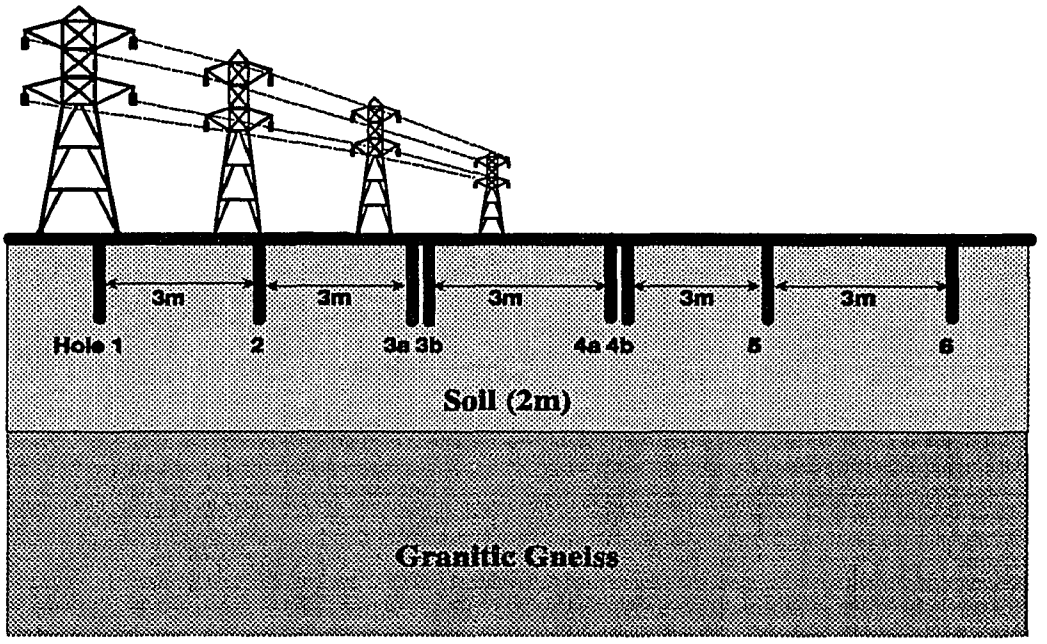

Figure 3.6 Schematic of Site 1 at Chester, NJ. Each tube is 1 m long. Holes 1 through 6 are each placed linearly 3m apart. Holes 3a and 3b are approximately 6 cm apart, as are 4a and 4b.

Six sampling locations with an additional sampling tube placed at two of the locations are present at Site 1 (referred to as holes 1, 2, 3a, 3b, 4a, 4b, 5 and 6). Holes 3a and 3b are approximately 6 cm apart, as are Holes 4a and 4b.

Site 2 is located near the border of the coastal plain and piedmont provinces in Matawan, NJ (Figure 3.7).

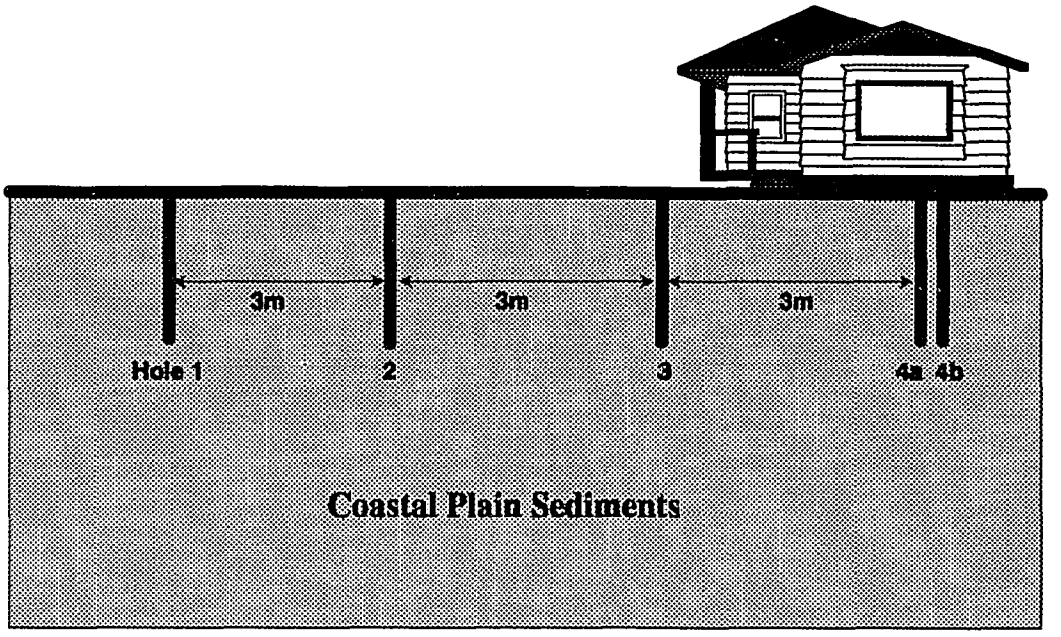

Figure 3.7 Schematic of Site 2 at Matawan, NJ. Each tube is 1 m long. Holes 1 through 4 are each linearly spaced 3m apart. Holes 4a and 4b are approximately 6 cm apart. The coastal plain sediments are at least 20m deep at this location.

At this site, there is only one layer of coastal plain sediments, mainly sand, to a maximum determination depth of the electrical resistivity method of about 20 m. There are five sampling locations with one additional sampling tube at Site 2 (referred to as Holes 1, 2, 3, 4a and 4b). The sampling tubes at Holes 4a and 4b are approximately 5 cm apart. There is virtually no vegetation in the area surrounding the holes.

Site 3 is located within Cheesequake State Park in Cheesequake, NJ. Cheesequake State Park is also near the border of the coastal plain and piedmont provinces, but is situated more within the piedmont province than is Site 2 (Figure 3.8).

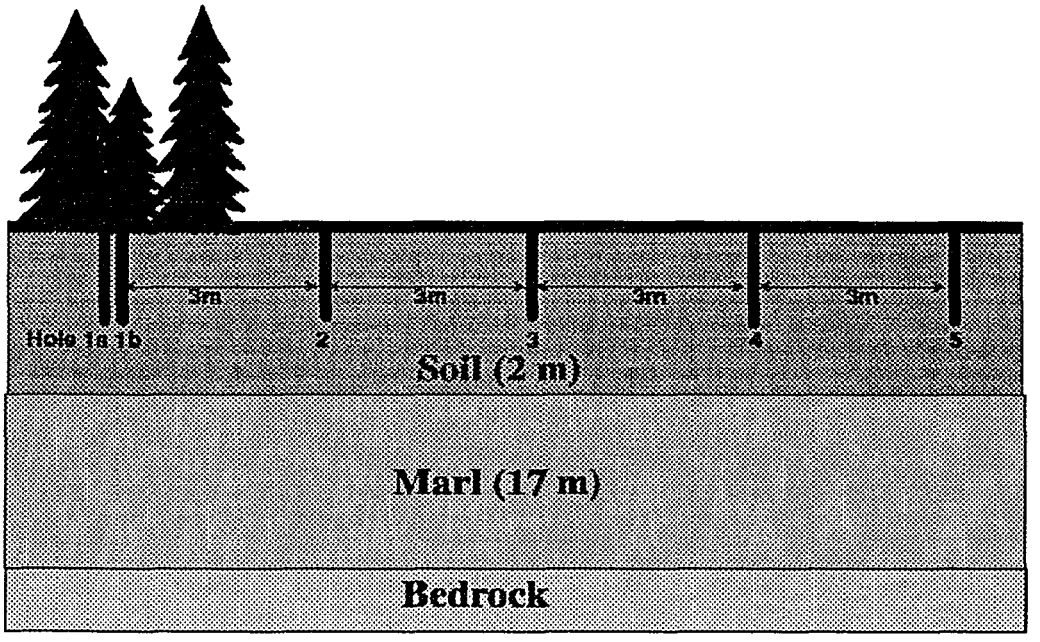

Figure 3.8 Schematic of Site 3 at Cheesequake, NJ. Holes 1 through 5 are linearly spaced 3m apart. Holes 1a and 1b are approximately 6 cm apart.

As shown in Figure 3.8, the site is underlain by a 2 m deep soil, on top of a 17 m layer of marl and sand, which is situated on top of a metamorphic bedrock. At this site, there are five sampling locations, with one additional sampling tube at one of the sampling locations (referred to as Holes 1a, 1b, 2, 3, 4 and 5). The sampling locations are located in a wooded area of the park, with a relatively thick (several cm's) organic layer at the surface.

3.6 Quality Assurance

The quality assurance (QA) of the data presented in this report is governed by the U.S. Department of Energy's Environmental Measurements Laboratory mandate on QA. All research projects conducted under the auspices of EML must have on file a QA self-assessment report. Additionally, it is a requirement that all reports, journal articles, or other project descriptions arising from EML programs must include a section on QA.

In general, quality assurance of the soil gas data was assessed by the following tasks:

(1) Background measurements performed on all cells for every measurement;

(2) Duplicate measurements performed at a rate of 1 in 10;

(3) Calibrations performed on a semi-annual basis in the EML Radon, Thoron, and Progeny Exposure Facility following standard procedures (HASL-300, 1990);

(4) Error analysis for determination of random errors in soil gas ^{222}Rn and ^{220}Rn measurements.

The discussion and interpretation of these items were incorporated into section 3.1, the manuscript submitted for publication. Quality Assurance data are presented in Appendix C.

Chapter 4

DATA PRESENTATION

4.1 Site 1 Soil Gas ^{222}Rn and ^{220}Rn

In order to understand the reproducibility of any observed seasonal variations, it was desired to have at least two years of data for any sampling location. Soil gas ^{222}Rn measurements were obtained from Site 1 beginning in September, 1989 and continue in some manner to the present (for subsequent associated studies within the EML soil gas program). Soil gas ^{220}Rn measurements began in March, 1990 at this site. To meet the two year objective, reported measurements are included until May, 1992. The ^{222}Rn and ^{220}Rn data collected from Site 1 are presented in Figures 4.1 - 4.18. A composite graph of the ^{222}Rn data and a composite graph of the ^{220}Rn data for all the holes are followed by ^{222}Rn and ^{220}Rn graphs for each hole. At Site 2, soil gas ^{222}Rn and ^{220}Rn measurements were obtained from May, 1990 until May, 1992. Data for Site 2 are presented in Figures 4.19 - 4.30, with composite graphs of ^{222}Rn and ^{220}Rn for all holes followed by ^{222}Rn and ^{220}Rn graphs by individual hole. At Site 3, soil gas ^{222}Rn and ^{220}Rn measurements were started in November 1991, however, samples could not be obtained for most of the Winter and Spring due to saturated soil conditions. Attempting to obtain a sample resulted in a muddy mixture being sucked up the sampling tube into the sampling apparatus. Therefore, due to the sparse data set, only the composite ^{222}Rn and ^{220}Rn graphs are shown for this site (Figures 4.31 and 4.32). Error bars are not included in the following figures for clarity. The total uncertainty in the ^{222}Rn measurements was determined to be 10.8%. The total uncertainty in the ^{220}Rn measurements was determined to be 20.7%. These values were derived from an analysis of duplicate samples (see preceding sections for discussion).

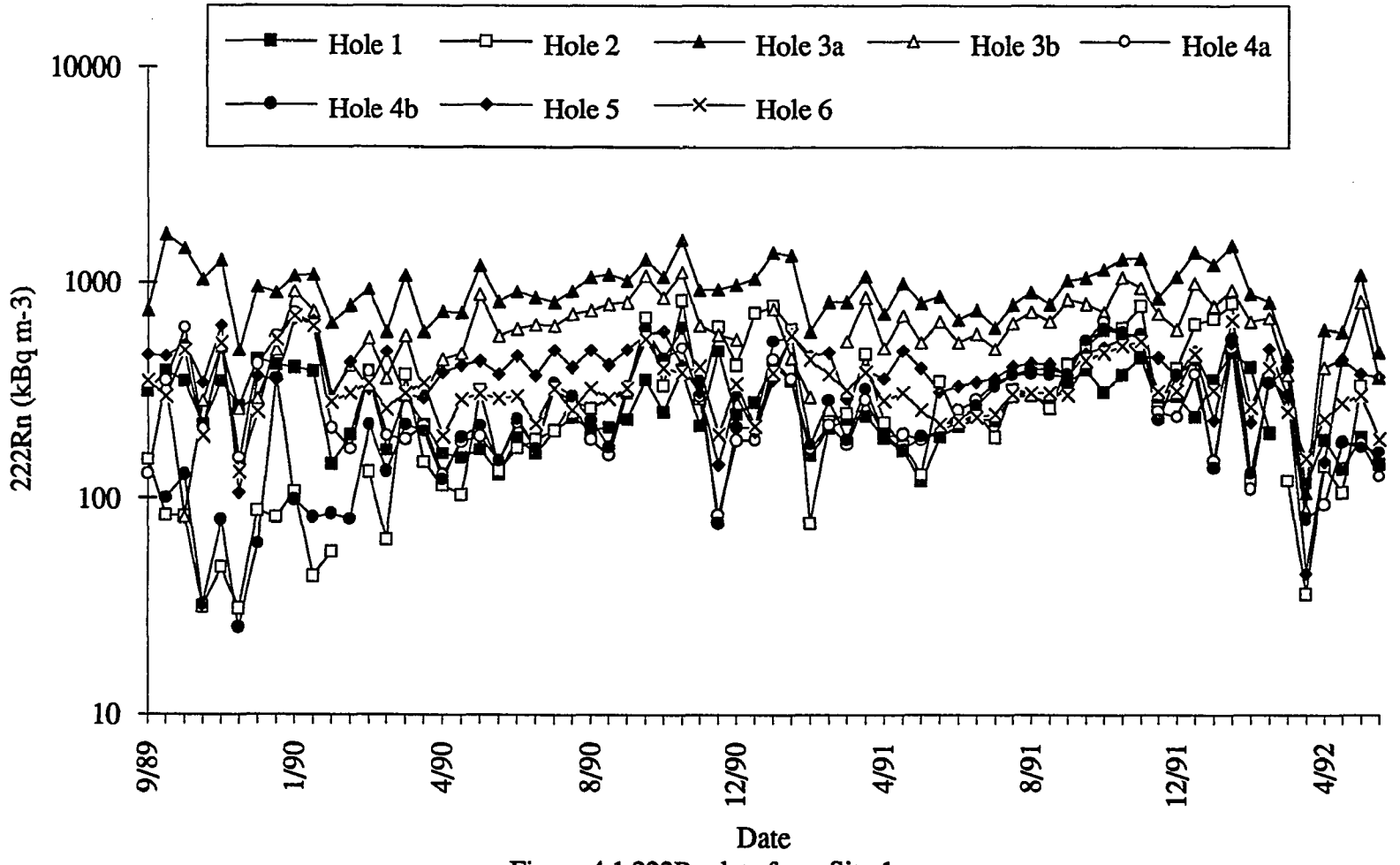

Figure 4.1 222Rn data from Site 1

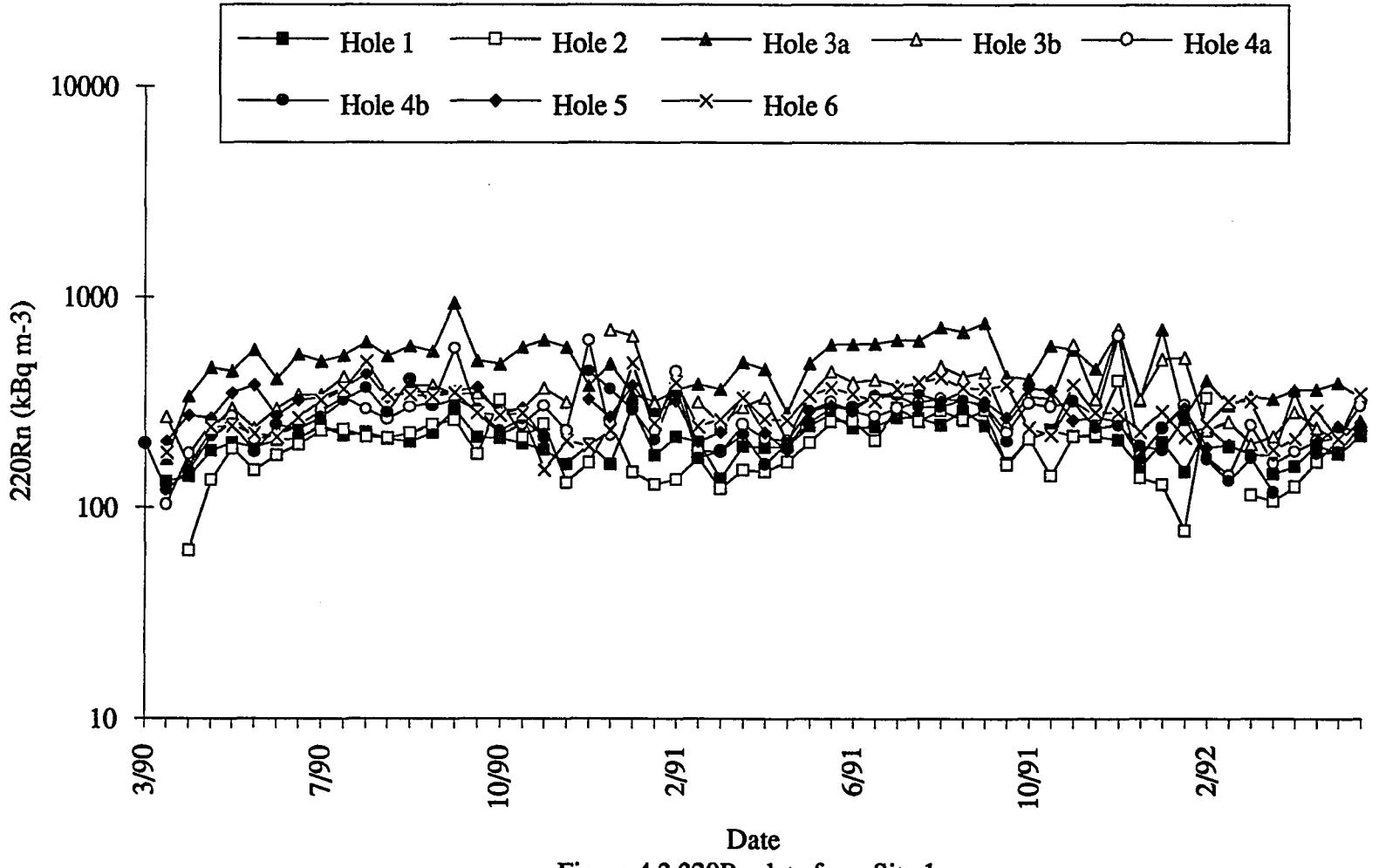

Figure 4.2 220Rn data from Site 1

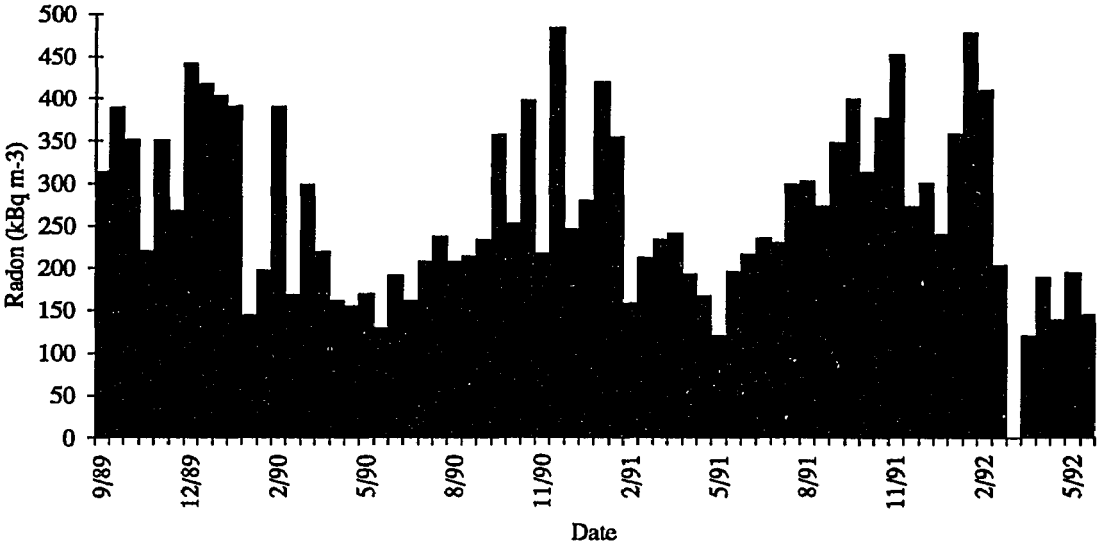

Figure 4.3 ^{222}Rn data from hole 1, Site 1.

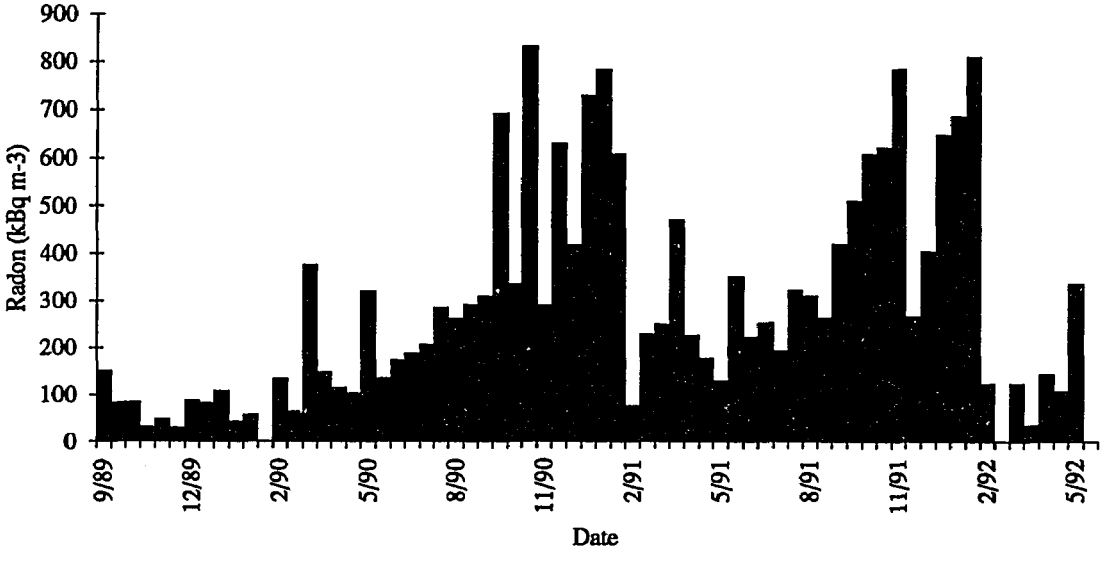

Figure 4.4 ^{222}Rn data from hole 2, Site 1.

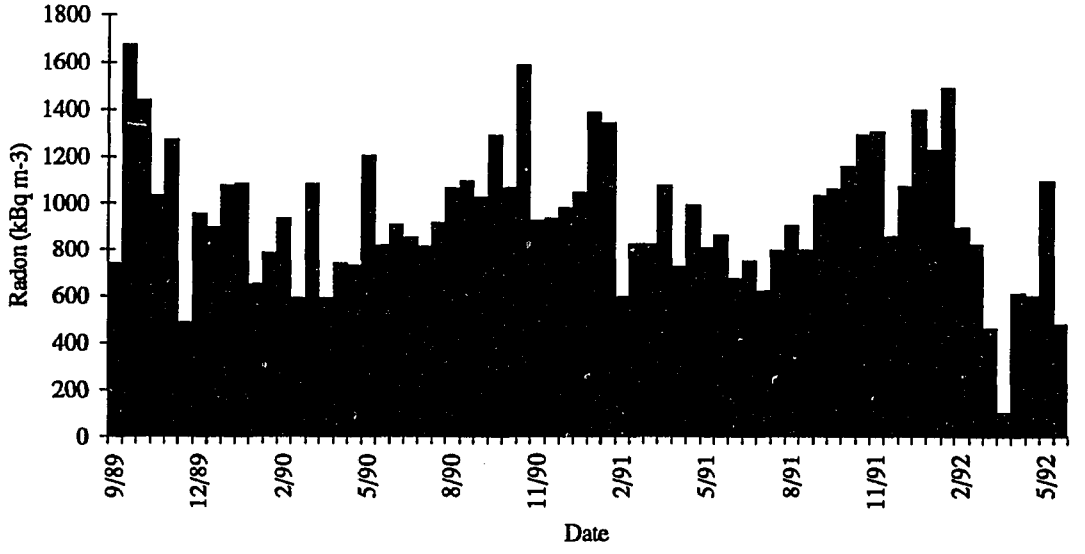

Figure 4.5 ^{222}Rn data from hole 3a, Site 1.

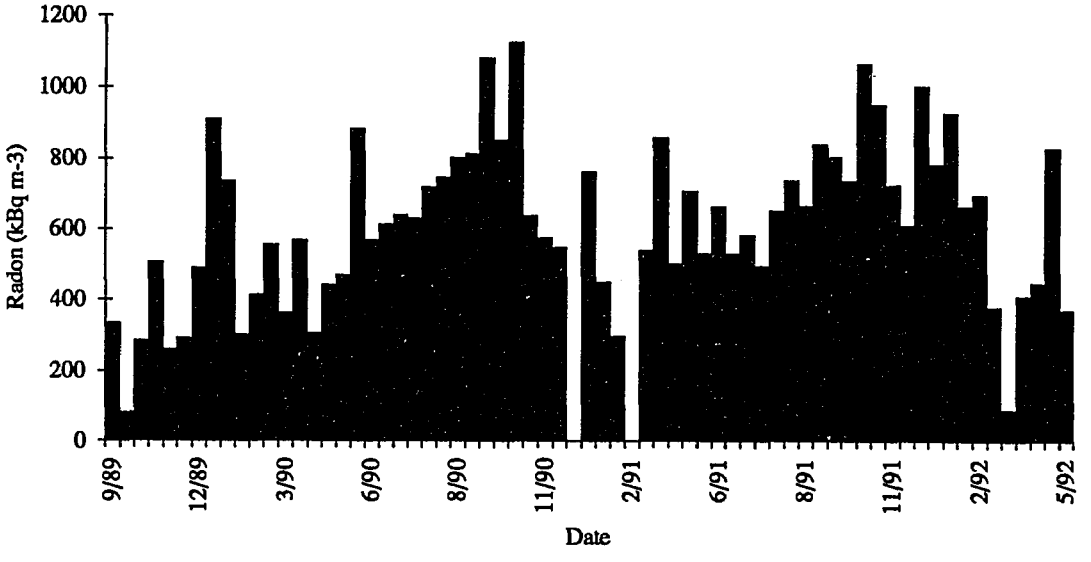

Figure 4.6 ^{222}Rn data from hole 3b, Site 1.

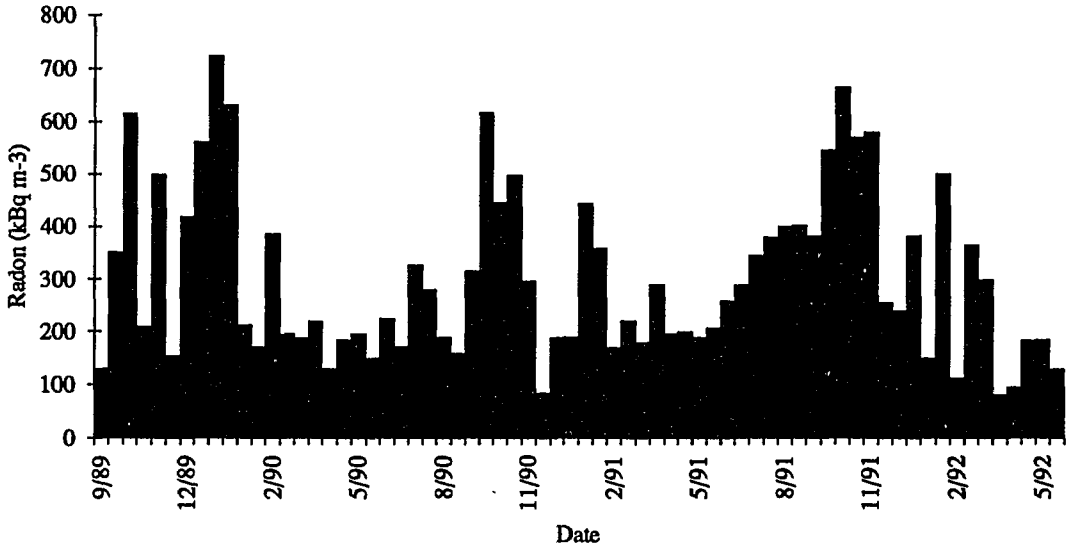

Figure 4.7 ^{222}Rn data from hole 4a, Site 1.

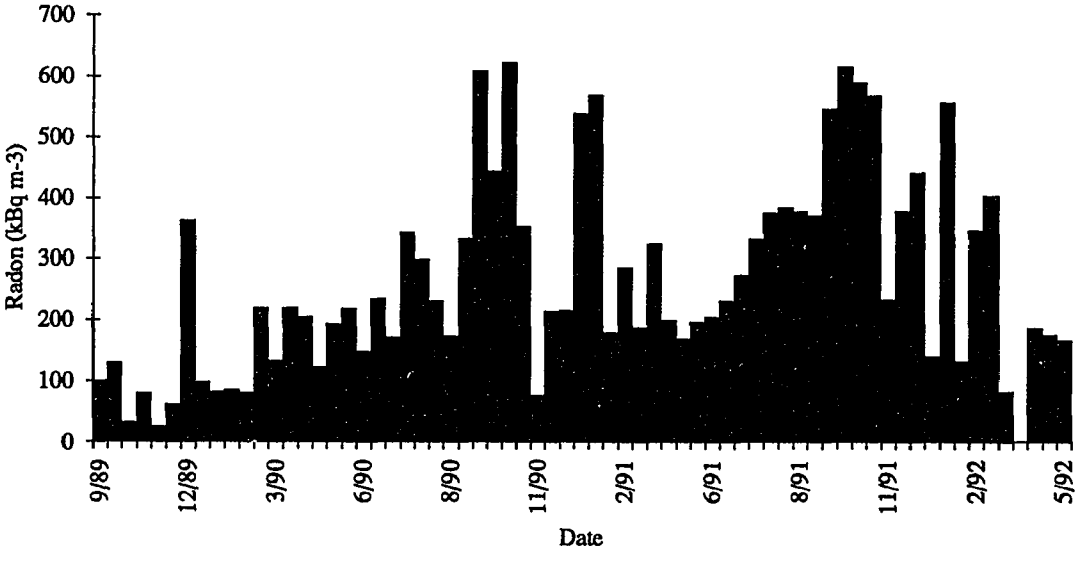

Figure 4.8 ^{222}Rn data from hole 4b, Site 1.

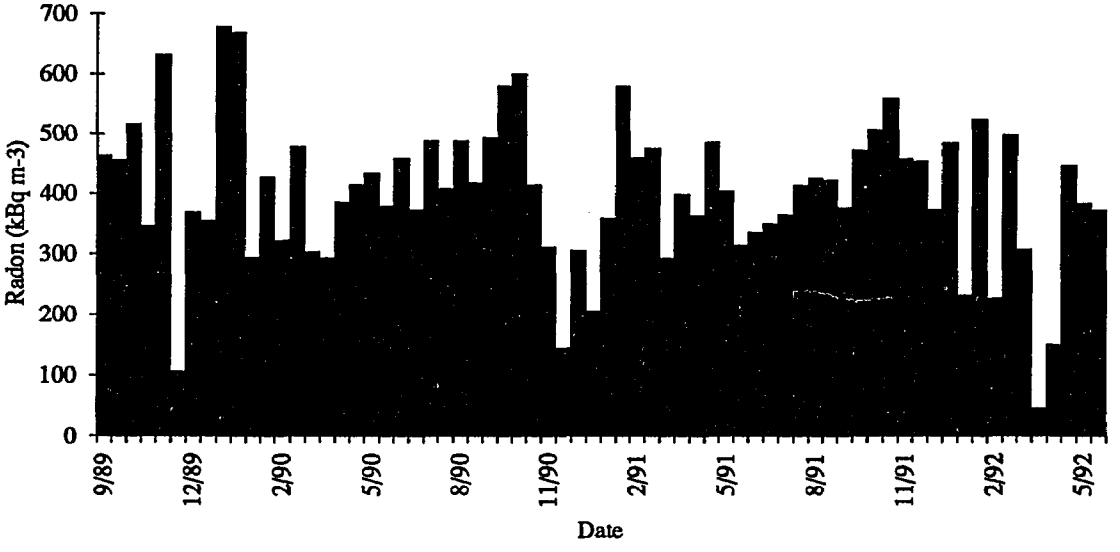

Figure 4.9 ^{222}Rn data from hole 5, Site 1.

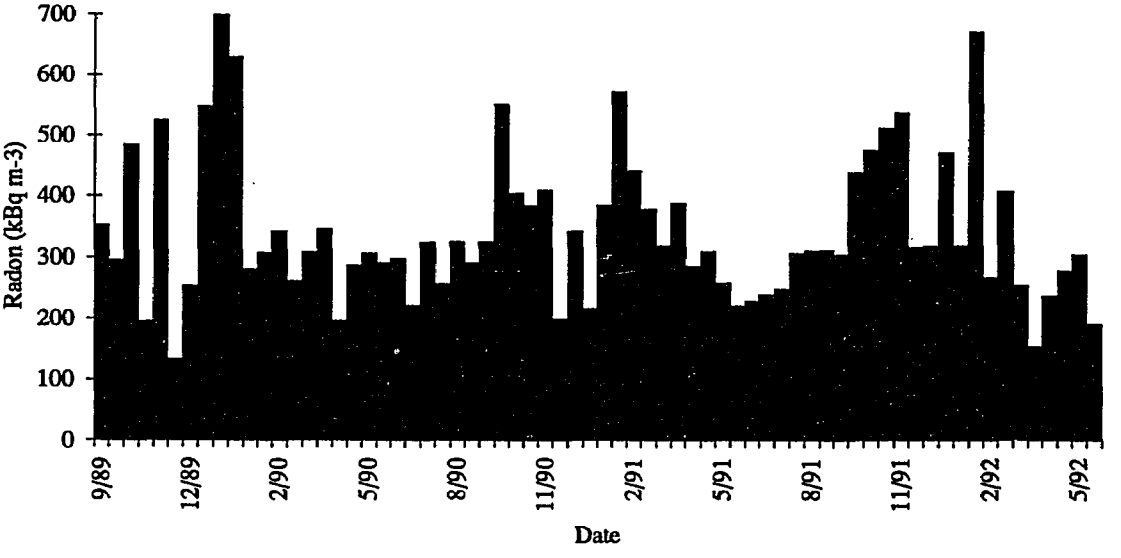

Figure 4.10 ^{222}Rn data from hole 6, Site 1.

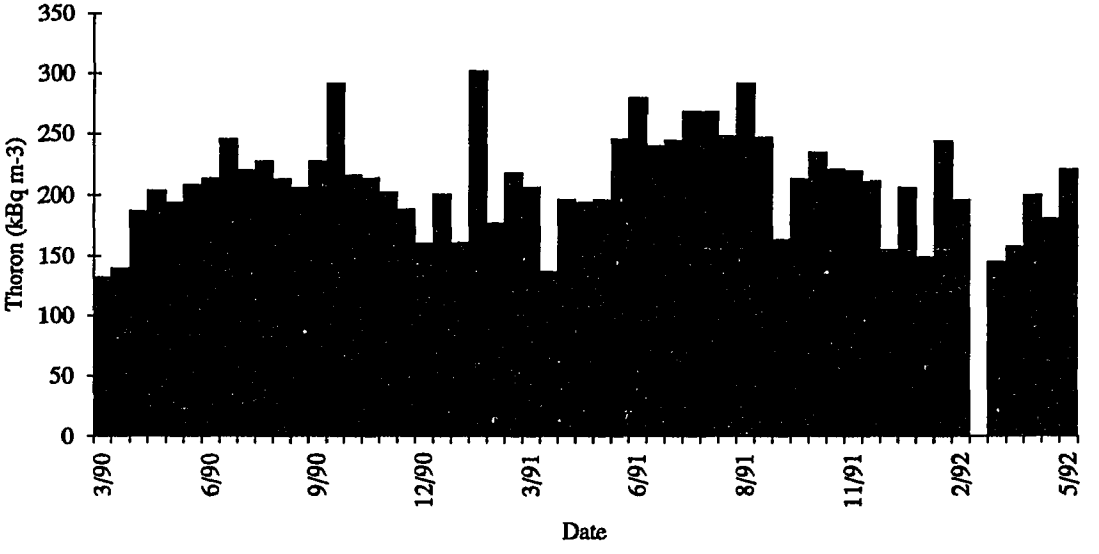

Figure 4.11 ^{220}Rn data from hole 1, Site 1.

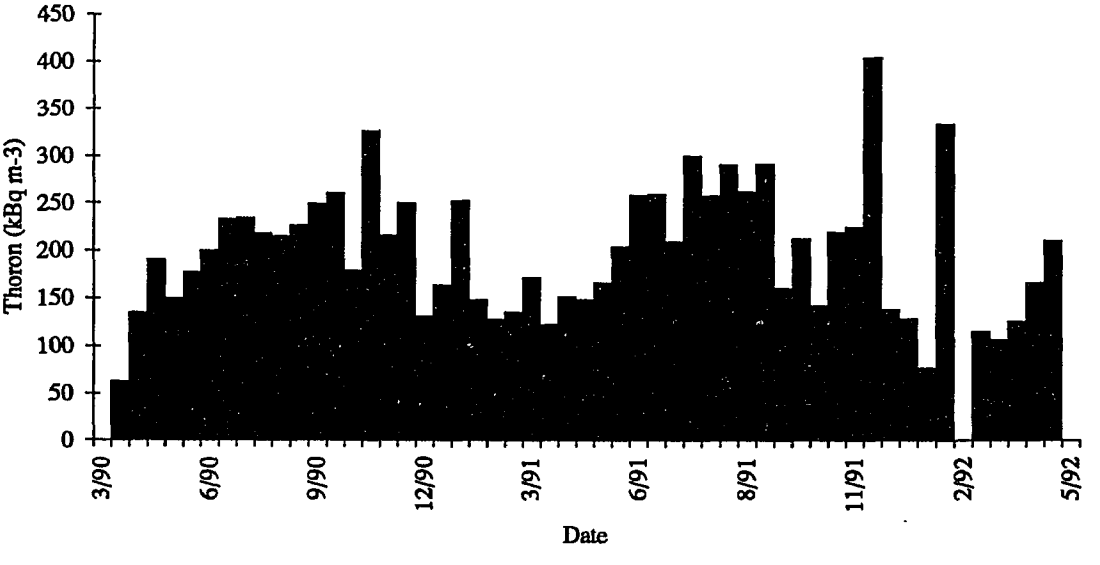

Figure 4.12 ^{220}Rn data from hole 2, Site 1.

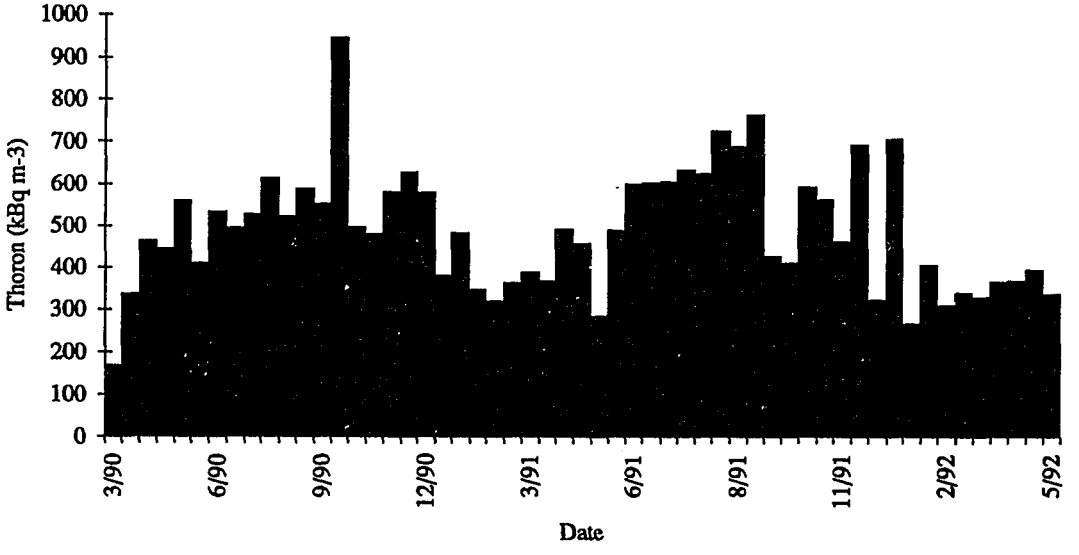

Figure 4.13 ^{220}Rn data from hole 3a, Site 1.

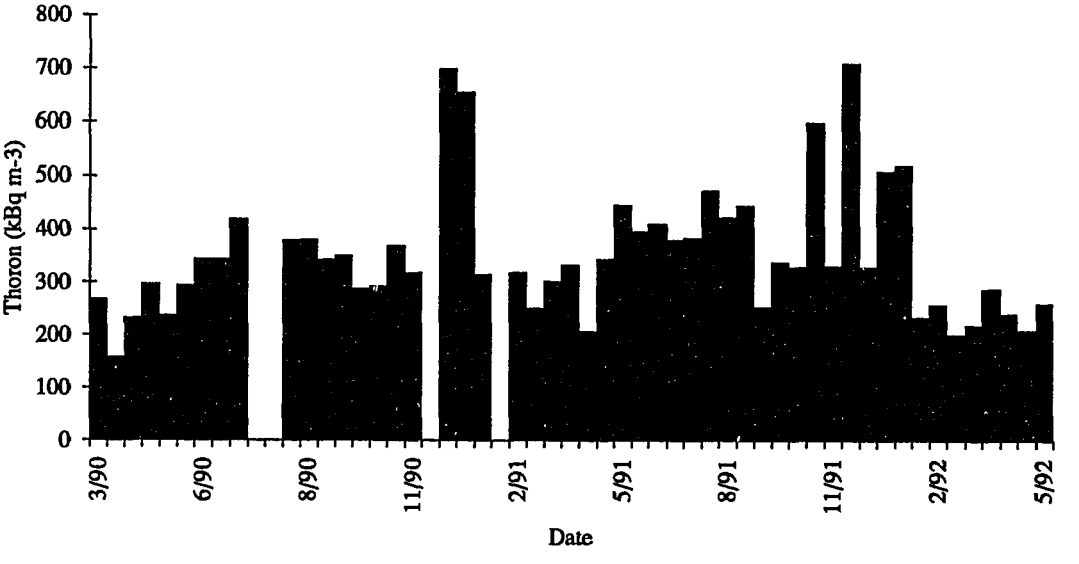

Figure 4.14 ^{220}Rn data from hole 3b, Site 1.

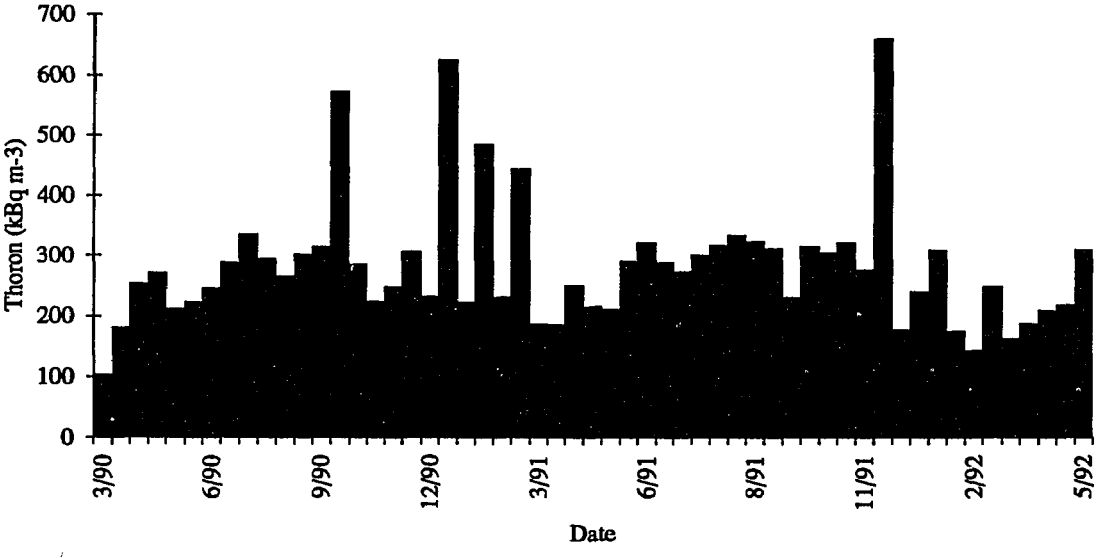

Figure 4.15 ^{220}Rn data from hole 4a, Site 1.

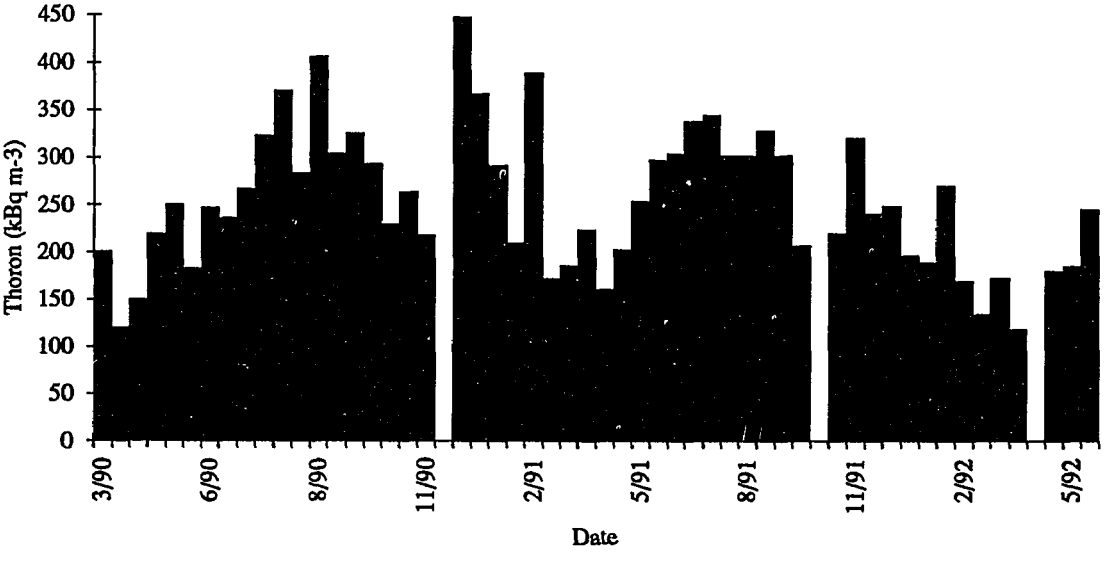

Figure 4.16 ^{220}Rn data from hole 4b, Site 1.

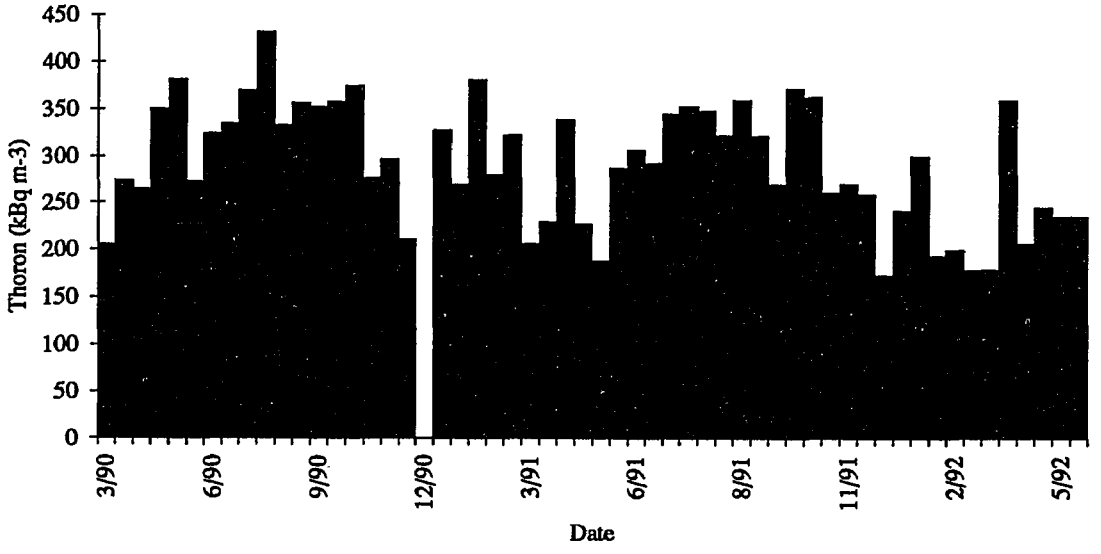

Figure 4.17 ^{220}Rn data from hole 5, Site 1.

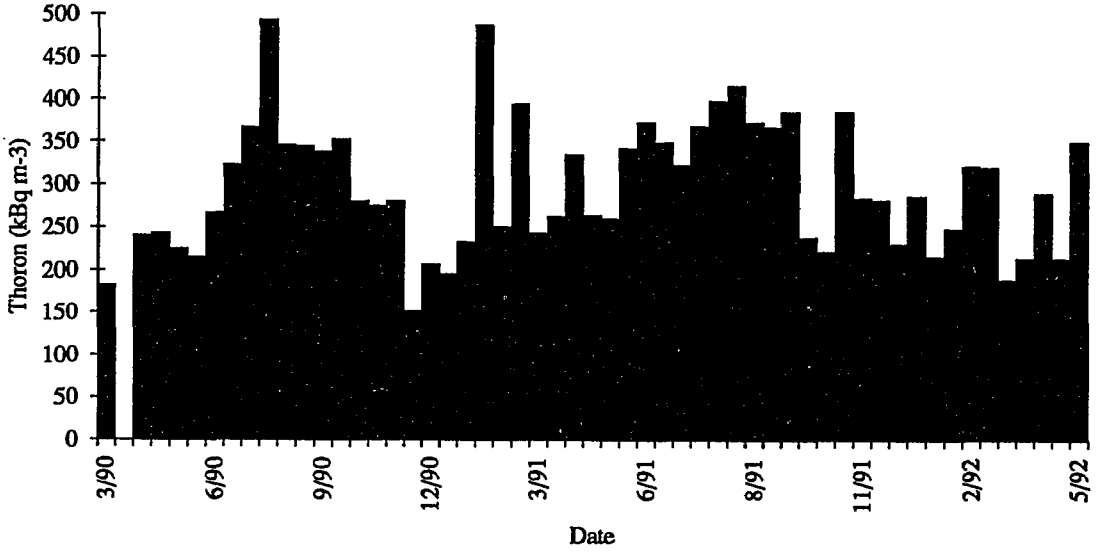

Figure 4.18 ^{220}Rn data from hole 6, Site 1.

4.2 Site 2 Soil Gas ^{222}Rn and ^{220}Rn

Soil gas samples were collected from the Matawan, NJ site (Site 2) from May 1990 through May 1992 on a bi-weekly basis. The collective soil gas ^{222}Rn concentration data for Site 2 are presented in Figure 4.19. Figure 4.20 shows the collective ^{220}Rn soil gas data for Site 2. Figures 4.21 to 4.25 show the ^{222}Rn concentration data for Site 2 holes 1, 2, 3, 4a, and 4b, respectively. Figures 4.26 to 4.30 show the ^{220}Rn concentration data for Site 2 holes 1, 2, 3, 4a, and 4b, respectively. Error bars are not included in the following figures for clarity. The total uncertainty was determined to be 10.8% for ^{222}Rn measurements and 20.7% for ^{220}Rn measurements. These values were derived from an analysis of duplicate samples (see preceding sections for discussion).

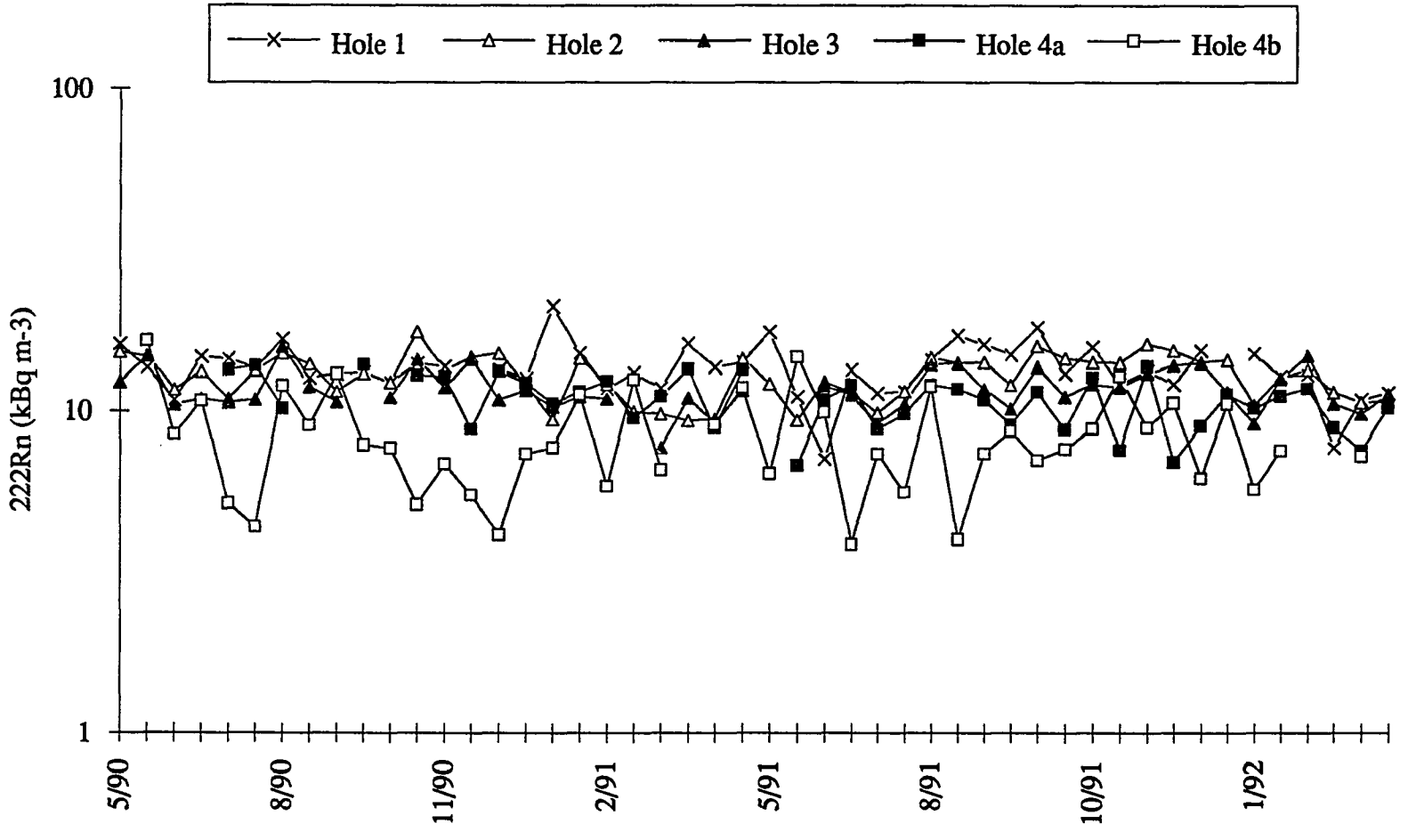

Figure 4.19 222Rn data from Site 2

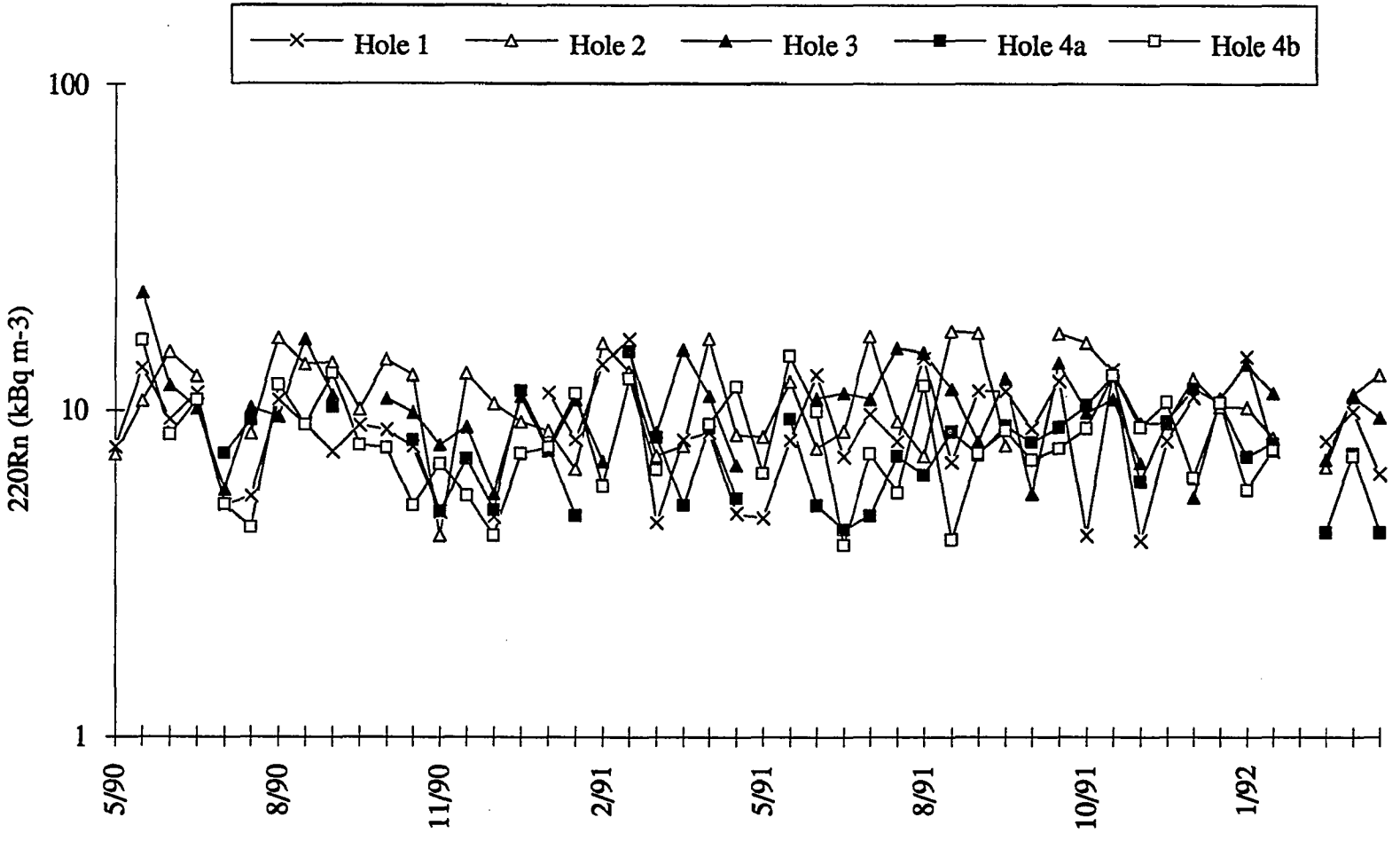

Figure 4.20 220Rn data from Site 2

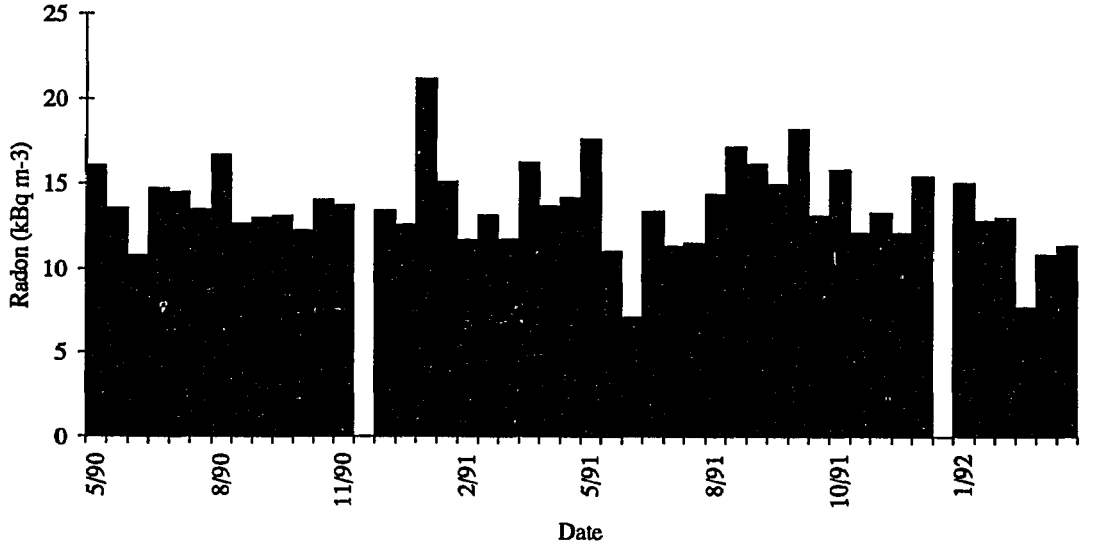

Figure 4.21 ^{222}Rn data from hole 1, Site 2.

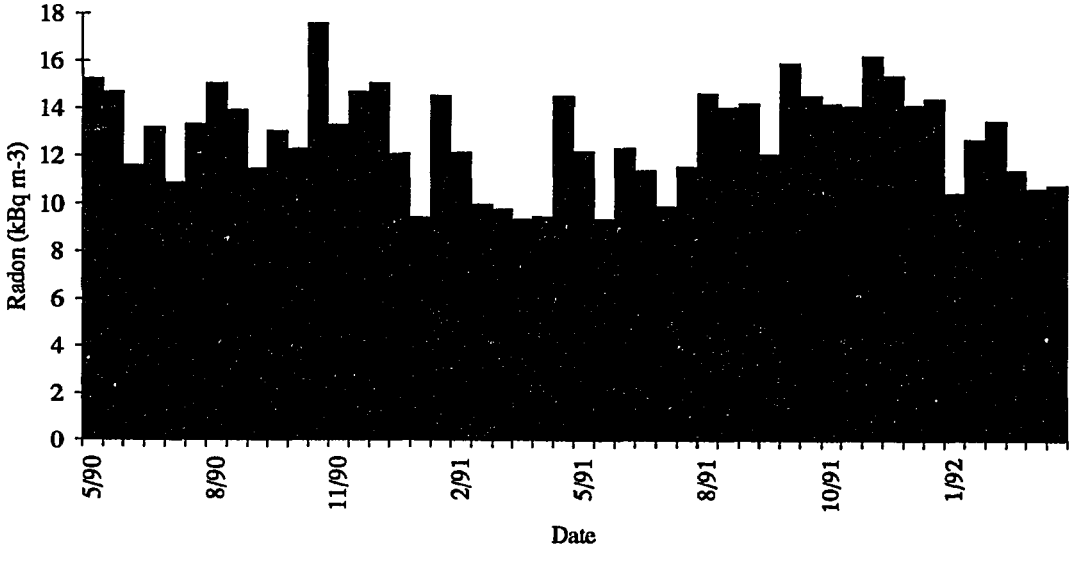

Figure 4.22 ^{222}Rn data from hole 2, Site 2.

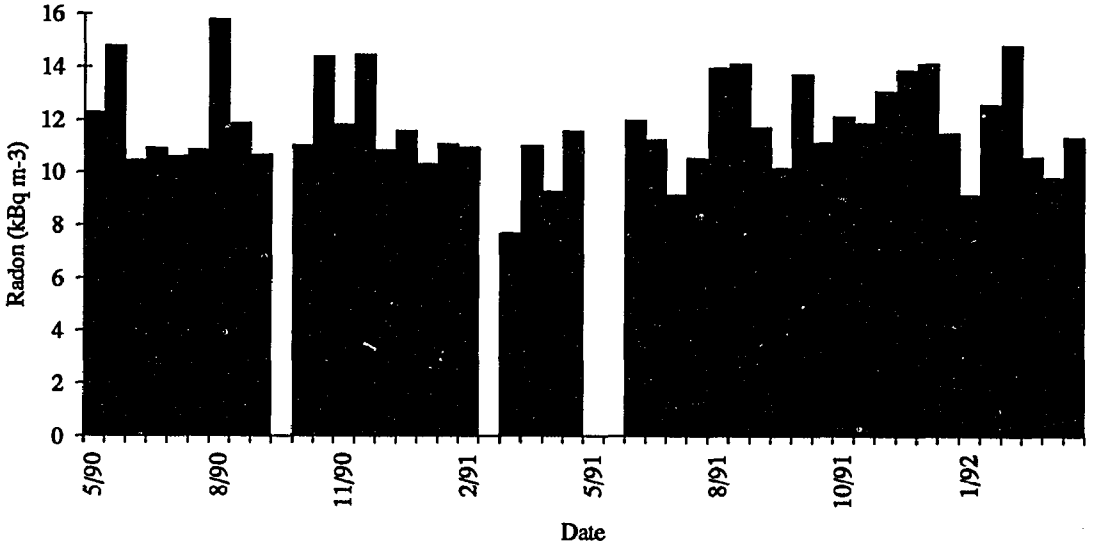

Figure 4.23 ^{222}Rn data from hole 3, Site 2.

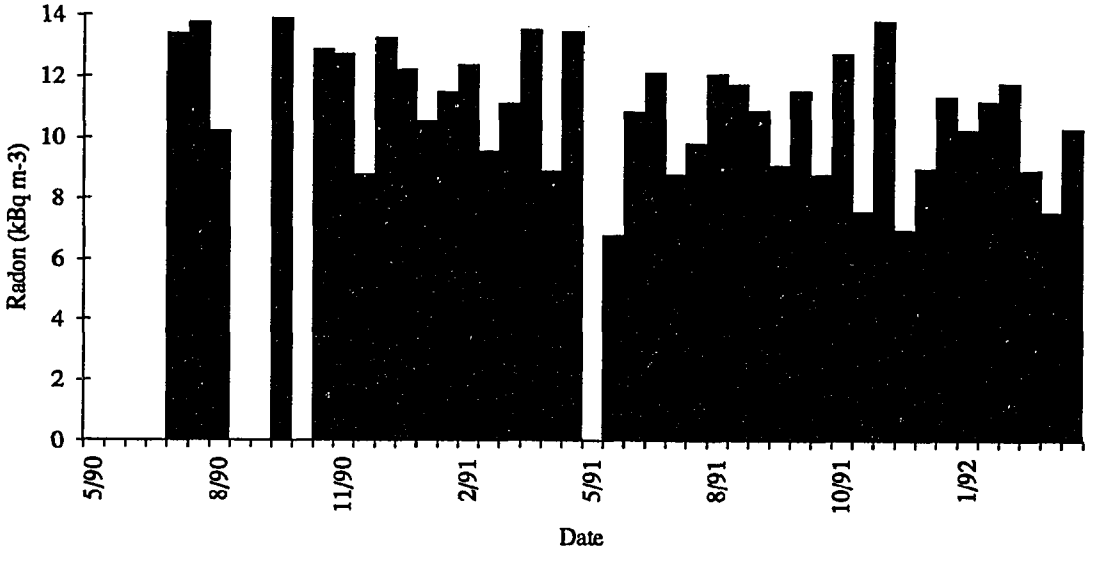

Figure 4.24 ^{222}Rn data from hole 4a, Site 2.

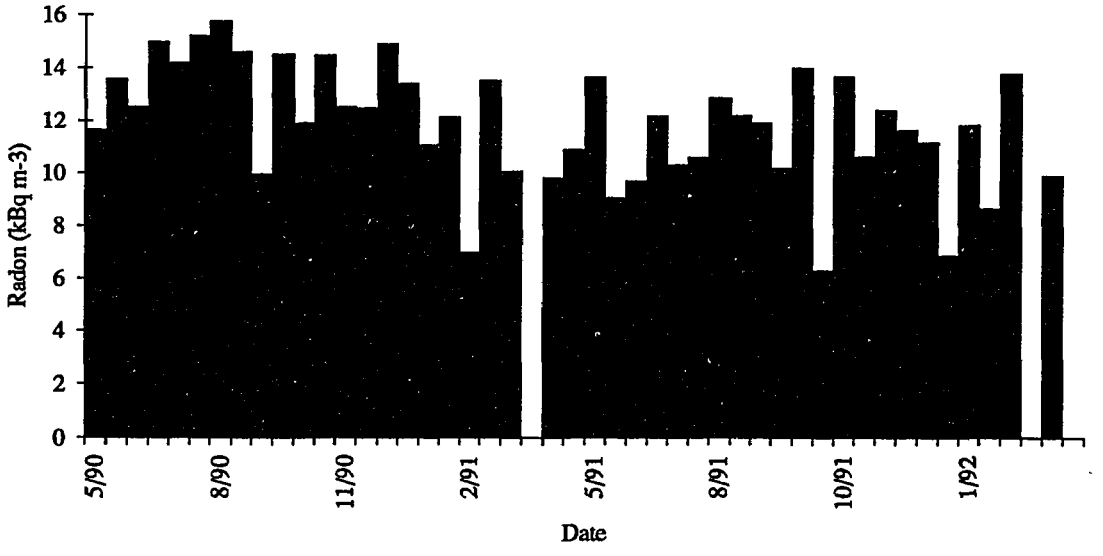

Figure 4.25 ^{222}Rn data from hole 4b, Site 2.

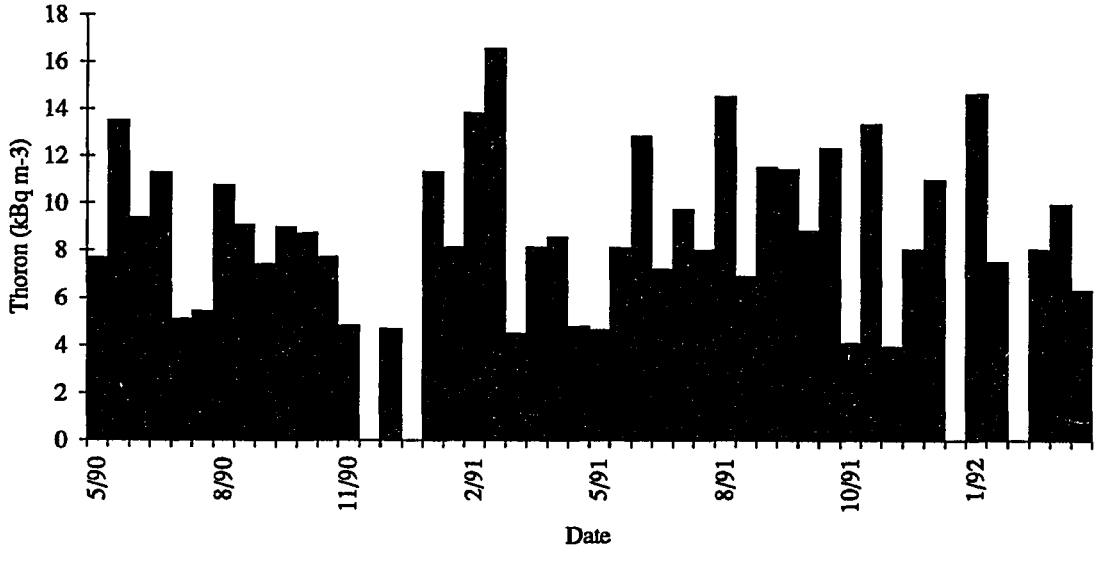

Figure 4.26 ^{220}Rn data from hole 1, Site 2.

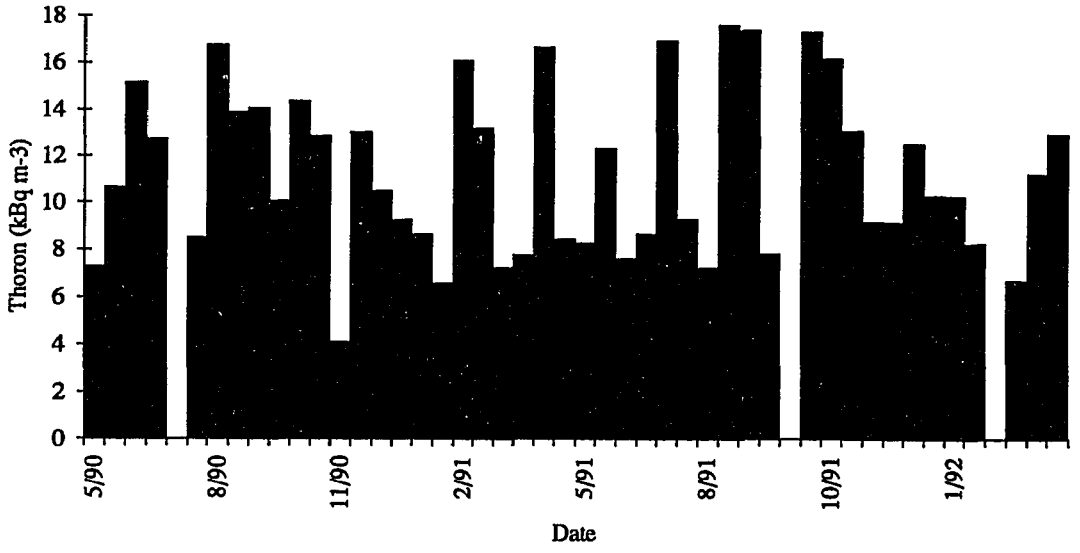

Figure 4.27 ^{220}Rn data from hole 2, Site 2.

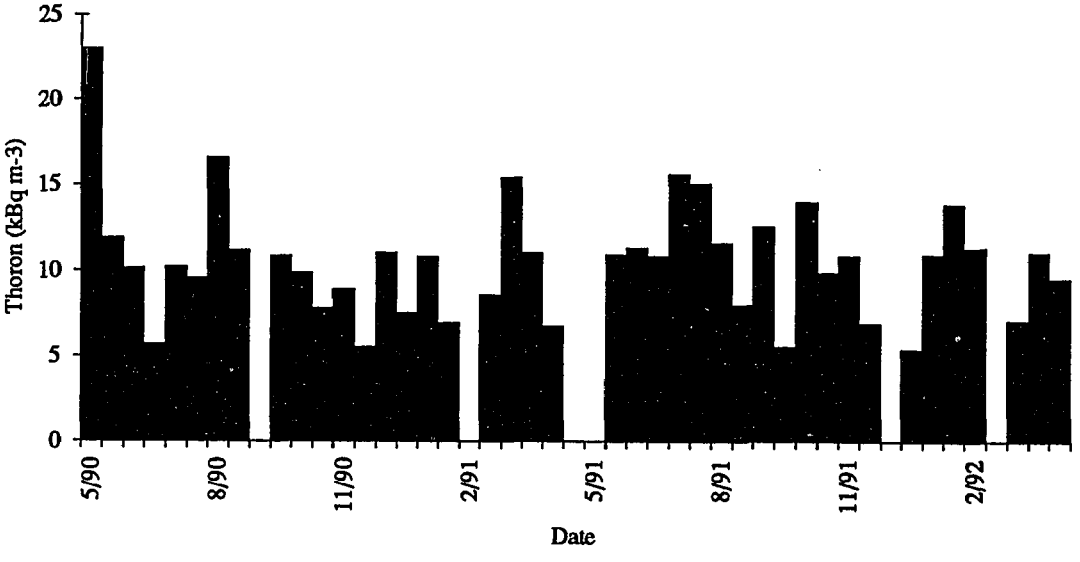

Figure 4.28 ^{220}Rn data from hole 3, Site 2.

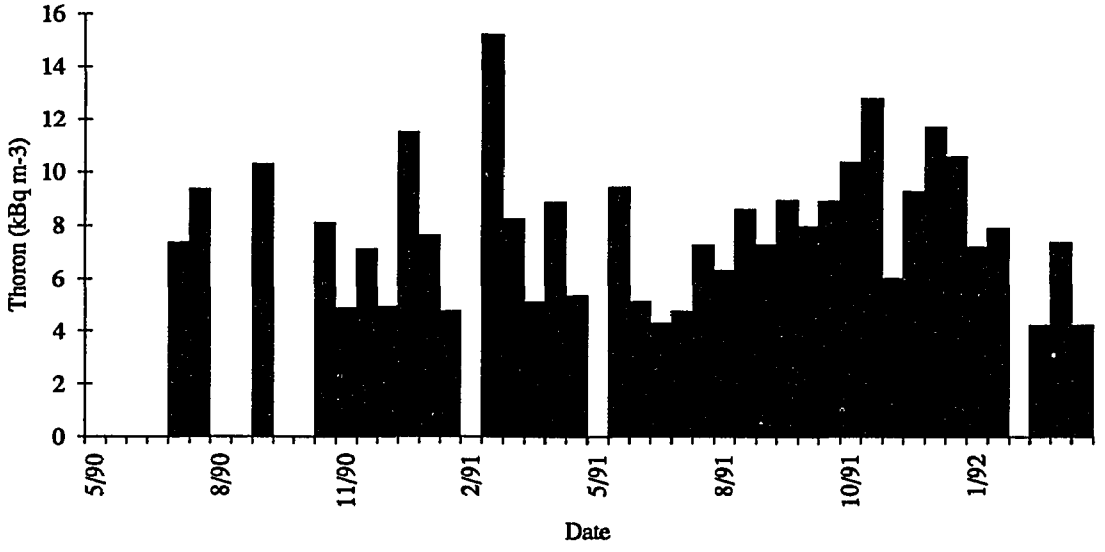

Figure 4.29 ^{220}Rn data from hole 4a, Site 2.

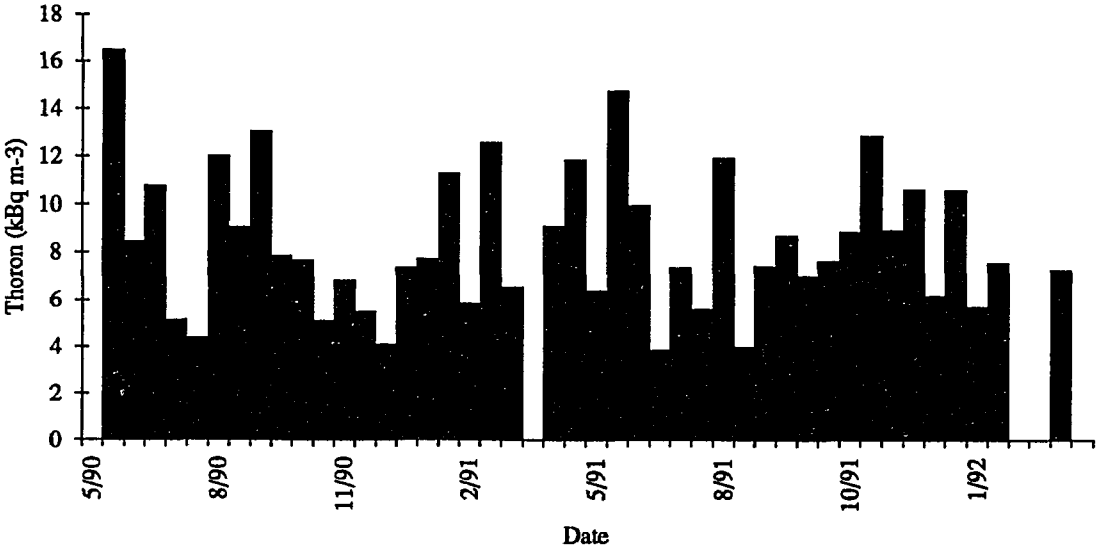

Figure 4.30 ^{220}Rn data from hole 4b, Site 2.

4.3 Site 3 Soil Gas ^{222}Rn and ^{220}Rn

The collective soil gas ^{222}Rn concentration data for Site 3 are presented in Figure 4.31. Figure 4.32 shows the collective ^{220}Rn soil gas data for Site 3. Soil gas samples at this site could not be collected from late December through May or June of 1992 and 1993 due to saturated soil conditions. Attempting to obtain a sample resulted in the pressure meter going off scale and the flow rate going to zero, as well as having a mud being sucked into the sampling apparatus.

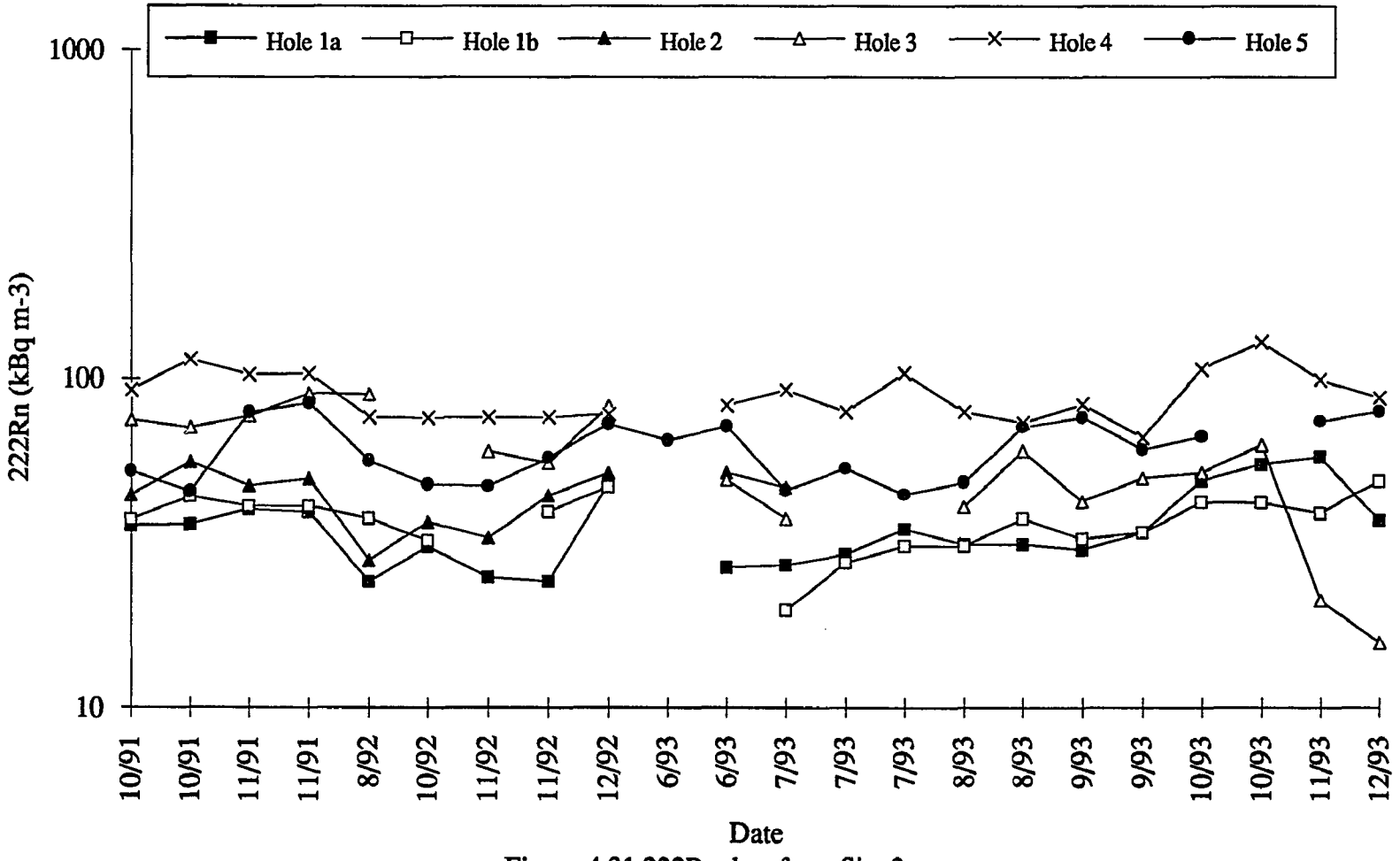

Figure 4.31 222Rn data from Site 3

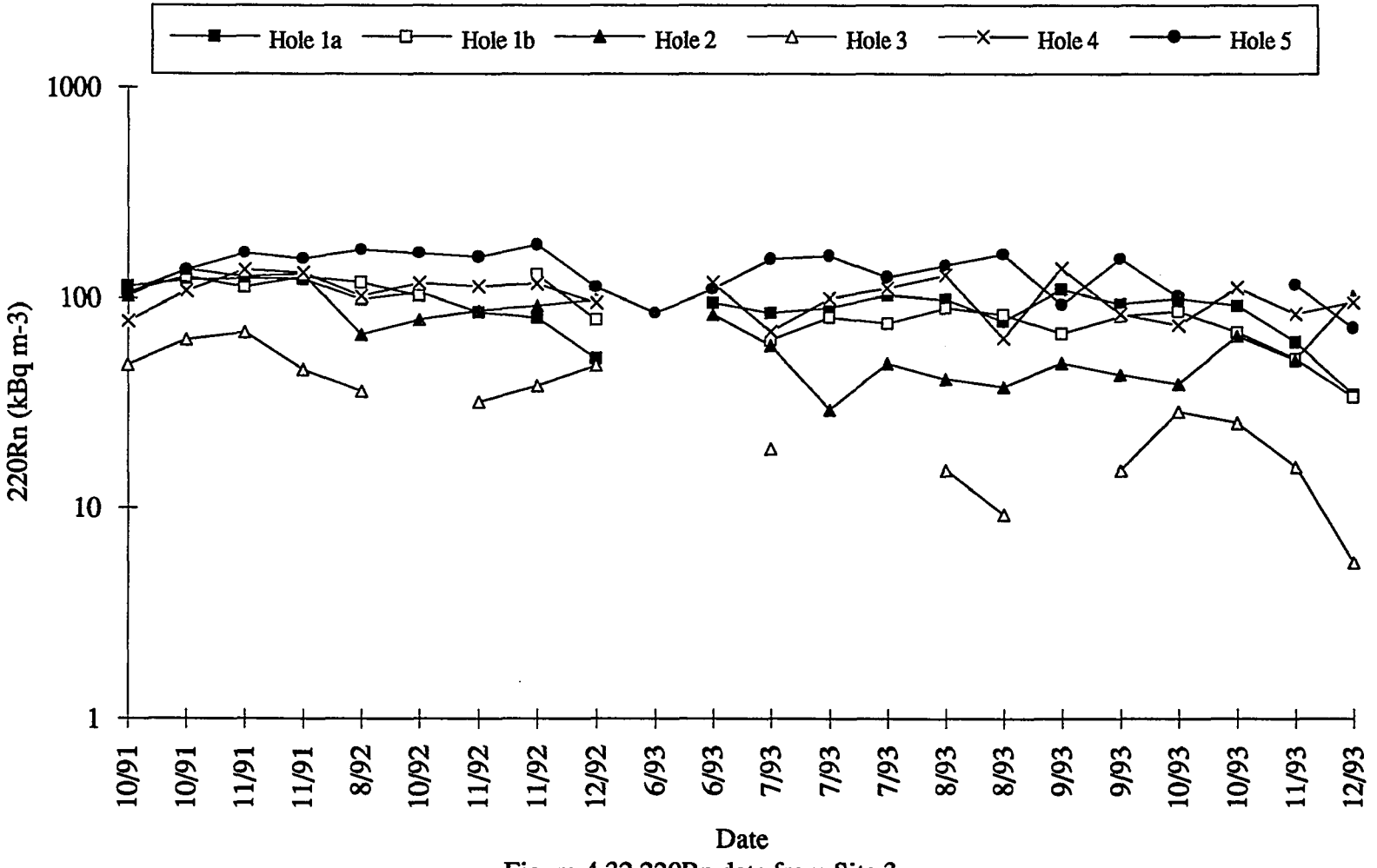

Figure 4.32 220Rn data from Site 3

4.4 ^{220}Rn to ^{222}Rn Ratio

Figure 4.33 shows the time series of the $^{220}Rn/^{222}Rn$ ratios data for Site 1 from April 1990 through May 1992. Figure 4.34 shows the time series of the $^{220}Rn/^{222}Rn$ ratios data for Site 2 from May 1990 through May 1992. Site 3 $^{220}Rn/^{222}Rn$ ratio time series for all available data from October 1991 through September 1993 are shown in Figure 4.35. The soil gas data from Site 3 are not continuous bi-weekly samples, as is the case for Sites 1 and 2, because soil gas samples could not be collected from late December through May or June of 1992 and 1993 due to saturated soil conditions. Attempting to obtain a sample resulted in a mud being sucked into the sampling apparatus. The error in the $^{220}Rn/^{222}Rn$ value is estimated to be 23.3%, determined from a propagation of the uncertainties determined in the ^{222}Rn and ^{220}Rn measurements, 10.8% and 20.7%, respectively as follows:

$$\varepsilon_{ratio} = ((\varepsilon_{222})^2 + (\varepsilon_{220})^2)^{1/2} \qquad (4.1)$$

where: ε_{ratio} = error in $^{220}Rn/^{222}Rn$ ratio; ε_{222} = error in ^{222}Rn measurement, = 10.8%; ε_{220} = error in ^{220}Rn measurement, = 20.7%, so that ε_{ratio} = 23.3%.

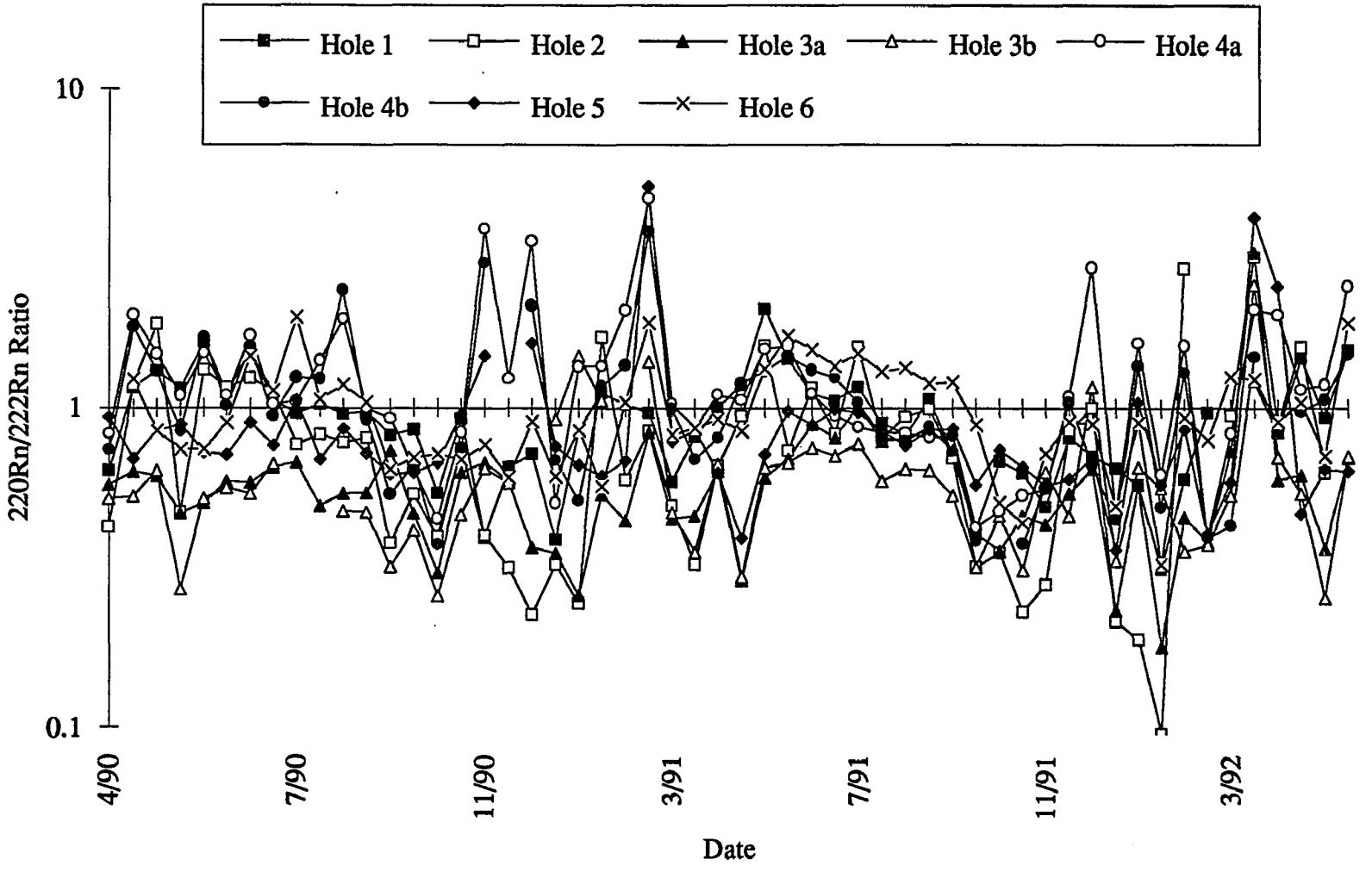

Figure 4.33 220Rn/222Rn ratios from Site 1

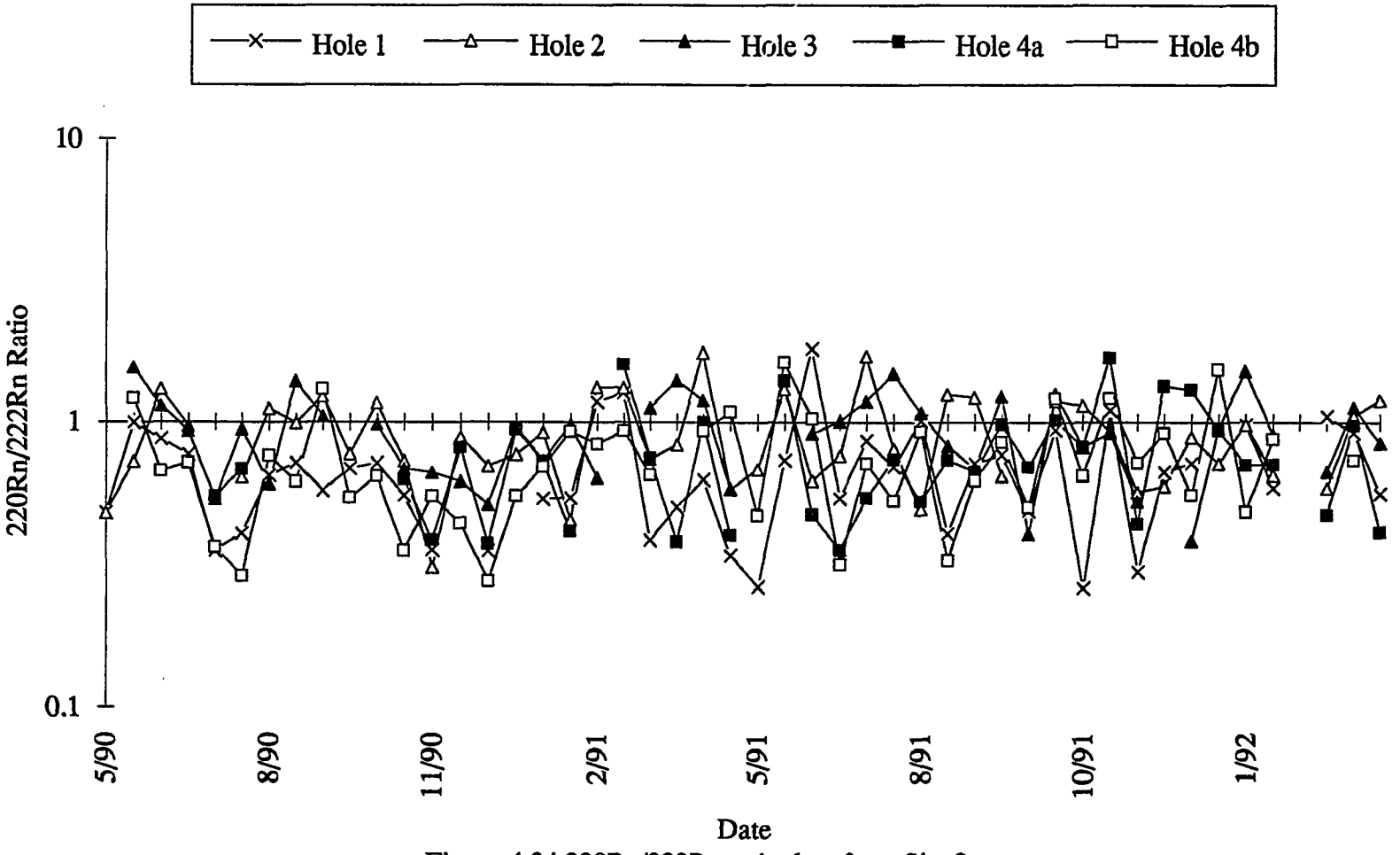

Figure 4.34 220Rn/222Rn ratio data from Site 2

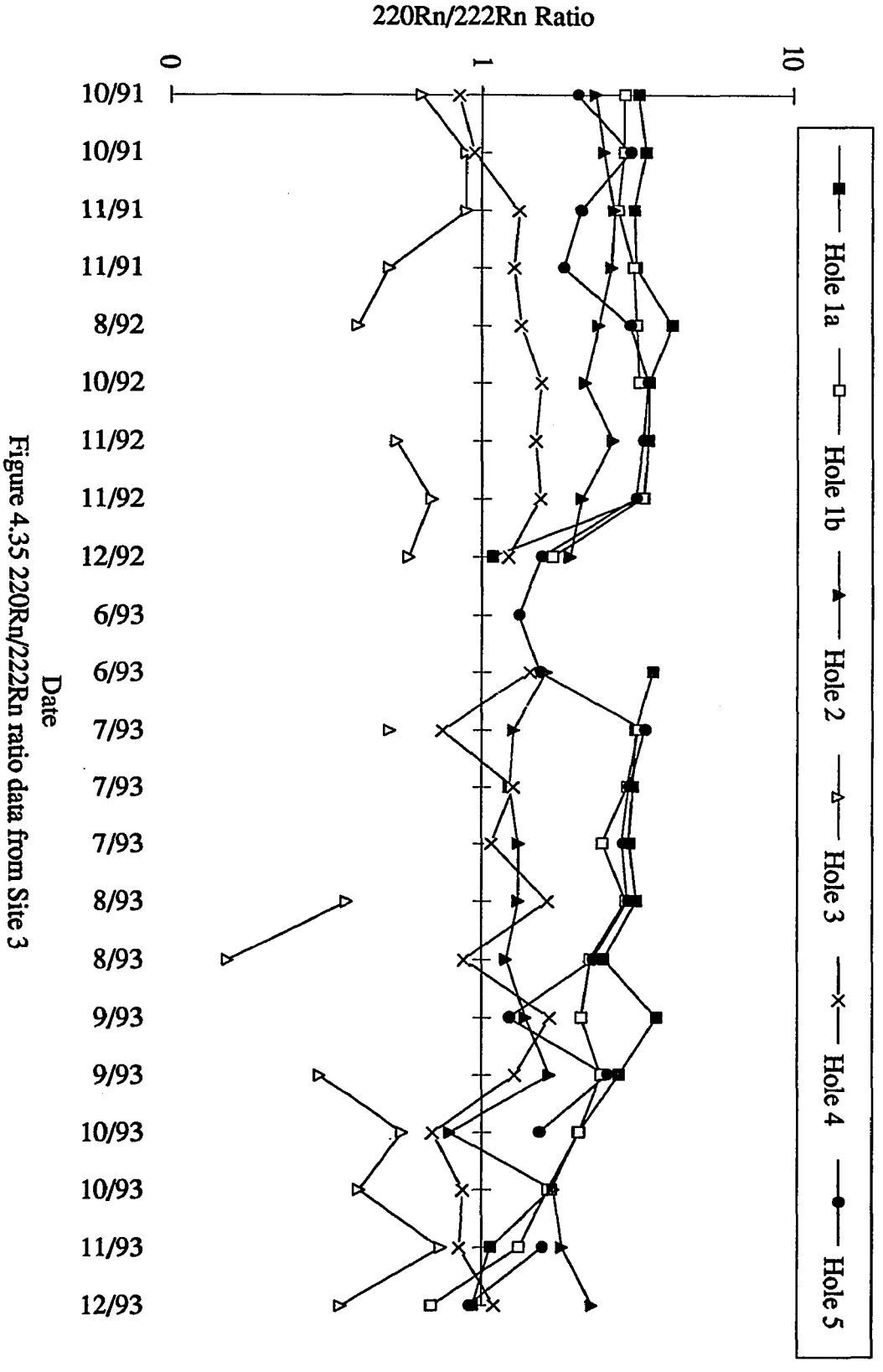

Figure 4.35 220Rn/222Rn ratio data from Site 3

Chapter 5

DATA DISCUSSION AND INTERPRETATION

The discussions that follow pertaining to the data obtained for this dissertation project attempt to summarize and synthesize the data, trends and observations such that the questions posed in the Statement of the Problem in the Introduction section can be effectively answered. These answers addressing the hypotheses are discussed throughout the following section, and are summarized in a subsequent chapter of this dissertation.

5.1 ^{222}Rn and ^{220}Rn

A first inspection of the data shows some obvious trends. At Site 1, the ^{222}Rn concentrations vary among all the holes by nearly two orders of magnitude. The values observed ranged from a low of 25 kBq m^{-3} to a high of 1672 kBq m^{-3} (675 to 45,000 pCi l^{-1}). Hole 3a had the highest ^{222}Rn concentration for each measurement day, followed closely by 3b, two holes that were only about 5 cm apart. Holes 4a and 4b, on the other hand, typically had the lowest ^{222}Rn values. Holes 5 and 6 were normally higher than Holes 1 and 2. These consistent spatial variations are probably due to inhomogeneous parent nuclide concentration in the soil, rather than due to variables such as soil moisture changes.

Seasonal trends in the ^{222}Rn concentrations can be seen when the ^{222}Rn data are observed for all the holes at Site 1 (Figure 4.1), with the lowest values occurring approximately from December until March, with increasing values from early Spring until late Autumn when the highest values are observed. ^{222}Rn concentrations from hole 2 show the strongest seasonal trend (Figure 4.4), with a greater than ten-fold increase from low to high seasons. Holes 4a and 4b (Figures 4.7 and 4.8) show about a five-fold variation, whereas, Holes 1, 3a, and 3b (Figures 4.3, 4.5 and 4.6, respectively) show more

modest seasonal variations on the order of two to three fold-variations from high to low values. Holes 5 and 6 show suspect, at best, seasonal trends (Figures 4.9 and 4.10, respectively).

Another observation with important consequences occurs in the data when the tubes were first emplaced in the Fall of 1989. As can be seen until the winter period ended in Figure 4.1, the variations observed in the soil gas ^{222}Rn concentrations are very large compared to after this period. The concentration variations seemed to decrease after this period into a much smoother and regular pattern. The widely variable concentrations are thought to be due to the disruption of the soil structure after the tubes were emplaced. During these first few months, there may have been cavities along the outside tube walls, allowing soil gas from shallower depths to reach the end of the tube and be sampled. The shallower depth soil gas would be expected to have lower concentrations, as has been observed in subsequent measurements (Hutter, unpub. data, 1993). After the freezing and thawing of the soil occurred during the winter months, the soil may have settled sufficiently to seal these cavities and ensure that the soil gas being sampled was near the 1.0 m depth of the tube end. These data indicate that a one-time sampling of the soil gas by emplacing a tube into the soil and taking a sample of the soil gas may not give an accurate assessment of the soil gas ^{222}Rn concentration from the depth of sampling.

The ^{220}Rn data for Site 1 (Figure 4.2) look very different than the ^{222}Rn data. The spatial variations among the holes are much less, with a total range of 62 to 944 kBq m^{-3} (1675 to 25526 pCi l^{-1}), and also with less temporal variations. However, seasonal trends can be observed at some of the holes, especially holes 2, 3a and 4b (Figures 4.12, 4.13 and 4.16, respectively), with two to three-fold variations. The other holes do not show statistically significant seasonal variations. The holes that exhibit seasonal variations also show a somewhat dissimilar pattern to the ^{222}Rn seasonal variations. The ^{220}Rn concentrations are again lowest during the winter months, but are highest in the summer months, gradually decreasing during the fall, as compared to the ^{222}Rn concentration

variations that increased virtually all summer and into the fall. The magnitude and timing of these observed seasonal trends in the ^{220}Rn concentration closely match the predicted trends from equation 2.17, a diffusion-only model. However, even if advective processes were occurring in the soil gas at Site 1, the transport distance of ^{220}Rn is so short, due to its short half-life, that the seasonal trends would still be expected to follow equation 2.17.

The magnitude of the ^{222}Rn and ^{220}Rn concentrations obtained at Site 2 are approximately one order of magnitude less than those observed at Site #1. In addition, both spatial and temporal variations are much smaller. Figure 4.19 shows essentially constant ^{222}Rn concentration values from 5/90 until 5/92. Figure 4.20 shows a broader band of ^{220}Rn concentration variation at Site 2, but this observation is based on the higher error associated with the ^{220}Rn measurements compared to the ^{222}Rn measurements. Figures 4.21 to 4.25 show the ^{222}Rn concentrations for Site 2 by hole, and no seasonal variations are present at any of the measurement holes. There are also no seasonal variations present in the ^{220}Rn values obtained at Site 2 (Figure 4.26 - 4.30). The lack of seasonal variations in the ^{222}Rn and ^{220}Rn concentrations is thought to be due to the soil being very well-drained, and thus, having little seasonal variation in the soil moisture content, thought to be a major factor affecting variations in soil gas ^{222}Rn and ^{220}Rn.

From the limited data that was able to be obtained at Site 3, 12 bi-weekly samples for each of 6 holes, there appear to be no temporal trends observed in the soil gas ^{222}Rn and ^{220}Rn concentrations, however, spatial variations are significant (Figures 4.31 and 4.32). Specifically, the arithmetic mean of the ^{222}Rn concentrations at hole 4 is 88 ± 14 ($1\ \sigma$) kBq m^{-3} (2375 ± 388 pCi l^{-1}) compared to 33 ± 8 ($1\ \sigma$) kBq m^{-3} (890 ± 219 pCi l^{-1}) at hole 1a. The arithmetic mean of the ^{220}Rn concentrations at hole 5 is 140 ± 31 ($1\ \sigma$) kBq m^{-3} (3774 ± 851 pCi l^{-1}) compared to 47 ± 13 ($1\ \sigma$) kBq m^{-3} (1277 ± 350 pCi l^{-1}) for hole 3. These large spatial variations in the arithmetic means of the ^{222}Rn and ^{220}Rn concentrations are likely to be due to inhomogeneity in the parent material in the

soil. Obtaining soil samples from the bottom of each hole and analyzing for the parent nuclide would give definitive evidence for this explanation of these variations, however, as stated previously, the holes can not be disturbed until other associated studies within the EML soil gas radon and thoron program are completed. Another plausible explanation for these variations has been widely held to be differing soil permeabilities. However, as discussed later in this section, soil permeability shows no correlation with ^{222}Rn or ^{220}Rn variations at this or other sites.

It is useful to study the distribution of the ^{222}Rn and ^{220}Rn concentrations taken as a whole by site in order to understand large-scale differences among the sites. The histograms of the ^{222}Rn and ^{220}Rn concentration measurements for each site are shown in Figures 5.1 - 5.6. The Site #1 ^{222}Rn and ^{220}Rn histograms show patterns approaching log-normal distributions, expected due to the strong seasonal variations observed. At Site 2, the 1σ of the ^{222}Rn and ^{220}Rn measurements taken as a whole are 19% and 36%, respectively, i.e., 12.2 ± 2.3 kBq m^{-3} (331 ± 63 pCi l^{-1}) and 9.43 ± 3.4 kBq m^{-3} (255 ± 92 pCi l^{-1}). These values are approximately twice the 1σ error of the duplicate measurements used for the total soil gas measurement error. The population of the ^{222}Rn and ^{220}Rn measurements taken at Site 2 seem to be normally distributed. Therefore, the variations in the ^{222}Rn and ^{220}Rn values at Site 2 are interpreted to be normal variations from a normally distributed sample population (Figures 5.3 and 5.4), i.e., there are no systematic spatial or temporal variations. This analysis suggests that the Site 2 soil characteristics, such as parent nuclide concentration and distribution, soil moisture dynamics, etc., are homogeneous. This is important as Site 2 is to be used as a control site needed to help discern background temporal variations in the soil gas ^{222}Rn and ^{220}Rn from variations at other sites that may be due to geologic features, soil inhomogeneity, etc. At Site 3, despite a small number of samples, the ^{222}Rn histogram shows a bi-modal distribution (Figure 5.5), interpreted to be due to the spatial variations

arising from the inhomogeneous parent nuclide concentration in the soil at this site. The histogram of ^{220}Rn data for Site 3 (Figure 5.6) shows a distribution approaching normal.

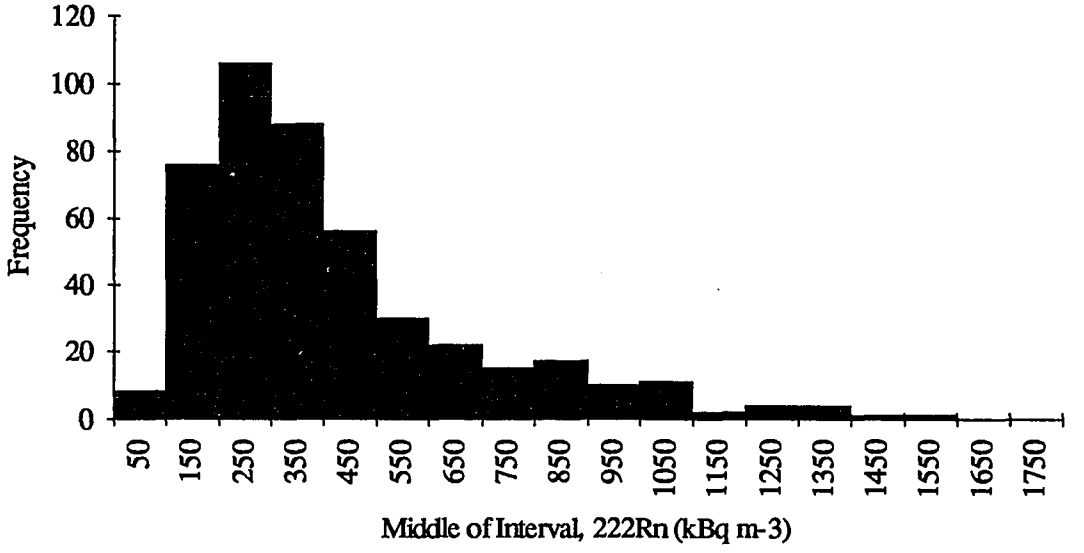

Figure 5.1 ^{222}Rn histogram from Site 1.

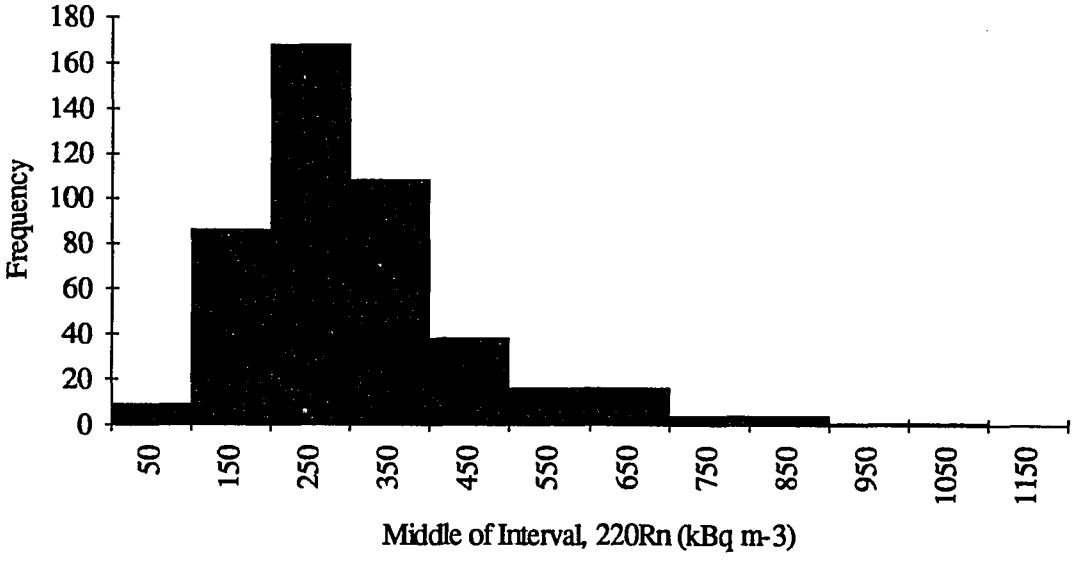

Figure 5.2 ^{220}Rn histogram from Site 1.

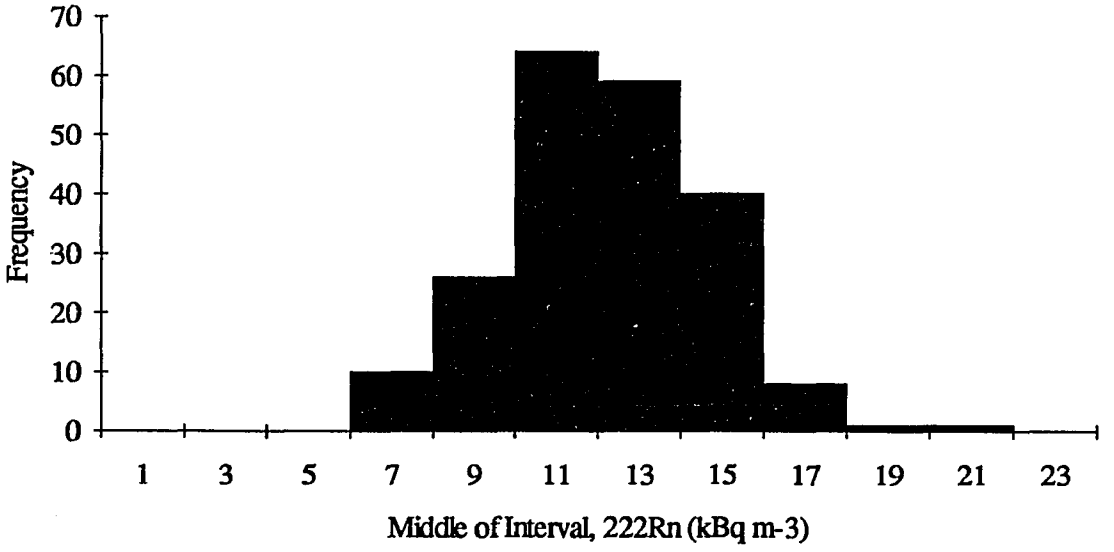

Figure 5.3 ^{222}Rn histogram from Site 2.

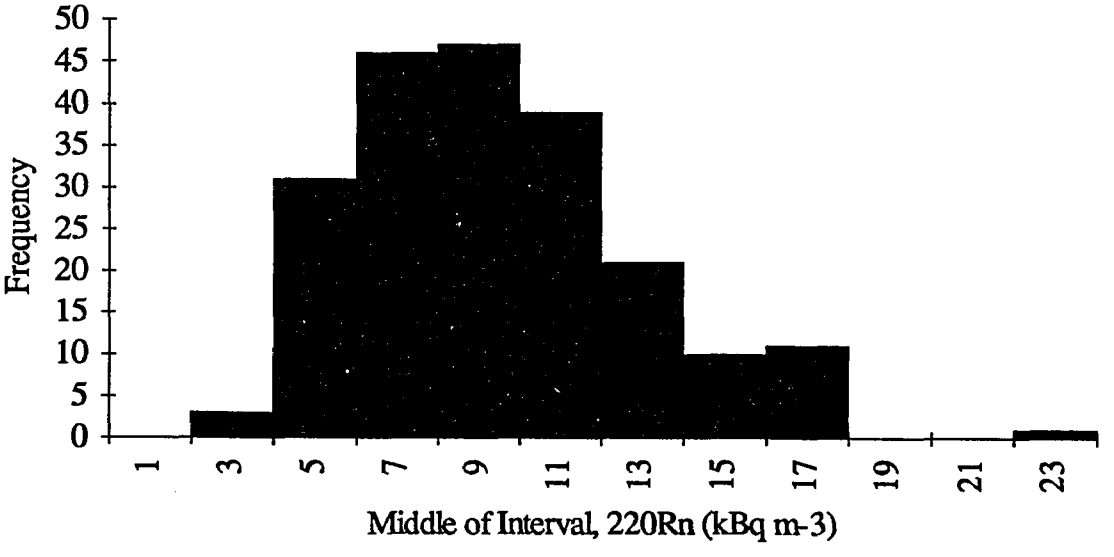

Figure 5.4 ^{220}Rn histogram from Site 2.

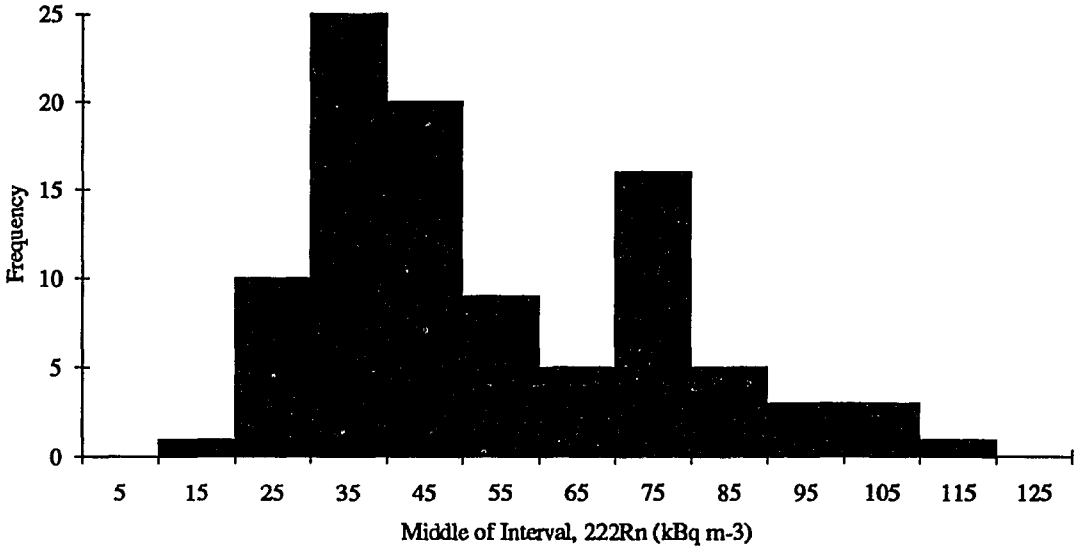

Figure 5.5 ^{222}Rn histogram for Site 3.

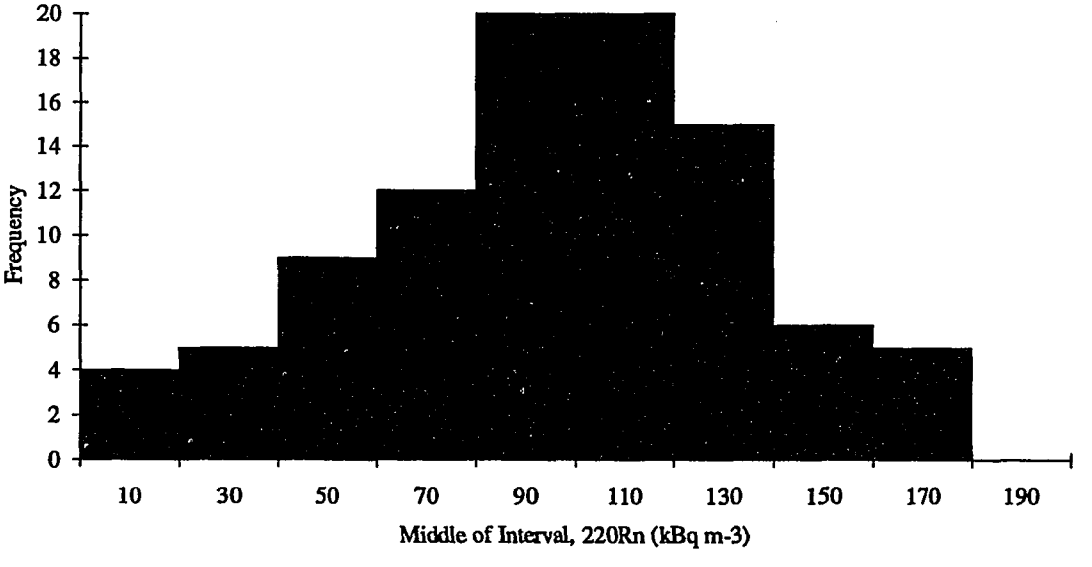

Figure 5.6 ^{220}Rn histogram for Site 3.

The data discussed above for the three sampling sites show that the geologic setting is a critical factor in determining some of the characteristics and dynamics of soil gas ^{222}Rn and ^{220}Rn. The histograms show different distributions for ^{222}Rn and ^{220}Rn concentrations among the sites, indicating large-scale causes and/or controls. The magnitudes of ^{222}Rn and ^{220}Rn concentrations at the sites, with Site 1 having concentrations an order of magnitude higher than those observed at Sites 2 and 3, corroborate the importance of geologic setting as a critical control. Furthermore, seasonal and spatial variations of ^{222}Rn and ^{220}Rn concentrations are very different for the sites. These inter-site comparisons and syntheses of the histogram, temporal and spatial variations data and trends indicate that the geologic setting has quantitative affects on the soil gas ^{222}Rn and ^{220}Rn concentration characteristics and dynamics.

5.2 $^{220}Rn/^{222}Rn$ Ratio

As discussed in the Data Presentation section, the error associated with $^{220}Rn/^{222}Rn$ ratios is approximately 23% (1σ), determined from propagating the errors associated with duplicate ^{222}Rn and ^{220}Rn measurements, 10.8% and 20.7%, respectively.

One of the major concerns with using this new method of ^{220}Rn determination in the soil gas is that the technique is largely based on a relatively small difference between two large numbers. It was conceivable that the technique, though theoretically valid, would be empirically shown to be compromised, i.e., have very high errors so as to limit the ability to interpret, due to this requisite calculation. If this was a major problem it would manifest in producing low ^{220}Rn numbers when the ^{222}Rn concentration was high, and vice versa. Therefore, in order to see if such a problem developed, a simple correlation was performed on all paired ^{222}Rn and ^{220}Rn data. If such a problem existed, an inverse correlation would be expected. On the other hand, a strong positive

correlation would also hint that the ^{222}Rn and ^{220}Rn are not independent of one another at that site, and that the technique of using $^{220}Rn/^{222}Rn$ ratios for transport determination would be shown to not apply to that site, that is, that ^{222}Rn is not being transported long distances, but is only from near the end of the sampling tube. A weak positive or no correlation is expected, if the calculation caveat is shown not to a problem, since factors affecting ^{222}Rn will also affect ^{220}Rn proportionately and at the same times.

As can be seen in the scatterplots of paired ^{222}Rn and ^{220}Rn at Sites 2 and 3 (Figures 5.7 and 5.8), there are virtually no correlations ($r = 0.01$ and -0.002, respectively) between ^{222}Rn and ^{220}Rn, corroborating the interpretation that there are no statistically significant seasonal or spatial trends in the soil gas data, indicating that diffusion is the dominant transport mode throughout the year at these sites. However, at Site 1 (Figure 5.9), a weak positive correlation exists ($r = 0.47$), so that transport mode changes may help explain these variations, via $^{220}Rn/^{222}Rn$ ratios. In all cases, there was no evidence of a negative correlation between ^{222}Rn and ^{220}Rn, showing the technique not to be significantly affected by the arithmetic calculation necessity of using a relatively small difference between two large numbers. The larger estimated random error in the ^{220}Rn measurement, when compared to ^{222}Rn, is almost entirely due to this necessary calculation, as discussed in the Methods section. Correlation coefficients between ^{222}Rn and ^{220}Rn at each hole, discussed later in this chapter and shown in Tables 5.7 and 5.8, for Sites 1 and 2, respectively, all show very small positive trends, corroborating the above arguments.

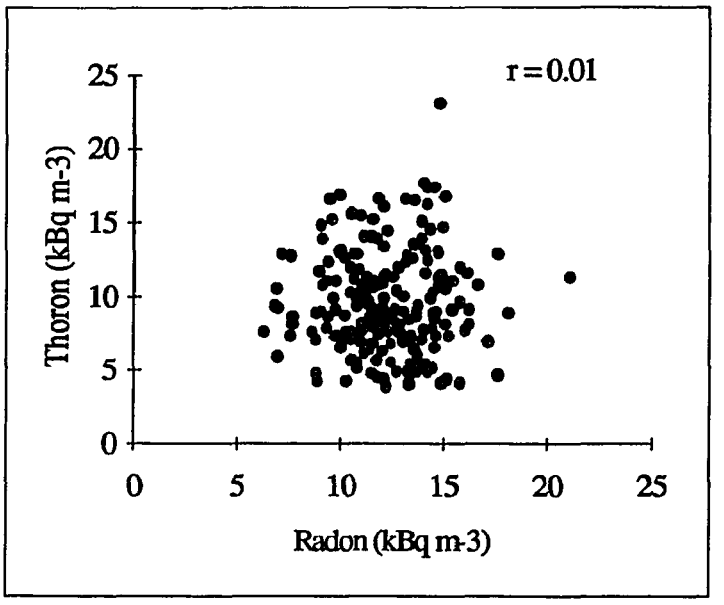

Figure 5.7 - ^{220}Rn vs. ^{222}Rn for Site 2, n = 211

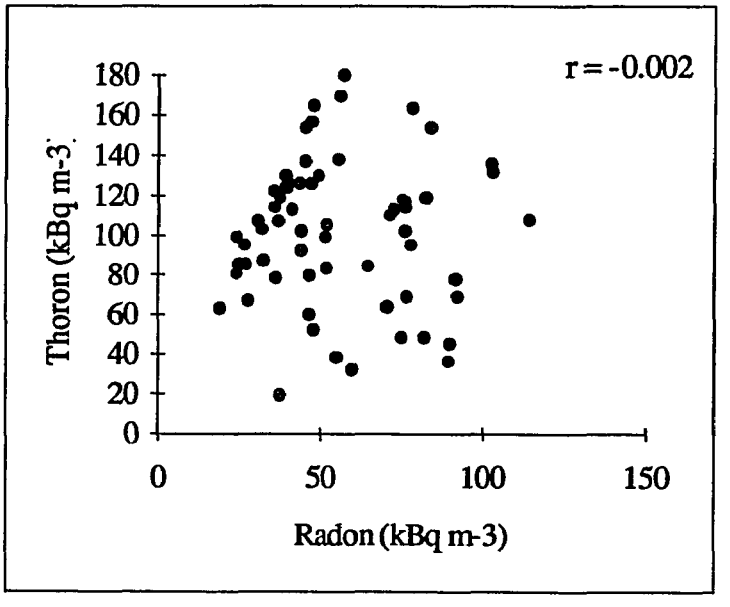

Figure 5.8 - ^{220}Rn vs. ^{222}Rn for Site 3, n = 63

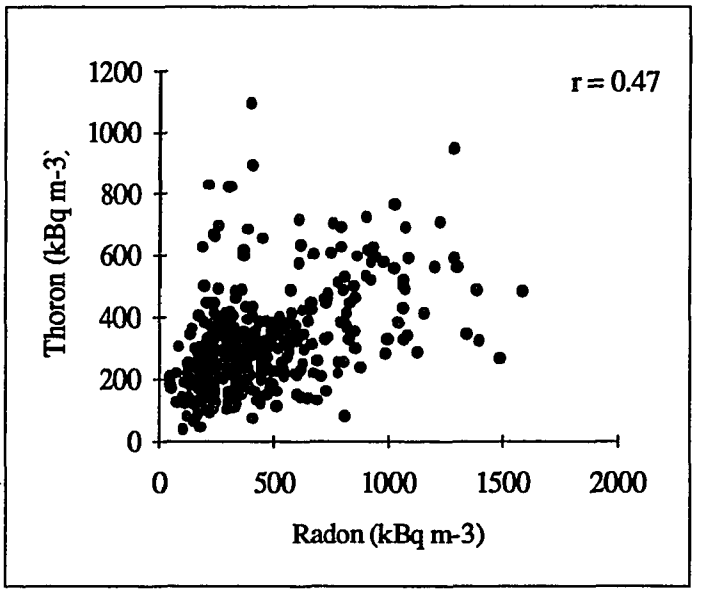

Figure 5.9 - ^{220}Rn vs. ^{222}Rn for Site 1, n = 450

According to Washington and Rose (1990), equation 2.15 describes soil gas ^{222}Rn temporal variations at sites by a factor of up to 3.8. However, at site 1, ^{222}Rn variations occur that are larger than can be explained by equation 2.15, the equation assuming diffusion-only transport process. The seasonal trends observed in the soil gas ^{220}Rn concentration can be effectively explained by equation 2.17, the analogous diffusion-only equation for ^{220}Rn, even if advective soil gas transport may be occurring at certain times of the year, as is suspected, since the migration distance of ^{220}Rn under all circumstances is very short. That the ^{220}Rn concentrations variations can be explained by equation 2.17 adds strength to the argument for using $^{220}Rn/^{222}Rn$ ratios as a means of discerning the soil gas transport mode, because if that equation for soil gas can not explain the ^{222}Rn concentration variations but can for the ^{220}Rn, then the assumptions of that equation as it relates to ^{222}Rn, namely, diffusional-only transport, need to be questioned for validity. Therefore, it is apparent that at Site 1, diffusion may not be the only soil gas migration process occurring, as has been widely thought.

At Site 1, statistically significant $^{220}Rn/^{222}Rn$ ratio variations are observed and appear to have a seasonal pattern. The ratios vary over an order of magnitude at each hole, as shown in Table 5.1.

Table 5.1 Range of $^{220}Rn/^{222}Rn$ ratio for Site 1.

Hole #	Minimum	Maximum
1	0.31	2.04
2	0.095	1.84
3a	0.16	2.78
3b	0.26	3.95
4a	0.42	4.54
4b	0.37	3.57
5	0.25	4.93
6	0.32	1.92

As can be observed from the $^{220}Rn/^{222}Rn$ ratios at Sites 2 and 3 (Figures 4.34 and 4.35), no temporal pattern of variations can be discerned (ranges for each hole are shown in Table 5.2)

Table 5.2 Range of $^{220}Rn/^{222}Rn$ ratios for Sites 2 and 3

	Site 2		Site 3	
Hole #	Minimum	Maximum	Minimum	Maximum
1a	0.26	1.81	1.08	4.09
1b	N/A	N/A	1.68	3.30
2	0.31	1.76	1.27	2.66
3	0.38	1.56	0.40	0.89
4a	0.35	1.68	0.74	1.55
4b	0.28	1.62	N/A	N/A
5	N/A	N/A	1.54	3.43

If diffusion was the only transport process at a site, the $^{220}Rn/^{222}Rn$ ratio should remain constant over time. At Site 1, the $^{220}Rn/^{222}Rn$ ratios change over time, however, in a complex manner not easily explainable (Figure 4.33). It appears that seasonal trends develop, with consistent lows occurring for all the sampling holes during September and October, the same months in which the highest ^{222}Rn values are observed. If high ^{222}Rn values were only due to local seasonal effects of soil conditions, then the ^{220}Rn concentration would be expected to rise as well. However, the $^{220}Rn/^{222}Rn$ ratio changes during this time. It is during the times of low $^{220}Rn/^{222}Rn$ ratio that it is interpreted that soil gas may be migrating via advection from an area of higher ^{222}Rn concentration, presumable from depth. Lateral soil gas migration has also been observed in some soils (Tanner, pers. comm., 1991), and could be a possibility at Site 1. Hole 2, which shows the strongest seasonal trend, is immediately adjacent to hole 3a, with the highest ^{222}Rn concentrations, and elevated ^{222}Rn concentrations observed at hole 2 may be due to ^{222}Rn migrating from the vicinity of hole 3a, approximately 3 m away.

Additionally, soil gas migration may be from an area of low ^{222}Rn concentration, and this scenario would be reflected in high $^{220}Rn/^{222}Rn$ ratios.

The question of whether diffusion is the only transport mechanism present is addressed by interpreting the magnitude of the ^{222}Rn concentration variations and the $^{220}Rn/^{222}Rn$ ratio variations. At Site 1, the apparent occurrence of non-diffusional transport processes is indicated by ^{222}Rn concentration variations larger than can be explained by diffusion equations, and corroborated by seasonal trends in the $^{220}Rn/^{222}Rn$ data, indicating changing transport processes in the soil gas.

In order to better understand the variations observed at the sites, correlations among all the holes at each site for ^{222}Rn, ^{220}Rn and $^{220}Rn/^{222}Rn$ (220/222) ratios were calculated. Tables 5.3 - 5.8 show the correlation coefficients (r) for ^{222}Rn and ^{220}Rn among the holes at Sites 1 and 2. These correlations were not performed for Site 3 due the lack of a sufficient number of samples to draw significant conclusions.

Table 5.3 ^{222}Rn correlation coefficients (r) between sampling locations (hole #) at Site 1.

Hole #	1	2	3a	3b	4a	4b	5
2	0.81						
3a	0.64	0.85					
3b	0.52	0.69	0.85				
4a	0.57	0.67	0.59	0.66			
4b	0.61	0.73	0.69	0.64	0.96		
5	0.10	0.27	0.54	0.57	0.65	0.65	
6	0.53	0.67	0.71	0.56	0.75	0.80	0.68

Table 5.4 ^{220}Rn correlation coefficients (r) between sampling locations (Hole #) for Site 1

Hole #	1	2	3a	3b	4a	4b	5
2	0.52						
3a	0.59	0.58					
3b	0.28	0.32	0.33				
4a	0.54	0.47	0.57	0.61			
4b	0.52	0.45	0.52	0.56	0.48		
5	0.57	0.16	0.47	0.25	0.45	0.66	
6	0.69	0.32	0.40	0.40	0.46	0.59	0.53

Table 5.5 $^{220}Rn/^{222}Rn$ ratio correlation coefficients (r) between sampling locations (Hole #) for Site 1

Hole #	1	2	3a	3b	4a	4b	5
2	0.54						
3a	0.26	0.60					
3b	0.14	0.43	0.73				
4a	0.16	0.25	0.27	0.46			
4b	0.27	0.28	0.24	0.28	0.87		
5	0.08	0.32	0.65	0.69	0.68	0.64	
6	0.49	0.37	0.42	0.32	0.41	0.52	0.47

Table 5.6 ^{222}Rn correlation coefficients (r) between sampling locations (hole #) at Site 2

Hole #	2	3	4a	4b
1	0.22	0.25	0.28	0.34
2		0.74	0.26	0.37
3			0.09	0.38
4a				0.46

Table 5.7 ^{220}Rn correlation coefficients (r) between sampling locations (hole #) at Site 2.

Hole #	2	3	4a	4b
1	0.24	0.41	0.37	0.34
2		0.01	0.33	0.01
3			0.06	0.42
4a				0.33

Table 5.8 $^{220}Rn/^{222}Rn$ ratio correlation coefficients (r) between sampling locations (Hole #) at Site 2.

Hole #	2	3	4a	4b
1	0.22	0.30	0.34	0.41
2		0.18	0.38	0.18
3			0.11	0.28
4a				0.46

Table 5.3 shows the highest correlations in the ^{222}Rn concentrations at site 1 to be between duplicate pair holes, specifically, Holes 4a and 4b, and Holes 3a and 3b, with correlation coefficients (r) of 0.96 and 0.85, respectively. A trend of decreasing ^{222}Rn correlation coefficients with increasing distance between the holes is also observed, as one might intuitively expect. For example, the highest ^{222}Rn correlation coefficient for Hole 1 is with Hole 2, with the next nearest holes, 3 and 4, having smaller correlation coefficients, and holes 5 and 6 even less.

The ^{220}Rn correlation coefficients for Site 1 do not show the same trends as observed for ^{222}Rn (Table 5.4). In general, the ^{220}Rn correlation coefficients are smaller than for ^{222}Rn, and show no obvious trend with spatial distribution, which is to be expected since there were fewer temporal and spatial variations in the cumulative data for ^{220}Rn than for ^{222}Rn (Figure 4.2).

Both the ^{222}Rn and the ^{220}Rn correlation coefficients for Site 2, as shown in Tables 5.6 and 5.7, are very small compared to Site 1, and in general show no statistical correlation, except for the ^{222}Rn between Holes 2 and 3. No trends among the ^{222}Rn and ^{220}Rn concentrations at Site 2 were expected since all measurements are interpreted to have been sampled from a normally distributed population.

In an attempt to understand the relative importance of the measured factors affecting the observed ^{222}Rn and ^{220}Rn variability in the soil gas, correlations were performed for all of the holes at Site 1 for all of the measured parameters (^{222}Rn, ^{220}Rn, $^{220}Rn/^{222}Rn$ ratio, and permeability) with available on-site meteorological data, i.e., temperature, barometric pressure, wind speed, and relative humidity (Tables 5.9). It was expected that poor correlations among the meteorological parameters and soil gas ^{222}Rn and ^{220}Rn concentrations would be found, as the measurement technique was devised to minimize atmospheric and meteorological affects on the soil gas ^{222}Rn and ^{220}Rn concentrations, mainly by the sampling depth of 1.0 m. For Sites 2 and 3, where no on-

site meteorological data were available, Tables 5.10 and 5.11 show the correlations for ^{222}Rn, ^{220}Rn, $^{220}Rn/^{222}Rn$ ratio, and permeability for each sampling hole.

Table 5.9 Site 1 correlation coefficients (r) for each sampling location (hole #)

		222Rn	220Rn	220/222	TEMP	P	RH	ws
Hole 1	**222Rn**				-0.38	-0.28	0.06	0.10
	220Rn	0.07			0.50	0.06	-0.38	0.10
	220/222	-0.84	0.31		0.53	0.22	-0.10	-0.08
	perm	0.00	-0.05	0.00	-0.16	0.21	-0.12	0.14
Hole 2	**222Rn**				-0.52	-0.24	0.12	-0.15
	220Rn	0.05			0.28	-0.29	-0.22	0.14
	220/222	-0.75	0.18		0.27	0.06	-0.15	0.31
	perm	-0.18	0.10	0.34	-0.28	0.10	-0.01	0.12
Hole 3a	**222Rn**				-0.59	-0.36	0.14	0.01
	220Rn	0.21			0.39	0.09	-0.08	-0.18
	220/222	-0.64	0.12		0.70	0.20	-0.12	-0.15
	perm	-0.26	-0.29	0.05	-0.19	0.08	0.07	0.00
Hole 3b	**222Rn**				-0.23	-0.38	0.22	-0.29
	220Rn	0.06			-0.28	0.25	-0.31	0.16
	220/222	-0.66	0.60		-0.13	0.30	-0.29	0.32
	perm	0.01	-0.03	-0.11	-0.01	0.22	-0.27	0.08
Hole 4a	**222Rn**				0.11	-0.32	0.35	-0.40
	220Rn	0.24			-0.25	0.26	-0.32	0.11
	220/222	-0.68	0.26		-0.37	0.37	-0.41	0.34
	perm	0.05	0.01	-0.07	0.17	0.17	-0.12	-0.07
Hole 4b	**222Rn**				-0.10	-0.43	0.25	-0.13
	220Rn	0.27			0.25	0.15	-0.41	-0.07
	220/222	-0.74	0.09		0.06	0.40	-0.39	0.11
	perm	0.05	-0.18	-0.12	0.05	0.08	0.00	0.20
Hole 5	**222Rn**				0.05	-0.14	0.34	-0.20
	220Rn	0.41			0.42	0.02	0.06	-0.16
	220/222	-0.76	-0.16		0.06	0.23	-0.30	0.02
	perm	0.11	-0.27	-0.09	0.02	0.12	0.24	-0.31
Hole 6	**222Rn**				-0.46	-0.12	0.29	0.00
	220Rn	0.11			0.39	-0.01	-0.28	0.10
	220/222	-0.70	0.53		0.68	0.13	-0.38	0.00
	perm	0.09	0.02	-0.01	-0.04	0.17	0.12	-0.20

Table 5.10 Site 2 correlation coefficients (r) for each sampling location (hole #)

		^{222}Rn	^{220}Rn	220/222
Hole 1	**^{220}Rn**	-0.05		
	220/222	-0.55	0.82	
	perm	-0.30	-0.18	0.01
Hole 2	**^{220}Rn**	0.06		
	220/222	-0.43	0.86	
	perm	-0.02	-0.11	-0.08
Hole 3	**^{220}Rn**	0.06		
	220/222	-0.39	0.88	
	perm	-0.02	-0.15	-0.15
Hole 4a	**^{220}Rn**	-0.28		
	220/222	-0.70	0.85	
	perm	-0.35	0.00	0.19
Hole 4b	**^{220}Rn**	-0.10		
	220/222	-0.60	0.83	
	perm	-0.16	0.13	0.16

Table 5.11 Site 3 correlation coefficients (r) for each sampling location (hole #)

		^{222}Rn	^{220}Rn	220/222
Hole 1a	^{220}Rn	0.08		
	220/222	-0.76	0.56	
	Perm	-0.96	0.02	0.84
Hole 1b	^{220}Rn	0.63		
	220/222	-0.43	0.42	
	Perm	0.61	0.94	0.39
Hole 2	^{220}Rn	0.74		
	220/222	0.20	0.79	
	Perm	0.48	0.79	0.62
Hole 3	^{220}Rn	0.54		
	220/222	-0.06	0.81	
	Perm	0.59	0.81	0.55
Hole 4	^{220}Rn	0.14		
	220/222	-0.59	0.71	
	Perm	-0.22	0.24	0.38
Hole 5	^{220}Rn	-0.07		
	220/222	-0.80	0.64	
	Perm	-0.36	-0.07	0.27

The mechanism of transport mode has been previously hypothesized to be largely dependent upon the permeability of the soil, i.e., soil permeability is a good indicator of the dominant transport mode. At permeabilities greater than 10^{-11} m^2, advective transport is thought to occur, whereas diffusive transport is thought to be dominant at permeabilities less than 10^{-11} m^2 (Nazaroff, 1992). Owing to this relationship, it was expected that a correlation of either ^{222}Rn and/or $^{220}Rn/^{222}Rn$ ratio with permeability may show these dependencies, and help to show that changing transport mode is an

important consideration as a control of soil gas ^{222}Rn variations. However, as noted in Table 5.9, the correlations are, for the most part, very poor.

The best correlation with permeability is the $^{220}Rn/^{222}Rn$ ratio at hole 2, at a value of 0.34. Any correlation of permeability and $^{220}Rn/^{222}Rn$ ratio would be expected to be negative, if the soil air advecting to a sampling hole had higher ^{222}Rn concentrations than during periods dominated by a diffusion mechanism. It is also possible, but less probable, that the soil gas may be of lesser concentration during times of advection, or alternatively, longer diffusional transport due to a change in the diffusion coefficient. For instance, if the permeability of the soil was such that the soil gas is able to be transported farther than usual, it is conceivable that soil air from depths shallower than 1.0 m, and known lesser concentrations, may be reaching the end of the sampling tube. Normal diffusional and advective transport are thought to be in the direction towards the surface, however.

Overall, soil permeability does not seem to show any strong affect on either soil gas ^{222}Rn, ^{220}Rn, or the $^{220}Rn/^{222}Rn$ ratio. These poor correlations suggest that either the permeability is not a good indicator of parameters thought to have strong affects on the soil gas ^{222}Rn and ^{220}Rn concentrations, such as soil moisture, porosity, water saturation fraction, etc., or that these factors themselves are not as important in controlling the soil gas ^{222}Rn and ^{220}Rn concentrations as has been thought. With the data obtained it is not possible to test the latter possibility, but, of the two scenarios, it is much less likely to be true. Therefore, apparently the soil permeability is not a valid indicator of soil characteristics controlling soil gas ^{222}Rn and ^{220}Rn concentrations at Site 1, contrary to previously accepted hypotheses. If the soil permeability measurements are not indicative of the true soil permeability, i.e., if the measurements were performed correctly, then the interpretations concerning the lack of correlation of soil permeability with any of the measured parameters are moot. The permeability measurements were performed according to published methods (see Methods section for discussion), however,

there is no mechanism for calibration of such measurements, so there is still some question as to whether the measurements are an accurate reflection of the actual soil gas permeability. Nevertheless, while this possibility exists, there is no reason to discredit the soil permeability data, so that the interpretations should not be compromised.

In order to objectively show that the transport mode is a contributing factor to the observed ^{222}Rn variations in the soil gas, it was expected that a good negative correlation would exist between the permeability and $^{220}Rn/^{222}Rn$ ratio. However, no correlations seem to exist, so definitive conclusions concerning the precise timing of dominant transport mode, e.g., when diffusion or advection is dominant process, can not be strongly made with the observations obtained in this study. However, data exist that circumstantially show promise for using $^{220}Rn/^{222}Rn$ ratios as indicators of the dominant soil gas transport processes occurring during different times of the year. $^{220}Rn/^{222}Rn$ ratios at Sites 2 and 3 do not show any seasonal trend, indicating that changes in migration process do not occur. The $^{220}Rn/^{222}Rn$ ratio variations at Site 1 seem to indicate times of the year when ^{222}Rn migration distance changes, either by a change in migration process from diffusion to advection or by an increase in diffusion length due to changing soil conditions. For instance, during late summer/early fall the $^{220}Rn/^{222}Rn$ ratios for all holes at Site 1 are at minimum values. It is during this period that it is interpreted that soil conditions are such that soil gas migration distances are larger than winter periods when $^{220}Rn/^{222}Rn$ ratios are high.

In looking at the correlations for the $^{220}Rn/^{222}Rn$ ratio at each site (Tables 5.9, 5.10 and 5.11), the parameter apparently controlling the $^{220}Rn/^{222}Rn$ ratio is different for different sites. If ^{222}Rn is the principle driving force in the changing $^{220}Rn/^{222}Rn$ ratios, then it is interpreted that the transport mode or distance can be an important factor in the seasonal variations observed in the ^{222}Rn soil gas concentrations. If ^{220}Rn is the controlling factor in the $^{220}Rn/^{222}Rn$ ratios, then it is clear that the soil gas transport distance is very short, and does not change throughout the year. At Site 1, the ^{222}Rn

concentration is shown to be controlling the changes in the $^{220}Rn/^{222}Rn$ ratio, expected if it is thought that ^{222}Rn migration distances change throughout the year as a cause of the ^{222}Rn variations. These trends are observed in Table 5.9, as the arithmetic average of the correlations between ^{222}Rn and the $^{220}Rn/^{222}Rn$ ratio for all the holes is -0.72 ± 0.06 whereas the correlations between ^{220}Rn and $^{220}Rn/^{222}Rn$ ratio for all the holes arithmetically average 0.24 ± 0.24. This difference in the controlling variable of the $^{220}Rn/^{222}Rn$ ratios implies that the transport distance of ^{222}Rn changes throughout the year and contributes to the seasonal variations observed that are too large to be explained by a diffusion model. At Site 2, the ^{220}Rn concentration appears to be controlling the $^{220}Rn/^{222}Rn$ ratio (Table 5.10), as the arithmetic average of the correlations between ^{220}Rn and the $^{220}Rn/^{222}Rn$ ratio for all the holes is 0.85 ± 0.02, compared to the arithmetic average of the correlations between ^{222}Rn and the $^{220}Rn/^{222}Rn$ ratio being -0.53 ± 0.13. The correlation between ^{220}Rn and the $^{220}Rn/^{222}Rn$ ratios at Site 2 strongly implies that the migration distance of the soil gas at this site is very short, and moreover, does not significantly change throughout the year, as is the case at Site 1. The interpretation of these $^{220}Rn/^{222}Rn$ data show that at Site 1 the transport mode, either diffusion or advection, changes throughout the year, whereas at Site 2, the transport mode is likely to be diffusion-dominant throughout the year. This use of $^{220}Rn/^{222}Rn$ ratios to show the importance of transport mode dynamics in soil gas as a controlling factor in seasonal ^{222}Rn concentration variations may have important applications for use in detecting and monitoring soil gas transport for other important environmental concerns, such as pollutant dispersal at waste facilities.

There are very few correlations among the meteorological parameters supporting strong cause and effect relationships (Table 5.9). A few of the correlations of temperature with ^{222}Rn and/or ^{220}Rn are about 0.5, expected due to the seasonal variations in the ^{222}Rn and ^{220}Rn concentrations observed at some of the holes. The barometric pressure shows virtually no strong correlations with any of the measured parameters, evidence that

the 1.0 m sampling depth is below the depth affected by this parameter. Relative humidity affects on the soil gas ^{222}Rn and ^{220}Rn concentration are minimal, at best.

Chapter 6

SUMMARY, CONCLUSIONS AND RECOMMENDATIONS

6.1 Summary of Investigation

The investigation described in this dissertation can be summarized as follows:

1. Measurements of soil gas ^{222}Rn on a bi-weekly basis were obtained at a site in Chester, NJ (Site 1).
2. After an analysis of six months of ^{222}Rn data showed spatial and temporal variations larger than could be explained by previous diffusion equations.
3. The hypothesis of using $^{220}Rn/^{222}Rn$ ratios as indicators of changing soil gas transport mode and migration distance was developed.
4. A method to measure soil gas ^{220}Rn accurately, precisely and rapidly was developed.
5. Bi-weekly ^{220}Rn, ^{222}Rn, and permeability measurements were obtained at two geologically different sites (Site 1 - Metamorphic gneiss in Chester, NJ; Site 2 - Coastal Plain and Quaternary Sediments in Matawan, NJ) for a period of two years.
6. A site was sought where soil gas advective-dominant migration processes were likely to occur, such as in the vicinity of fractures or faults. Access to the property for the purpose of obtaining ^{220}Rn and ^{222}Rn measurements was denied.
7. The use of a site was approved with similar geologic characteristics as Site 2. (Site 3 - Coastal Plain/Piedmont, in Cheesequake State Park, NJ). Soil gas ^{220}Rn, ^{222}Rn, and permeability measurements were obtained on a bi-weekly basis whenever possible. Saturated soil conditions for much of the year prohibited soil gas sampling.

6.2 Summary of Results

The results of this project can be summarized as follows:

1. A one-time sample of soil gas for the purpose of measuring ^{222}Rn and ^{220}Rn concentrations as a source term for indoor concentrations may only be accurate to an order of magnitude due to seasonal effects and the disruption of the soil structure.
2. A precise, accurate and quick technique for environmental ^{220}Rn measurement has been developed. The lowest ^{220}Rn concentration that can be measured using this method while maintaining a reasonable error is approximately 500 Bq m^{-3} (13 pCi l^{-1}). At this concentration level, the total error (90% confidence level) is about 30% for single measurements, decreasing to about 20% if multiple samples are obtained in series and an arithmetic mean is reported. At higher concentrations, e.g., those normally found in the soil gas, the 1σ error is approximately 20%.
3. ^{222}Rn concentrations at Site 1 are highest during September/October, corroborating previous research and supporting the speculation that wintertime testing of indoor ^{222}Rn concentrations may not give the most conservative estimate of exposure, as is desired.
4. At other sites, no seasonal ^{222}Rn variations are observed, also corroborating previous research.
5. Some ^{222}Rn variations are too large to be adequately explained by diffusion-only models.
6. Long-term trends in the soil gas ^{220}Rn concentration have been established at three sites, and differences are observed among the sites.
7. The highest soil gas ^{220}Rn concentrations occur during mid-summer, coinciding with predictions from diffusion-only models. The controlling parameters of

these variations are thought to be due to local soil conditions, such as soil moisture, water saturation fraction and temperature, among others.

8. Spatial variations in the soil gas ^{222}Rn and ^{220}Rn concentrations can be large (up to an order of magnitude) over distances of 10 to 12 m.
9. Permeability measurements, thought to be a major soil indicator of parameters controlling soil gas ^{222}Rn variations, show no correlation with ^{222}Rn, ^{220}Rn, or $^{220}Rn/^{222}Rn$ ratio at any of the sites.
10. There are trends in the $^{220}Rn/^{222}Rn$ ratio at some of the sampling holes at Site 1 that give credence to the mode of transport being a controlling factor in the seasonal ^{222}Rn variations observed in the soil gas. At the other sites, the $^{220}Rn/^{222}Rn$ ratios do not show any seasonal trends, indicating that diffusion is the dominant migration process throughout the year at these sites. An interpretation of long-term trends in the $^{220}Rn/^{222}Rn$ ratios and the controlling factors give an indication of any seasonal changes in soil gas migration processes, either diffusion-dominant or advection-dominant, or times of the year when transport distances significantly change. These analyses may thus provide information on transport distances of soil gas, vitally important in many environmental concerns, both within DOE and elsewhere.

6.3 Conclusions

The Data Discussion of this dissertation provide the bases from which to answer the questions posed in the Introduction section, Statement of Problem. Each are discussed below:

(1) Do differing geologic settings have quantitative affects on ^{222}Rn concentrations?

Data obtained at the three sampling sites show that the geologic setting is a critical factor in determining some of the characteristics and dynamics of soil gas ^{222}Rn and ^{220}Rn. Histograms of the Site 1 data show log-normal distributions for ^{222}Rn and ^{220}Rn concentrations, whereas Site 2 and Site 3 data show normal distributions. The magnitude of ^{222}Rn and ^{220}Rn concentrations at Site 1 are an order of magnitude higher than those observed at Sites 2 and 3. Furthermore, seasonal trends are present in the Site 1 ^{222}Rn and ^{220}Rn concentrations, whereas none are present at Sites 2 and 3. Spatial variations are clearly present at Sites 1 and 3, but not at Site 2. These spatial variations are thought to be due to inhomogeneous parent concentrations in the soil dependent upon weathering, soil type, etc. The inter-site comparisons and syntheses of the histogram, temporal and spatial variations data and trends indicate that the geologic setting produces quantitative affects on the soil gas ^{222}Rn and ^{220}Rn concentration characteristics and dynamics on a site-wide basis.

(2) Can the relative importance of the numerous variables affecting soil gas transport (e.g., soil moisture, porosity, type, atmospheric conditions, etc.) be determined from multi-variate statistical analysis of the data?

The mechanism of the changing transport mode has been previously hypothesized to be largely dependent upon the permeability of the soil. Multi-variate statistical analyses of all available data in the current study, i.e. correlations among soil gas ^{222}Rn, ^{220}Rn and soil permeability, do not show the relative importance of the parameters thought to affect soil gas transport. This situation suggests that either the permeability is not a good indicator of parameters thought to have strong affects on the soil gas ^{222}Rn and ^{220}Rn concentrations, such as soil moisture, porosity, water saturation fraction, etc., or that these factors themselves are not as important in controlling the soil gas ^{222}Rn and ^{220}Rn concentrations as has been thought. With the data obtained it is not possible to test the

latter possibility, and of the two scenarios, is much less likely to be true. Therefore, apparently the soil permeability is not a valid indicator of soil characteristics controlling soil gas ^{222}Rn and ^{220}Rn concentrations, contrary to previously accepted hypotheses. Because of this situation, the use of $^{220}Rn/^{222}Rn$ ratios were developed as an alternate means of discerning whether changing soil gas transport mode may be important in controlling soil gas ^{222}Rn variations.

(3) Is, as commonly thought, diffusion the dominant mechanism of soil gas transport or are other mechanisms operative thereby causing unexpected variations in ^{222}Rn concentrations?

If diffusion equations for soil gas can not explain the ^{222}Rn concentration variations but can for the ^{220}Rn, then the assumptions of that equation as it relates to ^{222}Rn, namely, diffusional-only transport, need to be questioned for validity. Therefore, it is apparent that at Site 1, diffusion may not be the only soil gas migration process occurring, since ^{222}Rn concentration variations are in excess of the maximum able to be predicted from diffusion-only equations.

(4) Can variations in the isotopic ratio $^{220}Rn/^{222}Rn$ over time be used to distinguish between steady-state diffusion-dominant migration of soil gas versus advection-dominant?

At Site 1, the apparent occurrence of non-diffusional transport processes, shown by ^{222}Rn concentration variations larger than can be explained by diffusion equations, is corroborated by trends in the $^{220}Rn/^{222}Rn$ data. If diffusion was the only transport process at a site, the $^{220}Rn/^{222}Rn$ ratio should remain constant over time. It is during

the times of low $^{220}Rn/^{222}Rn$ ratio that it is interpreted that soil gas may be migrating via advection from an area of higher ^{222}Rn concentration, presumable from depth. Furthermore, the major controlling parameter in the $^{220}Rn/^{222}Rn$ variations may also indicate if changing transport modes is an important consideration as a control for ^{222}Rn variations. If ^{222}Rn is the principle driving force in the changing $^{220}Rn/^{222}Rn$ ratios, then it is possible that the transport mode or distance is an important factor in controlling the seasonal variations observed in the ^{222}Rn soil gas concentrations, as the half-life of ^{222}Rn is such that long-distance transport is possible. On the other hand, if ^{220}Rn is the controlling factor in the $^{220}Rn/^{222}Rn$ ratios, then it is clear that the soil gas transport distance is very short, and does not change throughout the year, as the half-life of ^{220}Rn prohibits transport more than a cm or so. At Site 1, the $^{220}Rn/^{222}Rn$ ratios are controlled by the ^{222}Rn variations, whereas at Site 2, the $^{220}Rn/^{222}Rn$ ratios are controlled by ^{220}Rn.

(5) Finally, can a predictive model for ^{222}Rn soil gas concentrations be developed from the measurement of an array of time dependent soil, atmospheric, and geologic variables?

There have been previous studies attempting to develop a predictive model for soil gas ^{222}Rn concentrations, however, these do not adequately account for the data, trends and observations obtained for this dissertation. The data and proposed hypotheses obtained herein provide a new component for inclusion into subsequent models that will better predict soil gas ^{222}Rn variations over a broader range of circumstances. Specifically, parameters controlling soil gas transport mode changes throughout the year may help predict variations in ^{222}Rn concentrations, as indicated in this dissertation by $^{220}Rn/^{222}Rn$ ratio dynamics.

6.5 Recommendations

The findings and conclusions reached in the present investigation provide substantiated belief that $^{220}Rn/^{222}Rn$ ratios provides useful information concerning changing dominant transport process occurring the soil gas. Specifically, anomalous seasonal variations in the soil gas ^{222}Rn concentration may be due to soil gas being advected from areas of higher ^{222}Rn concentration (thought to be from depth, but could be lateral migration as well). In order to more fully study the potential of using $^{220}Rn/^{222}Rn$ ratios in this regard, a fractured or sheared site is needed where the likelihood of advective flow from depth is enhanced. At this site, long-term trends in the $^{220}Rn/^{222}Rn$ ratio should be obtained, as well as profiling the $^{220}Rn/^{222}Rn$ ratios.

Profiling of the soil gas ^{222}Rn and ^{220}Rn concentrations in the soil gas was started at two of the sampling locations at Site 1 during August 1992, and at Sites 2 and 3 during 1993. Depths of the new sampling tubes into the soil were 0.25, 0.5, and 1.5 m. The data from these sampling locations show some expected trends, e.g., decreasing concentration towards the surface, and increasing variability towards the surface, but are not included in this thesis because trends can not be established due to the disruption of the soil and widely varying ^{222}Rn and ^{220}Rn concentrations as was observed in the data presented for inclusion in this study. It is expected that these ^{222}Rn and ^{220}Rn profile data will further show the usefulness of using $^{220}Rn/^{222}Rn$ ratios as indicators of soil gas migration processes.

A rigorous mathematical analysis of profiles of long-term $^{220}Rn/^{222}Rn$ ratio trends in the soil gas may provide quantitative information on the migration distance of the soil gas. This information would prove to be valuable to the understanding of volatile pollutant dispersal and migration in the unsaturated zone, which is critical in helping to solve many problems facing environmental scientists and engineers.

Appendix A

^{222}RN AND ^{220}RN CALCULATION PROGRAM

```
' NEWCALCD.BAS BY ADAM HUTTER 10/92, REVISED OCCASIONALLY AS NEEDED
' THIS VERSION FOR USE WITH B+W MONITORS
'
' TO CALCULATE RADON:
'   FOR THE COUNT PERIOD AFTER ISOLATION OF THE SAMPLE IN
'   THE SCINTILLATION CELL, INTEGRATE THE EQUATION:
'
'1  (RN ACTIVITY + PO-218 ACTIVITY + BI-214 ACTIVITY)/INITIAL RN ACTIVITY
'
'   USING STANDARD DECAY EQUATIONS. THE RESULTANT NUMBER IS THEN DIVIDED
'   INTO THE NUMBER OF COUNTS OBTAINED IN THE COUNTING PERIOD, GIVING THE
'   INITIAL ACTIVITY OF RADON CONVERTED TO COUNTS AT THE MOMENT SAMPLING
'   ENDED. MULTIPLICATION BY A CALIBRATION FACTOR WILL GIVE RADON
'   CONCENTRATION.
'
' TO CALCULATE THORON:
'   AFTER CALCULATING RADON AT ANY TIME AFTER ISOLATION OF SAMPLE IN
'   SCINTILLATION CELL USING ABOVE METHOD, THE INITIAL ACTIVITY OF RADON
'   IN COUNTS IS KNOWN. NEXT, INTEGRATE THE ABOVE EQUATION FOR THE
'   TIME PERIOD WHERE COUNTS FOR THORON ARE AVAILABLE (USUALLY THE FIRST
'   MINUTE OR SO) AND MULTIPLY THE RESULT BY THE INITIAL RN
'   COUNTS TO GIVE THE COUNTS DUE TO RADON IN THIS PERIOD. SUBTRACT THE
'   COUNTS DUE TO RADON IN THIS PERIOD FROM THE TOTAL NUMBER OF COUNTS
'   OBSERVED - THE RESULTANT NUMBER WILL BE THE COUNTS DUE TO THORON AND
'   THORON PROGENY (SPECIFICALLY PO-216 SINCE IN THE FIRST FIVE OR SO
'   MINUTES AFTER ISOLATION,  VIRTUALLY NO PB-212 WILL HAVE BUILT UP).
'   INTEGRATE THE EQUATION:

'2      (RN220 ACTIVITY + PO216 ACTIVITY)/INITIAL RN220 ACTIVITY
'
'   FOR THE TIME PERIOD WHERE COUNTS ARE AVAILABLE USING STANDARD DECAY
'   EQUATIONS FOR THORON AND PROGENY. DIVIDE THIS NUMBER INTO THE COUNTS
'   DUE TO THORON TO GIVE INITIAL COUNTS DUE TO THORON AND MULTIPLY BY
'   CALIBRATION FACTOR TO GIVE THORON CONCENTRATION

' THIS PROGRAM CALCULATES THE RADON AND THORON CONCENTRATION AT ANY TIME
' AFTER SAMPLING INTO A SCINTILLATION FLASK, ASSUMING A FILTER HAS BEEN PLACED
' IN-LINE ASSURING THAT NO PROGENY WAS ENTERED INTO THE FLASK INITIALLY.
10   DIM FBKGR(200)
15   DIM CALIB(200)
40   TSAME = 1
52   PRINT
53   PRINT
56   PRINT
60   PRINT "                    NEWCALC"
70   PRINT "          A computer program brought to you by A.R.Hutter"
72   PRINT ""
85   PRINT "              Program to calculate 222Rn and 220Rn"
86   PRINT "                from scintillation cell counts"
```

```
87  PRINT
89  PRINT
90  PRINT "                    Last Update 6/8/93"
91  PRINT ""
92  PRINT
168  PRINT "                                    "
169  PRINT
170  PRINT "*********************************************************************************************"
171  PRINT "*********************************************************************************************"
172  PRINT "**********************ENTER DAY, HOURS, AND MINUTES
SEPARATED**********************"
173  PRINT "************************BY COMMAS USING A 24-HOUR
CLOCK************************"
175  PRINT "********************(e.g., 13,14,05 FOR 2:05 ON THE THIRTEENTH)********************"
176  PRINT "*********************************************************************************************"
177  PRINT "*********************************************************************************************"

178  PRINT
180  PRINT "ENTER THE NUMERICAL MONTH AND 2 DIGIT YEAR"
181  INPUT "(SEPARATED BY A COMMA)"; TMONTH, TYEAR
182  IF TMONTH = 1 THEN MONTH$ = "JANUARY"
185  IF TMONTH = 2 THEN MONTH$ = "FEBRUARY"
186  IF TMONTH = 3 THEN MONTH$ = "MARCH"
187  IF TMONTH = 4 THEN MONTH$ = "APRIL"
188  IF TMONTH = 5 THEN MONTH$ = "MAY"
189  IF TMONTH = 6 THEN MONTH$ = "JUNE"
190  IF TMONTH = 7 THEN MONTH$ = "JULY"
191  IF TMONTH = 8 THEN MONTH$ = "AUGUST"
192  IF TMONTH = 9 THEN MONTH$ = "SEPTEMBER"
193  IF TMONTH = 10 THEN MONTH$ = "OCTOBER"
194  IF TMONTH = 11 THEN MONTH$ = "NOVEMBER"
195  IF TMONTH = 12 THEN MONTH$ = "DECEMBER"

197  PRINT
199  PRINT "WHICH LOCATION ARE THESE SAMPLES FROM?"
200  PRINT " ENTER: 1 = CHESTER"
210  PRINT "        2 = ARH ESTATE"
220  PRINT "        3 = CHEESEQUAKE STATE PARK"
230  PRINT "        4 = ANOTHER SITE"
240  INPUT TLOCATE
242  IF TLOCATE = 1 THEN LOCATE$ = "CHESTER"
244  IF TLOCATE = 2 THEN LOCATE$ = "ARH ESTATE"
246  IF TLOCATE = 3 THEN LOCATE$ = "CHEESEQUAKE"
248  IF TLOCATE = 4 THEN LOCATE$ = "FIELD SITE"
250  IF TLOCATE = 1 GOTO 300
260  IF TLOCATE = 2 GOTO 550
270  IF TLOCATE = 3 GOTO 650
273  IF TLOCATE = 4 GOTO 720
275  IF TLOCATE > 4 GOTO 9700
280  GOTO 741
290
'CCCCCCCC FOR CHESTER
300  PRINT
310  PRINT "WHICH HOLE ARE YOU GOING TO CALCULATE THE RN"
```

```
320  PRINT "CONCENTRATION FOR?"
330  PRINT " ENTER: 1 FOR HOLE # 1"
340  PRINT "       2 FOR HOLE # 2"
350  PRINT "       3 FOR HOLE # 3A"
360  PRINT "       31 FOR HOLE # 3B"
370  PRINT "       4 FOR HOLE # 4A"
380  PRINT "       41 FOR HOLE # 4B"
390  PRINT "       5 FOR HOLE # 5"
400  PRINT "       6 FOR HOLE # 6"
420  INPUT THOLE
425  PDEPTH = 1#
430  IF TLOCATE = 1 AND THOLE = 2 GOTO 460
440  IF TLOCATE = 1 AND THOLE = 5 GOTO 460
450  GOTO 741
460  PRINT
470  PRINT "WHICH DEPTH ARE THESE SAMPLES FROM?"
480  PRINT " ENTER:  1.5 FOR 1.5 METER "
490  PRINT "        1.0 FOR 1.0 METER "
500  PRINT "        0.5 FOR 0.5 METER "
510  PRINT "        0.25 FOR 0.25 METER "
520  INPUT PDEPTH
530  GOTO 741
'CCCCCCCC FOR ARH ESTATE
550  PRINT
560  PRINT "WHICH HOLE ARE YOU GOING TO CALCULATE THE RN "
570  PRINT "CONCENTRATION FOR?"
580  PRINT " ENTER: 1 FOR HOLE # 1"
590  PRINT "       2 FOR HOLE # 2"
600  PRINT "       3 FOR HOLE # 3"
610  PRINT "       4 FOR HOLE # 4A"
620  PRINT "       41 FOR HOLE # 4B"
625  PDEPTH = 1#
630  INPUT THOLE
640  GOTO 741
'CCCCCCCC FOR CHEESEQUAKE
650  PRINT
660  PRINT "WHICH HOLE ARE YOU GOING TO CALCULATE THE RN "
670  PRINT "CONCENTRATION FOR?"
677  PRINT " ENTER: 1 FOR HOLE # 1A"
681  PRINT "       11 FOR HOLE # 1B"
685  PRINT "       2 FOR HOLE # 2"
691  PRINT "       3 FOR HOLE # 3"
699  PRINT "       4 FOR HOLE # 4"
703  PRINT "       5 FOR HOLE # 5"
707  PDEPTH = 1#
711  INPUT THOLE
712  GOTO 741
'CCCCCCCC FOR ANY OTHER HOLE
720  PRINT
721  PRINT "IS THIS AN AIR SAMPLE OR A SOIL GAS SAMPLE"
722  INPUT "ENTER A FOR AIR, S FOR SOIL GAS"; TYPE$
723  IF TYPE$ = "S" OR TYPE$ = "s" GOTO 727
724  PDEPTH = 0
725  GOTO 741
```

```
727  PRINT "WHAT DEPTH IS THE SAMPLE TAKEN FROM?"
728  INPUT "ENTER 0.25, 0.5, 1.0 OR 1.5 M"; PDEPTH
'CCCCCC COEFFICIENT A = 222Rn
'CCCCCC COEFFICIENT B = 218Po
'CCCCCC COEFFICIENT C = 214Pb
'CCCCCC COEFFICIENT D = 214Bi
'CCCCCC COEFFICIENT E = 214Po
'CCCCCC COEFFICIENT F = 210Pb
741  YA = .0000020985#
742  YB = .0037877#
743  YC = .00043106#
744  YD = .00058642#
745  YE = 4226.5#
746  YF = .0000000009991#
747  KA = .00012591#
748  KB = .22726#
749  KC = .0259#
750  KD = .035183#
751  KE = .00984#
752  KF = .000000059966#
753  CNINE = 2.71828#
769  PRINT
770  INPUT "ENTER THE DAY, HOUR, MINUTE OF SAMPLE START"; FSTARTD, FSTARTH, FSTARTM
772  PRINT
774  INPUT "ENTER THE DAY, HOUR, MINUTE OF COUNT START"; FCOUNTD, FCOUNTH, FCOUNTM
776  PRINT
778  INPUT "HOW LONG WAS THE SAMPLE PERIOD [in minutes]"; FSAMPT
780  FSTARTM = FSTARTM + FSAMPT
782  FCOUNTM = FCOUNTM + 60#
784  FCOUNTH = FCOUNTH - 1#
'CCCCC TAKING AWAY ONE HOUR AND ADDING 60 MINUTES ASSURES THAT FCOUNTM WILL
'CCCCC ALWAYS BE > FSTARTM
786  FTIMEM = (FCOUNTM - FSTARTM)
788  FTIMEH = (FCOUNTH - FSTARTH) * 60#
790  FTIMED = (FCOUNTD - FSTARTD) * 24# * 60#
792  FONE = (FTIMED + FTIMEM + FTIMEH) * 60#
'CCCCC FONE = TIME FROM END OF SAMPLING TO BEGINNING OF COUNT IN SECONDS
794  PRINT
796  PRINT "THE COUNT STARTED"; FONE; "SECONDS AFTER SAMPLING ENDED"
798  PRINT
800  INPUT "HOW MANY MINUTES LONG WAS THE COUNT ?"; FLONG
802  FTWO = FONE + (FLONG * 60#)
804  RNSTOP = YA * FTWO
806  RNSTART = YA * FONE
808  POSTOP = YB * FTWO
810  POSTART = YB * FONE
812  PBSTOP = YC * FTWO
814  PBSTART = YC * FONE
816  BISTOP = YD * FTWO
818  BISTART = YD * FONE
820  POBSTOP = YE * FTWO
822  POBSTART = YE * FONE
824  PBBSTOP = YF * FTWO
826  PBBSTART = YF * FONE
```

```
828  RADON = (1 / -YA) * (CNINE ^ (-RNSTOP) - CNINE ^ (-RNSTART))
830  POLO218 = ((KB / KA) * (YA / (YB - YA))) * (((1 / -YA) * (CNINE ^ (-RNSTOP) - CNINE ^ (-
RNSTART))) - ((1 / -YB) * ((CNINE ^ (-POSTOP)) - CNINE ^ (-POSTART))))
832  CPB2141 = (YA * YB) / ((YB - YA) * (YC - YA))
1030 CPB2142 = (YA * YB) / ((YA - YB) * (YC - YB))
1040 CPB2143 = (YA * YB) / ((YA - YC) * (YB - YC))
1050 PB214A = (KC / KA) * ((1 / -YA) * (CPB2141 * ((CNINE ^ (-RNSTOP)) - (CNINE ^ (-RNSTART)))))
1060 PB214B = (KC / KA) * ((1 / -YB) * (CPB2142 * ((CNINE ^ (-POSTOP)) - (CNINE ^ (-POSTART)))))
1070 PB214C = (KC / KA) * ((1 / -YC) * (CPB2143 * ((CNINE ^ (-PBSTOP)) - (CNINE ^ (-PBSTART)))))
1080 LEAD214 = PB214A + PB214B + PB214C
1090 BI2141 = (YA * YB * YC) / ((YB - YA) * (YC - YA) * (YD - YA))
1100 BI2142 = (YA * YB * YC) / ((YA - YB) * (YC - YB) * (YD - YB))
1110 BI2143 = (YA * YB * YC) / ((YA - YC) * (YB - YC) * (YD - YC))
1120 BI2144 = (YA * YB * YC) / ((YA - YD) * (YB - YD) * (YC - YD))
1130 BI214A = (KD / KA) * ((1 / -YA) * (BI2141 * ((CNINE ^ (-RNSTOP)) - (CNINE ^ (-RNSTART)))))
1140 BI214B = (KD / KA) * ((1 / -YB) * (BI2142 * ((CNINE ^ (-POSTOP)) - (CNINE ^ (-POSTART)))))
1150 BI214C = (KD / KA) * ((1 / -YC) * (BI2143 * ((CNINE ^ (-PBSTOP)) - (CNINE ^ (-PBSTART)))))
1160 BI214D = (KD / KA) * ((1 / -YD) * (BI2144 * ((CNINE ^ (-BISTOP)) - (CNINE ^ (-BISTART)))))
1170 BISM214 = BI214A + BI214B + BI214C + BI214D
1180 PO2141 = (YA * YB * YC * YD) / ((YB - YA) * (YC - YA) * (YD - YA) * (YE - YA))
1190 PO2142 = (YA * YB * YC * YD) / ((YA - YB) * (YC - YB) * (YD - YB) * (YE - YB))
1200 PO2143 = (YA * YB * YC * YD) / ((YA - YC) * (YB - YC) * (YD - YC) * (YE - YC))
1205 PO2144 = (YA * YB * YC * YD) / ((YA - YD) * (YB - YD) * (YC - YD) * (YE - YD))
1210 PO2145 = (YA * YB * YC * YD) / ((YA - YE) * (YB - YE) * (YC - YE) * (YD - YE))
1215 PO214A = (KE / KA) * ((1 / -YA) * (PO2141 * ((CNINE ^ (-RNSTOP)) - (CNINE ^ (-RNSTART)))))
1220 PO214B = (KE / KA) * ((1 / -YB) * (PO2142 * ((CNINE ^ (-POSTOP)) - (CNINE ^ (-POSTART)))))
1222 PO214C = (KE / KA) * ((1 / -YC) * (PO2143 * ((CNINE ^ (-PBSTOP)) - (CNINE ^ (-PBSTART)))))
1224 PO214D = (KE / KA) * ((1 / -YD) * (PO2144 * ((CNINE ^ (-BISTOP)) - (CNINE ^ (-BISTART)))))
1226 PO214E = (KE / KA) * ((1 / -YE) * (PO2145 * ((CNINE ^ (-POBSTOP)) - (CNINE ^ (-POBSTART)))))
1228 POLO214 = PO214A + PO214B + PO214C + PO214D + PO214E
1230 PB2101 = (YA * YB * YC * YD * YE) / ((YB - YA) * (YC - YA) * (YD - YA) * (YE - YA) * (YF - YA))
1232 PB2102 = (YA * YB * YC * YD * YE) / ((YA - YB) * (YC - YB) * (YD - YB) * (YE - YB) * (YF - YB))
1234 PB2103 = (YA * YB * YC * YD * YE) / ((YA - YC) * (YB - YC) * (YD - YC) * (YE - YC) * (YF - YC))
1236 PB2104 = (YA * YB * YC * YD * YE) / ((YA - YD) * (YB - YD) * (YC - YD) * (YE - YD) * (YF - YD))
1238 PB2105 = (YA * YB * YC * YD * YE) / ((YA - YE) * (YB - YE) * (YC - YE) * (YD - YE) * (YF - YE))
1240 PB2106 = (YA * YB * YC * YD * YE) / ((YA - YF) * (YB - YF) * (YC - YF) * (YD - YF) * (YE - YF))
1242 PB210A = (KF / KA) * ((1 / -YA) * (PB2101 * ((CNINE ^ (-RNSTOP)) - (CNINE ^ (-RNSTART)))))
1244 PB210B = (KF / KA) * ((1 / -YB) * (PB2102 * ((CNINE ^ (-POSTOP)) - (CNINE ^ (-POSTART)))))
1246 PB210C = (KF / KA) * ((1 / -YC) * (PB2103 * ((CNINE ^ (-PBSTOP)) - (CNINE ^ (-PBSTART)))))
1248 PB210D = (KF / KA) * ((1 / -YD) * (PB2104 * ((CNINE ^ (-BISTOP)) - (CNINE ^ (-BISTART)))))
1250 PB210E = (KF / KA) * ((1 / -YE) * (PB2105 * ((CNINE ^ (-POBSTOP)) - (CNINE ^ (-POBSTART)))))
1252 PB210F = (KF / KA) * ((1 / -YF) * (PB2106 * ((CNINE ^ (-PBBSTOP)) - (CNINE ^ (-PBBSTART)))))
1254 LEAD210 = PB210A + PB210B + PB210C + PB210D + PB210E + PB210F
1256 ANSWER = (RADON + POLO218 + BISM214) / 60
1258 PRINT
1260 PRINT "THE INTEGRATION FACTOR IS"; ANSWER
1270 PRINT "DISINTEGRATIONS FROM"; FONE; "TO"; FTWO; "SECONDS"
1280 PRINT "DIVIDED BY THE INITIAL RN ACTIVITY"
1290 PRINT
1300 INPUT "WHAT IS THE TOTAL NUMBER OF COUNTS IN THE TIME PERIOD SELECTED"; FCOUNT
1330 PRINT
1340 INPUT "WHAT CELL DID YOU USE"; X
1365 CALIB(1) = 4.32
1370 FBKGR(1) = 6
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1380 CALIB(2) = 4.32
1385 FBKGR(2) = 14
1395 CALIB(3) = 4.32
1400 FBKGR(3) = 31
1410 CALIB(4) = 4.32
1415 FBKGR(4) = 25
1425 CALIB(5) = 4.32
1430 FBKGR(5) = 11
1440 CALIB(6) = 4.32
1445 FBKGR(6) = 10
1455 CALIB(7) = 4.32
1460 FBKGR(7) = 14
1470 CALIB(8) = 4.32
1475 FBKGR(8) = 10
1490 CALIB(9) = 4.32
1495 FBKGR(9) = 8
1510 CALIB(10) = 4.32
1515 FBKGR(10) = 8
1525 CALIB(11) = 4.24
1530 FBKGR(11) = 4
1540 CALIB(12) = 4.17
1545 FBKGR(12) = 4
1555 CALIB(13) = 4.41
1560 FBKGR(13) = 3
1570 CALIB(14) = 4.3
1575 FBKGR(14) = 3
1585 CALIB(15) = 4.34
1590 FBKGR(15) = 3
1600 CALIB(16) = 4.37
1605 FBKGR(16) = 2
1615 CALIB(17) = 4.34
1620 FBKGR(17) = 2
1630 CALIB(18) = 4.38
1635 FBKGR(18) = 2
1645 CALIB(19) = 4.38
1655 FBKGR(19) = 2
1665 CALIB(20) = 4.34
1670 FBKGR(20) = 2
1680 CALIB(21) = 4.09
1685 FBKGR(21) = 2
1695 CALIB(22) = 4.09
1700 FBKGR(22) = 2
1710 CALIB(23) = 4.49
1715 FBKGR(23) = 2
1725 CALIB(24) = 4.43
1730 FBKGR(24) = 2
1740 CALIB(25) = 4.24
1745 FBKGR(25) = 2
1755 CALIB(26) = 4.24
1760 FBKGR(26) = 2
1770 CALIB(27) = 4.24
1775 FBKGR(27) = 2
1785 CALIB(28) = 4.3
1790 FBKGR(28) = 2
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1800 CALIB(29) = 4.39
1805 FBKGR(29) = 2
1815 CALIB(30) = 4.81
1820 FBKGR(30) = 2
1830 CALIB(31) = 4.38
1835 FBKGR(31) = 2
1845 CALIB(32) = 4.38
1850 FBKGR(32) = 2
1905 CALIB(33) = 4.35
1910 FBKGR(33) = 1
1920 CALIB(34) = 4.2
1925 FBKGR(34) = 1
1935 CALIB(35) = 4.61
1940 FBKGR(35) = 1
1955 CALIB(36) = 4.16
1960 FBKGR(36) = 1
1975 CALIB(37) = 4.32
1980 FBKGR(37) = 1
1995 CALIB(38) = 4.32
2000 FBKGR(38) = 1
2010 CALIB(39) = 3.95
2015 FBKGR(39) = 1
2025 CALIB(40) = 4.53
2030 FBKGR(40) = 1
2040 CALIB(41) = 4.26
2045 FBKGR(41) = 1
2055 CALIB(42) = 4.32
2060 FBKGR(42) = 1
2235 CALIB(103) = 4.5
2240 FBKGR(103) = 1
2250 CALIB(107) = 4.33
2255 FBKGR(107) = 1
2265 CALIB(151) = 4.6
2270 FBKGR(151) = 1
2280 CALIB(152) = 4.6
2285 FBKGR(152) = 1
2295 CALIB(153) = 4.34
2300 FBKGR(153) = 1
2480 PRINT
2560 PRINT
2580 PRINT " The background for cell"; X; "is"; FBKGR(X); "cpm"
2780 PRINT
2800 PRINT " The calibration for cell"; X; "is"; CALIB(X); "pCi/L per initial dpm"
' THESE CALIBRATIONS WERE LAST PERFORMED 2/22/93
2808 PRINT
2809 PRINT
2810 INPUT "WOULD YOU LIKE TO ENTER NEW BACKGROUND? Y OR N"; YESNO$
2820 IF YESNO$ = "Y" OR YESNO$ = "y" GOTO 2860
2830 IF YESNO$ = "N" OR YESNO$ = "n" GOTO 2910
2840 IF YESNO$ <> "Y" OR YESNO$ <> "N" GOTO 9700
2860 PRINT
2890 INPUT "ENTER NEW BACKGROUND IN CPM ======>>"; FBKGR(X)
2910 PRINT
3230 INPUT "WOULD YOU LIKE TO ENTER NEW CALIBRATION? Y OR N"; YESNO$
```

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3245 PRINT
3260 IF YESNO$ = "Y" OR YESNO$ = "y" GOTO 3300
3270 IF YESNO$ = "N" OR YESNO$ = "n" GOTO 3330
3280 IF YESNO$ <> "Y" OR YESNO$ <> "N" GOTO 9700
3300 PRINT "ENTER NEW CALIBRATION IN UNITS OF"
3310 INPUT "(pCi/L)/(INITIAL CPM)"; CALIB(X)
3330 FINAL = ((FCOUNT - (FBKGR(X) * FLONG)) / ANSWER) * CALIB(X)
3340 FINAP = FINAL * .037
3345 PRINT
3346 PRINT
3348 PRINT " On "; MONTH$; FSTARTD; "at"; FSTARTH; ":"; FMINUTE
3350 PRINT " The radon concentration at "; LOCATE$; " hole#"; THOLE; "at a depth of"; PDEPTH; "m"
3352 PRINT "                    = "; FINAL; "pCi/L"
3353 PRINT "                    = "; FINAP; "kBq/m3"
3356 PRINT
3364 FIRST$ = "      "
3365 SECOND$ = "            "
3366 THIRD$ = "                  "
3367 FOURTH$ = "                        "
3368 FIFTH$ = "                           "
3369 SIXTH$ = "                              "
3370 SEVENTH$ = "                                 "
3382 PRINT
3383 PRINT
3384 PRINT ""
3385 INPUT "DO YOU WANT TO WRITE THIS RADON VALUE TO A DATA FILE? (Y OR N)"; YESNO$
3386 PRINT " "
3390 IF YESNO$ = "Y" OR YESNO$ = "y" GOTO 3420
3400 IF YESNO$ = "N" OR YESNO$ = "n" GOTO 4230
3410 GOTO 9700
'CCCCCCCCC
3420 IF TLOCATE = 1 GOTO 3427
3421 IF TLOCATE = 2 GOTO 3434
3422 IF TLOCATE = 3 GOTO 3441
3423 IF TLOCATE = 4 GOTO 3449
3427 PRINT "IF YOU WOULD LIKE TO INPUT DATA TO SITE1.DAT"
3428 INPUT "TYPE A, OTHERWISE TYPE IN FILENAME"; FILENAME$
3430 IF FILENAME$ <> "A" THEN GOTO 3450
3431 IF FILENAME$ = "A" THEN FILENAME$ = "C:\SITE1.DAT"
3433 GOTO 3450
3434 PRINT "IF YOU WOULD LIKE TO INPUT DATA TO SITE2.DAT"
3435 INPUT "TYPE A, OTHERWISE TYPE IN FILENAME"; FILENAME$
3438 IF FILENAME$ <> "A" THEN GOTO 3450
3439 IF FILENAME$ = "A" THEN FILENAME$ = "C:\SITE2.DAT"
3440 GOTO 3450
3441 PRINT "IF YOU WOULD LIKE TO INPUT DATA TO SITE3.DAT"
3442 INPUT "TYPE A, OTHERWISE TYPE IN FILENAME"; FILENAME$
3443 IF FILENAME$ <> "A" THEN GOTO 3450
3444 IF FILENAME$ = "A" THEN FILENAME$ = "C:\SITE3.DAT"
3447 GOTO 3450
3449 INPUT "ENTER FILE NAME"; FILENAME$
3450 PRINT
3451 OPEN FILENAME$ FOR APPEND AS #1
3452 IF TLOCATE = 1 THEN LOCATE$ = "CHESTER"
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3454 IF TLOCATE = 2 THEN LOCATE$ = "ARH ESTATE"
3456 IF TLOCATE = 3 THEN LOCATE$ = "CHEESEQUAKE"
3458 IF TLOCATE = 4 THEN LOCATE$ = "FIELD SITE"
3459 IF TLOCATE = 4 GOTO 4145
3460 FMINUTE = FSTARTM - FSAMPT
3461 PRINT #1, "   "
3462 IF TSAME = 1 GOTO 3466
3463 IF TSAME = 0 GOTO 3467
3466 PRINT #1, "DATE = "; TMONTH; "/"; FSTARTD; "/"; TYEAR
3467 PRINT #1, "LOCATION = "; LOCATE$; " HOLE#"; THOLE; " DEPTH ="; PDEPTH; "m"
3470 PRINT #1, "TIME = "; FSTARTH; ":"; FMINUTE
3510 PRINT #1, "RADON = "; FINAL; "pCi/L"
3520 GOTO 4230
'CCCCCC FOR ANY OTHER PLACE
4145 IF TSAME = 1 GOTO 4160
4150 IF TSAME = 0 GOTO 4170
4160 PRINT "ENTER DESCRIPTION OF SITE FOR FUTURE REFERENCE"
4161 INPUT REFER$
4162 PRINT #1, REFER$
4164 PRINT #1, "DATE "; FIRST$; "      TIME"; FIRST$; "      RADON "
4165 PRINT #1, TMONTH; "-"; FSTARTD; "-"; TYEAR; FIRST$; FSTARTH; ":"; FMINUTE; FIRST$; FINAL
4167 GOTO 4230
4170 PRINT #1, SECOND$; "     "; FSTARTH; ":"; FMINUTE; FIRST$; FINAL
 'CC  CALCULATE THORON CONCENTRATION NOW
4230 CLOSE #1
4235 PRINT
4240 INPUT "IS THERE DATA FOR THORON ? (Y OR N)"; YESNO$
4260 IF YESNO$ = "Y" OR YESNO$ = "y" GOTO 4300
4270 IF YESNO$ = "N" OR YESNO$ = "n" GOTO 9501
4280 IF YESNO$ <> "Y" OR YESNO$ <> "N" GOTO 9700
4300 PRINT
4310 PRINT "ENTER THE NUMBER OF SECONDS ELAPSED AFTER SAMPLING"
4320 INPUT "TO THE TIME OF A COUNT FOR THORON"; FTHREE
4340 PRINT
4350 IF PDEPTH = 0 THEN FCOMP = 5
4355 IF PDEPTH = .25 THEN FCOMP = 10
4360 IF PDEPTH = .5 THEN FCOMP = 15
4365 IF PDEPTH = 1# THEN FCOMP = 20
4370 IF PDEPTH = 1.5 THEN FCOMP = 25
4400 FTHREE = FTHREE + FCOMP
4420 PRINT FCOMP; "SECONDS HAVE BEEN ADDED TO COMPENSATE FOR DECAY"
4430 PRINT "OF THORON CAPTURED IN CELL ENROUTE DURING SAMPLING"
4435 PRINT
4450 INPUT "ENTER THE NUMBER OF MINUTES OF COUNTS FOR THORON"; FFMIN
4470 FFOUR = (FFMIN * 60) + FTHREE
4480 RNSTOP2 = YA * FFOUR
4490 RNSTART2 = YA * FTHREE
4500 POSTOP2 = YB * FFOUR
4510 POSTART2 = YB * FTHREE
4520 PBSTOP2 = YC * FFOUR
4530 PBSTART2 = YC * FTHREE
4540 BISTOP2 = YD * FFOUR
4550 BISTART2 = YD * FTHREE
4560 POBSTOP2 = YE * FFOUR
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4570 POBSTART2 = YE * FTHREE
4580 PBBSTOP2 = YF * FFOUR
4590 PBBSTART2 = YF * FTHREE
4600 RADON2 = (1 / -YA) * (CNINE ^ (-RNSTOP2) - CNINE ^ (-RNSTART2))
4610 POLO2182 = ((KB / KA) * (YA / (YB - YA))) * (((1 / -YA) * (CNINE ^ (-RNSTOP2) - CNINE ^ (-
RNSTART2))) - ((1 / -YB) * ((CNINE ^ (-POSTOP2)) - CNINE ^ (-POSTART2))))
4620 PB214A2 = (KC / KA) * ((1 / -YA) * (CPB2141 * ((CNINE ^ (-RNSTOP2)) - (CNINE ^ (-RNSTART2)))))
4630 PB214B2 = (KC / KA) * ((1 / -YB) * (CPB2142 * ((CNINE ^ (-POSTOP2)) - (CNINE ^ (-POSTART2)))))
4640 PB214C2 = (KC / KA) * ((1 / -YC) * (CPB2143 * ((CNINE ^ (-PBSTOP2)) - (CNINE ^ (-PBSTART2)))))
4650 LEAD2142 = PB214A2 + PB214B2 + PB214C2
4660 BI214A2 = (KD / KA) * ((1 / -YA) * (BI2141 * ((CNINE ^ (-RNSTOP2)) - (CNINE ^ (-RNSTART2)))))
4670 BI214B2 = (KD / KA) * ((1 / -YB) * (BI2142 * ((CNINE ^ (-POSTOP2)) - (CNINE ^ (-POSTART2)))))
4680 BI214C2 = (KD / KA) * ((1 / -YC) * (BI2143 * ((CNINE ^ (-PBSTOP2)) - (CNINE ^ (-PBSTART2)))))
4690 BI214D2 = (KD / KA) * ((1 / -YD) * (BI2144 * ((CNINE ^ (-BISTOP2)) - (CNINE ^ (-BISTART2)))))
4700 BISM2142 = BI214A2 + BI214B2 + BI214C2 + BI214D2
4710 PO214A2 = (KE / KA) * ((1 / -YA) * (PO2141 * ((CNINE ^ (-RNSTOP2)) - (CNINE ^ (-RNSTART2)))))
4720 PO214B2 = (KE / KA) * ((1 / -YB) * (PO2142 * ((CNINE ^ (-POSTOP2)) - (CNINE ^ (-POSTART2)))))
4730 PO214C2 = (KE / KA) * ((1 / -YC) * (PO2143 * ((CNINE ^ (-PBSTOP2)) - (CNINE ^ (-PBSTART2)))))
4740 PO214D2 = (KE / KA) * ((1 / -YD) * (PO2144 * ((CNINE ^ (-BISTOP2)) - (CNINE ^ (-BISTART2)))))
4750 PO214E2 = (KE / KA) * ((1 / -YE) * (PO2145 * ((CNINE ^ (-POBSTOP2)) - (CNINE ^ (-POBSTART2)))))
4760 POLO2142 = PO214A2 + PO214B2 + PO214C2 + PO214D2 + PO214E2
4770 PB210A2 = (KF / KA) * ((1 / -YA) * (PB2101 * ((CNINE ^ (-RNSTOP2)) - (CNINE ^ (-RNSTART2)))))
4780 PB210B2 = (KF / KA) * ((1 / -YB) * (PB2102 * ((CNINE ^ (-POSTOP2)) - (CNINE ^ (-POSTART2)))))
4790 PB210C2 = (KF / KA) * ((1 / -YC) * (PB2103 * ((CNINE ^ (-PBSTOP2)) - (CNINE ^ (-PBSTART2)))))
4800 PB210D2 = (KF / KA) * ((1 / -YD) * (PB2104 * ((CNINE ^ (-BISTOP2)) - (CNINE ^ (-BISTART2)))))
4810 PB210E2 = (KF / KA) * ((1 / -YE) * (PB2105 * ((CNINE ^ (-POBSTOP2)) - (CNINE ^ (-POBSTART2)))))
4820 PB210F2 = (KF / KA) * ((1 / -YF) * (PB2106 * ((CNINE ^ (-PBBSTOP2)) - (CNINE ^ (-PBBSTART2)))))
4830 LEAD2102 = PB210A2 + PB210B2 + PB210C2 + PB210D2 + PB210E2 + PB210F2
4900 ANSWERP = (RADON2 + POLO2182 + BISM2142) / 60
6650 PRINT
6900 PRINT "WHAT IS THE TOTAL NUMBER OF COUNTS IN THE TIME PERIOD"
6920 PRINT FTHREE; "SECONDS TO"; FFOUR; " SECONDS"
6930 INPUT FCOUNT2
'CCC FRNCTS IS EQUAL TO THE NUMBER OF COUNTS DURING THORON COUNTING PERIOD
'CCC (FIRST MINUTE OR SO) THAT ARE DUE TO RADON
7000 FRNCTS = ((FCOUNT - (FBKGR(X) * FLONG)) / ANSWER) * ANSWERP
7010 PRINT
7020 PRINT "THE INTEGRATION FACTOR FOR 222RN IS "; ANSWERP; "DISINTEGRATIONS FROM";
FTHREE
7030 PRINT "TO "; FFOUR; "SECONDS AFTER SAMPLING ENDED DIVIDED BY THE INITIAL RN
ACTIVITY "
7035 PRINT
7070 PRINT "THE COUNTS DUE TO RADON FROM"; FTHREE; "TO"; FFOUR; "SECONDS ="; FRNCTS
'CCC FTHCTS IS EQUAL TO THE NUMBER OF COUNTS IN THE FIRST MINUTE OR SO THAT
'CCC ARE DUE EXCLUSIVELY TO THORON AND IT'S PROGENY, WHICH IS EQUAL TO THE
'CCC TOTAL COUNTS MINUS BACKGROUND MINUS COUNTS DUE TO RADON AND PROGENY
7090 FTHCTS = (FCOUNT2 - (FBKGR(X) * FFMIN) - FRNCTS)
7100 PRINT
7110 PRINT "THE COUNTS DUE TO THORON + PROGENY FROM"; FTHREE; "TO"; FFOUR; "SECONDS"
7130 PRINT " = TOTAL COUNTS IN PERIOD MINUS COUNTS DUE TO RADON AND BACKGROUND"
7150 PRINT " = "; FCOUNT2; "-"; FRNCTS; "-"; FBKGR(X); " = "; FTHCTS; "COUNTS"
7160 CTONE = -157.56#
7170 CTTWO = .209#
7180 CTEN = .0127#
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' CTEN = RN220 DECAY CONSTANT
7300 CELEVEN = 4.78#
' CELEVEN = PO216 DECAY CONSTANT
7400 FTHSTART = CTEN * FTHREE
7500 FTHSTOP = CTEN * FFOUR
7600 PO6START = CELEVEN * FTHREE
7700 PO6STOP = CELEVEN * FFOUR
7800 ADDFIVE = CTONE * (CNINE ^ (-FTHSTOP) - CNINE ^ (-FTHSTART))
' ADDFIVE = DECAY OF THORON AFTER TIME
7900 ADDSIX = CTTWO * (CNINE ^ (-PO6STOP) - CNINE ^ (-PO6START))
' ADDSIX = BUILDUP OF PO216 AFTER TIME
8000 ANSWER2 = (ADDFIVE + ADDSIX) / 60
8050 PRINT
8100 PRINT "THE INTEGRATION FACTOR FOR THORON IS "; ANSWER2; "DISINTEGRATIONS FROM";
FTHREE
8110 PRINT "TO"; FFOUR; " SECONDS DIVIDED BY THE INITIAL THORON ACTIVITY IN DPM"
8140 BTHIN = (FTHCTS / ANSWER2)
8150 BRADIN = ((FCOUNT - FBKGR(X)) / ANSWER)
8155 PRINT
8310 PRINT "THE INITIAL COUNTS DUE TO RADON ="; BRADIN
8330 PRINT "THE INITIAL COUNTS DUE TO THORON ="; BTHIN
8400 FINALTH = BTHIN * CALIB(X)
8450 FINALTHP = FINALTH * .037
8475 PRINT
8476 PRINT
8549 PRINT "  The thoron concentration at "; LOCATE$; " hole#"; THOLE; "on "; MONTH$; FSTARTD; ",";
TYEAR; "at"; FSTARTH; ":"; FMINUTE
8550 PRINT "                    ="; FINALTH; "pCi/L"
8560 PRINT "                    ="; FINALTHP; "kBq/m3"
8679 PRINT
8684 PRINT ""
8685 INPUT "DO YOU WANT TO WRITE THIS THORON VALUE TO A DATA FILE? (Y OR N)"; YESNO$
8686 PRINT ""
8690 IF YESNO$ = "Y" OR YESNO$ = "y" GOTO 8720
8700 IF YESNO$ = "N" OR YESNO$ = "n" GOTO 9500
8710 GOTO 9700
'CCCCCCCCC
8720 IF TLOCATE = 1 GOTO 8727
8721 IF TLOCATE = 2 GOTO 8734
8722 IF TLOCATE = 3 GOTO 8844
8723 IF TLOCATE = 4 GOTO 9000
8727 PRINT "IF YOU WOULD LIKE TO INPUT DATA TO SITE1.DAT"
8728 INPUT "TYPE A, OTHERWISE TYPE IN FILENAME"; FILENAME$
8730 IF FILENAME$ <> "A" THEN GOTO 8849
8731 IF FILENAME$ = "A" THEN FILENAME$ = "C:\SITE1.DAT"
8733 GOTO 8980
8734 PRINT "IF YOU WOULD LIKE TO INPUT DATA TO SITE2.DAT"
8735 INPUT "TYPE A, OTHERWISE TYPE IN FILENAME"; FILENAME$
8738 IF FILENAME$ <> "A" THEN GOTO 8849
8840 IF FILENAME$ = "A" THEN FILENAME$ = "C:\SITE2.DAT"
8841 GOTO 8980
8844 PRINT "IF YOU WOULD LIKE TO INPUT DATA TO SITE3.DAT"
8845 INPUT "TYPE A, OTHERWISE TYPE IN FILENAME"; FILENAME$
8846 IF FILENAME$ <> "A" THEN GOTO 8849
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8847 IF FILENAME$ = "A" THEN FILENAME$ = "C:\SITE3.DAT"
8848 GOTO 8980
8849 OPEN FILENAME$ FOR APPEND AS #2
8855 PRINT #2, "DATE = "; TMONTH; "/"; FSTARTD; "/"; TYEAR
8860 PRINT #2, "LOCATION = "; LOCATE$; " HOLE#"; THOLE; " DEPTH ="; PDEPTH; "m"
8870 PRINT #2, "TIME = "; FSTARTH; ":"; FMINUTE
8880 PRINT #2, "THORON = "; FINALTH; "pCi/L"
8890 GOTO 9090
8980 OPEN FILENAME$ FOR APPEND AS #1
8985 PRINT #1, "THORON = "; FINALTH; "pCi/L"
8990 GOTO 9500
'CCCCCC FOR ANY OTHER PLACE
9000 INPUT "ENTER FILE NAME"; FILENAME$
9002 OPEN FILENAME$ FOR APPEND AS #2
9006 IF TSAME = 1 GOTO 9020
9010 IF TSAME = 0 GOTO 9080
9020 PRINT "ENTER DESCRIPTION OF SITE FOR FUTURE REFERENCE"
9030 INPUT REFER$
9040 PRINT #2, REFER$
9050 PRINT #2, "DATE "; FIRST$; "      TIME"; FIRST$; "      THORON "
9060 PRINT #2, TMONTH; "-"; FSTARTD; "-"; TYEAR; FIRST$; FSTARTH; ":"; FMINUTE; FIRST$;
FINALTH
9070 GOTO 9090
9080 PRINT #2, SECOND$; "     "; FSTARTH; ":"; FMINUTE; FIRST$; FINALTH
9090 CLOSE #2
9091 GOTO 9501
9500 CLOSE #1
9501 PRINT
9502 INPUT "WOULD YOU LIKE TO CALCULATE ANOTHER CONCENTRATION ? (Y OR N)"; YESNO$
9550 TSAME = 1
9560 IF YESNO$ = "Y" OR YESNO$ = "y" GOTO 9580
9570 IF YESNO$ = "N" OR YESNO$ = "n" GOTO 9700
9580 PRINT
9590 PRINT "IS THIS FOR THE SAME DAY AND LOCATION AS PREVIOUSLY?"
9600 INPUT "(Y OR N)"; YESNO$
9620 IF YESNO$ = "Y" OR YESNO$ = "y" THEN
9630 TSAME = TSAME - 1
9640 GOTO 250
9650 ELSEIF YESNO$ = "N" THEN
9660 GOTO 178
9670 ELSE
9680 GOTO 9700
9690 END IF
9700 END
```

Appendix B

DATA TABLES

The following tables show all collected data for dates of soil gas sampling discussed in this dissertation. Meteorological data was available from the EML Chester, NJ station corresponding to Site 1 as discussed in this dissertation. On-site meteorological data was not available for Sites 2 and 3 (Matawan, NJ and Cheeesequake, NJ, respectively, both approximately 70 km from Site 1). In the meteorological data table, TEMP is the temperature at 1 m height in ^{o}Celsius; P is the atmospheric pressure in millibar, RH is the relative humidity in %, and WS is the wind speed in meters/second. Blanks in the meteorological data tables indicate that data was not available. In the soil gas data tables, blanks indicate that data was not available (e.g., blanks in the ^{220}Rn column before May 1990 when these measurements started), or was not able to be obtained due to equipment problems or failures.

Table B.1 Site 1 Data: Meteorological Data

Date	TEMP (^{0}C)	P (mb)	RH (%)	WS (m/s)
10/10/90		1013.00	99.60	5.70
10/25/90	12.70	884.90	97.90	2.80
11/7/90				
11/21/90				
12/10/90	6.10	979.40	59.80	4.70
12/27/90	-7.30	1011.60	36.30	1.20
1/9/91				
1/24/91	-4.50	986.10	25.80	5.90
2/6/91	6.10	994.70	96.10	1.60
2/22/91	12.70	977.10	18.40	5.20
3/12/91				
3/26/91				
4/15/91	5.30	988.80	96.50	3.30
4/27/91	16.70	985.70	80.90	2.50
5/9/91	11.60	997.10	46.10	1.40
5/24/91	18.60	991.20	88.30	1.60
6/7/91	23.30	996.30	31.30	2.50
6/21/91	25.30	985.30	37.90	3.30
7/2/91	21.00	983.30	37.50	2.80
7/19/91	30.40	986.90	54.70	0.90
8/2/91	28.40	987.60	30.90	0.60
8/15/91	20.20	984.10	97.70	1.20
8/29/91	26.90	992.00	70.70	2.00
9/12/91	13.90	988.00	38.30	1.60
9/26/91	8.80	972.70	97.70	1.20
10/11/91	11.20	977.10	80.90	1.40
10/25/91	12.70	998.60	97.70	0.00
11/7/91	1.80	991.20	34.00	1.60
11/22/91	14.30	987.30	97.70	0.60
12/5/91	-7.30	994.30	44.90	4.20
12/17/91	-8.80	991.60	47.70	1.70
12/31/91	-8.40	1004.50	58.20	2.00
1/23/92	2.20	982.20	96.90	2.30
2/4/92	-2.90	979.20	22.30	8.10
2/21/92	0.20	989.20		4.20
3/13/92				
3/28/92	-15.50	978.20		7.20
4/10/92	-8.00	989.12		2.70
4/24/92				
5/8/92				
5/22/92	25.70	994.70		2.80

Table B.1 Site 1 Data: Holes 1 and 2

Date	Hole 1 222Rn kBq m-3	Hole 1 220Rn kBq m-3	Hole 1 220/222	Hole 1 perm m2	Hole 2 222Rn kBq m-3	Hole 2 220Rn kBq m-3	Hole 2 220/222	Hole 2 perm m2
9/8/89	314				151			
9/28/89	389				83			
10/12/89	351				83			
10/25/89	221				31			
11/9/89	350				48			
11/22/89	268				31			
12/7/89	441				88			
12/21/89	418				82			
1/4/90	403				108			
1/17/90	390				44			
2/1/90	144				57			
2/14/90	197							
2/28/90	389				133			
3/15/90	168				64			
3/28/90	298	133	0.44		376			
4/12/90	220	140	0.64		148	62	0.42	
4/26/90	161	187	1.16		114	135	1.18	
5/10/90	155	204	1.31	1.50E-12	104	191	1.84	6.13E-13
5/22/90	169	194	1.15	1.02E-12	320	150	0.47	8.36E-13
6/6/90	129	209	1.61	1.03E-12	135	178	1.32	8.49E-13
6/20/90	191	214	1.12	1.06E-12	172	200	1.16	9.99E-13
7/3/90	162	245	1.52	1.03E-12	188	233	1.24	8.08E-13
7/18/90	207	220	1.06	1.50E-12	206	234	1.14	1.26E-12
7/30/90	237	227	0.96	1.56E-12	285	218	0.77	2.66E-12
8/15/90	207	213	1.03	1.03E-12	262	215	0.82	1.66E-12
8/31/90	214	206	0.96	1.24E-12	291	227	0.78	2.22E-12
9/14/90	233	227	0.97	1.36E-12	309	249	0.80	3.00E-12
9/27/90	356	292	0.82	1.31E-12	691	260	0.38	2.80E-12
10/10/90	253	216	0.85	1.31E-12	336	180	0.54	2.75E-12
10/25/90	396	214	0.54	9.20E-13	832	326	0.39	2.62E-12
11/7/90	218	202	0.93	1.32E-12	291	216	0.74	2.90E-12
11/21/90	483	189	0.39	1.20E-12	629	250	0.40	2.86E-12
12/10/90	246	160	0.65	1.06E-12	417	131	0.31	2.48E-12
12/27/90	280	200	0.72	9.69E-13	729	163	0.22	2.50E-12
1/9/91	419	161	0.38	1.24E-12	782	252	0.32	2.50E-12
1/24/91	354	302	0.85	1.21E-12	607	148	0.24	2.50E-12
2/6/91	158	177	1.12	1.33E-12	77	128	1.67	2.72E-12
2/22/91	212	218	1.03	1.75E-12	229	136	0.59	2.72E-12
3/12/91	214	206	0.97	1.69E-12	204	172	0.85	2.56E-12
3/26/91	234	137	0.59	1.25E-12	250	123	0.49	2.43E-12
4/15/91	241	196	0.81	1.40E-12	470	152	0.32	2.57E-12
4/27/91	193	194	1.01	1.03E-12	226	149	0.66	2.27E-12
5/9/91	167	196	1.18	1.29E-12	176	166	0.94	2.66E-12
5/24/91	120	245	2.04	1.40E-12	130	204	1.57	2.64E-12
6/7/91	195	280	1.43	1.48E-12	351	258	0.73	2.73E-12
6/21/91	216	240	1.11	1.61E-12	223	259	1.16	2.69E-12
7/2/91	232	244	1.05	1.54E-12	253	210	0.83	2.66E-12
7/19/91	230	268	1.16	1.61E-12	193	300	1.55	2.66E-12
8/2/91	299	267	0.90	1.56E-12	321	258	0.80	2.66E-12

Table B.1 Site 1 Data: Holes 1 and 2, continued

Date	Hole 1 222Rn kBq m-3	Hole 1 220Rn kBq m-3	Hole 1 220/222	Hole 1 perm m2	Hole 2 222Rn kBq m-3	Hole 2 220Rn kBq m-3	Hole 2 220/222	Hole 2 perm m2
8/15/91	302	248	0.82	1.71E-12	310	290	0.94	2.85E-12
8/29/91	274	292	1.07	1.71E-12	263	262	0.99	2.92E-12
9/12/91	348	247	0.71	1.61E-12	419	291	0.69	2.73E-12
9/26/91	399	164	0.41	1.06E-12	511	161	0.32	2.60E-12
10/11/91	313	213	0.68	1.81E-12	608	213	0.35	3.07E-12
10/25/91	377	235	0.62	2.85E-12	621	142	0.23	8.79E-12
11/7/91	451	221	0.49	3.07E-12	784	219	0.28	1.43E-11
11/22/91	273	219	0.80	3.00E-12	266	225	0.85	3.69E-11
12/5/91	300	211	0.70	2.11E-12	404	404	1.00	4.79E-11
12/17/91	241	155	0.65	1.57E-12	648	139	0.21	3.18E-12
12/31/91	358	205	0.57	2.10E-12	686	129	0.19	3.63E-12
1/23/92	477	149	0.31	1.71E-12	810	77	0.10	3.63E-12
2/5/92	410	244	0.60	3.63E-12	122	334	2.74	1.50E-11
2/21/92	203	196	0.96	3.36E-12				1.73E-11
3/13/92				2.66E-12	122	115	0.94	1.50E-11
3/28/92	119	145	1.22	2.35E-12	36	108	3.00	4.39E-11
4/10/92	189	158	0.83	3.07E-12	143	126	0.88	1.46E-11
4/24/92	139	199	1.05	3.63E-12	107	166	1.55	4.39E-11
5/8/92	194	180	0.93	3.99E-12	335	210	0.63	4.39E-11
5/22/92	145	221	1.52	3.63E-12				

Table B.1 Site 1 Data: Holes 3a and 3b

Date	Hole 3a 222Rn kBq m-3	Hole 3a 220Rn kBq m-3	Hole 3a 220/222	Hole 3a perm m2	Hole 3b 222Rn kBq m-3	Hole 3b 220Rn kBq m-3	Hole 3b 220/222	Hole 3b perm m2
9/8/89	740							
9/28/89	1672				335			
10/12/89	1438				82			
10/25/89	1033				284			
11/9/89	1272				504			
11/22/89	488				261			
12/7/89	956				291			
12/21/89	899				488			
1/4/90	1076				910			
1/17/90	1086				737			
2/1/90	652				301			
2/14/90	783				413			
2/28/90	933				556			
3/15/90	593				362			
3/28/90	1082	170	0.16		568	268	0.47	
4/12/90	592	338	0.57		305	158	0.52	
4/26/90	739	465	0.63		442	233	0.53	
5/10/90	729	445	0.61	3.00E-12	469	297	0.63	7.13E-13
5/22/90	1203	561	0.47	2.66E-12	883	238	0.27	9.99E-13
6/6/90	818	410	0.50	3.00E-12	569	295	0.52	9.22E-13
6/20/90	907	533	0.59	3.23E-12	614	344	0.56	1.12E-12
7/3/90	853	496	0.58	3.00E-12	638	343	0.54	7.07E-13
7/18/90	812	528	0.65	3.31E-12	630	420	0.67	1.26E-12
7/30/90	915	614	0.67	3.23E-12	718			1.71E-12
8/15/90	1065	523	0.49	3.00E-12	746			9.70E-13
8/31/90	1092	588	0.54	3.58E-12	801	380	0.47	9.71E-13
9/14/90	1024	554	0.54	3.30E-12	813	382	0.47	1.28E-12
9/27/90	1289	944	0.73	3.14E-12	1082	343	0.32	1.32E-12
10/10/90	1067	498	0.47	2.93E-12	853	350	0.41	1.26E-12
10/25/90	1586	481	0.30	3.00E-12	1125	289	0.26	1.03E-12
11/7/90	925	580	0.63	3.03E-12	636	293	0.46	9.45E-13
11/21/90	933	627	0.67	3.14E-12	575	371	0.64	1.03E-12
12/10/90	981	578	0.59	2.96E-12	547	319	0.58	1.04E-12
12/27/90	1045	380	0.36	2.85E-12	0			1.28E-12
1/9/91	1386	483	0.35	2.66E-12	760	699	0.92	1.06E-12
1/24/91	1340	347	0.26	2.66E-12	449	655	1.46	1.21E-12
2/6/91	597	320	0.54	3.18E-12	297	314	1.06	9.99E-13
2/22/91	820	362	0.44	2.81E-12	0			1.04E-12
3/12/91	463	387	0.84	2.80E-12	227	318	1.40	8.28E-13
3/26/91	819	367	0.45	2.85E-12	537	252	0.47	9.99E-13
4/15/91	1074	492	0.46		858	301	0.35	
4/27/91	723	458	0.63	2.41E-12	499	333	0.67	1.03E-12
5/9/91	991	285	0.29	2.66E-12	706	208	0.29	9.41E-13
5/24/91	807	488	0.60	2.78E-12	530	343	0.65	1.34E-12
6/7/91	864	598	0.69	2.66E-12	665	446	0.67	1.65E-12
6/21/91	677	602	0.89	2.75E-12	531	396	0.75	1.66E-12
7/2/91	750	605	0.81	2.66E-12	582	411	0.71	1.76E-12
7/19/91	622	632	1.02	2.66E-12	495	381	0.77	1.94E-12
8/2/91	795	623	0.78	2.66E-12	651	384	0.59	1.81E-12

Tabel B.1 Site 1 Data: Holes 3a and 3b, continued

Date	Hole 3a 222Rn kBq m-3	Hole 3a 220Rn kBq m-3	Hole 3a 220/222	Hole 3a perm m2	Hole 3b 222Rn kBq m-3	Hole 3b 220Rn kBq m-3	Hole 3b 220/222	Hole 3b perm m2
8/15/91	903	723	0.80	2.85E-12	736	473	0.64	2.00E-12
8/29/91	799	687	0.86	2.92E-12	665	424	0.64	1.94E-12
9/12/91	1030	762	0.74	2.80E-12	838	444	0.53	2.66E-12
9/26/91	1062	426	0.40	2.66E-12	803	253	0.32	1.13E-12
10/11/91	1155	409	0.35	2.85E-12	737	337	0.46	6.37E-13
10/25/91	1292	591	0.46	8.79E-12	1064	329	0.31	1.48E-12
11/7/91	1305	563	0.43	2.20E-11	949	598	0.63	2.60E-12
11/22/91	857	462	0.54	3.69E-11	723	330	0.46	3.49E-12
12/5/91	1073	690	0.64	1.50E-11	610	709	1.16	2.36E-12
12/17/91	1398	323	0.23	3.18E-12	999	330	0.33	1.39E-12
12/31/91	1225	704	0.57	3.63E-12	782	509	0.65	1.81E-12
1/23/92	1490	265	0.18	3.63E-12	927	519	0.56	1.46E-12
2/4/92	894	404	0.45	1.50E-11	663	234	0.35	2.85E-12
2/21/92	820	309	0.38	1.33E-11	694	257	0.37	3.45E-12
3/13/92	464	339	0.73	1.50E-11	378	201	0.53	
3/28/92	107	329	3.07	1.46E-11	90	219	2.43	
4/10/92	610	363	0.60	4.49E-11	409	287	0.70	3.63E-12
4/24/92	599	367	0.61	4.39E-11	446	240	0.54	3.63E-12
5/8/92	1094	394	0.36	4.49E-11	826	209	0.25	3.63E-12
5/22/92	481	336	0.70	4.49E-11	369	259	0.70	

Table B.1 Site 1 Data: Holes 4a and 4b

Date	Hole 4a 222Rn kBq m-3	Hole 4a 220Rn kBq m-3	Hole 4a 220/222	Hole 4a perm m2	Hole 4b 222Rn kBq m-3	Hole 4b 220Rn kBq m-3	Hole 4b 220/222	Hole 4b perm m2
9/8/89	130							
9/28/89	352				100			
10/12/89	615				129			
10/25/89	209				32			
11/9/89	500				79			
11/22/89	154				25			
12/7/89	418				62			
12/21/89	561				363			
1/4/90	723				98			
1/17/90	632				81			
2/1/90	211				85			
2/14/90	170				80			
2/28/90	386				219			
3/15/90	195				133	201	1.50	
3/28/90	187	103	0.55		219	120	0.55	
4/12/90	218	180	0.83		204	150	0.74	
4/26/90	129	252	1.96		122	219	1.79	
5/10/90	183	270	1.48	7.13E-13	192	250	1.30	5.99E-13
5/22/90	194	212	1.09	7.54E-13	217	183	0.84	6.80E-13
6/6/90	149	222	1.49	8.74E-13	148	247	1.67	8.28E-13
6/20/90	224	245	1.09	8.93E-13	233	236	1.01	8.70E-13
7/3/90	170	287	1.69	1.19E-12	171	266	1.55	5.81E-13
7/18/90	325	334	1.03	1.60E-12	342	323	0.94	7.99E-13
7/30/90	280	293	1.05	1.73E-12	298	370	1.24	9.66E-13
8/15/90	188	265	1.41	1.09E-12	230	283	1.23	8.77E-13
8/31/90	158	301	1.90	1.33E-12	173	406	2.34	9.18E-13
9/14/90	315	315	1.00	1.77E-12	332	304	0.91	5.26E-13
9/27/90	616	572	0.93	1.77E-12	607	326	0.54	6.80E-13
10/10/90	445	284	0.64	1.66E-12	444	293	0.66	7.90E-13
10/25/90	497	222	0.45	8.52E-13	621	230	0.37	8.52E-13
11/7/90	296	247	0.84	1.22E-12	353	263	0.75	7.10E-13
11/21/90	84	305	3.65	1.16E-12	77	218	2.84	8.74E-13
12/10/90	187	231	1.24	9.99E-13	214			9.46E-13
12/27/90	188	624	3.33	9.71E-13	214	447	2.09	9.20E-13
1/9/91	442	222	0.50	1.06E-12	538	367	0.68	9.70E-13
1/24/91	359	485	1.35	1.21E-12	568	291	0.51	9.18E-13
2/6/91	170	230	1.36	1.61E-12	179	210	1.17	9.59E-13
2/22/91	219	443	2.02	1.14E-12	285	389	1.36	7.87E-13
3/12/91	41	186	4.54	1.09E-12		172		8.70E-13
3/26/91	179	185	1.03	1.13E-12	187	186	0.99	9.70E-13
4/15/91	288	249	0.86		324	224	0.69	
4/27/91	195	214	1.10	8.28E-13	199	161	0.81	7.66E-13
5/9/91	198	210	1.06	1.21E-12	169	203	1.20	8.19E-13
5/24/91	188	289	1.54	1.63E-12	195	254	1.30	6.66E-13
6/7/91	205	320	1.56	1.90E-12	203	297	1.46	8.70E-13
6/21/91	258	287	1.11	1.94E-12	230	304	1.32	1.37E-12
7/2/91	288	272	0.95	2.05E-12	271	338	1.25	1.06E-12
7/19/91	344	299	0.87	2.05E-12	331	344	1.04	1.29E-12
8/2/91	379	317	0.84	1.85E-12	375	302	0.80	1.21E-12

Table B.1 Site 1 Data: Holes 4a and 4b, continued

Date	Hole 4a 222Rn kBq m-3	Hole 4a 220Rn kBq m-3	Hole 4a 220/222	Hole 4a perm m2	Hole 4b 222Rn kBq m-3	Hole 4b 220Rn kBq m-3	Hole 4b 220/222	Hole 4b perm m2
8/15/91	400	332	0.83	2.05E-12	383	302	0.79	1.29E-12
8/29/91	401	323	0.81	2.25E-12	377	328	0.87	1.32E-12
9/12/91	381	311	0.82	2.00E-12	370	302	0.81	1.16E-12
9/26/91	546	229	0.42	9.41E-13	545	207	0.38	8.15E-13
10/11/91	664	314	0.47	1.85E-12	614			4.40E-13
10/25/91	569	303	0.53	8.45E-13	588	220	0.37	2.78E-12
11/7/91	578	321	0.56	3.99E-12	568	320	0.56	1.03E-12
11/22/91	254	276	1.09	3.99E-12	233	241	1.04	1.43E-12
12/5/91	240	658	2.74	1.55E-12	378	248	0.66	1.10E-12
12/17/91	381	177	0.46	1.03E-12	441	197	0.45	6.04E-13
12/31/91	149	239	1.60	1.65E-12	139	189	1.36	8.91E-13
1/23/92	499	308	0.62	1.34E-12	555	270	0.49	9.41E-13
2/5/92	111	174	1.57	2.66E-12	132	170	1.29	2.85E-12
2/21/92	365	143	0.39	1.84E-12	345	135	0.39	1.22E-12
3/13/92	298	247	0.83	1.16E-12	403	172	0.43	1.24E-12
3/28/92	80	163	2.04	1.40E-12	81	118	1.45	1.13E-12
4/10/92	94	185	1.97	2.17E-12				1.21E-12
4/24/92	183	208	1.14	3.07E-12	185	180	0.98	1.40E-12
5/8/92	184	217	1.18	2.64E-12	174	185	1.06	1.40E-12
5/22/92	128	307	2.38	3.99E-12	166	245	1.48	

Table B.1 Site 1 Data: Holes 5 and 6

Date	Hole 5 222Rn kBq m-3	Hole 5 220Rn kBq m-3	Hole 5 220/222	Hole 5 perm m2	Hole 6 222Rn kBq m-3	Hole 6 220Rn kBq m-3	Hole 6 220/222	Hole 6 perm m2
9/8/89	462				352			
9/28/89	455				296			
10/12/89	516				484			
10/25/89	345				194			
11/9/89	632				525			
11/22/89	105				133			
12/7/89	368				253			
12/21/89	354				547			
1/4/90	678				699			
1/17/90	668				629			
2/1/90	294				281			
2/14/90	427				307			
2/28/90	322				342			
3/15/90	477				261			
3/28/90	302	205	0.68		308	182	0.59	
4/12/90	292	274	0.94		345			
4/26/90	385	265	0.69		195	241	1.24	
5/10/90	413	351	0.85	1.82E-12	287	244	0.85	7.77E-13
5/22/90	432	382	0.88	1.05E-12	307	225	0.73	7.43E-13
6/6/90	378	272	0.72	9.72E-13	291	216	0.74	8.74E-13
6/20/90	457	324	0.71	1.16E-12	298	267	0.90	1.03E-12
7/3/90	373	335	0.90	1.33E-12	222	322	1.45	4.61E-13
7/18/90	487	370	0.76	1.62E-12	324	367	1.13	8.52E-13
7/30/90	408	432	1.06	1.80E-12	257	494	1.92	9.22E-13
8/15/90	486	333	0.68	1.09E-12	325	346	1.06	9.72E-13
8/31/90	417	357	0.86	1.28E-12	292	345	1.18	9.71E-13
9/14/90	491	353	0.72	2.00E-12	325	338	1.04	5.08E-13
9/27/90	579	358	0.62	1.81E-12	550	353	0.64	8.52E-13
10/10/90	599	374	0.62	1.62E-12	404	280	0.69	1.03E-12
10/25/90	413	276	0.67	9.99E-13	384	275	0.72	9.99E-13
11/7/90	310	296	0.96	8.96E-13	409	280	0.69	7.66E-13
11/21/90	144	211	1.46	1.06E-12	198	152	0.76	9.22E-13
12/10/90	304			9.72E-13	342	207	0.60	9.72E-13
12/27/90	205	327	1.60	1.03E-12	217	196	0.90	9.71E-13
1/9/91	358	269	0.75	9.70E-13	384	233	0.61	6.88E-13
1/24/91	577	381	0.66	1.38E-12	572	487	0.85	1.68E-12
2/6/91	458	280	0.61	1.93E-12	441	251	0.57	1.93E-12
2/22/91	474	322	0.68	1.81E-12	378	394	1.04	1.93E-12
3/12/91	42	206	4.94	1.42E-12	131	243	1.85	1.58E-12
3/26/91	292	229	0.78	1.50E-12	319	262	0.82	1.63E-12
4/15/91	398	339	0.85	1.75E-12	387	335	0.86	1.83E-12
4/27/91	363	227	0.63	1.12E-12	286	264	0.92	1.12E-12
5/9/91	484	188	0.39	1.76E-12	309	260	0.84	1.68E-12
5/24/91	403	287	0.71	2.05E-12	257	342	1.33	1.94E-12
6/7/91	313	307	0.98	2.16E-12	221	373	1.69	1.95E-12
6/21/91	334	295	0.88	2.17E-12	228	349	1.53	2.05E-12
7/2/91	348	345	0.99	2.35E-12	238	322	1.35	2.05E-12
7/19/91	364	353	0.97	2.37E-12	248	368	1.48	2.05E-12
8/2/91	413	348	0.84	2.37E-12	306	398	1.30	1.94E-12

Table B.1 Site 1 Data: Holes 5 and 6, continued

Date	Hole 5 222Rn kBq m-3	Hole 5 220Rn kBq m-3	Hole 5 220/222	Hole 5 perm m2	Hole 6 222Rn kBq m-3	Hole 6 220Rn kBq m-3	Hole 6 220/222	Hole 6 perm m2
8/15/91	424	322	0.76	2.60E-12	311	416	1.34	2.05E-12
8/29/91	421	360	0.85	2.33E-12	312	373	1.20	2.25E-12
9/12/91	374	319	0.86	2.35E-12	304	367	1.21	2.22E-12
9/26/91	470	269	0.57	2.60E-12	437	386	0.88	2.11E-12
10/11/91	505	372	0.74	2.37E-12	474	238	0.50	1.89E-12
10/25/91	558	363	0.65	8.79E-12	512	222	0.43	4.09E-12
11/7/91	456	260	0.57	8.19E-12	537	386	0.72	5.71E-12
11/22/91	451	269	0.60	1.90E-11	315	284	0.90	7.99E-12
12/5/91	372	259	0.69	2.64E-12	318	282	0.89	
12/17/91	483	173	0.36	3.45E-12	470	231	0.49	1.94E-12
12/31/91	232	241	1.04	2.50E-12	319	286	0.90	2.50E-12
1/23/92	523	300	0.57	2.66E-12	670	216	0.32	2.50E-12
2/5/92	226	193	0.85	2.66E-12	268	249	0.93	3.41E-12
2/21/92	496	199	0.40	1.33E-11	407	321	0.79	3.45E-12
3/13/92	306	179	0.58	3.38E-12	254	319	1.25	2.05E-12
3/28/92	45	179	3.98	4.77E-12	153	189	1.23	2.10E-12
4/10/92	149	359	2.41	5.71E-12	236	214	0.91	3.07E-12
4/24/92	445	206	0.46	8.39E-12	278	290	1.04	3.63E-12
5/8/92	382	244	0.64	8.39E-12	304	214	0.70	4.44E-12
5/22/92	372	235	0.63	8.79E-12	190	350	1.84	4.88E-12

Table B.2 site 2 Data: Holes 1 and 2

Date	Hole 1 222Rn kBq m-3	Hole 1 220Rn kBq m-3	Hole 1 220/222	Hole 1 perm m2	Hole 2 222Rn kBq m-3	Hole 2 220Rn kBq m-3	Hole 2 220/222	Hole 2 perm m2
5/10/90	16.06	7.70	0.48	3.00E-12	15.24	7.29	0.48	2.64E-12
5/24/90	13.58	13.51	0.99	2.93E-12	14.69	10.66	0.73	2.75E-12
6/8/90	10.80	9.40	0.87	3.46E-12	11.62	15.17	1.31	3.00E-12
6/21/90	14.73	11.29	0.77	3.23E-12	13.21	12.73	0.96	2.80E-12
7/3/90	14.50	5.11	0.35	2.93E-12	10.88			2.75E-12
7/17/90	13.51	5.44	0.40	3.07E-12	13.36	8.51	0.64	2.66E-12
8/16/90	16.65	10.77	0.65	3.07E-12	15.06	16.76	1.11	2.80E-12
8/31/90	12.65	9.10	0.72	3.38E-12	13.95	13.88	0.99	3.00E-12
9/14/90	12.99	7.44	0.57	3.23E-12	11.47	14.06	1.23	2.73E-12
9/28/90	13.10	8.99	0.69	3.14E-12	13.06	10.06	0.77	2.75E-12
10/12/90	12.25	8.73	0.71	3.07E-12	12.28	14.36	1.17	2.80E-12
10/26/90	14.02	7.73	0.55	3.46E-12	17.61	12.84	0.73	2.56E-12
11/9/90	13.73	4.85	0.35	3.00E-12	13.32	4.11	0.31	2.66E-12
11/23/90				3.07E-12	14.69	12.99	0.88	2.86E-12
12/12/90	13.43	4.70	0.35	3.07E-12	15.06	10.51	0.70	2.64E-12
12/27/90	12.58			3.00E-12	12.06	9.25	0.77	2.66E-12
1/9/91	21.13	11.32	0.54	2.85E-12	9.44	8.66	0.92	2.73E-12
1/24/91	15.06	8.14	0.54	3.07E-12	14.54	6.59	0.45	2.85E-12
2/6/91	11.73	13.84	1.18	2.75E-12	12.14	16.10	1.33	2.90E-12
2/22/91	13.14	16.58	1.26	3.10E-12	9.99	13.17	1.32	3.04E-12
3/12/91	11.77	4.51	0.38	2.96E-12	9.81	7.25	0.74	2.64E-12
3/26/91	16.21	8.14	0.50	3.15E-12	9.36	7.77	0.83	2.66E-12
4/15/91	13.69	8.58	0.63	2.92E-12	9.47	16.65	1.76	2.78E-12
4/27/91	14.17	4.81	0.34	2.73E-12	14.54	8.44	0.58	2.48E-12
5/9/91	17.61	4.66	0.26	2.92E-12	12.17	8.29	0.68	2.66E-12
5/24/91	11.06	8.10	0.73	2.60E-12	9.40	12.28	1.31	2.18E-12
6/7/91	7.10	12.84	1.81	2.80E-12	12.32	7.62	0.62	2.73E-12
6/21/91	13.39	7.22	0.54	2.78E-12	11.40	8.66	0.76	2.60E-12
7/2/91	11.36	9.73	0.86	2.73E-12	9.92	16.91	1.71	2.50E-12
7/19/91	11.54	8.03	0.70	2.85E-12	11.54	9.29	0.80	2.60E-12
8/2/91	14.39	14.50	1.01	2.85E-12	14.65	7.25	0.49	2.51E-12
8/15/91	17.13	6.92	0.40	3.07E-12	14.06	17.61	1.25	2.78E-12
8/29/91	16.13	11.51	0.71	2.84E-12	14.21	17.39	1.22	2.84E-12
9/12/91	14.95	11.43	0.76	2.85E-12	12.06	7.81	0.65	2.50E-12
9/26/91	18.17	8.84	0.49	2.85E-12	15.91			2.85E-12
10/11/91	13.14	12.32	0.94	2.66E-12	14.58	17.32	1.19	2.60E-12
10/26/91	15.80	4.11	0.26	4.49E-11	14.21	16.17	1.14	1.46E-11
11/7/91	12.14	13.32	1.10	4.49E-11	14.10	13.06	0.93	1.46E-11
11/22/91	13.32	3.96	0.30	4.49E-11	16.21	9.18	0.57	4.39E-11
12/5/91	12.14	8.07	0.66	1.50E-11	15.39	9.14	0.59	1.50E-11
12/17/91	15.39	10.99	0.71	1.94E-12	14.17	12.51	0.88	1.94E-12
12/31/91				3.07E-12	14.43	10.29	0.71	3.07E-12
1/23/92	15.02	14.65	0.98	3.90E-12	10.47	10.25	0.98	3.23E-12
2/5/92	12.84	7.51	0.59	1.46E-11	12.69	8.25	0.65	1.46E-11
3/13/92	13.02			4.49E-11	13.51			4.49E-11
3/28/92	7.70	8.07	1.05	4.49E-11	11.43	6.70	0.59	4.49E-11
4/10/92	10.84	9.95	0.92	4.49E-11	10.66	11.21	1.05	4.49E-11
4/24/92	11.40	6.36	0.56	4.49E-11	10.80	12.91	1.20	4.49E-11

Table B.2 Site 2 Data: Holes 3 and 4a

Date	Hole 3 222Rn kBq m-3	Hole 3 220Rn kBq m-3	Hole 3 220/222	Hole 3 perm m2	Hole 4a 222Rn kBq m-3	Hole 4a 220Rn kBq m-3	Hole 4a 220/222	Hole 4a perm m2
5/10/90	12.28	0.00		3.46E-12				
5/24/90	14.80	23.01	1.56	3.21E-12				
6/8/90	10.47	11.91	1.14	3.53E-12				
6/21/90	10.92	10.14	0.93	3.38E-12				
7/3/90	10.62	5.70	0.54	3.21E-12	13.39	7.36	0.55	3.00E-12
7/17/90	10.84	10.21	0.94	3.30E-12	13.76	9.36	0.68	3.30E-12
8/16/90	15.80	9.58	0.61	3.38E-12	10.21			3.38E-12
8/31/90	11.88	16.58	1.40	3.66E-12				4.08E-12
9/14/90	10.66	11.17	1.05	3.51E-12		10.29		3.00E-12
9/28/90				3.14E-12	13.88			3.07E-12
10/12/90	11.03	10.84	0.98	3.14E-12				3.14E-12
10/26/90	14.39	9.88	0.69	3.38E-12	12.88	8.10	0.63	3.23E-12
11/9/90	11.84	7.84	0.66	3.28E-12	12.73	4.88	0.38	3.23E-12
11/23/90	14.47	8.92	0.62	3.23E-12	8.77	7.10	0.81	3.66E-12
12/12/90	10.84	5.55	0.51	3.00E-12	13.25	4.92	0.37	3.00E-12
12/27/90	11.58	11.03	0.95	3.23E-12	12.21	11.47	0.94	2.66E-12
1/9/91	10.32	7.51	0.73	3.15E-12	10.55	7.62	0.72	2.85E-12
1/24/91	11.06	10.80	0.98	3.41E-12	11.47	4.74	0.41	3.07E-12
2/6/91	10.95	6.96	0.64	3.09E-12	12.36			3.09E-12
2/22/91				2.90E-12	9.55	15.21	1.59	
3/12/91	7.70	8.58	1.12	3.07E-12	11.10	8.25	0.74	3.00E-12
3/26/91	10.99	15.39	1.40	2.92E-12	13.51	5.11	0.38	3.00E-12
4/15/91	9.29	11.06	1.19	2.78E-12	8.88	8.88	1.00	
4/27/91	11.58	6.77	0.58	2.73E-12	13.43	5.37	0.40	2.73E-12
5/9/91				2.66E-12				2.92E-12
5/24/91				2.88E-12	6.77	9.44	1.39	2.78E-12
6/7/91	11.99	10.92	0.91	2.73E-12	10.84	5.11	0.47	2.56E-12
6/21/91	11.25	11.32	1.01	3.07E-12	12.10	4.29	0.35	2.51E-12
7/2/91	9.18	10.84	1.18	2.92E-12	8.77	4.74	0.54	2.66E-12
7/19/91	10.55	15.61	1.48	2.85E-12	9.81	7.25	0.74	2.53E-12
8/2/91	13.95	15.06	1.08	2.60E-12	12.06	6.33	0.52	2.00E-12
8/15/91	14.10	11.62	0.82	3.00E-12	11.73	8.62	0.74	2.00E-12
8/29/91	11.73	8.03	0.68	2.84E-12	10.88	7.29	0.67	2.06E-12
9/12/91	10.18	12.54	1.23	2.85E-12	9.10	8.95	0.98	2.35E-12
9/26/91	13.73	5.55	0.40	3.07E-12	11.51	7.99	0.69	2.23E-12
10/11/91	11.14	13.99	1.26	2.85E-12	8.77	8.92	1.02	2.07E-12
10/26/91	12.14	9.88	0.81	4.49E-11	12.73	10.36	0.81	5.99E-12
11/7/91	11.88	10.84	0.91	4.39E-11	7.59	12.77	1.68	8.79E-12
11/22/91	13.10	6.88	0.53	4.49E-11	13.80	6.03	0.44	8.59E-12
12/5/91	13.88			1.50E-11	6.96	9.29	1.34	1.50E-11
12/17/91	14.13	5.40	0.38	2.17E-12	8.99	11.69	1.30	1.94E-12
12/31/91	11.47	10.92	0.95	3.07E-12	11.32	10.58	0.93	3.63E-12
1/23/92	9.18	13.88	1.51	3.49E-12	10.25	7.22	0.70	2.73E-12
2/5/92	12.58	11.32	0.90	1.46E-11	11.17	7.92	0.71	1.46E-11
3/13/92	14.84			4.49E-11	11.77			1.46E-11
3/28/92	10.58	7.07	0.67	4.49E-11	8.92	4.22	0.47	6.42E-12
4/10/92	9.81	11.06	1.13	4.49E-11	7.55	7.36	0.98	4.49E-11
4/24/92	11.32	9.55	0.84	4.49E-11	10.29	4.22	0.41	1.46E-11

Table B.2 Site 2 Data: Hole 4b

Date	Hole 4b 222Rn kBq m-3	Hole 4b 220Rn kBq m-3	Hole 4b 220/222	Hole 4b perm m2
5/10/90	11.66	0.00		2.53E-12
5/24/90	13.58	16.47	1.21	2.75E-12
6/8/90	12.54	8.44	0.67	2.93E-12
6/21/90	14.99	10.77	0.72	2.80E-12
7/3/90	14.21	5.14	0.36	2.50E-12
7/17/90	15.21	4.37	0.29	2.73E-12
8/16/90	15.76	11.99	0.76	2.86E-12
8/31/90	14.62	9.03	0.62	3.14E-12
9/14/90	9.95	13.02	1.31	2.73E-12
9/28/90	14.50	7.84	0.54	2.86E-12
10/12/90	11.91	7.66	0.64	2.93E-12
10/26/90	14.47	5.11	0.35	2.73E-12
11/9/90	12.51	6.85	0.55	2.82E-12
11/23/90	12.47	5.48	0.44	3.00E-12
12/12/90	14.91	4.11	0.28	2.80E-12
12/27/90	13.39	7.36	0.55	2.66E-12
1/9/91	11.06	7.70	0.70	2.66E-12
1/24/91	12.14	11.25	0.93	3.07E-12
2/6/91	6.99	5.85	0.84	2.90E-12
2/22/91	13.51	12.54	0.93	2.07E-12
3/12/91	10.06	6.55	0.65	1.85E-12
3/26/91				
4/15/91	9.81	9.10	0.93	2.71E-12
4/27/91	10.88	11.84	1.09	2.66E-12
5/9/91	13.62	6.40	0.47	2.92E-12
5/24/91	9.07	14.73	1.62	2.60E-12
6/7/91	9.69	9.95	1.03	1.69E-12
6/21/91	12.17	3.85	0.32	6.43E-13
7/2/91	10.32	7.36	0.71	7.20E-13
7/19/91	10.58	5.59	0.53	2.85E-12
8/2/91	12.84	11.91	0.93	1.61E-12
8/15/91	12.21	4.00	0.33	3.00E-12
8/29/91	11.88	7.40	0.62	2.84E-12
9/12/91	10.21	8.70	0.85	2.85E-12
9/26/91	13.99	7.03	0.50	2.85E-12
10/11/91	6.29	7.62	1.21	2.60E-12
10/26/91	13.65	8.84	0.65	1.50E-11
11/7/91	10.62	12.84	1.21	4.19E-11
11/22/91	12.40	8.92	0.72	8.59E-12
12/5/91	11.62	10.62	0.91	1.50E-11
12/17/91	11.17	6.18	0.55	1.71E-12
12/31/91	6.88	10.58	1.54	3.63E-12
1/23/92	11.80	5.70	0.48	3.81E-12
2/5/92	8.66	7.55	0.87	1.46E-11
3/13/92	13.76			4.49E-11
3/28/92		0.00		1.50E-11
4/10/92	9.88	7.25	0.73	4.49E-11
4/24/92				4.49E-11

Table B.3 Site 3 Data

Date	Hole 1a 222Rn kBq m-3	Hole 1a 220Rn kBq m-3	Hole 1a 220/222	Hole 1a perm m2	Hole 1b 222Rn kBq m-3	Hole 1b 220Rn kBq m-3	Hole 1b 220/222	Hole 1b perm m2
10/17/91	35.74	113.63	3.18	1.12E-12	37.30	107.00	2.87	9.2E-13
10/25/91	36.08	121.47	3.37	1.09E-12	43.88	126.10	2.87	1.21E-12
11/7/91	40.07	123.73	3.09	1.13E-12	41.03	112.67	2.75	1.24E-12
11/22/91	39.22	123.54	3.15	9.66E-13	40.89	125.91	3.08	1.03E-12
8/28/92	24.12	98.68	4.09	2.59E-13	37.56	118.70	3.16	1.01E-12
10/23/92	30.86	106.71	3.46	2.59E-13	32.12	103.12	3.21	1.09E-12
11/6/92	24.86	85.62	3.44	3.29E-13				9.55E-13
11/20/92	24.16	80.29	3.32	2.9E-13	39.33	129.65	3.30	1.23E-12
12/4/92	47.77	51.58	1.08	2.12E-13	46.92	79.03	1.68	4.2E-13
6/4/93								
6/18/93	26.64	94.94	3.56	5.1E-13				9.33E-13
7/2/93	27.16	85.21	3.14	5.58E-13	19.76	62.90	3.18	1.75E-13
7/16/93	29.19	89.76	3.07	6.07E-13	27.53	81.10	2.95	2.38E-13
7/30/93	34.89	103.79	2.97	7.16E-13	31.01	75.52	2.44	2.45E-13

Date	Hole 2 222Rn kBq m-3	Hole 2 220Rn kBq m-3	Hole 2 220/222	Hole 2 perm m2	Hole 3 222Rn kBq m-3	Hole 3 220Rn kBq m-3	Hole 3 220/222	Hole 3 perm m2
10/17/91	44.36	102.34	2.31	1.72E-12	74.81	47.66	0.64	6.24E-13
10/25/91	55.72	137.20	2.46	3.07E-12	71.00	63.31	0.89	7.61E-13
11/7/91	47.29	125.95	2.66	4.66E-12	77.11	68.52	0.89	9.18E-13
11/22/91	49.73	129.76	2.61	4.99E-12	90.28	45.33	0.50	9.66E-13
8/28/92	28.05	66.67	2.38	1.56E-13	89.61	35.67	0.40	3.47E-13
10/23/92	36.70	78.70	2.14	2.12E-13				3.95E-13
11/6/92	32.82	86.91	2.65	2.12E-13	60.09	31.78	0.53	3.57E-13
11/20/92	44.10	92.09	2.09	2.43E-13	55.43	38.00	0.69	4.08E-13
12/4/92	51.50	98.68	1.92	1.85E-13	82.51	47.88	0.58	3.47E-13
6/4/93								
6/18/93	51.84	83.55	1.61	1.33E-13	49.21			4.07E-14
7/2/93	46.81	59.27	1.27	1.75E-13	37.63	18.98	0.50	9.14E-14
7/16/93	23.83	29.19	1.23	7.73E-14				7.35E-14
7/30/93	37.11	48.62	1.31	7.35E-14				7.73E-14

Date	Hole 4 222Rn kBq m-3	Hole 4 220Rn kBq m-3	Hole 4 220/222	Hole 4 perm m2	Hole 5 222Rn kBq m-3	Hole 5 220Rn kBq m-3	Hole 5 220/222	Hole 5 perm m2
10/17/91	92.17	77.29	0.84	3.07E-12	52.24	105.23	2.01	4.58E-13
10/25/91	114.59	108.04	0.94	2.31E-12	45.47	136.42	3.00	3.42E-13
11/7/91	102.71	135.57	1.32	2.85E-12	78.88	163.39	2.07	3.84E-13
11/22/91	103.56	131.46	1.27	2.64E-12	84.25	153.59	1.82	3.84E-13
8/28/92	76.52	102.56	1.34	1.09E-12	56.35	169.61	3.01	5.70E-13
10/23/92	75.92	118.10	1.56	3.74E-12	47.73	163.76	3.43	7.21E-13
11/6/92	76.33	113.59	1.49	5.4E-12	47.21	156.47	3.31	8.61E-13
11/20/92	76.22	117.29	1.54	4.42E-12	57.42	179.67	3.13	8.61E-13
12/4/92	77.92	94.76	1.22	2.21E-12	72.78	113.11	1.55	5.21E-13
6/4/93					64.64	84.77	1.31	
6/18/93	82.99	118.77	1.43	1.1E-12	71.45	109.93	1.54	5.58E-12
7/2/93	92.65	69.01	0.74	1.86E-12	45.77	153.29	3.35	2.17E-11
7/16/93	79.25	100.05	1.26	1.41E-12	53.39	158.92	2.98	4.81E-12
7/30/93	104.16	111.63	1.07	1.77E-12	44.44	125.54	2.83	1.05E-11

Appendix C

QUALITY ASSURANCE DATA

Table C.1 - ^{222}Rn duplicate measurement data. 222Rn1 refers to first measurement obtained, 222Rn2 the second, typically sampled approximately 5 minutes later than 222Rn1; Mean is the arithmetic mean of 222Rn1 and 222Rn2; Diff is 222Rn1 minus Mean; % is the Diff/Mean.

Date	Site #	Hole #	222Rn 1 kBq m-3	222Rn2 kBq m-3	Mean kBq m-3	Diff kBq m-3	%
9/8/89	1	6	365.67	339.07	352.37	26.60	7.55
9/8/89	1	5	458.80	463.98	461.39	-5.18	-1.12
9/8/89	1	4	124.80	138.82	131.81	-14.02	-10.64
9/8/89	1	3	715.21	806.23	760.72	-91.02	-11.96
9/28/89	1	1	419.58	373.96	396.77	45.62	11.50
9/28/89	1	2	95.35	76.29	85.82	19.06	22.20
9/28/89	1	3a	1658.71	1693.86	1676.29	-35.15	-2.10
9/28/89	1	4a	340.88	372.22	356.55	-31.34	-8.79
9/28/89	1	5	407.00	505.05	456.03	-98.05	-21.50
9/28/89	1	6	284.64	308.77	296.70	-24.12	-8.13
2/1/90	1	4b	84.69	97.01	90.85	-12.32	-13.56
3/15/90	1	3a	659.45	644.17	651.81	15.28	2.34
7/3/90	1	3a	853.04	922.34	887.69	-69.30	-7.81
7/3/90	1	3b	637.99	640.40	639.19	-2.40	-0.38
7/3/90	1	4a	169.79	164.72	167.26	5.07	3.03
7/3/90	1	4b	171.79	191.62	181.71	-19.83	-10.91
9/14/90	2	1	12.99	13.02	13.01	-0.04	-0.28
11/21/90	1	1	483.26	514.41	498.83	-31.15	-6.25
11/21/90	1	2	659.23	599.29	629.26	59.94	9.53
11/21/90	1	3a	980.43	884.86	932.64	95.57	10.25
1/9/91	1	5	431.16	357.57	394.36	73.59	18.66
1/24/91	1	2	630.70	637.18	633.94	-6.48	-1.02
1/24/91	2	3	11.06	9.58	10.32	1.48	14.34
2/6/91	1	2	76.70	75.55	76.13	1.15	1.51
2/6/91	1	3a	597.11	652.94	625.02	-55.83	-8.93
2/6/91	1	4b	309.17	268.55	288.86	40.63	14.06
3/12/91	1	3a	457.43	462.69	460.06	-5.25	-1.14
8/2/91	1	3a	886.52	854.11	870.31	32.41	3.72
8/2/91	2	4a	12.47	12.65	12.56	-0.19	-1.47
8/15/91	1	1	301.99	277.57	289.78	24.42	8.43
8/15/91	1	2	315.80	314.87	315.33	0.93	0.29
8/29/91	1	2	263.40	249.23	256.32	14.17	5.53
8/29/91	1	3b	664.89	576.13	620.51	88.76	14.30
8/29/91	2	2	14.21	14.50	14.36	-0.30	-2.06
9/12/91	1	3a	1030.38	920.08	975.23	110.30	11.31
9/12/91	1	3b	837.79	762.09	799.94	75.70	9.46
9/12/91	2	4a	9.10	8.99	9.05	0.11	1.23

Table C.1 (continued)

Date	Site #	Hole #	222Rn1 kBq m-3	222Rn2 kBq m-3	Mean kBq m-3	Diff kBq m-3	%
9/26/91	1	3b	892.74	864.43	878.58	28.31	3.22
10/11/91	1	3a	1154.73	1171.16	1162.95	-16.43	-1.41
10/11/91	1	1	312.95	334.89	323.92	-21.94	-6.77
10/11/91	2	2	14.58	12.91	13.75	1.67	12.11
10/25/91	1	2	621.42	599.10	610.26	22.31	3.66
10/25/91	1	3a	1291.63	1219.26	1255.45	72.37	5.76
10/25/92	3	4	114.59	108.04	111.31	6.55	5.88
11/7/91	1	1	451.29	436.60	443.94	14.69	3.31
11/7/91	1	3a	1305.36	1308.25	1306.80	-2.89	-0.22
11/7/91	3	4	102.71	88.87	95.79	13.84	14.45
11/7/91	2	4b	10.62	10.47	10.55	0.15	1.40
11/22/91	1	2	265.62	273.58	269.60	-7.95	-2.95
11/22/91	3	3	90.28	74.00	82.14	16.28	19.82
12/5/91	1	3a	1072.63	1189.77	1131.20	-117.14	-10.36
2/4/92	1	4a	98.38	111.19	104.78	-12.80	-12.22
4/10/92	1	3a	610.46	663.60	637.03	-53.13	-8.34
4/10/92	1	3b	409.07	491.92	450.49	-82.84	-18.39
4/10/92	2	2	9.88	8.40	9.14	1.48	16.19
4/24/92	1	6	278.02	296.74	287.38	-18.72	-6.51
4/24/92	2	1	12.14	11.40	11.77	0.74	6.29
5/8/92	1	2	335.48	382.99	359.23	-47.51	-13.22
9/14/92	1	5	187.44	176.27	181.86	11.17	6.14
10/23/92	1	5	775.78	833.13	804.45	-57.35	-7.13
11/6/92	1	2	1157.18	934.62	1045.90	222.56	21.28
11/20/92	3	4	87.25	76.22	81.73	11.03	13.49
12/4/92	1	2	858.18	967.03	912.61	-108.85	-11.93
12/4/92	1	5	726.50	727.38	726.94	-0.89	-0.12
12/4/92	3	4	56.20	77.92	67.06	-21.72	-32.39
12/4/92	3	5	70.23	72.78	71.50	-2.55	-3.57
12/17/92	1	5	443.41	421.50	432.46	21.90	5.07
12/17/92	1	2	459.91	508.16	484.03	-48.25	-9.97
2/11/93	3	2	24.86	29.45	27.16	-4.59	-16.89
2/15/93	1	5	384.47	325.27	354.87	59.20	16.68
3/11/93	1	2	274.95	268.29	271.62	6.66	2.45
3/11/93	1	2	352.17	418.80	385.48	-66.64	-17.29
4/10/93	1	2	212.97	234.28	223.63	-21.31	-9.53
4/22/93	1	2	665.26	706.66	685.96	-41.40	-6.04
5/18/93	1	5	620.01	578.64	599.33	41.37	6.90
6/4/93	1	5	246.16	221.04	233.60	25.12	10.75
6/18/93	1	2	134.13	154.18	144.15	-20.05	-13.91
6/18/93	1	2	498.02	445.78	471.90	52.24	11.07
7/2/93	1	2	724.76	681.87	703.31	42.88	6.10
7/2/93	3	3	45.25	46.81	46.03	-1.55	-3.38
7/16/93	1	5	398.97	449.11	424.04	-50.14	-11.82
7/16/93	3	4	79.25	85.88	82.57	-6.62	-8.02
7/30/93	1	2	598.11	631.74	614.92	-33.63	-5.47

Table C.1 (continued)

Date	Site #	Hole #	222Rn1 kBq m-3	222Rn2 kBq m-3	Mean kBq m-3	Diff kBq m-3	%
7/30/93	3	5	44.44	41.85	43.14	2.59	6.00
8/13/93	1	5	506.64	452.95	479.80	53.69	11.19
8/13/93	3	5	48.54	41.18	44.86	7.36	16.41
8/27/93	1	5	530.47	467.38	498.93	63.09	12.64
8/27/93	1	2	731.82	731.34	731.58	0.48	0.07
8/27/93	3	4	73.67	76.89	75.28	-3.22	-4.28
9/10/93	1	2	753.76	918.90	836.33	-165.13	-19.74
9/10/93	1	5	586.97	512.89	549.93	74.07	13.47
9/10/93	3	4	83.84	76.37	80.11	7.47	9.33
9/10/93	3	5	44.96	50.14	47.55	-5.18	-10.89
9/23/93	1	2	978.28	821.62	899.95	156.66	17.41
9/23/93	1	5	562.03	497.54	529.78	64.49	12.17
9/23/93	3	5	61.05	53.95	57.50	7.10	12.36
10/7/93	1	2	982.31	1169.35	1075.83	-187.04	-17.39
10/7/93	1	5	858.66	861.06	859.86	-2.40	-0.28
10/22/93	1	2	1095.35	1116.07	1105.71	-20.72	-1.87
11/19/93	1	2	1101.64	1084.47	1093.05	17.17	1.57
		mean	457.36	456.26	456.81		0.64
		σ	374.79	374.94	373.83		10.82
		MIN	9.10	8.40	9.05		-32.39
		MAX	1305.36	1308.25	1306.80		21.28

Table C.2 - ^{220}Rn duplicate measurement data. 220Rn1 refers to first measurement obtained, 220Rn2 the second, typically sampled approximately 5 minutes later than 222Rn1; Mean is the arithmetic mean of 220Rn1 and 220Rn2; Diff is 220Rn1 minus Mean; % is the Diff/Mean.

Date	Site #	Hole #	220Rn1 kBq m-3	220Rn2 kBq m-3	Mean kBq m-3	Diff kBq m-3	%
7/3/90	1	3a	495.80	587.01	541.40	-91.21	-16.85
7/3/90	1	3b	343.47	320.61	332.04	22.87	6.89
7/3/90	1	4a	286.97	315.43	301.20	-28.45	-9.45
7/3/90	1	4b	266.40	294.37	280.39	-27.97	-9.98
9/14/90	2	1	7.44	7.33	7.38	0.11	1.50
11/21/90	1	1	223.63	188.55	206.09	35.08	17.02
11/21/90	1	3a	636.03	617.42	626.72	18.61	2.97
1/24/91	2	3	10.80	8.70	9.75	2.11	21.63
2/6/91	1	2	121.51	135.31	128.41	-13.80	-10.75
2/6/91	1	3a	334.70	305.66	320.18	29.05	9.07
2/6/91	1	4b	363.16	412.66	387.91	-49.51	-12.76
8/15/91	1	1	248.05	260.74	254.39	-12.69	-4.99
8/15/91	1	2	290.41	298.81	294.61	-8.40	-2.85
8/29/91	1	2	291.63	264.51	278.07	27.12	9.75
8/29/91	1	3b	261.55	355.46	308.51	-93.91	-30.44
9/12/91	1	3a	761.65	563.92	662.78	197.73	29.83
9/12/91	1	3b	444.48	455.14	449.81	-10.66	-2.37
9/12/91	2	4a	8.95	7.70	8.33	1.26	15.11
9/26/91	1	3a	425.83	439.01	432.42	-13.17	-3.05
9/26/91	1	3b	253.45	250.71	252.08	2.74	1.09
10/11/91	1	3a	409.48	386.69	398.08	22.79	5.73
10/11/91	1	1	212.94	247.09	230.01	-34.15	-14.85
10/11/91	2	2	17.32	11.17	14.25	6.14	43.12
10/25/91	1	3a	591.08	462.69	526.88	128.39	24.37
10/25/92	3	4	108.04	117.48	112.76	-9.44	-8.37
11/7/91	3	4	135.49	128.43	131.96	7.07	5.36
11/7/91	2	4b	12.84	10.25	11.54	2.59	22.44
11/22/91	1	2	224.55	196.91	210.73	27.64	13.12
11/22/91	3	3	45.33	40.37	42.85	4.96	11.57
12/5/91	1	3a	689.75	459.28	574.52	230.47	40.12
2/4/92	1	4a	174.09	175.97	175.03	-1.89	-1.08
4/10/92	1	3a	363.38	396.31	379.84	-32.93	-8.67
4/10/92	1	3b	286.64	206.65	246.64	79.99	32.43
4/24/92	1	6	289.60	229.84	259.72	59.76	23.01
4/24/92	2	1	5.00	6.36	5.68	-1.37	-24.10
5/8/92	1	2	210.42	151.81	181.12	58.61	32.36
9/14/92	1	5	322.90	218.63	270.77	104.27	38.51
10/23/92	1	5	271.54	258.67	265.11	12.88	4.86
11/6/92	1	2	560.51	543.83	552.17	16.69	3.02
11/20/92	3	4	102.42	117.29	109.85	-14.87	-13.54
12/4/92	1	2	596.66	480.52	538.59	116.14	21.56
12/4/92	1	5	186.04	224.18	205.11	-38.15	-18.60

Table C.2 (continued)

Date	Site #	Hole #	220Rn1 kBq m-3	220Rn2 kBq m-3	Mean kBq m-3	Diff kBq m-3	%
12/4/92	3	4	79.40	94.76	87.08	-15.36	-17.63
12/4/92	3	5	78.03	113.11	95.57	-35.08	-36.70
12/17/92	1	5	233.84	271.43	252.64	-37.59	-14.88
2/11/93	3	2	100.12	90.84	95.48	9.29	9.73
4/10/93	1	2	113.07	136.64	124.86	-23.57	-18.88
4/22/93	1	2	131.35	146.08	138.71	-14.73	-10.62
4/22/93	1	5	248.90	271.95	260.42	-23.05	-8.85
6/4/93	1	2	176.12	184.15	180.13	-8.03	-4.46
6/18/93	1	2	139.16	162.69	150.92	-23.53	-15.59
6/18/93	1	2	629.70	426.50	528.10	203.20	38.48
7/2/93	3	3	779.44	599.44	689.44	180.01	26.11
7/2/93	1	5	62.79	59.27	61.03	3.52	5.76
7/16/93	3	4	351.87	380.03	365.95	-28.16	-7.69
7/16/93	1	2	100.05	120.40	110.22	-20.35	-18.46
7/30/93	3	5	801.31	663.00	732.16	138.31	18.89
7/30/93	1	5	125.54	173.31	149.42	-47.77	-31.97
8/13/93	3	5	491.21	568.32	529.77	-77.11	-14.56
8/13/93	1	5	146.82	183.93	165.37	-37.11	-22.44
8/27/93	1	2	365.67	331.89	348.78	33.78	9.69
8/27/93	3	4	698.15	643.91	671.03	54.24	8.08
8/27/93	1	2	64.42	95.50	79.96	-31.08	-38.87
9/10/93	1	5	580.23	711.99	646.11	-131.76	-20.39
9/10/93	3	4	379.51	522.29	450.90	-142.78	-31.67
9/10/93	3	5	138.38	93.28	115.83	45.10	38.94
9/10/93	1	2	184.67	133.83	159.25	50.84	31.92
9/23/93	1	5	667.18	623.01	645.10	44.18	6.85
9/23/93	1	2	154.18	153.22	153.70	0.96	0.63
10/7/93	1	5	568.25	371.55	469.90	196.69	41.86
10/7/93	1	2	115.26	92.57	103.91	22.68	21.83
10/22/93	1	2	430.87	498.28	464.57	-67.41	-14.51
11/19/93	1	2	543.49	396.31	469.90	147.19	31.32
		mean	7986.26	7579.74	7579.74		2.82
		σ	5780.48	5091.92	5091.92		20.70
		MIN	135.00	172.00	172.00		-38.87
		MAX	21657.00	19243.00	19243.00		43.12

Table C.3 Calibration data for scintillation cells used in this study. All values are in Bq m^{-3} (initial cpm)$^{-1}$. New denotes the date the cells were obtained. The dates across the top of the charts indicate the date the calibrations were performed. Cells 1 - 10 were removed from use during August 1991 due to increasing background and not being able to obtain a consistent calibration.

cell #	new	8/1/89	8/2/89	8/15/89	8/16/89	4/1/90	4/2/90	6/29/90	6/14/91	6/21/91
1	8/1/89	159	174		162	161	167	164	160	179
2	8/1/89	159	158	160	162	155		176	162	
3	8/1/89	162	160	160	160	152	151	156		161
4	8/1/89	152	169	172	154		162	173	168	
5	8/1/89	171	167					162	158	
6	8/1/89	157	152					155	179	156
7	8/1/89	158	150					161		158
8	8/1/89	162	145						170	
9	8/1/89	164	156					165		173
10	8/1/89	160	166					179		174

cell #	new	6/29/90	6/14/91	6/21/91	8/14/91	1/16/92	2/3/92	4/15/92
11	6/7/90	153	148	155		153	161	157
12	6/7/90	184		175		155	153	154
13	6/7/90	178	182		171	159	167	163
14	6/7/90	184	175		166	169		159
15	6/7/90	147	150	152	176	176		161
16	8/14/91				180	166	158	162
17	8/14/91				169	160	173	161
18	8/14/91				148	156	168	162
19	8/14/91				172	157	168	162
20	8/14/91				174	154		161
21	8/14/91				150		149	151
22	8/14/91				158		155	156
23	1/16/92					166		170
24	1/16/92					164		161
25	1/16/92					138		150
26	1/16/92					140		150
27	1/16/92					132		146
28	1/16/92					165		157
29	1/16/92					162		155
30	1/16/92					178		169
31	1/16/92					162		160
32	1/16/92					162		155

Appendix D

SAMPLE CALCULATION FOR 222RN AND 220RN

The sample calculation shown below is for the sample collected at Site 1, Hole 5 on September 12, 1991.

1. Five-minute background of cell # 10 = 10 counts
2. Sample collection started: 10:51
3. Flow rate: 2.0 l min^{-1}
4. ΔP: 17 in. H_2O
5. Sample collection ended: 10:52
6. One-minute-count at 9 seconds after sampling ended = $C1_T$ = 4334 counts
7. Five-minute-count at 15:45 = C5 = 30968 counts
8. ^{222}Rn integration factor for five-minute-count period, obtained by integrating equation 3.1:

   $\beta_{222Rn5} = (^{222}Rn + ^{218}Po + ^{214}Po)/^{222}Rn_o$

   using computer program in Appendix A = 14.49 counts min^{-1}
9. Initial cpm (at t = 0) due to ^{222}Rn = cpm_{o222}

   = (C5-bkgr)/β_{222Rn5} = (30968-10)/14.49 = 2137
10. Calibration factor, determined using EML Radon, Thoron and Progeny Exposure Facility = 175 Bq m^{-3} $(cpm_{o222})^{-1}$
11. ^{222}Rn concentration = cpm_{o222} * calibration factor

    = 2137 * 175 = 374 kBq m-3
12. ^{222}Rn integration factor for one-minute-count beginning at 9 seconds (β_{222Rn1}), and using 20 seconds to account for soil gas to reach scintillation cell and decays during this time (see text for explanation), obtained by integrating equation 3.1 = 1.20

13. Total counts in one-minute-count ($C1_T$) comprised of counts due to ^{222}Rn and progeny ($C1_{222}$) and ^{220}Rn and progeny ($C1_{220}$).

    $C1_{222} = cpm_{o222} * \beta_{222Rn1} = 2137 * 1.20 = 2564$

14. $C1_{220} = C1_T$ - background - $C1_{222} = 4334 - 2 - 2564 = 1768$

15. ^{220}Rn integration factor for one-minute count beginning at 9 seconds and using 20 seconds to account for soil gas to reach scintillation cell and decays during this time (see text for explanation), obtained by integrating equation 3.3:

    $\beta_{220Rn} = (^{220}Rn + ^{216}Po$ activity$) / ^{220}Rn_o = 0.969$

16. Initial cpm $^{220}Rn = cpm_{o220} = C1_{220}/\beta_{220Rn} = 1768/0.969 = 1825$

17. ^{220}Rn concentration $= cpm_{o220} *$ calibration factor

    $= 1825 * 175 = 319$ kBq m^{-3}

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