To the Graduate Council:

I am submitting herewith a dissertation written by Mahmoud M. Abdelrazek entitled "Investigation of Parameters Affecting the Concentration of Radon and Its Daughters in Indoor Atmospheres." I recommend that it be accepted in partial fulfillment of the requirements for the degree of Doctor of Philosophy, with a major in Engineering Science.

William T. Snyder, Major Professor

We have read this dissertation
and recommend its acceptance:

Accepted for the Council:

Vice Chancellor
Graduate Studies and Research

INVESTIGATION OF PARAMETERS AFFECTING THE CONCENTRATION OF RADON AND ITS DAUGHTERS IN INDOOR ATMOSPHERES

A Dissertation

Presented for the

Doctor of Philosophy

Degree

The University of Tennessee, Knoxville

Mahmoud M. Abdelrazek

June 1980

DEDICATED

TO

NERMEEN , MARGIE , AHMAD,

LAILA AND FRED.

ACKNOWLEDGEMENTS

The author would like to express his appreciation to Dr. W. T. Snyder, chairman of his committee and head of the Department of Engineering Science and Mechanics, for the generous, valuable discussions and advice he offered the author during the course of this dissertation. Special thanks are also extended to Dr. Snyder for offering the author a teaching assistantship which enabled him to continue his graduate studies.

The author would also like to express his deepest appreciation and gratitude to Mr. Fred F. Haywood who generously offered every possible assistance to the author during the course of this work. Without his help this work could not have been completed. His interest and knowledge in the field of this study are immense, and his criticism was highly considered.

Thanks are also extended to Drs. Jerry E. Stoneking, Wayne T. Davis, and Robert D. Birkhoff for their willingness to serve on the author's committee. Dr. William A. Goldsmith of Oak Ridge National Laboratory (ORNL) deserves special thanks. He generously offered his time for discussions, criticism, and advice. His expertise in the field of radon technology was a significant factor in the output of this work. The author wishes to express his thanks to Dr. J. S. Bradley of the Mathematics Department for offering the author a teaching assistantship and also to the staff members of the Health and Safety Research Division of ORNL for the hospitality they offered to the author during the past seven years, and for providing the necessary instruments needed for the experimental measurements. Last, but not least, thanks to UTCC for providing the

computer time and to Lana Melton, ORNL, for typing this dissertation. Many thanks go to the author's family which has been extremely patient and given him every chance to complete this work.

ABSTRACT

Accumulation of the radioactive gas, radon (Rn-222), and its daughters in inhabited areas could significantly influence population exposure to natural radiations. Exposures to high concentrations of airborne radon daughters have been associated with the induction of lung cancer among uranium miners. This research is directed toward studying the parameters which affect the concentration of radon and its daughters inside a building contaminated with radium-226 deposits. Measurements were conducted in a building of an industrial park in southwestern Pennsylvania.

Ventilation rates inside the building were measured using carbon dioxide (as a tracer gas) which was detected by an infrared spectrometer. Ventilation rates were found to vary depending on the outdoor wind conditions. The technique of using carbon dioxide as a tracer gas coupled with infrared spectrometry proved to be an excellent and reliable technique.

Concentrations of radon gas inside the building at different locations were measured continuously using modified computer-based versions of the well-known Wrenn chambers. Measurements indicated that indoor radon concentrations vary according to the activities performed in the building as well as on the ventilation conditions. A pronounced increase in radon concentration was observed whenever airexchange rate was reduced.

Radon flux measurements were performed in order to quantify the individual indoor radon sources which contributed to the observed overall concentration. Radon flux emanating from the concrete floor

slabs, cracks, and floor drains was found to be the principal source of radon in the building. Radon flux measurements were accomplished by utilizing activated charcoal canisters as accumulators. Field measurements of this kind usually require a large number of canisters in order to obtain reliable results. The preparation and handling of large quantities of canisters became a time-consuming process. For this purpose, a crack monitor was designed and calibrated. This crack monitor is based on the idea of circulating the radon gas emanating from a certain crack length (one meter) and collected in a metal housing through an accumulator (charcoal canister). The accumulator thus collects the integral radon flux over the entire length of the crack monitor. Results showed that for a flow rate of 8 liters/minute the crack monitor records 70 percent of the actual radon flux. Radon flux emanating from drains in that building was measured also. A new method was developed for measuring radon flux emanating from drains without blocking natural convective flow into and from the drain. The method is referred to as the open-end, double-detector method, in which two canisters are connected back to back by a short brass pipe. The canister facing the drain will collect the integral flux emanating from the drain over the collection period, while the upper canister will collect the reverse flow if it may occur.

Radon daughter concentrations were measured using the technique devised (and later refined) by Kerr in which particulate radon daughters in building air are collected on a filter and then counted using a surface barrier alpha spectrometer. Measured radon daughter concentrations vary considerably and are controlled to some extent by the concentration of radon gas as well as by ventilation conditions. No

general correlation between radon concentrations and daughter concentrations was obtained in this particular situation.

An analytical model based on conservation laws was developed which incorporates the effect of parameters such as wall/floor material, air-exchange rates, variations in atmospheric pressure, and outdoor radon concentration. Indoor radon gas was assumed to be removed by air-exchange process, radioactive decay and interaction with walls. A linear model was introduced in order to evaluate the effect of variations in atmospheric pressure on the exhalation rate of radon from surfaces (wall/floor). A good agreement between experimentally measured radon concentrations and those calculated using this model for the west room of the building was obtained. Also measured and calculated working levels for the same room agreed very well.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF FIGURES

LIST OF FIGURES

CHAPTER I

INTRODUCTION

Presently increasing emphasis is being placed on reduction in energy consumption for heating and cooling of houses. Reduction of air-exchange rates in houses and utilization of tight thermal envelopes are being proposed to achieve energy conservation goals. It has been recognized that conserving energy by reducing air-exchange rates in houses will result in a significant increase in population exposure to indoor pollutants including the radioactive radon gas.

Most building materials contain small quantities of naturally occurring radioactive materials. The amount of radioactive material found in building materials varies according to the type of material involved. It is known that concrete and concrete products, stone, and some bricks contain more radioactivity than wood and paper products. Radium-226 (a member of uranium-238 decay series) is the radionuclide in building materials which decays to give the radioactive noble gas radon-222.

Some of the radon created within the crystal structure of the building materials and the underlying soil diffuses through air-filled pores to the indoor atmosphere. At the same time, radon gas diffuses from the surrounding soil to produce the outdoor radon gas. The exhalation process from building materials and underlying soil together with the infiltration of outside air containing radon-222 into a structure constitute the major source of indoor radon gas.

Under normal ventilation conditions, some of the radon gas is removed from the structures prior to undergoing radioactive decay. However, with less exchange between outside and inside air there will be some buildup in the concentration of radon gas and its daughters. Such a buildup in occupied spaces results in increases in the radiation dose to the lung which in turn increases the risk of developing lung cancer. Accordingly, to exploit fully the energy savings afforded by these conservation measures without, at the same time, risking adverse health effects, one must have a quantitative knowledge of the consequences of lowering air-exchange rates in houses and public buildings.

Project Objective

The principal objective of this research was to study the effect of air-exchange rates on the concentration of radon and its daughters inside houses. One major task of this work was to measure the (overall) concentration of radon and its daughters in houses under varying air-exchange conditions. The second major task of this work was to identify the individual radon sources which contribute to the observed radon concentration and to devise suitable means to measure these sources. The third major task of this research is to provide a mathematical model of the problem. The effect of parameters such as building materials, ventilation rates, and atmospheric pressure on the concentration of radon and its daughters in houses are incorporated in this model.

Project Significance

The evaluation of the population exposure to radiations in an area is significant in itself in that a potential source of radiation-induced health effects is investigated and quantified. Another argument for the

significance of a study of this nature is the demonstration that the source under investigation produces population doses comparable to better-known sources which have received considerable attention in the past. The significance of this last argument may be realized from the following discussion concerning population exposure due to the different radiation sources.

Radon and its daughters are present everywhere in the atmosphere, and accordingly, everyone receives a certain radiation dose due to this natural radioactivity. The magnitude of this exposure depends on the radon concentration in the environment in which a person resides or works and also on the concentration of radon daughters. Health effects of elevated concentrations of radon in air are due not to radon itself but to its short-lived, alpha-emitting daughters which, when inhaled, are deposited on the interior surfaces of the respiratory tract. The subsequent decay of these radionuclides results in the absorption of alpha particles in the bronchial epithelium region of the lung. Absorption of this alpha-particle energy leads to radiation exposure to the lung.

Assume that all people in the USA spend one-half of their time indoors. This results in a population-lung-dose per year of approximately 10^8 person-lung-rem per year (number of persons in USA population x average indoor radon daughter concentration x lung-dose conversion factor). A reduction in the ventilation rate of a structure by a factor of two may result in an increase in radon daughter concentration by a factor of about 2.5 [59]. Hence, if energy conservation measures were applied to all structures in this country, the resulting population-lung-dose could conceivably be as great as 2.5×10^8 person-lung-rem per

year. Likewise, if such measures were applied to structures in which only one percent of the population resided, there would be an increase of about 2.5×10^6 person-lung-rem per year over the natural background radiation exposure rate. On the other hand, population-lung-dose from diagnostic radiographs is estimated from the Environmental Protection Agency (EPA) data [2, 37, 57] to be 1.4×10^7 person-lung-rem per year. Also, it is estimated from Nuclear Regulatory Commission (NRC) data that the annual lung dose due to operations in the entire nuclear fuel cycle (mining, milling, fuel fabrication, reactor operations, etc.) is approximately 7×10^4 person-lung-rem per year. A comparison between the given population-lung-doses per year due to different radiation sources clearly shows the significance of a study of this nature.

Moreover, the methods of identifying and measuring the individual indoor radon sources presented in this research should be of significant importance for remedial actions which may be needed. Also, the generality of the mathematical model presented in this study makes it applicable to buildings under various conditions of ventilation and structure.

CHAPTER II

REVIEW OF RELATED RESEARCH

This chapter presents a literature review of articles related to the core subject of this project which is indoor radon. Natural radiation sources, together with radon properties and radon decay scheme, are briefly discussed. A great deal of attention was given to the effect of building materials, ventilation conditions, and outdoor radon on the concentration of indoor radon. The radium content of building materials and the process of radon exhalation and its dependence on atmospheric pressure and on temperature are also discussed.

Natural Radiation Sources

Human exposure to radiation is of great concern because of the potential harmful effects of radiation on the human body. Among the consequences of continuous low-level exposure to radiation are cancer, shortening of life span, and cataracts [1].

Radiations of natural origin are recognized as the largest source of human exposure to ionizing radiations. Radiation sources of natural origin include cosmic radiations, radiations from naturally occurring radionuclides in the earth, and radiation from radionuclides in the human body [2]. Because man now spends the greater part of his life inside buildings, the greater fraction of population exposure to natural radiation sources is provided by the radioactivity of the living quarters or place of work [2, 3].

Indoor radioactivity is basically associated with the existence of the radioactive radon gas (Rn-222) and its decay products in indoor atmospheres. It is well known that the increased risk of respiratory cancer mortality among uranium and other underground miners is related directly to the inhalation of radon and its decay products [4, 5]. From epidemiology studies of miner populations, it is estimated that for a miner who is exposed to the guideline value of radon daughter concentration ($\sim$60 pCi/m^3) for 2,000 hours per year, there is an increase in that person's risk of dying from lung cancer of about four percent [4, 5].

Radon Decay Scheme

Radon (Rn-222)* is a radioactive noble gas with a half-life of 3.82 days, and it is the immediate daughter of radium-226 (a member of uranium decay series). The following is the decay scheme of radium-226 where the type of emitted particles is shown together with the half-life time of the various decay products.

$$\text{Ra-226} \xrightarrow[1602\ \text{year}]{\alpha} \text{Rn-222} \xrightarrow[3.82\ \text{day}]{\alpha} \text{Po-218} \xrightarrow[3.05\ \text{min}]{\alpha}$$

$$\text{Pb-214} \xrightarrow[26.8\ \text{min}]{\beta} \text{Bi-214} \xrightarrow[19.7\ \text{min}]{\beta} \text{Po-214} \xrightarrow[164\ \mu\text{sec}]{\alpha}$$

$$\text{Pb-210} \xrightarrow[21\ \text{year}]{\beta} \text{Bi-210} \xrightarrow[5\ \text{day}]{\beta} \text{Po-210} \xrightarrow[138\ \text{day}]{\alpha} \text{Pb-206 (stable)}.$$

Historically, radon decay products (or radon daughters) are known as RaA (Po-218), RaB (Pb-214), RaC (Bi-214), RaC′ (Po-214), RaD (Pb-210), and RaE (Bi-210). Evans [6] gave an excellent summary of the physical and engineering considerations concerning radon and its

*The term radon will always be used to refer to the radioactive radon gas (Rn-222).

decay products. For radiation exposure purposes (i.e., dose calculations) only three decay products (namely, RaA, RaB, and RaC′) need to be considered. The beta emitters in this decay series do not contribute significantly to the total dose but may have to be taken into consideration because of their decay to an alpha emitter (RaC′) [3, 6-10].

Parameters Affecting Indoor Radon Concentration

Indoor radon concentration depends on the type of building material (including paints, decoration, and underlying soil or rock bed), ventilation conditions, and outdoor radon concentration [2, 10-13]. Recently it was reported that radon from natural gas and underground water could also contribute to indoor radon radioactivity [8, 14-18]. Moreover, recent proposed energy conservation means could introduce another indoor radon source through the use of heat storage medium coupled to air and to solar systems [2, 19-20]. The contribution from each of these sources to indoor radon concentration will be discussed briefly.

Building Materials

Most rocks and soils contain uranium as a minor contaminant in concentrations of typically 1-4 ppm [6]. Consequently, radium-226, as a member of uranium decay series, is found with various proportions in rocks and soils which are used directly as building materials or as ingredients for preparing building materials. Accordingly, radium content of building materials is of major interest with regard to indoor radioactivity. Radium can be viewed essentially as a constant-rate radon source (because of its long half-life time) as well as a source of other radiations being emitted by the constituent nuclei themselves [10-13, 20-21].

Radium content of soils and rocks. Radium content of soils and rocks has been extensively reported in the literature, and a very useful summary is given in reference [21]. From this reference, as well as others [22-24], the following generalized conclusions are obtained. The radioactivity in igneous rocks is related to its silicate content, being highest (∿4.7 ppm) in acidic variations (e.g., granites) and lowest (∿0.08 ppm) in the ultrabasic rocks (e.g., dunites). Sedimentary rocks, on the other hand, exhibit lower radioactivity than igneous rocks. Certain sedimentary rocks, including some shales and phosphate rocks, are highly radioactive (∿3.5 ppm), while others such as limestone halite and gypsum are low in their radionuclide content.

The radioactivity of soils depends not only on that of the parent rocks but also on soil formations, depth, and transport process. Typical radium content of a wide variety of soils in North America and Europe is in the range $0.5\text{-}2.0 \times 10^{-12}$ g/g soil.

Commonly used materials for private and public buildings are mainly concrete, bricks, and wood. It has long been recognized that there is little radium in wood and most other materials from which light frame houses are constructed [2, 10, 25-29].

Soltizki et al. [cited in reference 11] reported a series of measurements of radium content of raw ingredients of some building materials in the U.S.S.R. Radium content of sand, bricks, and granites was found to be within the ranges 4-8, 9-14, and pCi/g, respectively.

Radium content of building materials commonly used in Great Britain (clay bricks, calc silicate bricks, flint aggregate bricks, and gypsum by-products) was studied by Hamilton [30]. These results showed that the highest radium content (∿17 pCi/g) was observed in gypsum by-products

which are used in plaster boards. Granite bricks, however, were found to have a mean radium content of 6.9 pCi/g.

Pensko [31] in a recent study on the radon-emanating powers of different building materials commonly used in Poland gave some values for the radium content of those materials. By-product gypsum was found to have the highest ($\sim$11.8 pCi/g) radium content among those materials followed by fly ash with 2.6 pCi/g.

Another source of radium which can contribute to some extent to the indoor radon could result from the use of radium-bearing paints as reported by Haque [12] or from the use of some decoration materials as reported by Hoather [32].

Radon exhalation process. Radium being naturally existing in building materials, soils, and rocks will decay, and radon gas will be formed within the crystalline structure of these materials. Radon atoms, because of their inert gas structure and balanced charge, are insufficiently bounded within the crystalline structure of materials, and accordingly, they can escape into void spaces. On the other hand, radon daughters, because of their nature as being ionized atoms, have a very limited movement within the crystalline structure of materials, and they will be absorbed at the point of their birth. Some portion of the radon gas diffuses toward the surface along a concentration gradient where at the surface some gas escapes into the surrounding atmosphere before undergoing radioactive decay [23, 33-35]. The term emanating power (or exhalation power) is used to represent the ratio of the amount of radon that escapes to the amount produced at the equilibrium state. Baretto et al. [35] in their work on the emanation power of different rocks and soils concluded that the percentage of radon which escapes

from a mineral is not correlated at all with the uranium concentration, but it is largely dependent upon the stability of the mineral structure and its crystallinity.

Radon exhalation rate into room air from the surrounding walls and from the underlying floor is influenced by the wall and floor surface finishing, variations in barometric pressure, and temperature.

The effect of wall and floor surface finishing on radon exhalation rate is due to the fact that some materials act as radon barriers [11, 36-38]. Steinhausler [11] reported that plastic paints, thick wall papers, and metallic paneling applied to wall surfaces reduce the amount of radon exhaled into the room atmosphere. On the other hand, Culot et al. [36] were able to develop a radon barrier which is capable of impeding the diffusion of radon from the floor of an experimental building where uranium rich materials (uranium mill tailings) were used as a land filler. A very comprehensive list of experimental effectiveness of selected commercial polymeric sealants in stopping radon gas diffusion from walls of uranium mines was presented by Franklin and Nuzman [39].

The effect of atmospheric pressure on radon exhalation rate has been the subject of numerous investigations with conflicting results in some cases [40-45]. The studies involving the effect of atmospheric pressure indicated that pressure changes of the order of 1 percent or less may be responsible for variations in radon exhalation rate of as much as 50 to 100 percent depending on the rate and duration of the change of pressure [44, 45].

Kraner et al. [40] proposed a qualitative model to account for this effect. It states that during periods of decreasing atmospheric pressure,

radon rich air is drawn from the walls (or soil surface), and during periods of rising pressure air, low in radon, is forced into the walls (or soils).

Clements and Wilkening [44] introduced a simple analytical model to account for pressure effects on radon exhalation rates in soils. Model calculations showed that pressure changes of 1 percent produce changes in radon exhalation rate from a soil by 20 to 60 percent depending upon the rate of change of pressure and its duration. These results were experimentally confirmed by Clements [44] and Jonassen [45].

Experimental investigation of the effect of atmospheric pressure on radon exhalation from walls of Norwegian dwellings was conducted by Stranden et al. [46] who reported that the daily supply of activity in a small room with very poor ventilation from the sucking effect during a period of falling atmospheric pressure is about 2.7 pCi per liter/mm Hg of daily decrease in atmospheric pressure. Similar experimental results were reported by Jonassen et al. [47] who reported that for a room of an emanating area of 360 m^2 and air volume of 324 m^3 the emanation power per unit pressure drop is 91 atoms/m^2-sec-mm Hg.

Although the effect of pressure changes on radon exhalation rate is of significant importance, its contribution to any increase in indoor radon concentration is limited by the exchange of outdoor air (low in radon) and indoor air (rich in radon) (i.e., by air-exchange rates).

Temperature effect on radon exhalation rate from walls was measured by Gabrysh and Davis [20] where it was found that in a room of a radiantly heated dwelling, a concentration of about 1.41×10^{-10} curies of radon per liter of air could result, which is approximately 1.5 times the allowable radon concentration in a room.

Few other investigations concerning temperature effect on radon exhalation from soils and radioactive minerals were reported [23, 48-51]. This effect was attributed to the opening of the microfissures inside materials under the effect of heat and thus releasing radon from previously closed microfissures [51]. On the other hand, Giletti [48] attributed the increase in radon exhalation rate to the increase of the diffusion coefficient with temperature although the partial pressure of radon may also contribute.

Ventilation Conditions

Air is exchanged in residences either by natural infiltration or by ventilation. Natural infiltration results chiefly from leakage through cracks and gaps around doors and windows and is not controlled by residence occupants. It depends mainly on the wind speed in the open air and on the temperature difference between room air and outdoor air [52]. Increased wind speed and/or temperature differences from the inside to the outside increase ventilation by infiltration [11]. Ventilation, on the other hand, comprises the controlled displacement of air in residences either naturally through doors and windows or mechanically by using air conditioning systems [11, 52].

Ventilation rates (or air-exchange rates) in houses have been the subject of numerous investigations. A very useful summary of techniques and methods used to measure home ventilation rate was presented by Handley and Barton [52], and from this summary the following conclusion is reached. The concentration of a tracer gas in a house was found to decrease exponentially as indicated by the following equation:

$$C = Coe^{-It},$$

where

C = tracer gas concentration at time t,

Co = tracer gas concentration at time t = 0,

t = time in hours,

I = air-exchange rate per hour; assumed to be constant.

Tracer gases such as helium, ethane, sulfur hexafluoride, and inert radioactive gases have been used to measure air-exchange rates. Recently sulfur hexafluoride has been the preferred tracer gas because of the high sensitivity of detecting this nontoxic gas. However, helium is by far the most common tracer used although its use has been questioned because it has a higher diffusion rate than air. Recently, in a study on the levels of radon and other indoor pollutants in a tight, well-insulated residential structure, Haywood and Hawthorne [53] used carbon dioxide (CO_2) as a tracer gas, and measurements were made using a portable mass spectrometer.

Air-exchange rates in homes vary over a wide range because of the differences in construction quality and location. Measured air-exchange rates in a gas-heated house were found to vary from 0.24 to 0.83 per hour, while the range was 0.13 to 0.42 for an all-electric house [54]. It was suggested (by the reviewers) that a range of 0.5 to 1.5 air exchanges per hour be used for purposes of radiation exposure calculations in United States housing units.

Recently Spitz and Wrenn [55] were able to infer the ventilation rate of a structure from measurements of indoor (Ci) and outdoor (Co) radon concentrations. Assuming a constant emanation rate ($\overline{SA}$) and a

constant ventilation rate (λ_v), the following equation was derived for steady-state conditions:

$$Ci - Co = \frac{\overline{SA}}{(\lambda_r + \lambda_v)V} ,$$

where

λ_v = ventilation rate (1/hr),

λ_r = radon decay constant (1/hr),

Ci, Co = indoor, outdoor radon concentrations (Ci/liter),

$\overline{SA}$ = emanation rate (curie/hr),

V = volume of air contained in liters.

Continuous radon measurements indoor and outdoor of a residential structure were performed and from the above formula λ_v was found to be 0.3/hr. This calculated value of λ_v was found to be reasonable for the type of structure considered. It is worth mentioning that Spitz's approach bears some relation to that of Wilkening and Watkins [56] who assumed that under steady-state conditions the following relation could be used for cave atmospheres:

$$\frac{C\ max - C}{C} = \frac{Q(\theta)}{\lambda_v} ,$$

where

C max = maximum mean concentration of radon,

C = mean concentration of radon in cave air,

λ = decay constant of radon,

v = internal volume of cave,

$Q(\theta)$ = air-exchange rate between outdoor and cave at temperature θ.

This formula was derived for a time scale which is longer than the radon half-life and the turnover time of air in caves and also for cave temperatures less than outdoor temperatures.

Generally, outdoor radon concentration is less than indoor concentration. This has been reported by many authors [2, 3, 11-13, among others]. Accordingly, it will be expected that the function of home ventilation (natural and/or artificial) is to lower indoor radon concentrations through dilution with outdoor air. This will automatically lead to a reduction in the amount of the hazardous radon daughters. Furthermore, movement of indoor air masses (by ventilation) will considerably increase the probability of radon daughters attachment to walls and other surfaces and thus reduce the amount of radon daughters available for inhalation by occupants [57-59].

An early study [57] on the occurrence of nonequilibrium atmospheric mixtures of radon and its daughters in uranium mines revealed some useful results about ventilation effects on radon daughter concentrations in mines. Part of the results are shown in Table 1 as an illustration of ventilation effects on daughter concentration.

TABLE 1

EFFECT OF VENTILATION RATES ON THE CONCENTRATION OF RADON DAUGHTERS IN URANIUM MINES (FROM REFERENCE 57)

Ventilation rate (ft^3/min)	Ventilation period	Concentration (atom/Liter)		
		RaA	RaB	RaC
800	30 m	4.7×10^1	1.4×10^2	5.1×10^1
400	30 m	2.7×10^2	1.95×10^3	1.27×10^3
300	30 m	8.1×10^2	5.16×10^3	3.6×10^3
150	30	6.35×10^3	4.96×10^4	3.06×10^4

Cliff [58] recently measured the effect of ventilation rates on the concentration of radon daughters in a group of dwellings in Great Britain. These measurements showed that for a room emanating 0.54 pCi radon/liter per hr, the concentrations of RaA, RaB, and RaC were decreased by factors of 16.35, 28, and 41.33, respectively, by increasing the hourly ventilation rate from 0.1 to 2.0.

On the other hand, measurements of radon concentrations in different buildings (homes and industrial premises) were conducted by Haque et al. [12], which show clearly the effect of ventilation on indoor radon concentration. Ventilation rates were termed adequate or inadequate and no numerical values were given. The results show that radon concentration in sealed rooms varies according to exhalation rate in each room with an average value of 30×10^{-13} curie/liter. Average radon concentration inside an adequately ventilated room was found to be 1.64×10^{-13} curie/liter, while that outside was 0.42×10^{-13} curie/liter. In addition, average concentration inside an inadequately ventilated room was found to be 3.5×10^{-13} curie/liter, while outside concentration was 1.3×10^{-13} curie/liter. One can see that radon concentration in an adequately ventilated room was reduced by a factor of 18, while that of an inadequately ventilated room was reduced by a factor of 9.

Few other investigations concerning the effect of air-exchange rates on indoor radon concentrations were reported [8, 24, 59, 60] with the general conclusion that a reduction in radon concentration inside buildings can be achieved by increasing air-exchange rates.

Outdoor Radon Concentration

The continents are the only significant source of atmospheric radon (outdoor radon) because there is very little radium-226 dissolved in sea

water and also because radon created on the sea floor will decay before reaching the atmosphere [21, 41, 44, 61-66]. Recently Travis [66] estimated the release of radon in the United States in 1978 from various natural and technologically enhanced sources. Travis' results show that the annual release of radon from natural soil in the United States is 1.28×10^8 Ci.

Concentrations of outdoor radon depend on the concentration and distribution of radium-226 in the soil, the emanating power of radon from its point of formation and the diffusion properties of the soil. Soil covers such as snow or vegetation can affect the rate of radon escape from the surface of the ground [21, 22, 41, 44, 61-65]. Soil moisture has significant effect on radon diffusion and exhalation processes. The observed decrease in the outdoor radon concentration after rain has been attributed to the excess moisture in the soil [41, 61, 63, among others].

At a specific location, outdoor radon concentrations depend not only on soil properties but also on the prevailing atmospheric or meteorological conditions. Diurnal and seasonal variations of outdoor radon concentrations have been observed and mostly can be attributed to changes in atmospheric stability conditions [22]. Maximum ground-level concentrations are exhibited in the early morning hours when stable atmospheric conditions occur [22, 65]. On the other hand, wind associated with pronounced turbulent motion has been observed to cause a reduction in the outdoor radon concentration [11, 22, 41]. This effect is most likely a result of mixing of the air and radon. Other meteorological parameters such as temperature and pressure influence the outdoor radon concentration due to changes in radon exhalation with

temperature and pressure. This effect has been discussed previously in connection with building materials.

Indoor radon concentrations can be affected by variations in outdoor concentration mainly through the process of air exchange between indoor and outdoor atmospheres.

CHAPTER III

EXPERIMENTAL MEASUREMENTS

This chapter is concerned with the details of experimental measurements and instrumentation. A brief description of the building where measurements were conducted is given. The methods of measuring air-exchange rates, radon concentrations in air, and radon flux emanated from various indoor sources are discussed and references are cited whenever necessary. Also, the method of measuring the concentration of radon daughters is discussed briefly. Instruments and detection devices described in this chapter are provided by the Off-Site Pollutant Measurements Group of Oak Ridge National Laboratory's Health and Safety Research Division. This group has been actively involved in the field of radon assessment and measurements technology. Detailed descriptions, calibration, and field testing have been reported in numerous places [67-69].

Experimental Building

Measurements of radon concentration in air, air-exchange rates, and indoor radon flux were conducted in a building (Building 1), located in a small industrial park in southwestern Pennsylvania. The industrial park occupies an area of 18 acres and was formerly used under a U.S. Government contract for the extraction of uranium. This area is bounded by a creek, a railroad, a former pottery company, and a former street railway right-of-way. The various buildings in the park are leased to tenant companies with a total of approximately 110

employees. During the period from 1911 to 1957, this park area was an industrial facility for extracting radium and uranium from their natural ores. As a result, the area became contaminated mostly with radium and uranium products. The history of the operations conducted in the park area, as well as radiological surveys in the park area, are reported in the United States Department of Energy reports concerned with the remedial action program for formerly utilized Atomic Energy Commission sites [70].

Building 1 (built in 1951) is a two-story building with concrete block walls, steel framing, concrete floors, and a precast concrete slab roof (tar and gravel covered). The building dimensions are 92.33 ft x 31.83 ft, giving a floor area of 5880 square feet. The ground level, which was used as a laboratory during uranium extraction operations, consists of two wide rooms and a small entrance leading to the upper level which is entirely an office area. The rooms on the ground floor are designated east and west according to their location with respect to the center wall (see Figures 1 and 2). Because of its previous use as a laboratory, the ground level contains many drains in the floor; some of these are completely open, whereas others are plugged with concrete or wood. The floor of the ground level has cracks in numerous places together with expansion joints. The length of the cracks in the floor of the ground level is approximately 70 meters.

Currently, the building is being used by an industrial laundry as a clothes delivering station with 11 employees involved. The ground level of the building is ventilated naturally through doors, windows, and some holes in the walls. The upper level is also ventilated naturally although a few offices have added electric fans. Heating

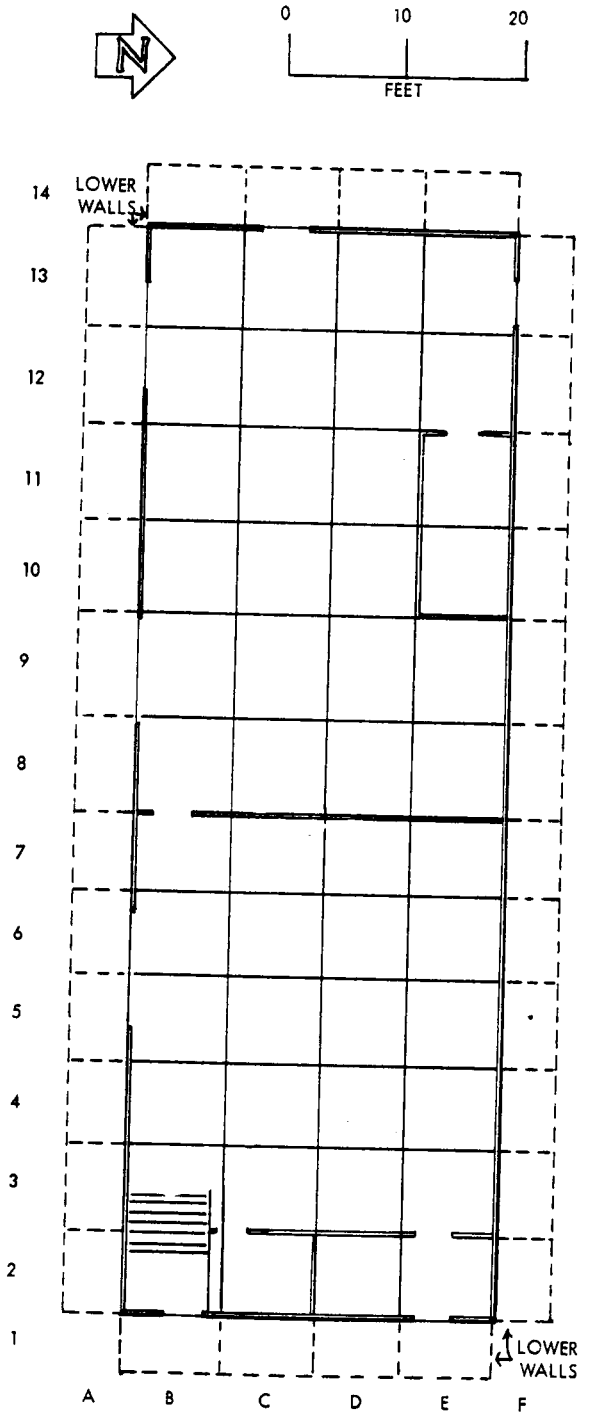

Figure 1. The ground level of Building 1 of the Canonsburg Industrial Park.

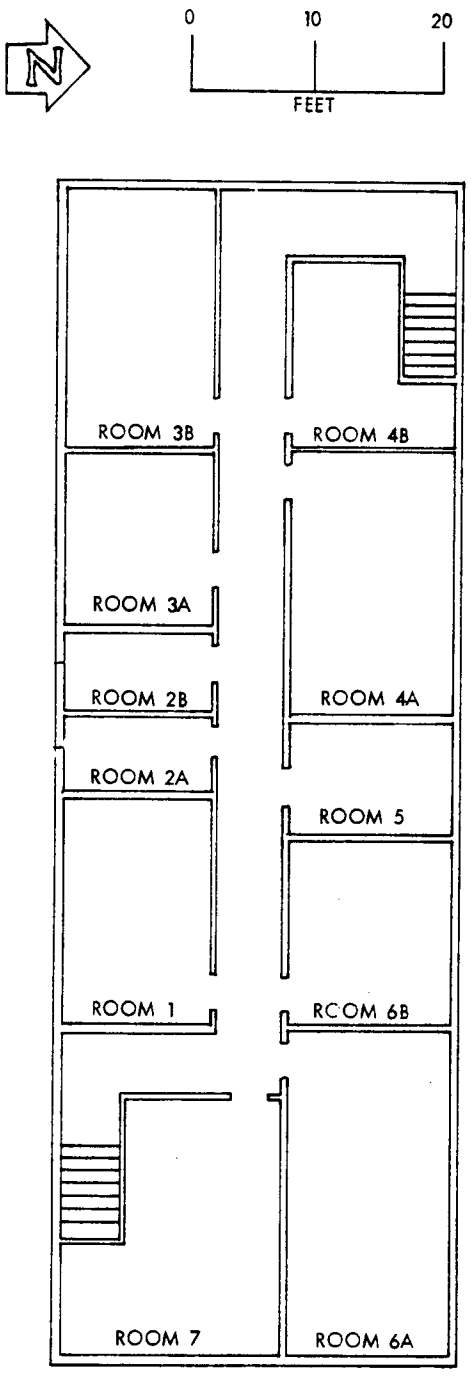

Figure 2. Upper level of Building 1.

of the building during winter is accomplished by using (old type) jet heaters on the ground level and small portable heaters on the upper level.

Measurement of Air-Exchange Rates

Air-exchange rates were measured inside the building during normal operations; no attempt was made to regulate these activities. The method used to measure air-exchange rate is that of Haywood and Hawthorne [53, 71]. In that method carbon dioxide (CO_2) is used as the tracer gas, and a computerized, infrared spectrometer (Wilks, MIRAN-80) is used as the detector. The following is a brief description of the spectrometer based on information reported by Hawthorne [71].

The MIRAN-80 has a wavelength range of 2.5 μm to 14.5 μm with 0.4 μm accuracy and 0.05 μm reproducibility. It is a single-beam instrument with a lithium tantalate ($LiTaO_3$) pyroelectric detector and a nichrome wire source capable of detecting 0.0001 absorbance unit. The heart of this instrument is a microprocessor which controls the wavelength, records transmission data, and calculates concentrations from stored matrix coefficients that account for compound interference. The stored coefficients must be obtained from a matrix inversion operation performed on calibration data by a computer other than the MIRAN-80.

In routine operation a mixture of five compounds can be analyzed quantitatively in approximately one minute. The computer is capable of handling up to 11 interfering compounds. The instrument has an operator keyboard to handle communication with the computer and also has a printer to give a permanent copy of the data analysis.

For the purpose of measuring air-exchange rates inside the various rooms of Building 1, the MIRAN-80 was preset to detect only carbon

dioxide gas and to print out the data each minute. The instrument was allowed to warm up for a period of 30 minutes, and background CO_2 readings were obtained during this period. Carbon dioxide gas from pressurized bottles was then released into the atmosphere of the room under investigation. To obtain a nearly uniform distribution of the gas into the room air, the pressurized bottle was turned around into different directions during gas release. The buildup of gas concentration could be observed from the printed data and, as soon as a constant concentration was reached, gas release was stopped. Air-exchange rates are then calculated from the time-decay curve of the concentration as has been discussed before in connection with ventilation conditions.

Measurement of Radon Concentration in Air

Several techniques have been developed for measuring the concentration of radon and its daughters in various media, including air. A review of these techniques is given by Budnitz [72]. Generally, there are two quite different approaches to the specific measurement of radon. In the first, equilibrium can be assumed to have been established between radon and its daughters; in the second, the daughters are initially filtered out from the sample gas, after which either the decay of radon itself is detected or the daughters are allowed to ingrow again for detection. This second approach has been utilized by many investigators in developing environmental radon-measuring instruments [73-78].

Spitz and Wrenn [73] designed a continuous radon-measuring instrument (known as Wrenn chamber) based on the detection of positive ions (RaA) resulting from the decay of radon atoms into a sensitive volume. Radon daughters are first filtered out from the incoming gas

before entering the sensitive volume by using a polyurethane foam layer. Like most plastics, this foam has an internal charge which serves to trap particulates and allows only radon gas to diffuse through.

In the ORNL model, curved quadrants of foam are glued at the junctions and placed over a hemispherical metallic screen. This hemispherical screen provides outer structural support for the foam and functions as the anode for the electrostatic field applied within the sensitive volume of the chamber. A hemispherical light pipe coated with zinc sulfide scintillator and covered with a thin aluminized mylar film to provide a conducting surface (cathode) constitutes the alpha particle detector in this instrument as shown in Figure 3. The potential between the aluminized mylar film and the hemispherically shaped metallic screen creates a strong electric field which serves to attract the charged ions. The ions thus attracted remain on the surface of the mylar film and continue their radioactive decay to other radon daughters. The principal radiations detected by a radon monitor of this type are alpha particles from polonium-218 (RaA) and polonium-214 (RaC´). Light scintillations produced by the interaction of alpha particles with the zinc sulfide scintillator are then converted to voltage pulses through the use of a photomultiplier. Pulses from the photomultiplier are then amplified and fed to a counter after passing through a suitable threshold-discriminating circuit which excludes pulses other than those resulting from alpha particles. Periodic calibration of the chambers in use at ORNL showed that the average conversion factor is 2.36 counts-liter/pCi-minute and the average background is 0.28 counts/minute [69].

Modified Wrenn chamber units in use at ORNL are all fitted with MDT-COMP-8 microcomputers and printers. Communications with the

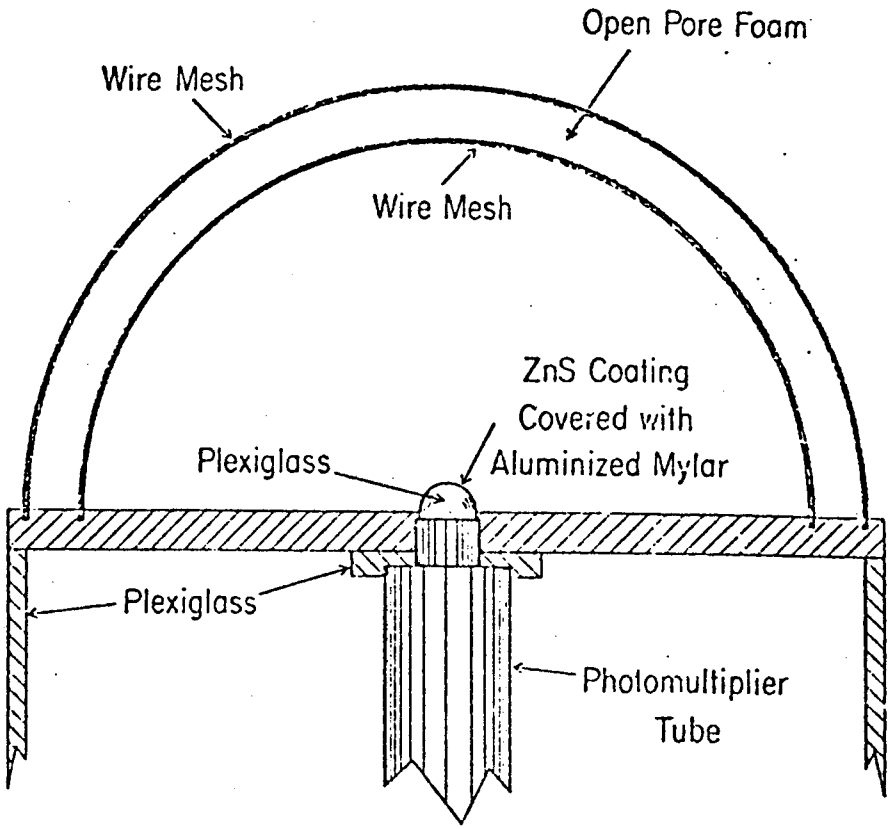

Figure 3. A cross section of the detector portion of a Wrenn chamber.

computers are handled by using an external, portable teletype where data such as date, time of day, chamber number, background, conversion factor, and counting interval are fed into the computers. At the end of each counting interval (usually 30 minutes), the total counts and the radon concentration are printed automatically, the system is then reset, and counting for the next period is initiated.

Measurement of Radon Daughter Concentration in Air

An alpha spectrometry technique has been refined by Kerr for the measurement of radon daughter concentrations in air. From one integral count of the Po-218 alpha activity and two integral counts of the Po-214 alpha activity, the concentrations in air of Po-218, Bi-214, and Pb-214 may be calculated.

Particulate radon daughters attached to airborne dust are collected on a membrane filter with a pore size of 0.4 μm. A sampling time of 5 to 10 minutes and a flow rate of 10 to 20 LPM are used. This filter sample is then placed under a silicon surface barrier detector and counted. Usually, counting of this kind is performed with a vacuum between the sample and the detector, which requires a complicated sample holder and time-consuming, sample-changing methods. Experiments at ORNL have shown that ease in sample handling is obtained with little loss in resolution when helium is used as a chamber fill gas. In this counter, helium is flowed between the diode and the filter sample, which are separated by a distance of 0.5 cm. One integral count of the Po-218 alpha activity is obtained from 2 to 12 minutes, and two integral counts of the Po-214 activity are obtained from 2 to 12 minutes and 15 to 30 minutes, respectively. All counting intervals are referenced to $t = 0$ at the end of sampling.

The equations describing the radon daughter atoms collection rates on the filter are of the form:

$$\frac{dn_i(t)}{dt} = C_i v + \lambda_{i-1}\, n_{i-1}(t) - \lambda_i n_i(t),$$

where

n_i = number of the ith species of atom on the filter as a function of time,

λ_i = radioactive decay constant of the ith species (minute^{-1}),

C_i = concentration of the ith species (atoms l^{-1}),

v = air sampling flow rate (liters minute^{-1}).

From the general form of the solution, specific equations can be obtained describing the number of each radon decay product collected on the filter as a function of time. Also, by letting v = 0 in the previous equation, a set of equations describing the decay on the filter of each radon daughter can be obtained. The equations describing the decay of radon daughters on the filter can be integrated and related to the integral counts obtained experimentally. Values for the total activities of Po-218, Pb-214, and Bi-214 on the filter at the end of sampling are obtained by applying matrix techniques. The airborne concentrations are obtained by solving the equations describing the atom collection rates on the filter. A computer program has been written by Kerr [67-68] to perform these matrix operations and to calculate the air concentrations of the radon progeny.

Measurement of Radon Flux

Several methods have been used to measure radon flux emanating from surfaces. The most common method is the direct accumulation of radon in a closed container resting on the surface. This method has been

used extensively in the United States to measure radon flux from soil surfaces [40, 43]. Megumi and Mamuro [49] developed another method whereby the radon emanating from soil is adsorbed on a layer of granular activated charcoal spread directly on the ground. After exposure, the charcoal is bagged and returned to the laboratory where the analysis involves measuring the gamma activity from bismuth-214 (RaC).

Countess [75] presented a new procedure based on the method described by Megumi and Mamuro [49] in which a U.S. Army gas mask canister (M11) containing activated charcoal is placed directly in contact with the emanating surface. The original army canisters are modified by removing the section containing a pleated paper filter and fitting a metallic lid on the other threaded end. The remaining cylindrical metal canister is 6 cm deep by 10.5 cm in diameter, weighs 225 grams, and contains 148 grams of activated charcoal. At the end of an exposure period the canisters are removed from the emanating surface, bagged in plastic bags, and returned to the laboratory for analysis.

The above method of measuring radon flux has been used extensively in this research. Some modifications have been added to account for various components of the radon source (e.g., cracks and drains) which could exist in normal living quarters or places of work. At ORNL, the canisters are usually depleted by heating them for a 24-hour period in an oven set at 100°C. After cooling down, the canisters are bagged in tight plastic bags to avoid moisture accumulation which could affect their adsorptive properties. Before exposing these canisters they are counted on a gamma-ray spectrometer to determine the background activity. During exposure, canisters are sealed to emanating surfaces using a clay-like material known commercially as sealflex (a product

of the M and W Electric Manufacturing Company, East Palestine, Ohio). After exposure, canisters are rebagged and transferred to the laboratory for analysis.

In this research, the charcoal canisters were analyzed using a gamma-ray spectrometer consisting of a sodium iodide crystal coupled to a multichannel pulse height analyzer. A schematic diagram of the gamma-ray spectrometer is shown in Figure 4. In practice, the exposed canister is placed on the NaI detector with the exposed side down and is counted for 10 minutes any time later than 4 hours after the end of the exposure period. This delay time is required to allow radon daughters to reach equilibrium with the adsorbed radon. The integration limits (windows) of the multichannel analyzer are set to include only the gamma-ray activity from lead-214 (0.242 MeV, 0.294 MeV, and 0.325 MeV) and from bismuth-214 (0.609 MeV). The multichannel analyzer displays the results in the form of two integral counts. A calibration coefficient of the counting system is obtained by counting a standard NBS radium-226 solution of known activity. Periodic calibration of the counting system resulted in a coefficient of 0.213 cpm/pCi.

It is well known that the decay or growth of a radionuclide is governed by the basic exponential law [6]. Simple mathematical manipulations of the radon decay equation yield the following formula which is used to calculate radon flux emanating from surfaces:

$$J = \frac{\lambda_1^2 N}{EA(1 - e^{-\lambda t})(e^{-\lambda t_1} - e^{-\lambda t_2})},$$

ORNL-DWG.80-10912R1

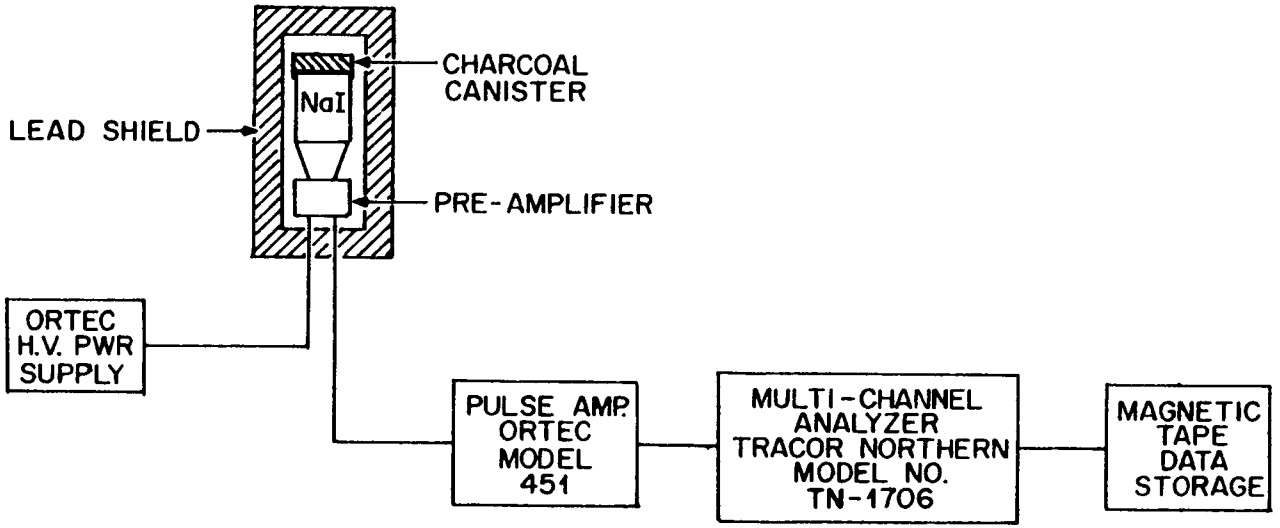

Figure 4. Block diagram of the gamma-ray spectrometer used to measure the gamma activity of the charcoal canisters.

where

J = radon flux, pCi/m^2-min,

N = net counts,

A = area of the canister, m^2,

E = calibration factor of the counting system, cpm/pCi,

t = exposure time, hours,

t_1 = time from the end of the exposure period to the beginning of counting (delay time), hours,

$t_2 = t_1$ + counting interval, hours,

λ, λ_1 = radon decay constant in hours and minutes, respectively.

It is important to recognize that the previous radon flux formula can be used only when surface (i.e., area) sources are considered. In order to adopt this method for measuring radon flux emanating from cracks (line source), drains and/or vents, the previous flux formula needs to be modified.

Cracks resulting from the rupture of wall and/or floor materials provide an easy passway for radon because the air medium inside these cracks has a much higher diffusion coefficient ($\sim$0.12 cm^2/sec) than the surrounding wall and/or floor materials. Therefore, a charcoal canister placed over a crack will essentially see the radon flux as a line source instead of an area source. This is due to the fact that radon input from cracks and joints is often several orders of magnitude greater than that from adjoining surfaces. Because of the small dimensions of the canister, it can be assumed that the canister will only see the radon flux emanating from a crack of length equal to its diameter, and the resulting radon flux will be given in units of picocuries per meter of crack length per unit time. Thus, the canister area (A) has to be

replaced by the diameter (D) in cases where line sources are considered. The total radon flux (in pCi/min) from cracks can thus be calculated by multiplying the flux (J) per meter of crack length by the total length of the cracks.

In field measurements, a large number of canisters may be needed in order to obtain results which are typical of the location under investigation. The handling of such a large number of canisters becomes a rather tedious and time-consuming process. For this reason a crack monitor was designed (Figure 5) which reduces considerably the number of canisters to be used for monitoring radon flux emanating from cracks and also to give more reliable results than those from single canisters. The basic idea of this crack monitor is to enclose the radon gas emanating from a crack of one meter length into a relatively small volume. Concentration buildup inside this volume is avoided by circulating the radon gas enclosed in this volume through a charcoal canister where radon gets adsorbed. The circulation in this volume is accomplished by utilizing a small air pump to cycle the air through the canister and back into the accumulator volume. A small fan inside the enclosure assures mixing of radon and air. The gas flow rate inside the crack monitor is of prime importance because high flow rates may produce sucking effect and low flow rates may produce concentration buildup. It was reported [79] that flow rates up to 16 liters/minute will not affect the adsorption (i.e., collection) efficiency of the charcoal canisters. Consistent and reproducible results were obtained with a flor rate of 8 liters/minute. Calibration results showed that the crack monitor records 70 percent of the actual radon flux as measured by individual canisters.

Figure 5. A photograph of the crack monitor showing the canister (right corner) and the air pump.

In actual field measurements crack monitors are placed on cracked areas and sealed to floors using the sealflex compound. Before placing the canister in its position on the crack monitor, the air pump and fan are allowed to run for one minute to remove the radon gas which may accumulate during the period of sealing the crack monitor to the floor.

On the other hand, the use of charcoal canisters to accumulate radon emanating from drains (due to contaminated sewer system) pose a problem which is different from that of cracks or surfaces. In this case the canister will essentially collect a volume source, and therefore, the flux becomes independent of the canister's area (A). Thus the flux emanating from drains (J_D) can be calculated from

$$J_D = \frac{\lambda_1^2 N}{E(1 - e^{-\lambda t})(e^{-\lambda t_1} - e^{-\lambda t_2})},$$

where λ_1, λ, t, t_1, t_2, and E are the same as described before, and J_D will be given in units of pCi/minute. In field measurements it is desirable to perform experimental measurements without disturbing the environment (i.e., the source in this case). This condition will put severe limitations on the method which could be applied to measure radon flux emanating from drains. The charcoal canister in its usual form (sealed from one side) will block the natural convective flow of air into or from drains. Accordingly, it became necessary to adopt a modified arrangement of the canisters which will permit normal air flow currents and also will cause minimum disturbance to the source. An arrangement which satisfies this condition is called the open-end double canister. For this method, two open-end charcoal canisters are connected together by a short brass pipe. One canister is placed with its wide base facing the drain, while the other canister faces the room

atmosphere. The lower canister will accumulate radon emanating from a drain through molecular as well as through convective diffusion processes. The upper canister works as a trap for any gas that may escape from the lower canister and also as an accumulator for any radon in room air which flows into the drain. To determine whether or not reverse flow occurs, another canister (called monitor) is placed in air near the drain and a comparison between the "monitor" reading and that of the upper canister will clearly identify the source recorded by the upper canisters.

CHAPTER IV

MATHEMATICAL MODEL

This chapter is concerned with the development of a mathematical model which is capable of predicting the concentration of radon and its daughters in the atmosphere of a single room. Parameters such as radioactive decay, air-exchange rates, and wall interactions are fully incorporated in this model to account for the removal of radon and its daughters from indoor atmospheres. The dependence of indoor radon concentrations on radon exhalation from inner surfaces is included. Special attention is given to the effect of variations in atmospheric pressure on radon exhalation rates from inner surfaces.

Calculation of Indoor Radon Concentrations

In the following development, it is assumed that radon accumulates in the indoor atmospheres due to exhalation from inner surfaces (walls and floor) and also due to the infiltration of outdoor air which may contain radon. The process of radon exhalation from walls and floor was discussed in Chapter II where it was mentioned that this process depended on the radium content of the emanating surfaces as well as on variations in atmospheric conditions such as pressure and temperature. The process of air exchange between indoor and outdoor atmospheres tends to reduce the indoor radon concentration if it is higher than that out of doors. However, this process could be reversed if the outdoor concentration was greater than that indoors. On the other hand, indoor radon is removed by radioactive decay, ventilation, and by wall

interactions. Removal of radon by radioactive decay is governed by its decay constant (0.0075/hr), and it is expressed as the product of the radon concentration (C_{Rn}) and the radioactive decay constant (λ_{Rn}). This is in accordance with the basic laws which govern the decay (or growth) of radioactive materials. The radon removal rate due to ventilation is controlled by the air-exchange rate (λ_V), and it can be expressed as the product of this parameter and the radon concentration.

The removal of radon due to wall interactions is introduced for the first time to account for the possible loss of gas molecules due to collision with inner surfaces (walls). The fact that radon gas can be adsorbed with high efficiency by activated charcoal led to the assumption that materials other than charcoal may interact similarly with radon. It is not necessary to restrict wall interaction processes to adsorption only since other processes such as diffusion or trapping of gas molecules into voids and fractures in walls could also occur. Accordingly, the term wall interaction will refer to all processes which result in a reduction in the gas concentration due to collision with wall surfaces. No experimental results were found in the literature concerning this type of interaction.

Approximate values of wall removal coefficient (λ_w) can be calculated using the basic laws of the kinetic theory of gases. According to these laws, the rate per unit surface area at which gas molecules strike a surface S (enclosed by a volume V) is proportional to the mean molecular speed [80]. Assuming that only a fraction β of the molecules which strike the wall will be removed, the wall removal coefficient (λ_w) can be evaluated using the following expression:

$$\lambda_w = \beta \times S/V \times (8RT/\pi M)^{1/2},$$

where

R = gas constant (8.317×10^7) ergs/mole-degree),

T = absolute temperature (°K),

M = gas molecular weight ($M = 222$).

The values of the fraction β will determine the dependence of the wall removal coefficient on the structural properties of different building materials. In this work, β is assigned the value of 0.000001 which could be modified according to future experimental evaluation.

On the basis of the previous discussion and by assuming a rather homogeneous concentration distribution of radon in the atmosphere of a simple room of surface area S and volume V, the following differential equation can be written to represent the time rate of change of the indoor radon concentration:

$$\frac{dC_{Rn}}{dt} = E \times S/V + \lambda_v C_o - (\lambda_{Rn} + \lambda_v + \lambda_w) \times C_{Rn},$$

where

C_{Rn} = time dependent indoor radon concentration (pCi/m^3),

C_o = constant or time dependent outdoor radon concentration (pCi/m^3),

E = exhalation rate (pCi/m^2hr),

λ_{Rn} = radon radioactive decay constant (1/hr),

λ_v = air-exchange rate (1/hr),

λ_w = wall removal coefficient (1/hr).

The exhalation rate, E, presented in the previous differential equation is assumed to be dependent on variations in atmospheric pressure. The dependence of radon exhalation rate on variations in

atmospheric pressure was discussed in Chapter II, where it was concluded that a drop in the atmospheric pressure will increase the exhalation rate in proportions which depend on the magnitude and on the duration of the pressure drop. To incorporate this dependence, the following simple model is introduced in which an incremental change of exhalation rate is proportional to the pressure variation [i.e., $dE = -\alpha dp(t)$]. The negative sign is introduced to compensate for the reverse effect of pressure variations on exhalation rate. The term, $dp(t)$, represents the change in pressure (drop or increase) measured from the normal atmospheric pressure. The parameter α represents the exhalation rate per unit pressure, and it has the units, pCi/m^2-hr-mm Hg. This parameter has to be determined experimentally for different building materials. Only a few values of α for concrete walls have been reported in the literature [42, 45-47]. Future experimental measurements of α for building materials other than concrete are required in order to use this model. In this work, α is taken to have a value of 18 pCi/m^2-h-mm Hg for concrete walls as has been reported by Jonassen [47].

Now, assuming that exhalation rate at normal (constant) atmospheric pressure is E_o and that the time rate of change of pressure variations is $\dot{p}$, then the integration of dE over the duration time (DT) of pressure variations yields the following result:

$$E = E_o - \alpha \int_{DT} \dot{p}\, dt.$$

According to this result, the exhalation rate E depends not only on the pressure-independent exhalation rate E_o but also on the functional behavior of the time rate of change of pressure variations. This functional behavior could be chosen to represent real pressure fronts.

At this point it is important to indicate that the basic difference between the present model and other reported models [60, 81, 82] concerned with the study of indoor radon is in the use of pressure dependent exhalation rate and in the use of wall removal interactions. The significance of pressure effect on exhalation rate is well documented in the literature, while the significance of wall removal processes requires further experimental verification.

Calculation of Radon Daughters Concentration

The importance of calculating the concentration of radon daughters in an environment can be attributed to the fact that health effects of elevated concentrations of radon in air are due not to the radon itself but to its short-lived alpha-emitting daughters which are deposited on the interior surfaces of the respiratory tract. The radiation dose to the basal cells of the bronchial epithelium is a function not only of the concentration of radon in air but also the degree of radioactive equilibrium between radon and its first four decay products. Radioactive equilibrium between radon and its daughters is expressed as the ratio of the concentration of various daughters to that of the parent radon concentration.

Accumulation of radon daughters in the atmosphere of inhabited areas is due to the physical decay of indoor radon and also due to contributions from outdoor atmosphere through the process of air exchange. Removal of radon daughters, however, is due to their radioactive decay, air exchange, and attachment to surfaces and aerosol particles. The removal of daughters by radioactive decay is controlled by their distinct decay coefficients (see radon decay scheme in

Chapter II). The effect of indoor-outdoor air-exchange process on radon daughters concentration follows the same rule discussed before in calculating the indoor radon concentration.

Radon daughters could exist in an atmosphere as free particles (i.e., not attached to aerosol particles) and/or attached particles (i.e., attached to aerosol particles). Both fractions could be removed by sedimentation and/or by deposition on walls and other indoor surfaces (e.g., furniture). Accordingly, the process of daughters' attachment to walls and aerosols is of significant importance in studying their behavior in the atmospheres of inhabited areas. The process of daughters' attachment to aerosol particles depends on the diffusion coefficient of the free daughters and also on the particle size distribution of the aerosol in the considered volume [60, 81, 83]. On the other hand, the deposition of daughters (free or attached) on walls and other indoor surfaces depends on the size and geometry of the region under consideration (i.e., on surface/volume) and also on the deposition velocity of radon daughters [81, 83].

Many experimental studies concerning the attachment of radon daughters to aerosol particles have been reported. Wilkening [84] showed experimentally that radon daughters were attached to submicron-size aerosols with mean diameters of 0.009 and 0.018 micrometer. Another experimental investigation of the interactions that occur between radon daughters and aerosols was reported by Raabe [83] who found that the activity is proportional to the surface area of the particles with the majority of concentration being associated with particles less than 0.1 micrometer diameter. On the other hand calculated values of radon daughters attachment rates were reported by

Jacobi [81]. In these calculations Jacobi used Lassen's semi-empirical formula (cited in Jacobi's paper) to calculate the coagulation constant of the daughters. According to these calculations, an attachment rate of the order of 40 per hour could be expected for normal particle size distributions of atmospheric aerosol in the atmosphere near the ground where aerosol concentration is of the order of 10^4 particles per cm^3. In similar calculations reported by Porstendörfer [60] an attachment rate of the order of 86 per hour was used in studying the behavior of indoor free radon daughters.

Removal of radon daughters due to wall deposition has been studied by few investigators. Wrenn et al. [85] used the diffusion theory to calculate the wall deposition of free radon daughters due to pure molecular diffusion in a cylindrical volume of static, clean air. These results showed a strong influence of wall deposition on the degree of disequilibrium between radon and its daughters. On the other hand, Jacobi [81] used the following formula to calculate the wall deposition rate constants for either the free or the attached fractions.

$$\lambda_{d,i} \simeq U_{d,i} \; S/V,$$

where

$\lambda_{d,i}$ = wall deposition rate constant (1/hr) for attached (i = 1) or for free (i = 2) fractions,

$U_{d,i}$ = deposition velocity characteristic of each fraction (meter/hr),

S/V = surface area to volume ratio (1/meter).

A wall deposition rate of 0.7 per hour for attached daughters was calculated by Jacobi on the basis of a deposition velocity of the order

of 0.01 cm/sec for particles of 0.05-0.2 micrometer diameter at an air velocity of 10 cm/sec and for a surface-to-volume ratio of 2 meter^{-1}. At a size range of 0.1-0.2 micrometer, attached daughters deposition velocities of the order of 0.001 cm/sec were measured by Clough [86]. These last results were used by Porstendörfer [60] in evaluating wall deposition rates of attached daughters.

To calculate wall deposition rates of free radon daughters, it is necessary to know the deposition velocity of free daughters. In this regard Jacobi [81] assumed that the ratio of the deposition velocity of free ions to that of attached ions is equal to the square root of the ratio of their respective diffusion coefficients. On the basis of this formulation, for a deposition velocity of attached daughters of the order of 0.01 cm/sec, the corresponding deposition velocity of free daughters is within the range of 0.5-1.5 cm/sec. On the other hand, Möller and Schumann [87] measured experimentally the deposition velocities of different particles using a wind tunnel with wind speeds of 3 through 8 meter/sec. Their results were best fitted by a relation in which the deposition velocity is proportional to the diffusion coefficient to the two-thirds power. Wall deposition rate of 100 per hour for free radon daughters was used by Porstendörfer et al. [60] in their investigation of indoor radon daughters.

The previous discussion outlined the important factors which could affect the concentration of radon daughters in various atmospheres. Two approaches have been used in assessing the behavior of radon daughters in certain atmospheres. In the first approach, the removal of radon daughters is achieved by radioactive decay as well as by air-exchange process [13, 57, 59]. This approach may be useful in situations where

the effect of air exchange on the concentration of radon daughters dominates all other removal processes (i.e., in cases where air-exchange rates are relatively high). In the second approach (such as that of Jacobi and Porstendörfer) radon daughters are divided into two fractions (attached and free) and each fraction is treated separately according to its distinct properties. In this approach, free radon daughters are removed by radioactive decay, air exchange, wall deposition and attachment to aerosol particles while attached daughters are removed by the same processes (with different rates) but not including attachment to aerosol particles. In models of such kind the characteristic removal processes (with different rates) but not including attachment to aerosol particles. In models of such kind the characteristic removal processes (except radioactive decay) of each fraction are considered to be the same for all daughters. In the development of these models each fraction is calculated separately and then the disequilibrium fraction of each daughter is calculated by summing up its two individual fractions.

Experimental studies conducted by Jonassen [45] showed that the removal of radon daughters due to filtration and diffusion cannot be described by a single removal coefficient. These results also showed that in an unventilated room radium-A was removed at a higher rate than radium-B or radium-C. Moreover, the same results showed that the removal rate of radium-B is not the same as that of radium-C. Although these results may be characteristic of the place and the conditions where they were measured, they suggest the existence of some differences in the removal of various radon daughters due to filtration and diffusion. Taking this suggestion into consideration, it seems reasonable to assume that each radon daughter has its own wall removal rate with no

distinction between free and attached fraction. In other words, this approach represents the gross end results of the models which are based on the second approach as discussed before.

In the present development it is assumed that radon daughters are removed by radioactive decay, air exchange, and by wall deposition. The mathematical form of daughters removal by radioactive decay and by air exchange is similar to that of radon with the appropriate decay coefficients to be used.

To calculate wall deposition rates for various radon daughters, it is necessary to set up certain criteria regarding the deposition velocities of daughters. This can be achieved by introducing what could be called "characteristic deposition length." The principle of a characteristic length is well known in problems associated with turbulent boundary layers. Landau and Levich [88] indicated that the characteristic length in the turbulent boundary layer is proportional to the deposition velocity. Accordingly, it seems possible to correlate between the deposition velocities of radon daughters by using this previous assumption. Assuming that radon daughters possess only one characteristic length, then the relation between the various deposition velocities can be represented by the following expression:

$$T_i \times U_{d,i} = L, \qquad i = 1,2,3,$$

where

T_i = half-life time of ith daughter,

$U_{d,i}$ = deposition velocity of ith daughter.

The previous expression will require the specification of only one deposition velocity from which the other pair can be evaluated. It is important to indicate that the accuracy obtained from the previous

expression depends on the choice of the reference deposition velocity of a certain daughter. Although it is difficult to assign an exact deposition velocity to a certain daughter because of the lack of published information yet the results of Möller and Schumann [87] suggest that radium-B is a good choice. Accordingly, in the following development the deposition velocity of radium-B will be taken as the reference velocity. Using the expression for the characteristic length, then it is possible to calculate the wall deposition rates using the following formula:

$$\lambda_{d,i} = S/V \times U_{d,i},$$

where

$\lambda_{d,i}$ = wall deposition rate (1/hr),

S/V = surface-to-volume ratio as has been defined before (1/meter).

Combining all the information and using the laws of conservation, the following differential equation can be written to represent the time rate of change of the ith radon daughter concentration in a room of surface area S and of volume V.

$$\frac{dC_i}{dt} = \lambda_i C_{i-1} + \lambda_v C_i^o - (\lambda_i + \lambda_v + \lambda_{d,i}) C_i.$$

In this expression λ_i, λ_v, and $\lambda_{d,i}$ are the same as has been described before. Also in this expression C_o (for i = 1) is equal to C_{Rn} which is the concentration of indoor radon gas.

Analytical solutions to the two differential equations describing the behavior of indoor radon and its daughters can be easily obtained if the outdoor concentrations are constant. However, it is difficult to find an analytical solution to these differential equations if outdoor concentrations are time dependent. Accordingly, no effort was made to

solve these equations analytically. Instead numerical solutions were obtained by utilizing the famous Runge-Kutta method of numerical integration. This method is discussed in almost every numerical analysis textbook. A computer program based on Runge-Kutta numerical integration method was developed and was used in calculating the theoretical results.

CHAPTER V

RESULTS AND DISCUSSION

This chapter is concerned with the results and discussion of some experimental measurements conducted in a building (Building 1), located in a small industrial park in southwestern Pennsylvania. This is the site of a facility once used under contract by the U.S. Government for the extraction of uranium from ores. Results of air-exchange and radon flux measurements are presented in this chapter. The results of radon concentration as well as daughter concentrations are also given. In addition, theoretical results obtained from the mathematical model presented in Chapter IV are given here. Results are presented in the form of tables and/or graphs followed by appropriate discussions.

Results of Air-Exchange Measurements

Air-exchange rates were measured at various intervals over a period of five days starting on July 19, 1979. The intervals of measurements were chosen to cover as many periods of the day (e.g., morning, evening) as possible because it was impractical to conduct such measurements over the entire 24-hr period.

Analysis of the experimental data is based on the assumption that the concentration of the tracer gas (CO_2) used in the measurements decays exponentially with time after thorough mixing in room air has occurred. Also, it is assumed that the air-exchange rate is constant during the period of measurement. The natural logarithm of the measured concentrations of the tracer gas were plotted against time and fitted to

a straight line whose slope gives an air-exchange rate which is characteristic of the place where measurements are taken. Typical results obtained using this particular measuring technique are presented in Figures 6-8. An example of results of air-exchange rate measurements in room 6B are presented in Figure 6. At this particular time, the air-exchange rate was found to be 1.4 air changes per hour. Similarily in Figures 7 and 8, results are presented of the air-exchange measurements taken in the west and east rooms of the ground floor of Building 1. The corresponding air-exchange rates were found to be 5.4 and 0.75 air changes per hour, respectively. There is no particular reason for choosing only these three figures except to show the behavior of the tracer gas at different air-exchange rates. In Tables 2 and 3, the results of air-exchange measurements over the entire period of this investigation are presented. From these tables it can be seen that air-exchange rates inside the experimental building vary over a wide range with a maximum of $\sim$6 air exchanges per hour to a minimum of $\sim$0.4 air exchanges per hour. High air-exchange rates were observed on windy days. Lower air-exchange rates, however, were obtained by closing and sealing windows and doors of the building for a period of 24 hours. During these experiments, it was observed that outdoor wind conditions were the most significant factor affecting the air-exchange rates in the building. This is mainly because the building is not furnished with mechanical ventilation devices and also because the building is fitted with numerous large-sized windows and doors which were kept open most of the time when measurements were taken.

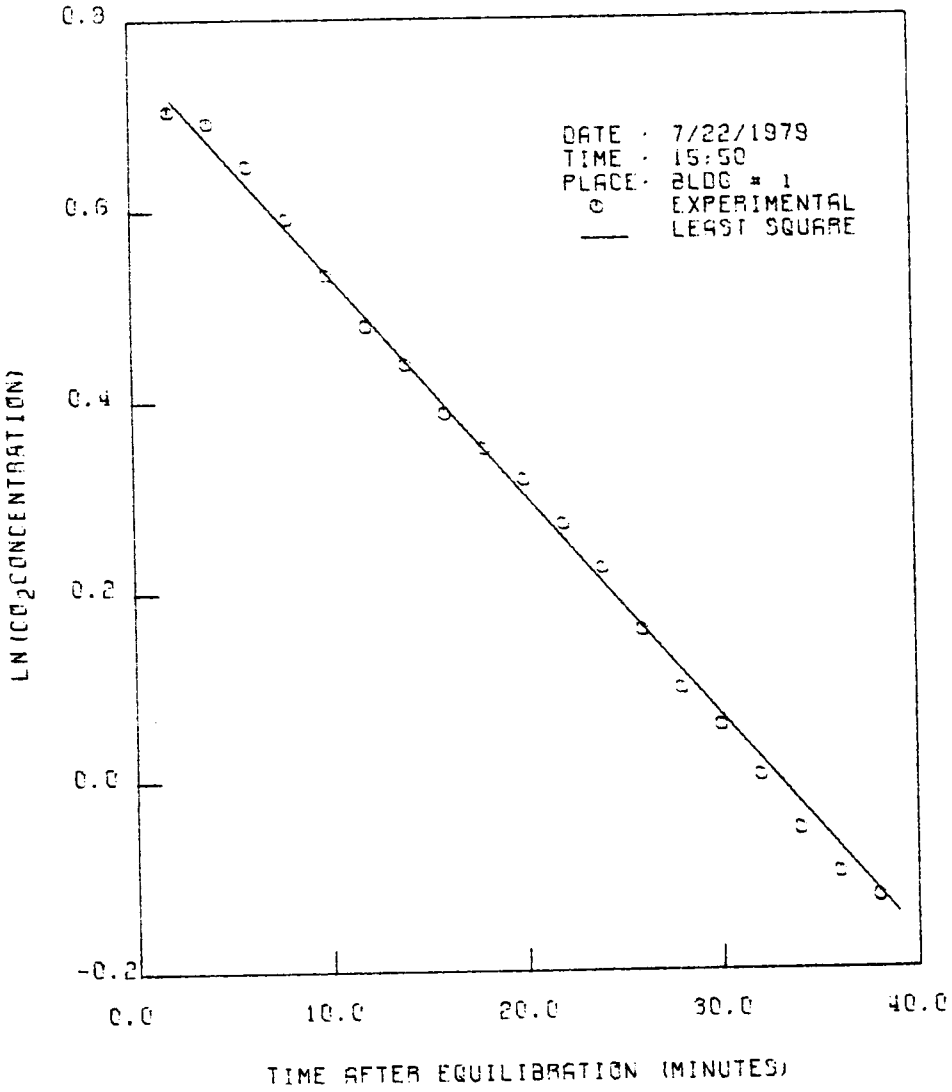

Figure 6. Variation of tracer gas (CO_2) concentration with time measured in Room 6B using the MIRAN-80 infrared spectrometer.

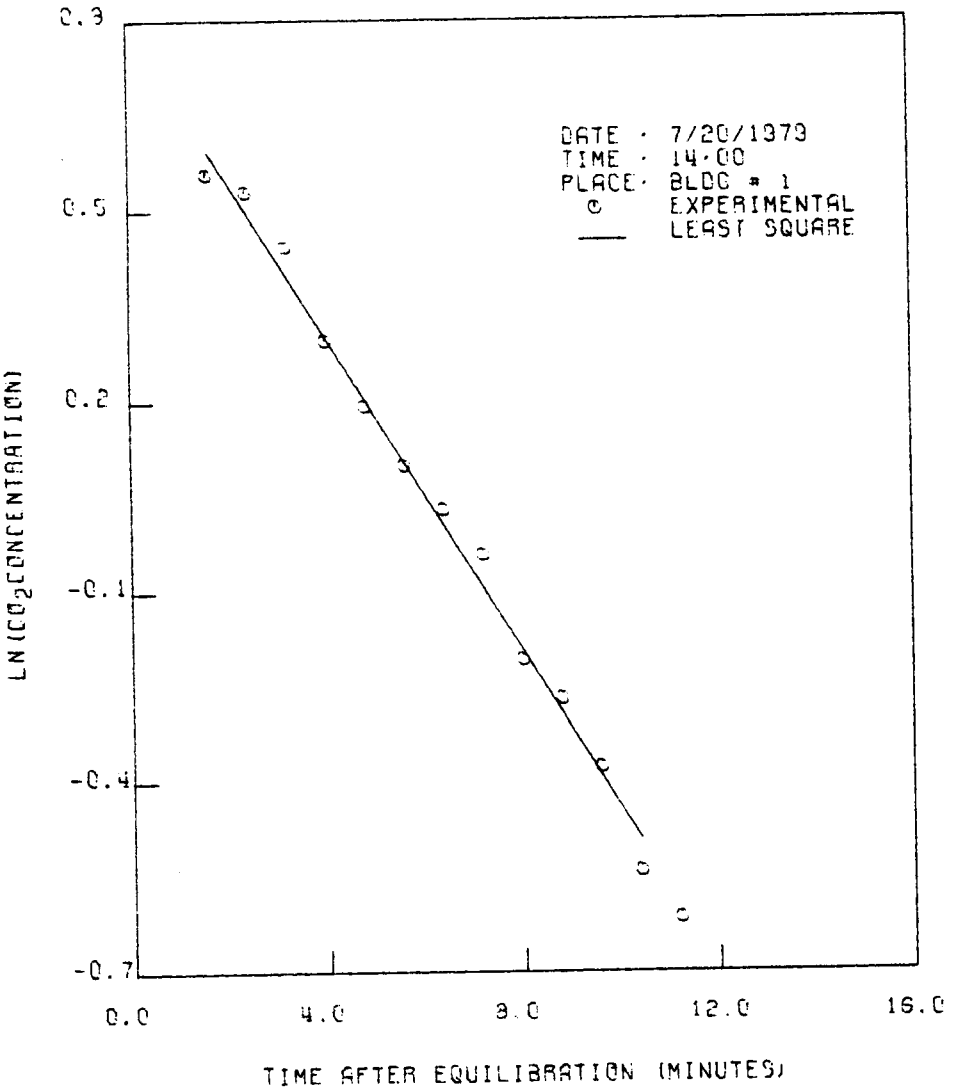

Figure 7. Variation of tracer gas (CO_2) concentration with time measured in the west room using the MIRAN-80 infrared spectrometer.

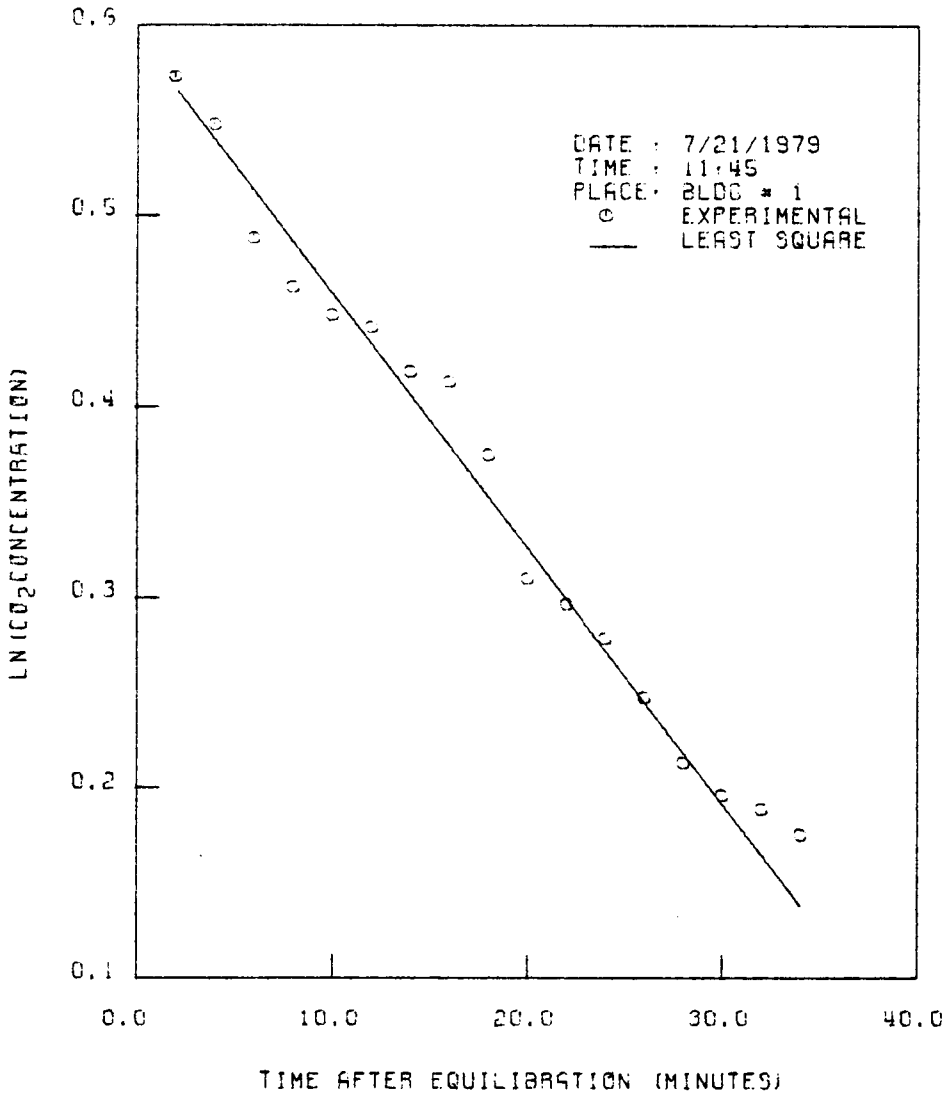

Figure 8. Variation of tracer gas (CO_2) concentration measured in the east room using the MIRAN-80 infrared spectrometer.

TABLE 2

MEASURED AIR-EXCHANGE RATES IN THE GROUND FLOOR
EAST ROOM OF THE EXPERIMENTAL BUILDING
DURING JULY 1979

Date	Time	Air-exchange rate (1/hr)
7/19	21:00	3.0
7/20	14:00	5.4
	22:00	2.8
7/21	4:00	1.9
	6:30	1.0
	12:00	0.8
7/22	18:00	0.4
7/23	16:40	4.5
	19:20	6.1

TABLE 3

MEASURED AIR-EXCHANGE RATES IN THE GROUND FLOOR
WEST ROOM OF THE EXPERIMENTAL BUILDING
DURING JULY 1979

Date	Time	Air-exchange rate (1/hr)
7/19	22:00	3.1
7/20	14:00	5.4
	22:00	3.6
7/21	4:30	2.0
	6:00	2.2
	8:00	1.4
	12:00	0.8
7/22	18:00	0.4
7/23	18:05	3.0

Results of Radon Flux Measurements

Radon flux was measured on the ground level of the experimental building during July 1979 and continued for four days. During those four days, three complete sets of measurements were taken. Flux detectors (canisters) were exposed for approximately 24 hours to radon flux emanating from different indoor radon sources. Appropriate flux values were then calculated on the basis of the discussions presented in Chapter III. Locations at which radon flux was measured were distributed across the area of the ground level in such a way that representative results for the flux distribution inside the building could be collected. The west room (see Figure 9) took more attention than other places inside the building because more potential radon sources (cracks and drains) are concentrated in that room. Locations at which flux measurements were taken are shown in Figure 9. In this figure the letter S refers to locations where flux emanating from concrete floor slabs was measured. Also the letter C refers to locations where flux emanating from cracks was measured. The letter D refers to locations where flux emanating from drains was measured. The concrete floor slabs in this building are uncovered and unpainted. The surface area of the concrete slabs in the east room was found to be 132 square meters while that of the west room was found to be 139 square meters. On the other hand, the length of visible cracks were measured in both rooms, and it was found that the total length of cracks in the east room is approximately 40 meters while that in the west room is approximately 32 meters. Moreover, 18 observable drains (plugged and unplugged) were counted in both rooms with 14 of them located in the west room. Among those drains, there are eight unplugged (open) drains. Seven of these are located in the west room and one is in the east room.

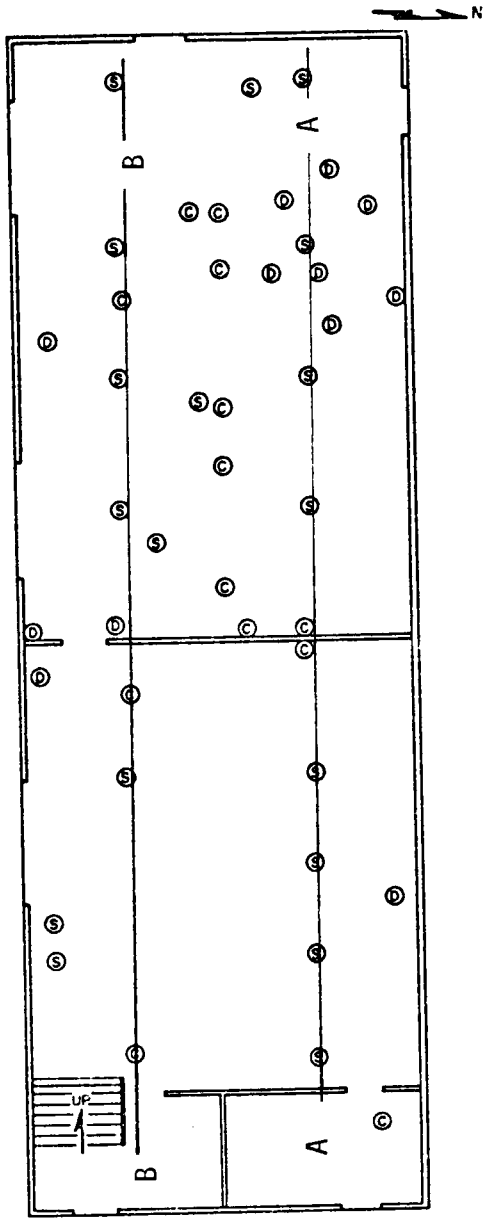

Figure 9. Locations of flux measurements in the ground level of Building 1.

Radon flux emanating from concrete slabs was measured at different locations in each room and was averaged over the area of the specific room. The total radon influx from slabs in each room can thus be calculated by multiplying this average flux by the area of the room. Similarily, the radon flux emanating from cracks in each room was averaged and normalized to a crack length of one meter. The total influx of radon from cracks in each room is obtained by multiplying this average flux by the total length of cracks in each room. The total influx from drains was calculated by adding the contributions of all drains. It is necessary to mention that during the July 1979 measurements, the technique used to measure radon flux emanating from drains failed to give reasonable results. For this reason, another set of measurements was taken during November 1979. These measurements employed a new technique of an open-end double-canister described in Chapter III. This was used successfully to measure the radon flux emanating from drains. A correlation factor between the two sets of measurements was obtained by repeating some of the measurements at the same locations as those of July measurements.

The averaging process used to find the total radon influx either from cracks or from slabs is necessary in field measurements of this kind. This is because it is practically impossible to cover the entire floor surface area with flux detectors (canisters) and also because the concentration of radium-226 in soil beneath the concrete floor is not distributed homogeneously as can be seen in Figures 10 and 11. The variations of radon flux from slabs as a function of the distance from the east wall of the building and along the line AA is shown in Figure 10. It can be seen from this figure that radon flux emanating from concrete

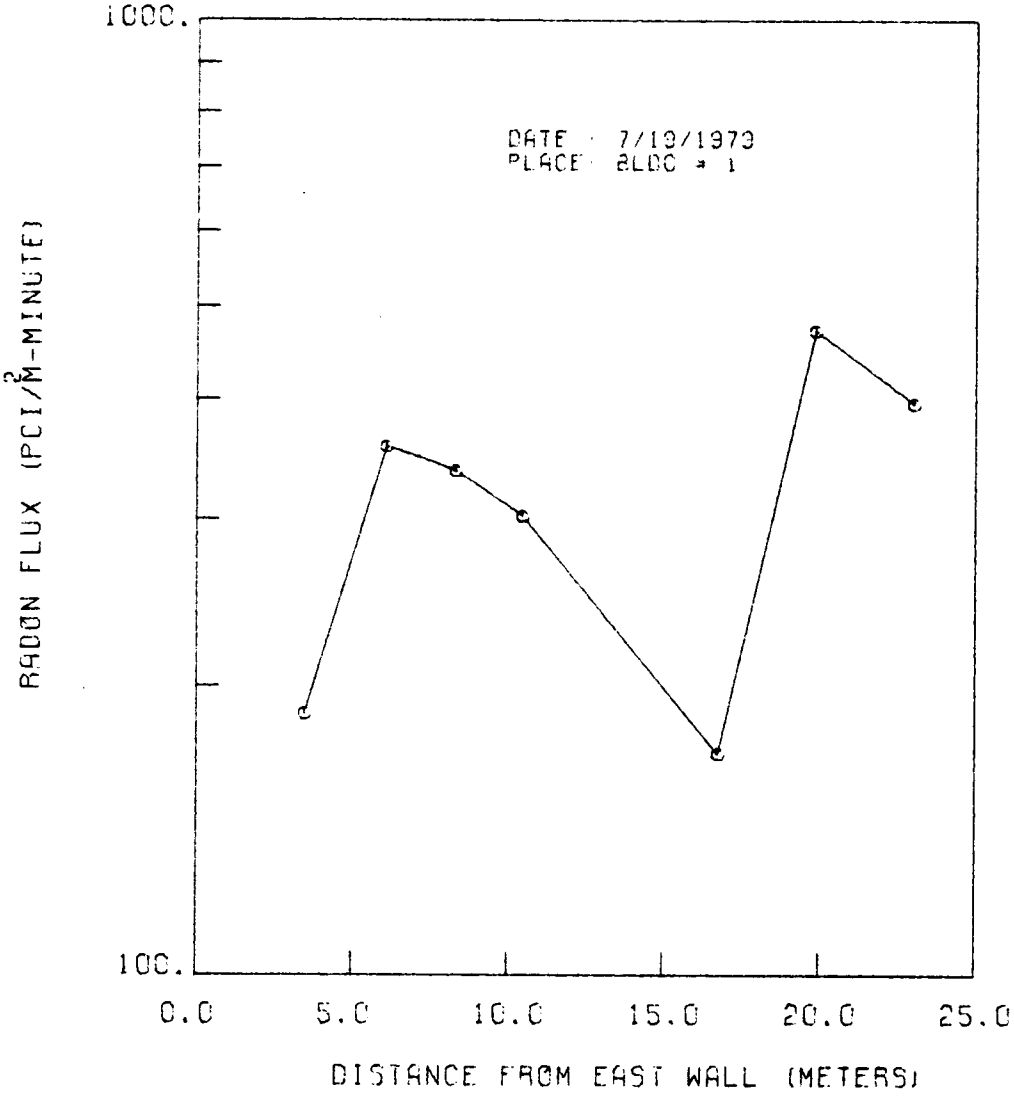

Figure 10. Measured radon flux emanating from the concrete slabs of Building 1 (along AA) as a function of the distance from the east wall of the building.

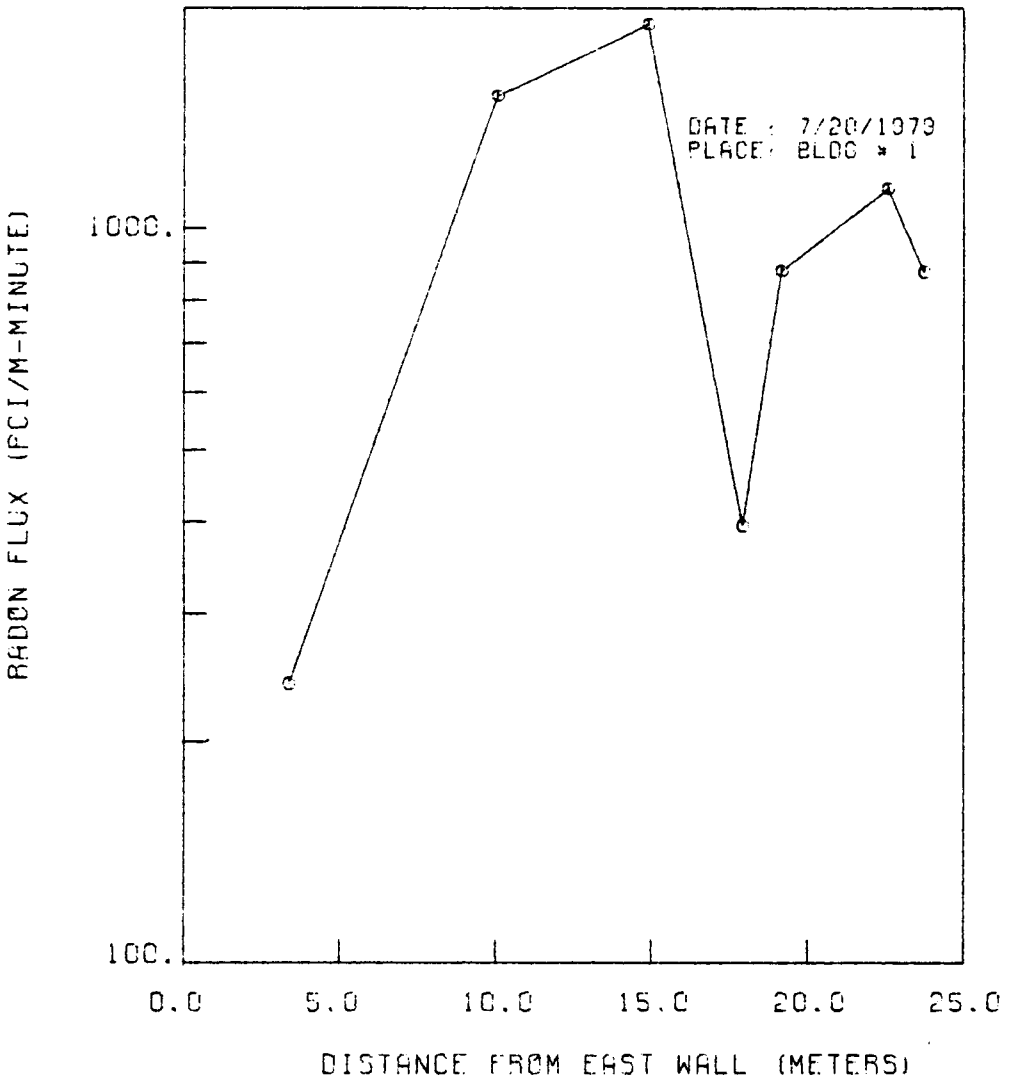

Figure 11. Measured radon flux emanating from visible cracks in the concrete floor of Building 1 (along BB) as a function of the distance from the east wall of the building.

floor slabs varies in an irregular manner. It is important to mention that the radon flux measurements shown in Figure 10 were all obtained at the same time; therefore, the effect of atmospheric variables was a common factor among them. On the other hand, the variation in radon flux emanating from cracks in the floor as a function of the distance from the east wall along the line BB is shown in Figure 11. Also seen in this figure is that the radon flux from cracks varies in an irregular manner across the entire length of the building.

The cumulative results of radon influx measurements for various indoor sources in both the east and west rooms are summarized in Tables 4 and 5. Both tables show the period during which flux was measured as well as the contribution from each individual source. The total radon influx in each room (last row in tables) represents the sum of all contributing sources during each period. From Table 4 it can be seen that the radon influx from concrete slabs and from cracks in the floor contribute the most to the total influx in the east room. Also, it can be seen from this table that the total radon influx in the east room is not constant but varies with time. On the other hand, Table 5 shows the same type of result with the exception that the total radon influx in the west room is much higher than that in the east room.

It is important to mention that the Atomic Energy Commission (AEC) initiated a program in 1974 for determination of the current radiological condition of sites formerly utilized under contract by the Manhattan Engineer District (MED) and the AEC for work involving the handling of radioactive materials. Based upon the findings of radiological investigations of these sites, measures will be undertaken to assure the public health and safety. The experimental building where

TABLE 4

MEASURED AVERAGE RADON INFLUX (pCi/Minute) FROM SLABS, CRACKS, AND DRAINS IN THE EAST ROOM

Source/Day	7/19-7/20	7/20-7/21	7/21-7/22
Slabs	38680.0	No data	42535.0
Cracks	25795.0	No data	18880.0
Drains	1326.0	No data	1687.0
Total	65801.0	No data	63102.0

TABLE 5

MEASURED AVERAGE RADON INFLUX (pCi/Minute) FROM SLABS, CRACKS, AND DRAINS IN THE WEST ROOM

Source/Day	7/19-7/20	7/20-7/21	7/21-7/22
Slabs	59775.0	46925.0	38266.0
Cracks	34240.0	33078.0	31552.0
Drains	2760.0	2667.0	2543.0
Total	96775.0	82670.0	72361.0

radon flux measurements were taken is among those sites being considered for remedial actions. Accordingly, these experimental findings will be of significant importance in determining what remedial action could be taken.

Results of Radon Concentration Measurements

Radon concentrations inside the experimental building were measured continuously for five consecutive days starting on July 19, 1979. The measurements were taken downstairs in the east and west rooms and also in rooms 4A and 6B upstairs. On the other hand, outdoor radon concentration was measured outside the building across from the north wall of the west room. During the period when these measurements were taken, surface weather data was obtained from the National Weather Service Station located at Pittsburgh International Airport.

The experimental results of radon measurements inside and outside the experimental building are shown in Figures 12-16. These figures also show the variation of the atmospheric pressure with time during the period of collecting the data. The variations in outdoor radon concentration as a function of time are shown in Figure 12. It can be seen from this figure that outdoor radon concentration at this particular location is low enough to be neglected in relation to indoor radon concentrations. On the other hand, Figures 13 and 14 show the variations in indoor radon concentration with time as measured in the east and west rooms, respectively. Variations in radon concentrations with time measured in rooms 4A and 6B (upstairs) are shown in Figures 15 and 16.

The results of the measurements of radon concentration inside the building show that the concentration of radon in the west room is the

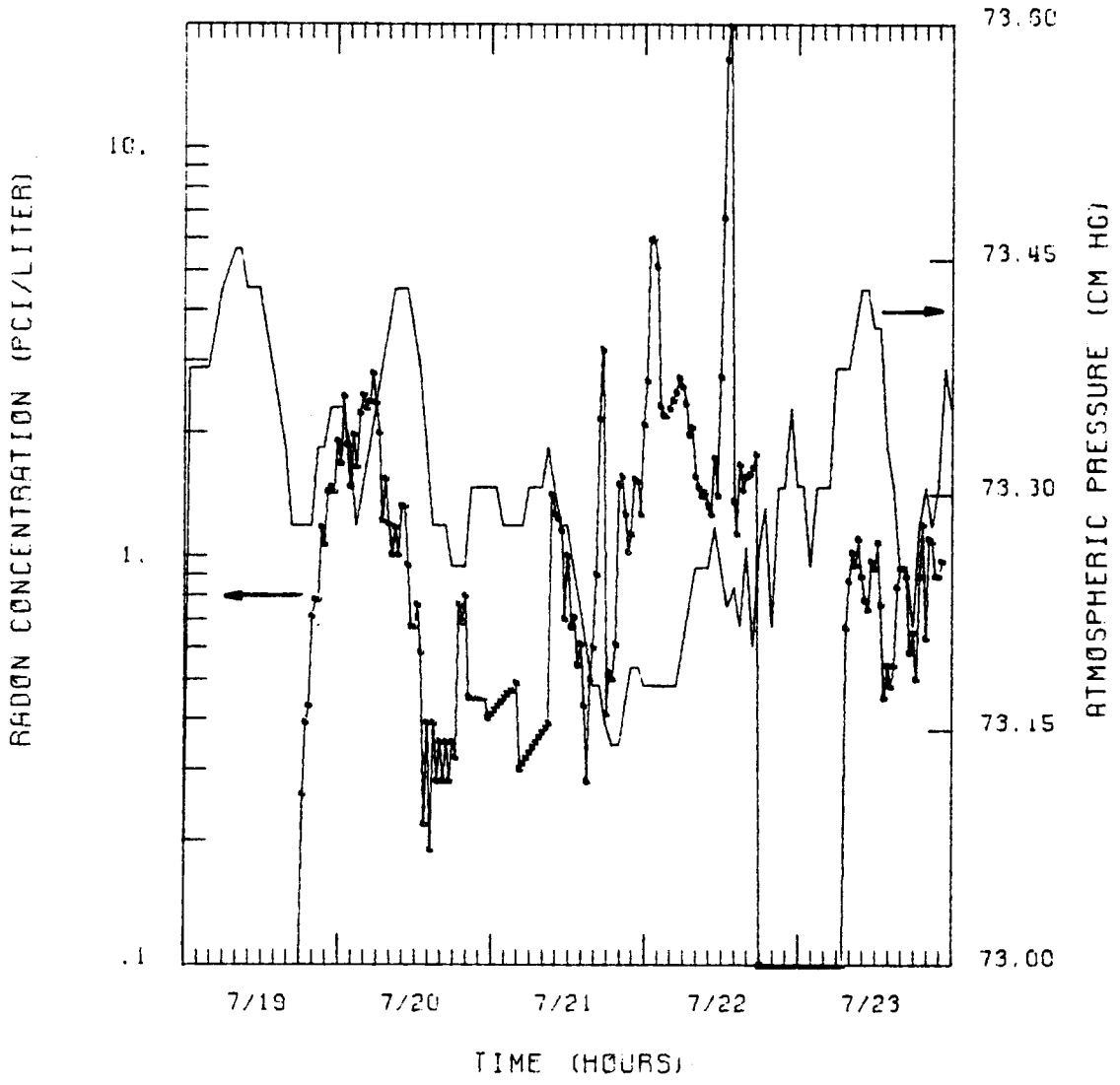

Figure 12. Variation of radon concentration with time measured outdoors near experimental building for five days during July 1979 plotted together with the corresponding variation of atmospheric pressure.

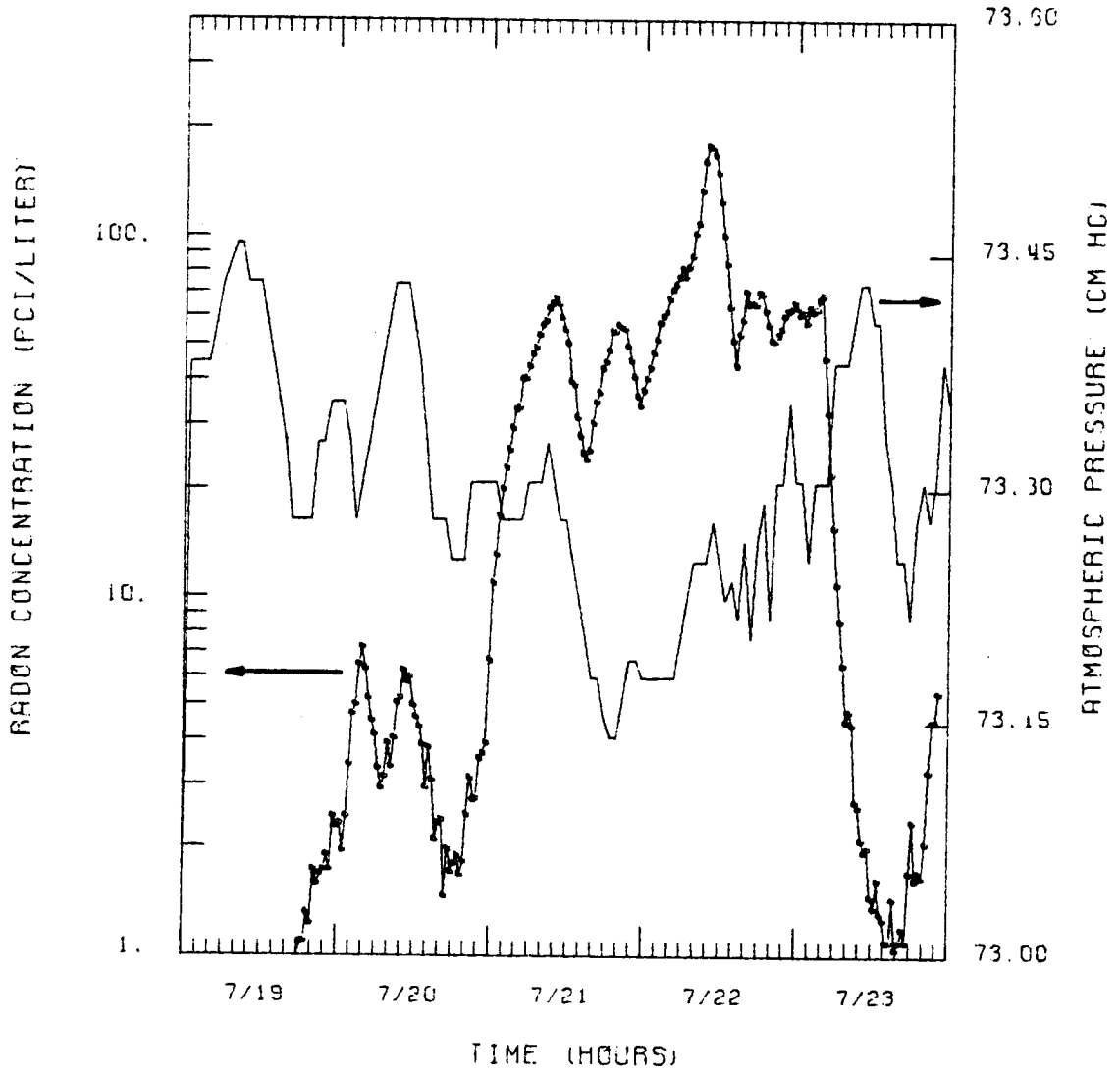

Figure 13. Variation of radon concentration with time measured in the east room for five days during July 1979 plotted together with the corresponding variations of atmospheric pressure.

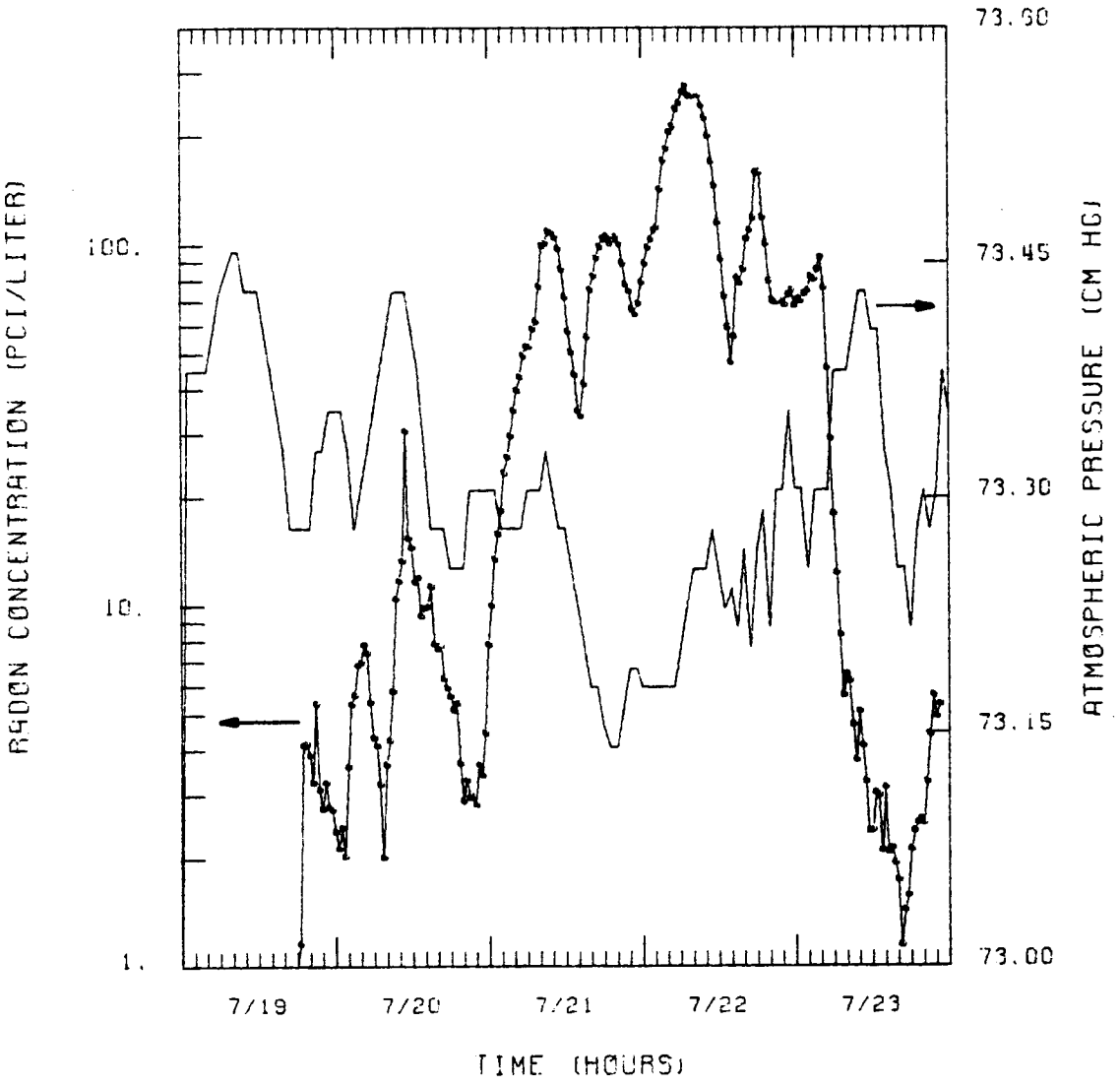

Figure 14. Variation of radon concentration with time measured in the west room for five days during July 1979 plotted together with the corresponding variations of atmospheric pressure.

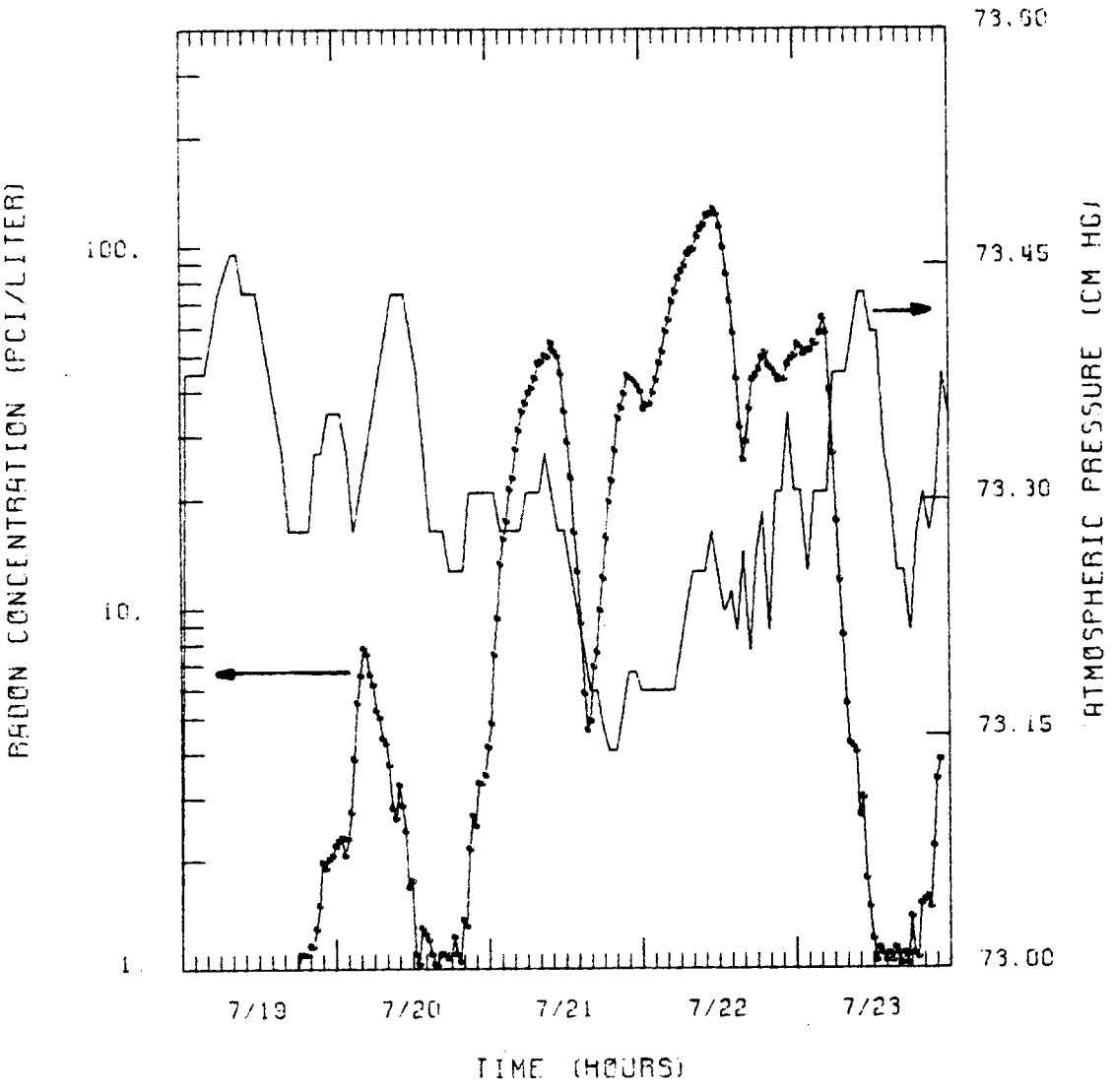

Figure 15. Variation of radon concentration with time measured in Room 4A for five days during July 1979 plotted together with the corresponding variations in atmospheric pressure.

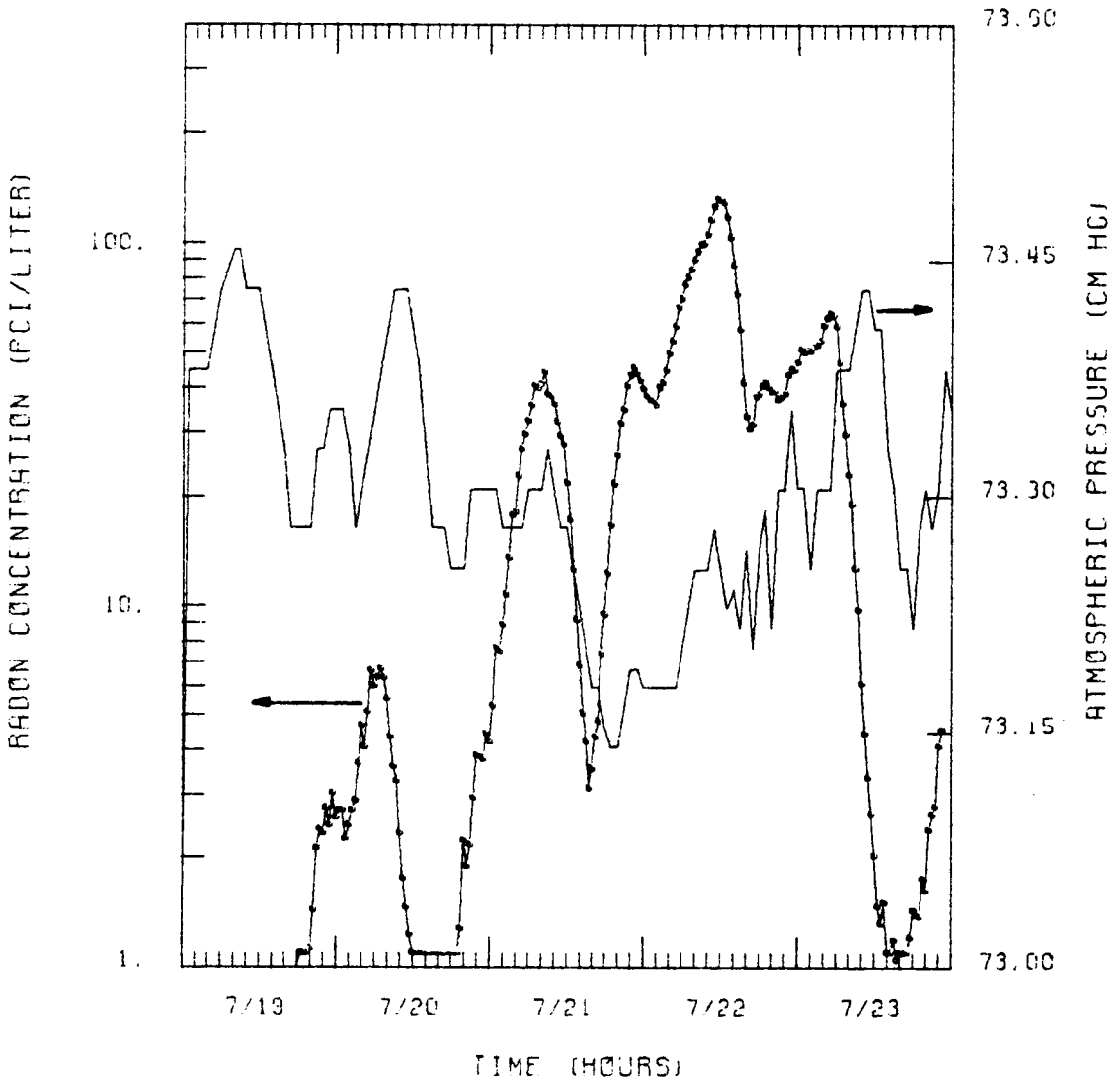

Figure 16. Variation of radon concentration with time measured in Room 6B for five days during July 1979 plotted together with the corresponding variations in atmospheric pressure.

highest among all places in the building. This can be attributed to the fact that most of the indoor radon sources are located in that room (see Table 6). Moreover, these results also show the effect of variations in atmospheric pressure on radon exhalation process. From Figures 13-16 it can be seen that the pressure drop which occurred on July 21 and continued through July 22 caused an observable increase in the concentration of indoor radon. Unfortunately, it is not possible with the data at hand to evaluate quantitatively the effect of variations in atmospheric pressure on radon exhalation rates. One main reason is that the building during those two days (7/21, 7/22) was closed so that lower air-exchange rates could be measured. Accordingly, the observed increase in the concentration of indoor radon during those two days is associated with the drop in the atmospheric pressure as well as with the reduction of air-exchange rates. There is a slight time lag between the time of the occurrence of the pressure drop and the time of recording the maximum concentration; yet these results still demonstrate the effect of pressure variations on the accumulation of indoor radon. At this point it is important to indicate that the previous argument may contradict the results of total radon influx measurements (Tables 4 and 5). The total radon influx was highest when the observed radon concentration was near its lowest value. This contradiction can be removed if the effect of air-exchange rates on radon exhalation rate is considered. At higher air-exchange rates radon is removed quickly from the building and accordingly a concentration gradient across the floor is created which enhances the diffusion of radon from the floor to the room air. This will cause an increase in the amount of the exhaled radon while at the same time the air-exchange process will cause a

TABLE 6

CONCENTRATION OF RADON AND ITS DAUGHTERS (pCi/Liter)
MEASURED IN THE EAST ROOM DURING JULY 1979

	Nuclei			
Day	Rn	RaA	RaB	RaC
July 19		No data		
July 20	4.0	1.1	0.42	0.17
	2.3	1.7	0.32	0.06
July 21	65.0	44.4	28.3	23.3
July 22	64.0	24.1	16.4	13.0
	135.0	61.1	36.1	27.0
July 23	1.3	0.08	0	0

reduction in the observed radon concentration. This explanation agrees with the idea that radon exhalation rate increases slowly as air-exchange rate increases [81]. Moreover, the drop in the observed radon concentration during the period July 21 to July 22 can be ascribed to the effect of higher air-exchange rate as well as to the effect of increasing atmospheric pressure.

On the other hand, the results of radon measurements in rooms 4A and 6B (Figures 15 and 16) illustrate an important phenomenon which can be called the inverse effect of air-exchange process on indoor radon concentration. In most cases outdoor radon concentration is less than indoor radon concentration and accordingly the air-exchange process becomes a diluting process where air low in radon is brought into the indoor atmosphere and at the same time air rich in radon is taken out of the indoor atmosphere. Accordingly, it is expected that indoor radon concentrations will decrease as air-exchange rate increases. This can be seen from Figure 17 where average radon concentrations measured in the east and in the west rooms are plotted against the corresponding measured air-exchange rates. This figure shows clearly that reduction in air-exchange rate causes a significant increase in the indoor radon concentration. This increase in concentration depends to some extent on the strength of the radon source [60]. The effect of the source strength can also be seen from Figure 17 where the rate of decrease in radon concentration with air-exchange rate is faster in the east room (with low radon influx) than in the west room (with high radon influx). This situation can be reversed whenever outdoor radon concentration becomes higher than indoor concentration which is the case of rooms 4A and 6B. Those two rooms have negligible radon sources, and thus any radon

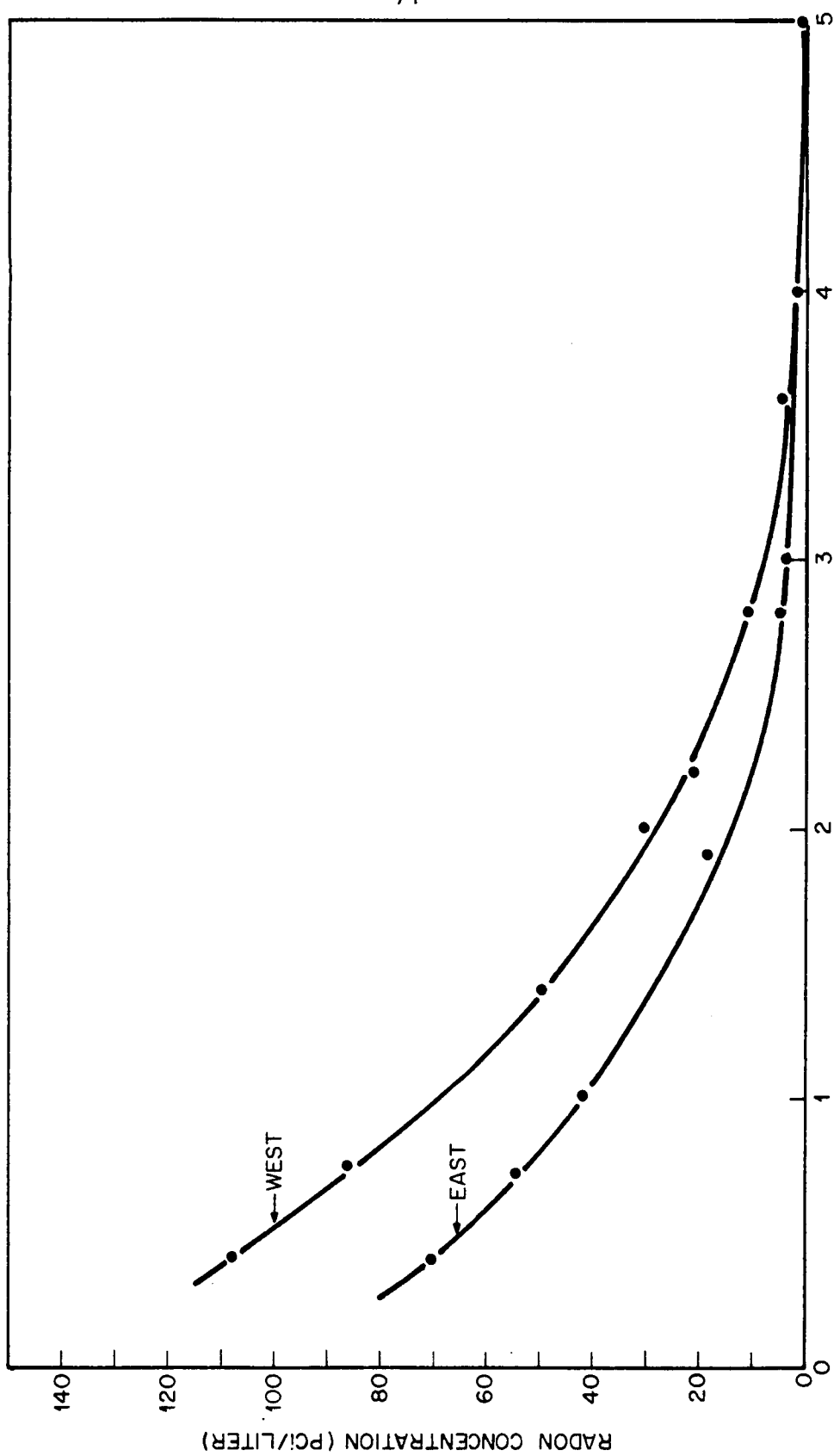

Figure 17. Effect of air-exchange rates on radon concentrations measured in the east and west rooms during July 1979.

accumulation inside them has to be supplied by some outside sources which in the present case could be those located in the ground level of the building. From Figure 12 it can be concluded that outdoor radon concentration cannot contribute significantly to the radon concentration inside the building. Thus the accumulation of radon in rooms 4A and 6B is due mainly to the exchange of air between the ground level which is rich in radon and that of the upper level which contains less radon. The confirmation of this argument is obvious from the comparison of the variation patterns of radon concentration in both the ground and upper levels of the building (Figures 13-16). Unfortunately there is not enough data on the air-exchange rates in rooms 4A and 6B which permit further analysis of this phenomenon. Nevertheless these data provide strong evidence of the importance of air-exchange processes on the accumulation of radon in the atmospheres of inhabited areas.

Results of Radon Daughter Measurements

Radon daughter concentrations (RaA, RaB, and RaC) were measured inside the building at the same locations where radon concentrations were measured. The techniques of reducing the experimental results have been described in Chapter III. Results of radon daughter measurements are given in Tables 6-9. These tables show the concentration of radon and its daughters at the time when measurements were taken.

The results show that the concentrations of radon daughters inside the building vary over a wide range. In the ground floor west room, the highest daughter concentration was observed on the early morning of July 21, 1979. This was associated with an elevated radon exhalation from one of the drains in the west room. This elevated exhalation

TABLE 7

CONCENTRATION OF RADON AND ITS DAUGHTERS (pCi/Liter)
MEASURED IN THE WEST ROOM DURING JULY 1979

	Nuclei			
Day	Rn	RaA	RaB	RaC
July 19	3.2	1.13	0.55	0.36
July 20	16.0	6.2	1.21	0.59
	3.1	1.24	0.41	0.21
July 21	115.0	81.0	34.3	48.6
July 22	58.0	23.8	13.8	10.6
	118.0	48.9	22.0	13.4
July 23	2.2	0.77	0.12	0.11

TABLE 8

CONCENTRATION OF RADON AND ITS DAUGHTERS (pCi/Liter)
MEASURED IN ROOM 4A DURING JULY 1979

	Nuclei			
Day	Rn	RaA	RaB	RaC
July 19		No data		
July 20	1.5	1.23	0.32	0.3
	1.0	0.70	0.22	0.22
July 21	47.0	27.3	18.2	16.0
July 22	85.0	59.5	42.6	38.8
	33.0	19.7	19.7	10.0
July 23	1.0	0.36	0.0	0.0

TABLE 9

CONCENTRATION OF RADON AND ITS DAUGHTERS (pCi/Liter)
MEASURED IN ROOM 6B DURING JULY 1979

	Nuclei			
Day	Rn	RaA	RaB	RaC
July 19	3.4	2.7	1.1	0.8
July 20	3.1	1.64	0.74	0.6
	2.9	4.74	1.9	1.05
July 21	34.7	23.1	19.7	16.7
July 22	109.0	53.8	44.3	42.0
	35.0	43.1	36.0	27.3
July 23	1.0	0.6	0.3	0.12

affected the daughter concentration inside the building. On the other hand, the next major increase in the concentration of radon daughters was observed in the late afternoon of July 22, 1979. This can be related to the low air-exchange rates inside the building at that time because the building was sealed for several hours before these measurements were taken. It is interesting to compare the concentration of radon daughters measured upstairs (rooms 4A and 6B) with those measured downstairs (east and west rooms) where most of the radon sources are located. Taking the data of July 22 as the basis of this comparison, it can be seen, on the average, that daughter concentrations upstairs are slightly higher than those downstairs. This difference in daughter concentrations cannot be completely related to the effect of the air-exchange process because measured air-exchange rates in the rooms upstairs on that day ($\sim$1.0-1.4) were higher than those measured downstairs ($\sim$0.75). The other reason which may cause this observed difference in concentrations is that the effective surface area and hence the wall deposition rate of radon daughters is much larger downstairs than upstairs. On the other hand, the effect of wall deposition on daughter concentrations downstairs will be more effective because of the low air-exchange rate at the time of measuring these results. The effect of air-exchange rate on wall deposition will be encountered later on in the discussions of the theoretical results.

During the period of collecting the experimental data, activities inside the building on July 22, 1979, were minimal because the building had been closed and sealed for several hours. Accordingly, results of the measurements taken on this day may provide a better source of information. Results of measurements on this day were used to calculate

the gross wall removal rates for the different radon daughters assuming that approximately steady-state conditions were achieved inside the the building--particularly in the ground-level rooms. At steady-state conditions, the differential equation presented in Chapter IV for radon daughters gives the following result:

$$C_i = \frac{\lambda_i C_{i-1} + \lambda_v Ci}{\lambda_i + \lambda_v + \lambda_{w,i}}, \qquad i = 1,2,3.$$

The parameters λ_i, λ_v, and $\lambda_{w,i}$ are the same as described in Chapter IV, and C_{i-1} at $i = 1$ represents the concentration of the parent radon (C_{Rn}) at the time of measurements. The term for radon daughters in outdoor air (C_i^o) can be neglected in this case since outdoor concentrations were much less than those indoors. Therefore, the previous relation can be written in a different form using the basic definition of the disequilibrium fraction as follows:

$$K_i = \frac{\lambda_i}{\lambda_i + \lambda_v + \lambda_{w,i}},$$

where K_i is the disequilibrium fraction (C_i/C_{Rn}). Thus, if steady-state conditions are assumed, it is possible to estimate $\lambda_{w,i}$ for certain values of λ_v. Results of measurements taken in ground-level rooms on July 22, 1979 (given separately in Table 10), were used to calculate $\lambda_{w,i}$, and the results are shown in Table 11. It is important to recognize that the data in Table 11 are approximate since exact steady-state conditions ($dC_i/dt = 0$) may not have existed. Also, it is important to realize that these data are characteristic of the place where they were measured. Nevertheless these results show that there could be a difference in the wall deposition rates among different radon daughters.

TABLE 10

MEASURED CONCENTRATIONS OF RADON AND ITS DAUGHTERS IN THE EAST AND WEST ROOMS OF BUILDING 1 USED TO ESTIMATE APPROXIMATE WALL REMOVAL RATES GIVEN IN TABLE 11

	Concentration (pCi/Liter)			
Nuclei	East(1)	East(2)	West(1)	West(2)
Rn	64	140	58	118
RaA	24.13	61.1	23.8	48.9
RaB	16.4	36.1	13.7	22.0
RaC	13.0	27.3	10.6	13.4

TABLE 11

APPROXIMATE WALL REMOVAL RATES OF RADON DAUGHTERS IN THE GROUND FLOOR OF BUILDING 1 CALCULATED FROM THE DATA GIVEN IN TABLE 10

	Concentration (pCi/Liter)			
Nuclei	$\lambda_w(1)$	$\lambda_w(2)$	$\lambda_w(1)$	$\lambda_w(2)$
RaA	22.3	17.2	18.8	19.1
RaB	0.32	0.66	1.65	0.74
RaC	0.3	0.42	0.94	0.21

Another important effect which has to be considered in studying the behavior of radon daughters in indoor atmospheres is that of the air-exchange process. According to the discussions presented previously in connection with the effect of air-exchange processes on indoor radon concentration, it is expected that a reduction in air-exchange rate will cause an increase in the concentration of indoor radon daughters. This can be seen in Figure 18 where the concentrations of radon daughters measured in the east and west rooms and expressed in units of working level* are plotted against air-exchange rates. The data in this figure show a nonuniform decrease in the concentration of radon daughters with increasing air-exchange rates. This can be attributed to the fact that the experimental measurements which produced these results were conducted in an uncontrolled building where the effect of other variables could not be eliminated. Regardless of their scattered form, these results still show the significant effect of the air-exchange process on the concentration of radon and its decay products in indoor atmospheres.

Theoretical Results

Theoretical results presented here are calculated on the basis of the model which was described in Chapter IV. The discussions and arguments presented in connection with the experimental results are also used in connection with the theoretical results. It was assumed that indoor radon concentration was initially zero while the outdoor

---

*The working level (WL) is defined as any combination of short-lived radon decay products in 1 liter of air that will result in the ultimate emission of 1.3×10^5 MeV of alpha energy. The WL unit was proposed about 1957 following recognition that airborne radiation exposure of the human lung if predominantly due to the radon decay products deposited in the lungs from inspired air [6].

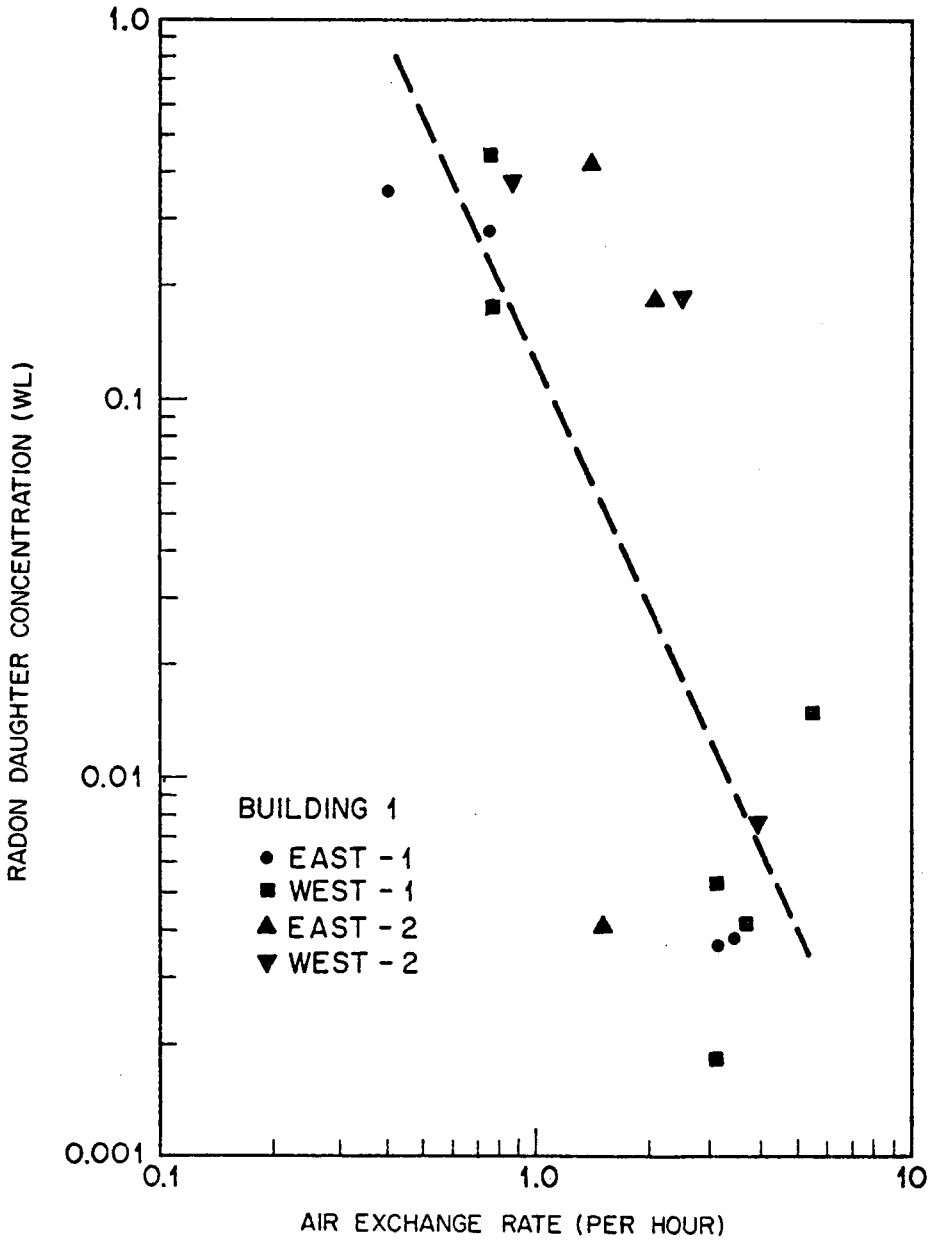

Figure 18. Effect of air-exchange process on the concentration of radon daughters measured in the east and in the west rooms of the building.

concentration was 100 pCi/m^3. Furthermore, a reference deposition velocity of 0.001 cm/sec for radium-B is being used to calculate the wall deposition rate of the different radon daughters. This reference deposition velocity was chosen on the basis of an interpolation of the data presented by various investigators [86, 87].

The variation of radon concentration with time at air-exchange rates ranging from zero to 1.5 per hour is shown in Figure 19. In calculating these results a constant, pressure-independent, exhalation rate of 100 pCi/m^2-hr was assumed. Also, the surface-to-volume ratio (S/V) was considered to be 2 $meter^{-1}$. It can be seen from this figure that for λ_v = 0 steady-state conditions were not established even at 50 hours after the release of the radon source. On the other hand, steady-state conditions were established much faster at higher air-exchange rates. At the same time it can also be seen that the steady-state concentration was reduced by a factor of two by increasing the air-exchange rate from 0.5 to 1.5 air exchanges per hour. Similar calculations at different exhalation rates showed that the functional behavior of the concentration with time is the same as that shown in Figure 19.

The effect of air-exchange process as well as the effect of exhalation rate on the concentration of indoor radon can be seen in Figure 20 where the steady-state radon concentrations are plotted against air-exchange rates for different exhalation rates. This figure shows the expected effect of the air-exchange process on the indoor radon concentration. At the same time this figure also illustrates the effect of exhalation rates (at various air-exchange rates) on the concentration of indoor radon. At higher exhalation rates, the reduction of radon concentration associated with an increase in air-exchange

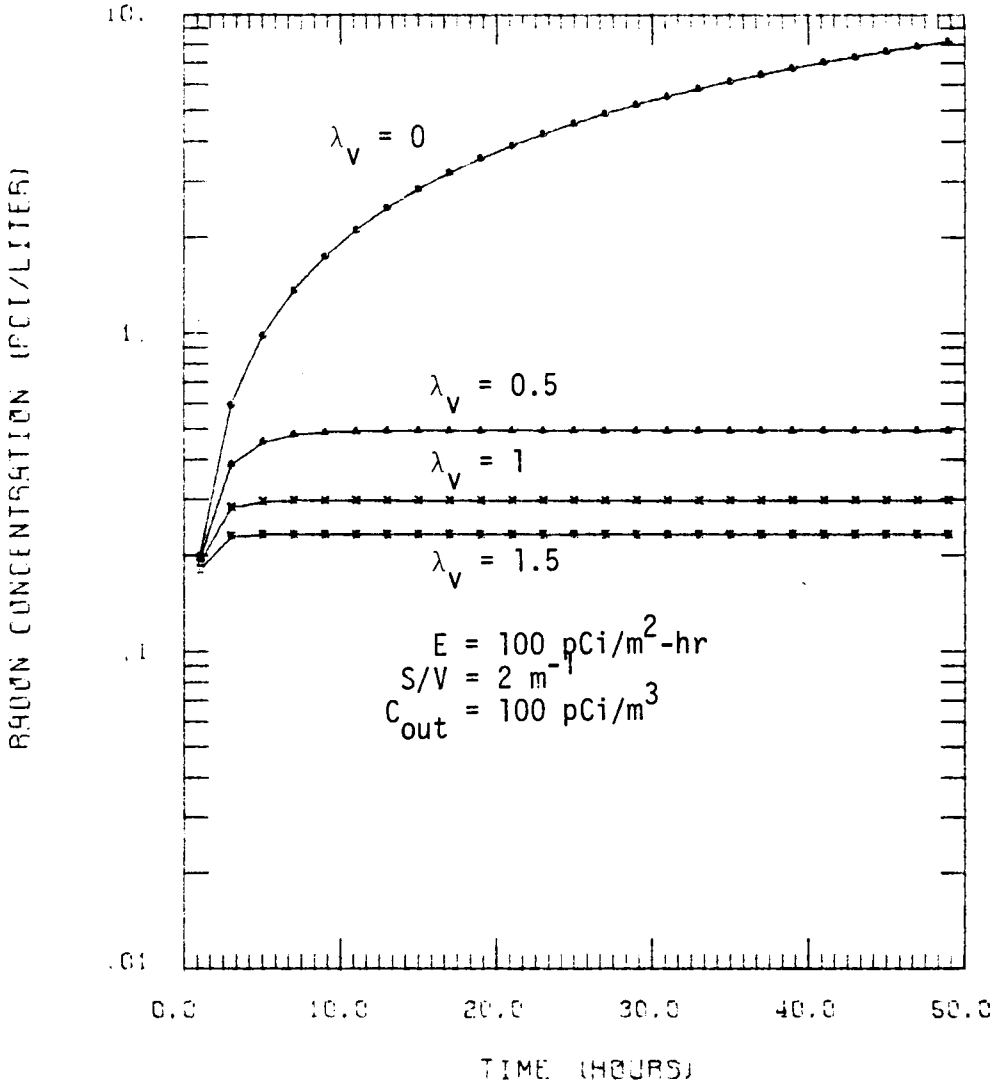

Figure 19. Variation of calculated radon concentration with time at different air-exchange rates for a constant exhalation rate.

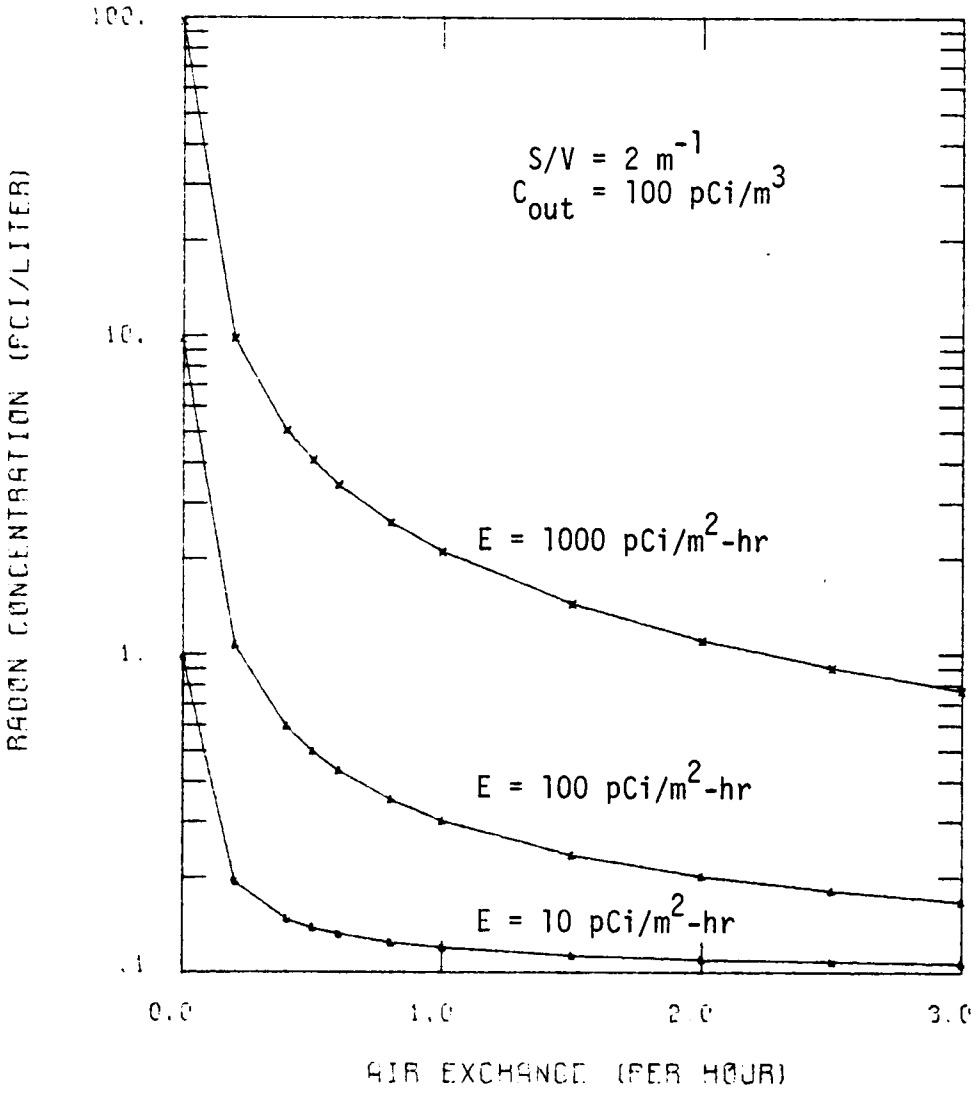

Figure 20. Variation of calculated radon concentrations with air-exchange rates for different exhalation rates.

rate is larger than for lower exhalation rates. For an exhalation rate of 1000 pCi/m^2-hr, a reduction in steady-state concentration by a factor of $\sim$4 is achieved by increasing the air-exchange rate from 0.5 to 2.5 air exchanges per hour. On the other hand, a reduction in the concentration by a factor of $\sim$1.3 is achieved for the same increase in air-exchange rate for an exhalation rate of 10 pCi/m^2-hr. This same phenomena was observed in the experimental results shown in Figure 17 (pp. 79).

Another parameter which was introduced in the discussions of Chapter IV is that concerned with the removal of radon due to some interactions on the walls. The effect of these wall interactions (λ_w) can be seen from Figure 21 where calculated radon concentrations at $\lambda_v = 0$ are plotted against time for the case where λ_w was set equal to zero (upper curve), and λ_w was set equal to 0.15 per hour. This numerical value of λ_w was calculated on the basis that the fraction β (defined in Chapter IV is equal to 10^{-6}. It is important to indicate that the choice of this value of β is just an approximation since these wall interactions up to the present time have not been studied experimentally. On the other hand, the calculated results given in Figure 22 show that the effect of λ_w is less pronounced at higher air-exchange rates than at lower air-exchange rates. This is true not only for the effect of λ_w but also for the effect of λ_{Rn}. At higher rates the effect of air-exchange process dominates over all other removal processes.

To study the effect of variations in atmospheric pressure on the concentration of radon, a hypothetical pressure front was assumed. The pressure front has a form similar to that of the positive (or negative) portion of a sine function whose amplitude is 20 mm Hg. This amplitude represents the maximum pressure drop (or increase) during an interval of

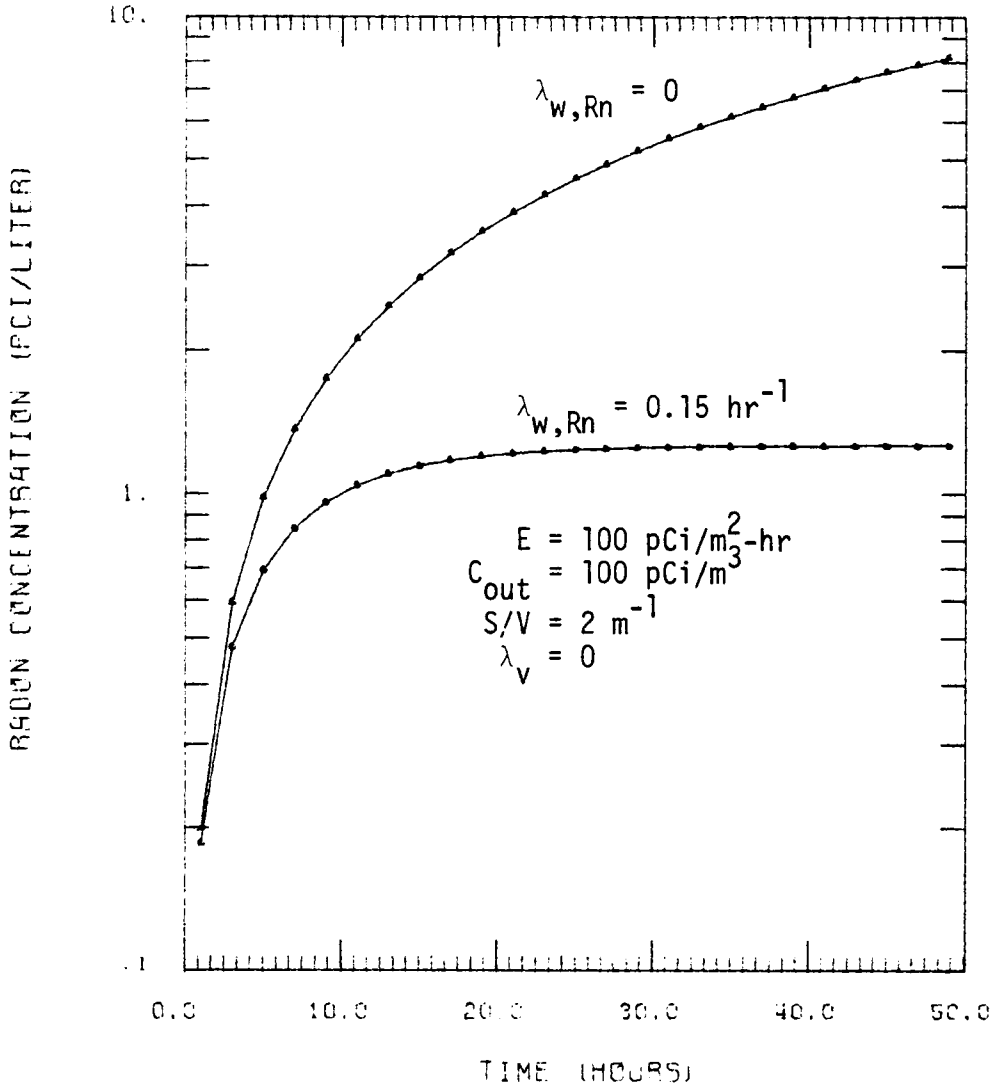

Figure 21. Variations of calculated radon concentrations with time for λ_v = 0 with and without the effect of radon removal by wall interactions.

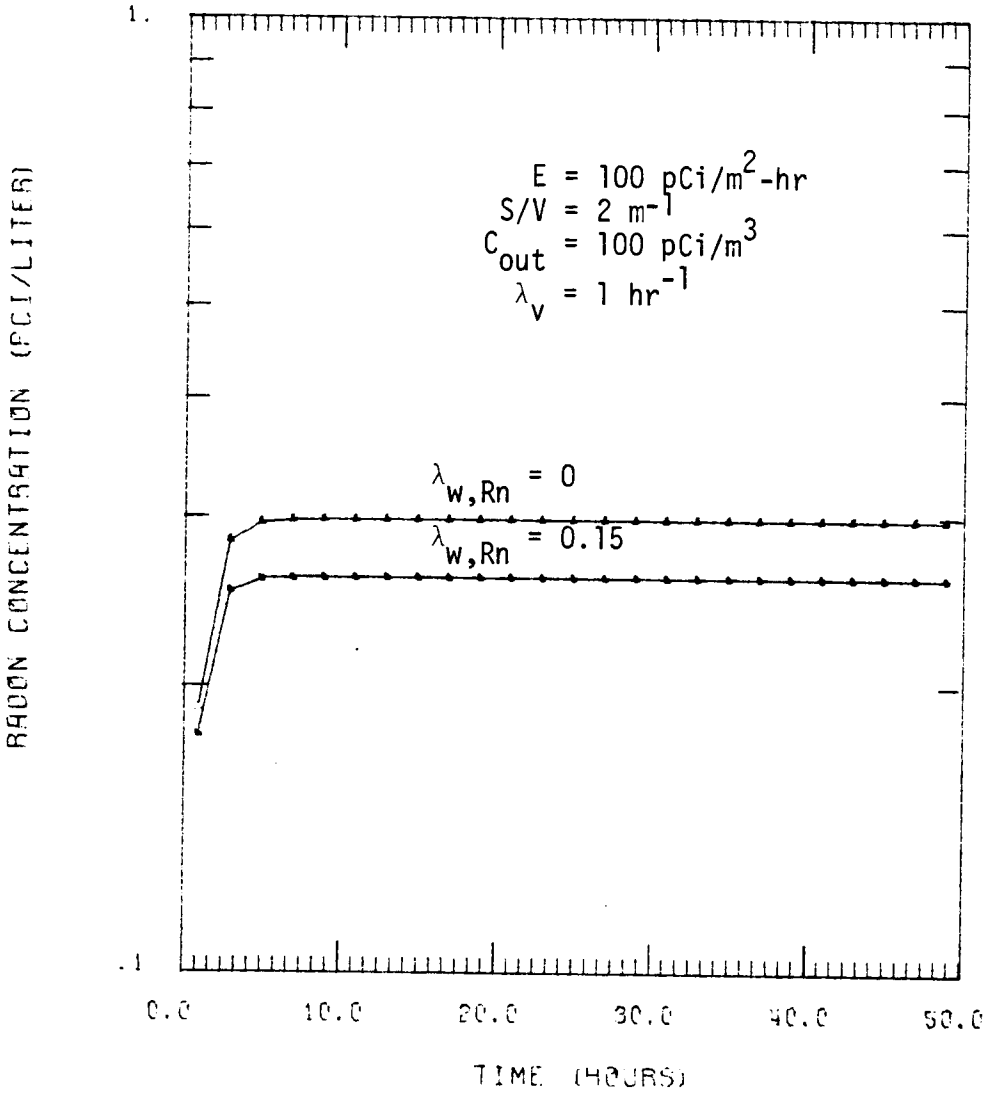

Figure 22. Variations of calculated radon concentration with time for λ_V = 1 air exchanges per hour with and without the effect of radon removal by wall interactions.

96 hours. The results of this simulation are shown in Figure 23 where the variation of radon concentration with time is shown for cases where pressure drop, pressure increase, and no pressure are considered. The results in Figure 23 were calculated for E_o = 500 pCi/m^2-hr and λ_v = 1 air exchanges per hour. This form of a pressure front was chosen because it will be easy to see the effect of both pressure drop and increase. Any other form could, of course, be incorporated in the model as has been described in Chapter IV. It can be seen from Figure 23 that there is an increase in the radon concentration which is associated with the decrease of atmospheric pressure (upper curve). The reverse phenomena can also be seen in the same figure (lower curve) where the concentration drops as the atmospheric pressure increases beyond its normal value. The effect of pressure variations on the concentration of radon was also observed in the experimental data (Figures 13-16, pp. 64-67). Quantitatively, it can be seen from Figure 23 that for this particular pressure front there is a maximum of 50 percent increase (or decrease) in the radon concentration at the time when the maximum variation in pressure occurs. It is to be emphasized that this increase (or decrease) in radon concentration is not universal, but it is a characteristic of this assumed pressure front at this particular air-exchange rate. Moreover, it is important to indicate that the amount of concentration increase (or decrease) associated with pressure variations as being calculated in this model depends on the value of α to be used. As mentioned earlier in Chapter II, both theoretical and experimental results related to the effect of variations in atmospheric pressure on radon exhalation rate showed that a pressure change of 1 percent could change the exhalation rate by 20 to 60 percent depending on the rate and

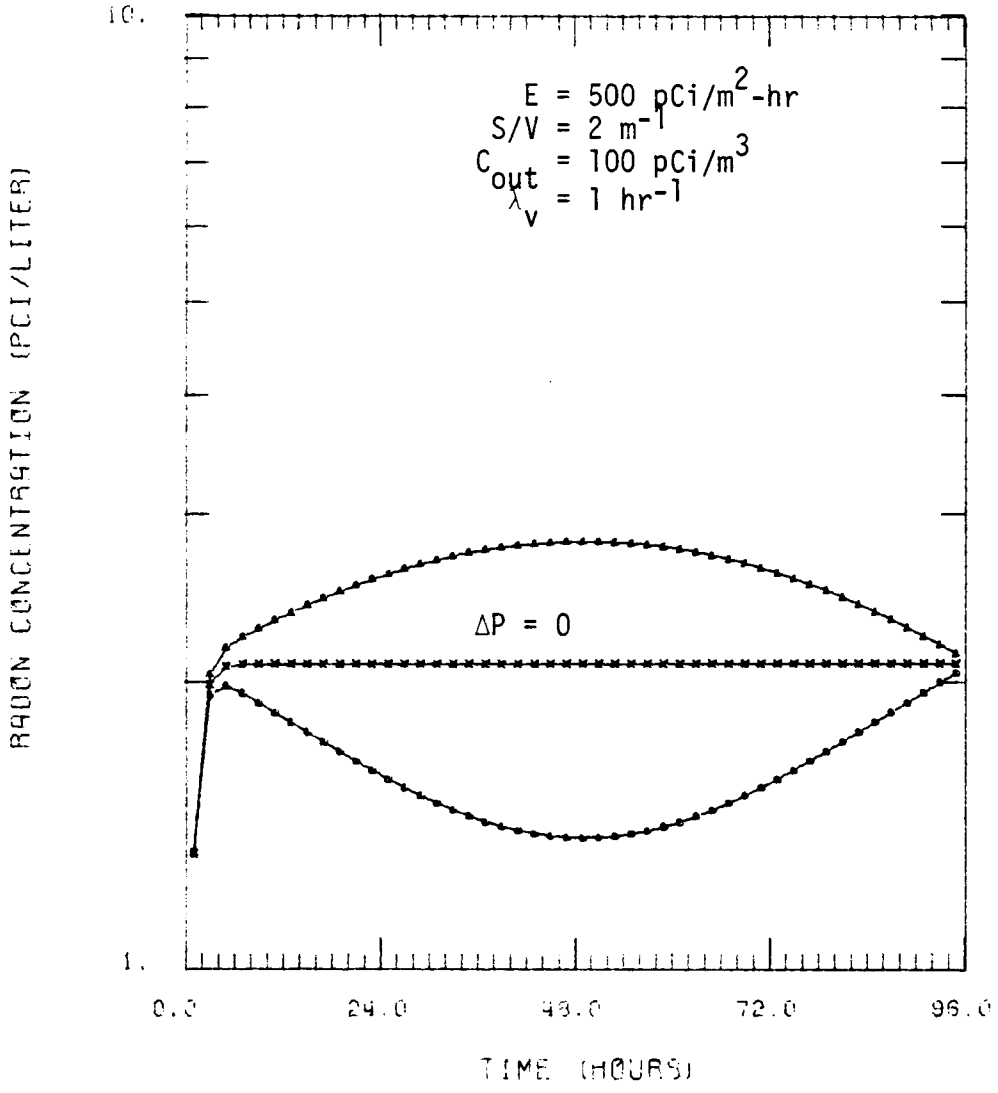

Figure 23. Variation of calculated radon concentration with time under the effect of pressure increase (lower curve) and pressure drop (upper curve) for λ_v = 1 hr^{-1} and E - 500 pCi/m^2-hr.

duration of the change in pressure [44-47]. Accordingly, the change in concentration under the influence of this hypothetical pressure front as calculated by this model agrees well with these reported results.

After calculating the effect of the various parameters on the concentration of indoor radon, it is important to study the behavior of radon daughters associated with these conditions. However, under such idealized conditions the daughter concentrations are expected to behave in a manner which is consistent with their parent radon. In other words, any relative increase in the concentration of indoor radon will be accompanied by a relative increase in the daughter concentrations. This is true since radon gas represents the only source term in the daughter's basic differential equation (Chapter IV). Accordingly, the variation of daughter concentrations with time will follow closely that of the radon gas except with different removal rates. It is thus more important to study the influence of the most effective parameter, which is air-exchange processes, on the concentration of the different radon daughters. It is necessary to recognize that these calculations are performed on the basis that short-lived radon daughters have different deposition velocities and hence different wall deposition rates as has been discussed in Chapter IV. In accordance with the discussions of Chapter IV and on the basis of the chosen reference deposition velocity, the wall deposition rates of radium-A, radium-B, and radium-C were found to be 0.6, 0.07, and 0.1, respectively, for $S/V = 2m^{-1}$. As mentioned before, the approach of using different wall deposition rates for the different radon daughters is being introduced for the first time. Accordingly, it is not possible to compare the wall deposition rates

calculated on the basis of this approach with other data. However a single wall deposition rate of 0.7 per hour for attached radon daughters was reported for Jacobi [81]. This value seems very close to that of radium-A which was calculated on the basis of this new approach.

The effect of wall deposition on the disequilibrium fraction (K) can be seen by comparing the results given in Figures 24 and 25. The effect of wall deposition is being neglected in Figure 24 and thus the removal of daughters is achieved through their radioactive decay and through air-exchange process. On the other hand, Figure 25 shows the combined effect of all removal processes. It can be seen from these figures that the removal of radon daughters by wall deposition will be more significant at lower air-exchange rates. At $\lambda_v = 0.0$, this effect is very clear for all daughters (Figure 25).

The effect of the air-exchange process is also shown in Figure 25 where the disequilibrium fraction (K) (for RaA, RaB, and RaC) is plotted against air-exchange rates. It can be seen from this figure that the influence of air-exchange rate on the disequilibrium fraction of radium-A is minimum because of the large physical decay rate of radium-A ($\lambda_2 = 13.6\ hr^{-1}$). On the other hand, this effect is much more pronounced for radium-B and radium-C since their physical decay rates are much smaller than that of radium-A. A reduction in K for radium-B by a factor of two can be obtained by increasing the air-exchange rate from 0.5 to 3.0 air exchanges per hour. However, increasing the air-exchange rate for the same value will reduce the value of K for radium-C by a factor of $\sim$7. This phenomenon once again shows the importance of the air-exchange process in controlling the concentration of radon daughters in indoor atmospheres.

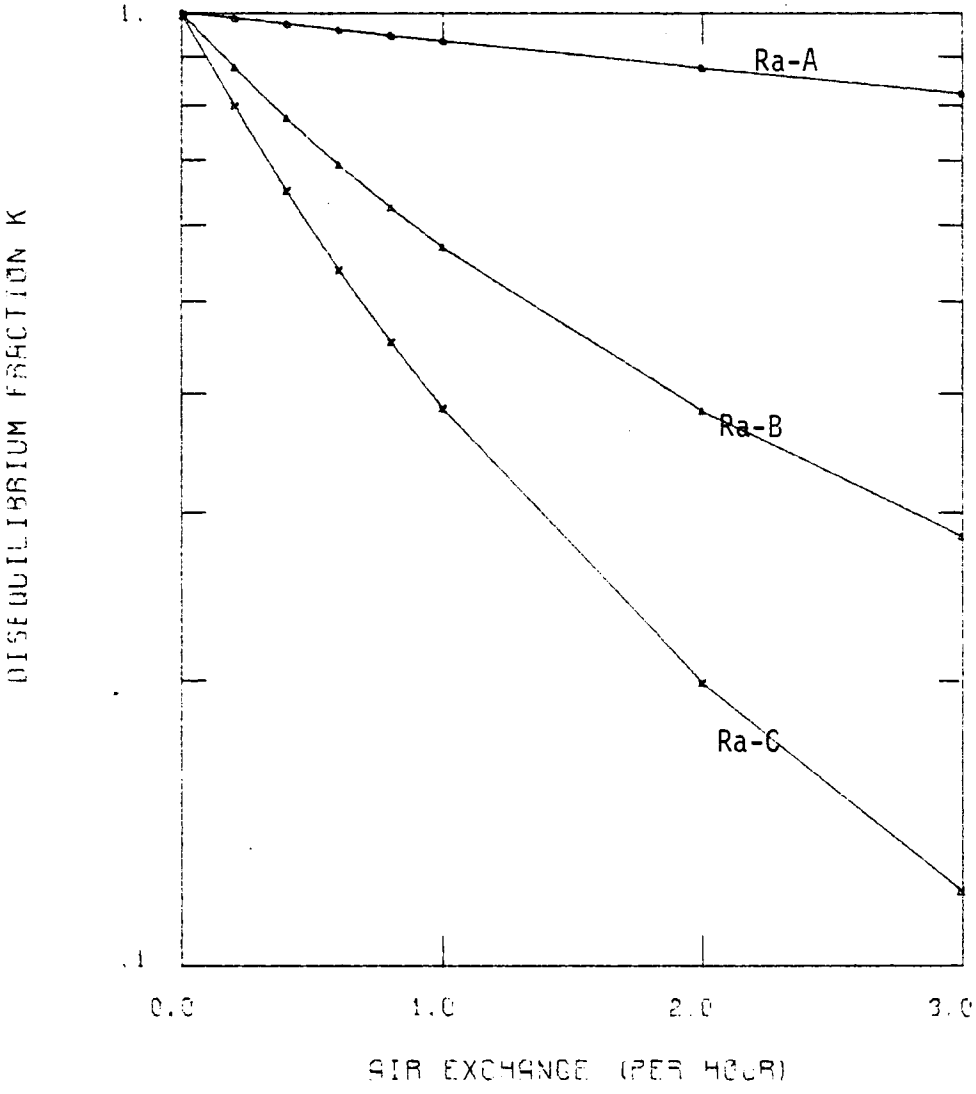

Figure 24. Variation of Radon Daughter Disequilibrium Fractions With Air-Exchange Rates Without Wall Deposition.

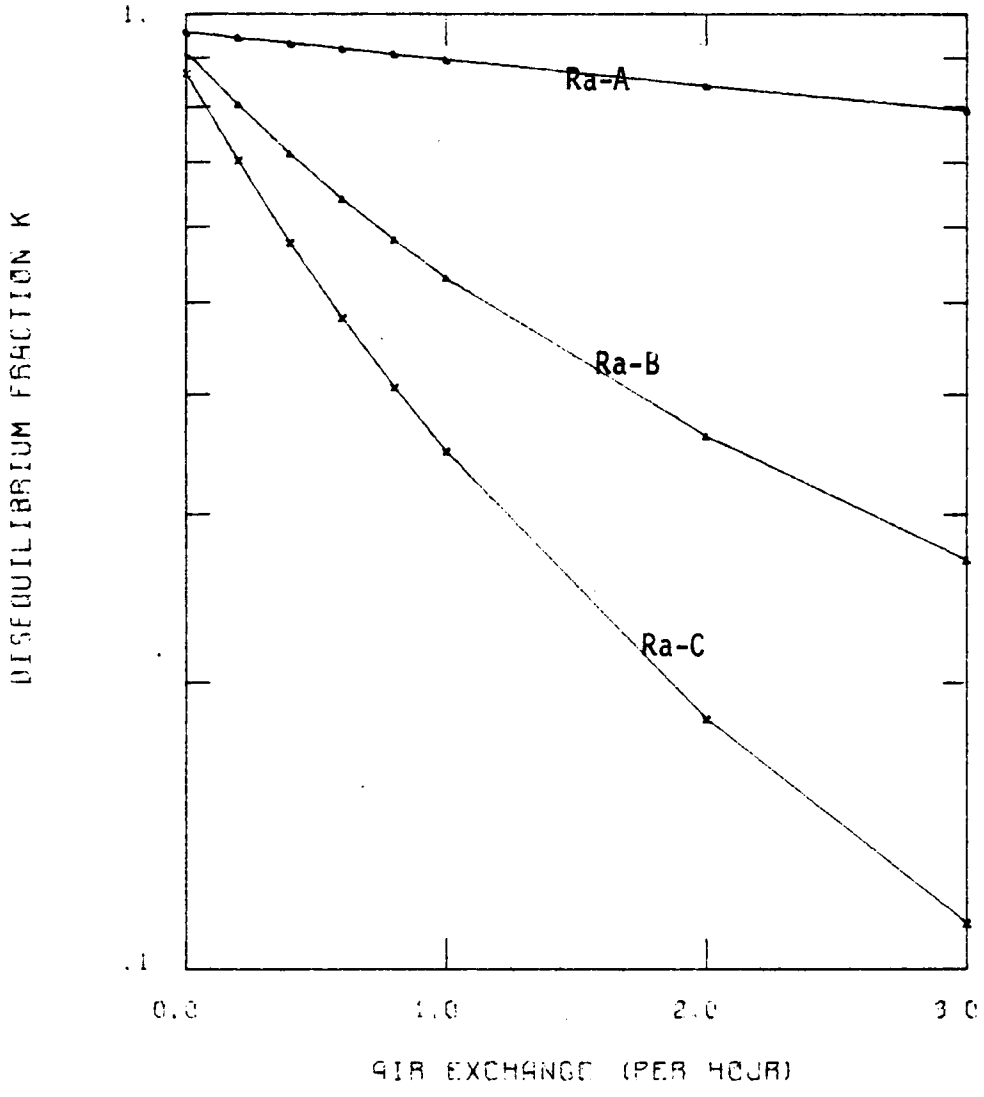

Figure 25. Variation of Radon Daughter Disequilibrium Fractions With Air-Exchange Rates With Wall Deposition.

The proposed model was used to calculate the concentration of radon in the west room of the building on the basis of the available experimental data. Air-exchange rates were assumed whenever experimental data were unavailable. The results of this calculation are given in Figure 26 where measured and calculated concentrations are plotted against time. Although these calculations are approximate yet they show some agreement between calculated and measured concentrations. A comparison between calculated and measured values of the radon daughter working level in the west room for the period July 21-22 provided a better agreement. During those two days, the measured working level varied from a maximum of 0.43 to a minimum of 0.19 giving an average working level of 0.26. Calculated working levels for the same period varied from a maximum of 0.29 to a minimum of 0.25 giving an average working level of 0.27. A comparison between those two numbers shows that there is a good agreement between the calculated and measured working levels. This indicates that the model is also capable of predicting radon daughter concentrations in indoor atmospheres.

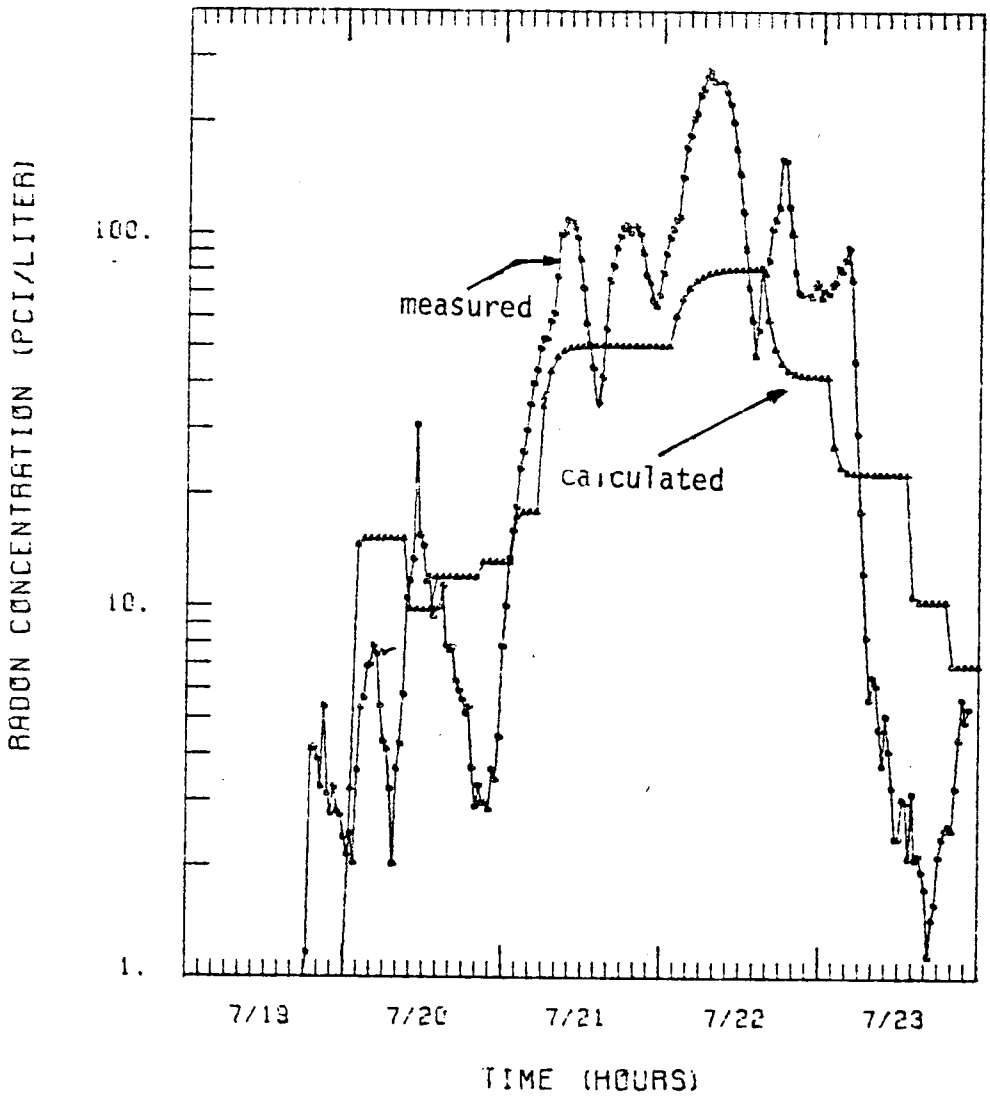

Figure 26. Variations in measured and calculated radon concentrations in west room of the building during july 1979.

CHAPTER VI

SUMMARY AND CONCLUSIONS

This chapter is concerned with the summary of this work. Conclusions derived from results obtained in this research are given together with suggestions for further studies in this field of study.

Summary

This research was designed to study a natural radiation source which contributes significantly to population exposures to ionizing radiation. This radiation field is associated with the accumulation of the noble gas, radon, and its decay products in the atmospheres of inhabited areas. The problem of radon accumulation in indoor atmospheres was studied experimentally as well as theoretically. Experimental measurements were conducted in a building of an industrial park in southwestern Pennsylvania. This building represents a unique experimental facility because it contains a variety of indoor radon sources. To carry on this research, it was necessary to investigate the various processes which could affect the accumulation of radon and its daughters in indoor atmospheres. To study these processes, it was necessary to measure quantities such as air-exchange rates, radon flux, and the concentration of airborne radon and its daughters. The techniques used to measure these variables have been published and are well established. However, in some cases it was necessary to modify some of the measurement techniques, namely, that of measuring radon flux to accommodate the experimental conditions.

Air-exchange rates were measured using CO_2 as a tracer gas which was detected by an infrared spectrometer known as MIRAN-80. This instrument is fitted with a microprocessor which controls the functioning of the instrument and also records and analyze the data. This spectrometer proved to be a reliable tool which was easy to handle and was semiportable.

Radon concentration inside and outside the building was measured continuously using modified computer-based versions of a continuous radon monitor designed by Wrenn. The five units which were used during these measurements were all fitted with NDT-COMP-8 microcomputers (designed and built at ORNL) which are preprogrammed to analyze the data according to specific input data concerning the characteristic properties of each unit. These units provided an excellent, reliable, and quiet instrument and required practically no attention after it was placed into operation. Indoor radon concentrations were measured at four locations inside the building and at one location outside the building. Radon concentrations were measured continuously for a period of five days during July 1979.

Concentrations of radon daughters were measured using an alpha spectrometry technique in which particulate radon daughters were collected on a membrane filter and counted using a silicon surface barrier detector. Daughter concentration measurements were taken where radon monitors were located.

Radon flux was measured at numerous locations in the ground level of the building. Hundreds of charcoal canisters (radon collectors) were used for measuring radon flux emanating from the different sources inside the building. The locations at which radon flux was measured

were chosen in such a way to produce representative results characteristic of this building. Two gamma spectrometers were used to count the charcoal canisters after being exposed to radon for a period of about 24 hours.

Three main sources of indoor radon were identified. The source which contributed most to the observed radon concentration in the building is the concrete floor slabs. In measuring the radon flux emanating from these concrete slabs the usual method of using the charcoal canisters was employed. The second major source of radon was determined to be the cracks in the concrete floor. Numerous cracked areas were observed in the floor of the building's ground level. Using distributed canisters to measure the flux emanating from cracks would have required a large number of these canisters in order to get representative results. In order to save time and effort of handling a large number of canisters, a crack monitor was designed specifically to measure radon flux emanating from a crack of 1 meter length. Radon gas emanating from a crack of 1 meter length is enclosed in a relatively small volume and circulated through a charcoal canister where the radon is adsorbed. Gas circulation was accomplished by using a small air pump which cycles the air through the canister and back to the accumulator volume. To prevent the stagnation of radon near the floor surface, a small battery-operated fan was installed inside the crack monitor housing. Consistent and reproducible results were obtained at a flow rate of 8 liters/minute. Calibration results showed that the crack monitor records 70 percent of the actual radon flux. This simple device was introduced for such measurements for the first time and facilitated

the measurements of radon emanating from cracks. Because of its weight, the monitor requires little effort to be sealed to a flat surface.

The third source of indoor radon which was identified and successfully measured is that from drains which are scattered throughout the ground level and in particular in the west room of the building. Measurements of radon flux emanating from drains were made using two charcoal canisters positioned back to back and placed so as to collect all radon emanating from the drain. The flux recorded by these two canisters together with a room background canister provide enough information to estimate the radon flux emanating from drains. This method was successfully used in this research.

Theoretical consideration of the problem was presented in the form of an analytical model which could be used to calculate the concentration of radon and its daughters in the atmospheres of inhabited areas. In these theoretical considerations it was assumed that exhalation of radon from indoor sources together with contributions from outdoors causes the accumulation of radon in indoor atmospheres. Removal of radon was assumed to be due to the effect of radioactive decay, air-exchange processes, and wall interactions. The removal of radon by wall interaction is presented here for the first time and based on the kinetic theory of gases, an expression was derived which permitted the evaluation of the rate of removal due to this process. The effect of this removal process on indoor concentration at various air-exchange rates was thoroughly discussed.

Radon daughters accumulating due to decay of their parent radon were assumed to be removed by radioactive decay, air exchange with the outdoor atmosphere, and by wall deposition. It was assumed that all

short-lived radon daughters possess one characteristic length which defines their deposition velocity. Based on this assumption, it was possible to calculate different deposition velocities for different radon daughters. The effect of wall deposition on the behavior of daughters was thoroughly studied. The working level calculated on the basis of this model agree well with those derived from experimental results.

Conclusions

This work addressed the problem of the accumulation of indoor radon in very broad and natural terms. Air-exchange processes were found to be the most influential factor which can control the indoor radon concentration. Although this is not a new discovery, the experimental results of this research strongly verified this conclusion. This observation has to be considered seriously when reduction in air-exchange rates are to be considered as means to conserve energy.

The model presented in this work competently predicted the concentration of radon and its daughters within the accuracy of the input results. Because of its general nature, this model could be used in other structures so that it could predict increases in indoor radon concentration associated with reductions in air-exchange rates for energy conservation purposes. Moreover the introduction of this model posed several interesting research points which require further investigation. Removal of radon due to wall interactions should be studied. Dependence of these interactions on the type of wall material need to be identified. Another interesting point would be the measurement of wall deposition rates for the different radon daughters as proposed in this

model and also their dependence on wall material. Finally, it would be interesting to study the effect of variations in atmospheric pressure on radon exhalation rate for materials other than concrete.

LIST OF REFERENCES

REFERENCES

1. Cember, H. (1969). Introduction to Health Physics, Pergamon Press, New York.

2. Yeates, D. B., Goldin, A. S., and Moeller, D. W. (1972). Natural Radiation in the Urban Environment, Nuclear Safety, Vol. 13, p. 275.

3. Toth, A. (1972). Determining the Respiratory Dosage from RaA, Rab, and RaC Inhaled by the Population in Hungary, Health Phys., Vol. 23, p. 281.

4. Archer, Victor E., Wagoner, Joseph K., and Lundin, Frank E. (1973). Lung Cancer Among Uranium Miners in the United States, Health Phys., Vol. 25, p. 351.

5. Sevc, J., Kunz, E., and Placek, V. (1976). Lung Cancer in Uranium Miners and Long-Term Exposure to Radon Daughter Products, Health Phys., Vol. 30, p. 433.

6. Evans, Robley D. (1969). Engineers Guide to the Elementary Behavior of Radon Daughters, Health Phys., Vol. 17, p. 229.

7. Haque, A. K. M. M. and Collinson, A. J. L. (1967). Radiation Dose to the Respiratory System Due to Radon and Its Daughter Products, Health Phys., Vol. 13, p. 431.

8. Barton, C. J., Moore, R. E., and Rohwer, P. S. (1973). Contribution of Radon in Natural Gas to the Dose from Airborne Radon Daughters in Homes, in Noble Gases (Moghissi, A. A. and Stanley, R. E., Eds.), p. 134, U.S. Government Printing Office, Washington, D. C.

9. Cliff, K. D. (1978). The Measurement of Low Concentrations of Radon-222 Daughters in Air, with Emphasis on RaA Assessment, Physics in Medicine and Biology, Vol. 23(1), p. 55.

10. Auxier, J. A. (1976). Respiratory Exposure in Buildings Due to Radon Progeny, Health Phys., Vol. 31, p. 119.

11. Steinhäusler, Friedrich (1975). Long Term Measurements of Rn-222, Rn-220, Pb-214, and Pb-212 Concentrations in the Air of Private and Public Buildings and Their Dependence on Meteorological Parameters, Health Phys., Vol. 29, p. 705.

12. Haque, A. K. M. M., Collinson, A. J. L., and Brooke, C. O. S. (1965). Radon Concentration in Different Environments and the Factors Influencing It, Physics in Medicine and Biology, Vol. 10(4), p. 505.

13. Hultquist, B. (1956). Studies on Naturally Occurring Ionizing Radiation Doses in Swedish Houses of Various Types [cited in Reference 14].

14. Snihs, J. O. (1973). The Significance of Radon and Its Progeny as Natural Radiation Sources in Sweden, in Noble Gases (Moghissi, A. A. and Stanley, R. E., Eds.), U.S. Government Printing Office, Washington, D. C., p. 115.

15. Gesell, Thomas F. (1975). Occupational Radiation Exposure Due to Rn-222 in Natural Gas and Natural Gas Products, Health Phys., Vol. 29, p. 681.

16. Gesell, Thomas F. and Prichard, Howard M. (1978). The Contribution of Radon in Tap Water to Indoor Radon Concentration, proc. symp. on The Natural Radiation Environment III, Houston, Texas.

17. Gesell, Thomas F. and Prichard, Howard, M. (1975). The Technologically Enhanced Natural Radiation Environment, Health Phys., Vol. 28, p. 361.

18. Prichard III, Howard Milton (1978). Population Radiation Doses Attributable to Radon-222 in Ground Waters in Greater Houston, Texas, Ph.D. Dissertation, University of Texas, Houston.

19. U.S. Energy Research and Development Administration, Division of Construction Planning and Support and Division of Solar Energy (1977). Solar Design Handbook, ERDA 77-65.

20. Gabrysh, A. F. and Davis, F. J. (1955). Radon Release from Concrete in Radiant Heating, Nucleonics, Vol. 13(1), p. 50 [also Civil Engineering, Vol. 24, p. 839 (1954)].

21. National Council on Radiation Protection and Measurements (1975). Natural Background Radiation in the United States, NCRP Report No. 45.

22. National Council on Radiation Protection and Measurements (1976). Environmental Radiation Measurements, NCRP Report No. 50, Chapt. 2.

23. Pearson, John E. and Jones, Gary E. (1965). Emanation of Radon-222 from Soils and Its Use as a Tracer, J. Geophys. Res., Vol. 70, p. 5279.

24. Windham, S. T., Savage, E. D., and Phillips, C. R. (1977). The Effects of Home Ventilation Systems on the Indoor Radon-Radon Daughters Levels, U.S. Environmental Protection Agency Report EPA-520/5-77-011.

25. Toth, A., Feher, I., Keszthelyi, B., Lakatos Novotny, S., and Koszorus, L. (1978). Fitting Normal Distributions to the Observed Thorium, Radium, Emanating Radium, Potassium and Emanating Power Data for the Concretes and Bricks in Hungary, proc. symp. on The Natural Radiation Environment III, Houston, Texas.

26. Wollenberg, Harold A. and Smith, Alan R. (1966). A Concrete Low-Background Counting Enclosure, Health Phys., Vol. 12, p. 53.

27. Swedjemark, Gun Astri (1978). Radon in Swedish Dwellings, proc. symp. on The Natural Radiation Environment III, Houston, Texas.

28. Eichholz, G. G., Clarke, F. J., and Kahn, B. (1978). Radiation Exposure from Building Materials, proc. symp. on The Natural Radiation Environment III, Houston, Texas.

29. Wachsmann, F. and Regulla, D. F. (1978). Methods and Results of Measurements of the Environmental Radiation in Bavaria, F.R.G., proc. symp. on The Natural Radiation Environment III, Houston, Texas.

30. Hamilton, E. I. (1971). The Relative Radioactivity of Building Materials, Amer. Indus. Hygiene Assoc. J., Vol. 32, p. 398.

31. Pensko, Jerzy and Stpiczynska, Zofia (1978). Emanating Power of Rn-222 Measured in Building Materials, proc. symp. on The Natural Radiation Environment III, Houston, Texas.

32. Hoather, W. H. (1952). Radioactive Building Tiles, Nucleonics, Vol. 10(12), p. 73.

33. Israel, H. (1962). In Kernstrahlung in Der Geophysik (Nuclear Radiation in Geophysics), (Israel, H., Krebs, A., and Adams, R. D., Eds.), Academic Press, Inc., New York.

34. Tanner, Allan B. (1964). Radon Migration in the Ground: A Review, in The Natural Radiation Environment (Adams, J. A. S. and Lowder, W. M., Eds.), University of Chicago Press.

35. Barretto, Paulo M. C., Clark, Ronald B., and Adams, John A. S. (1975). Physical Characteristics of Radon-222 Emanation from Rocks, Soils and Minerals: Its Application to Temperature and Alpha Dose, proc. symp. on The Natural Radiation Environment III (Adams, Lowder and Gessell, Eds.), p. 731.

36. Culot, Michel V. J., Schiager, Keith J., and Olson, Hilding G. (1978). Development of a Radon Barrier, Health Phys., Vol. 35, p. 375.

37. Auxier, J. A. (1972). Contribution of Natural Terrestrial Sources to the Total Radiation Dose to Man, Ph.D. Dissertation, Georgia Institute of Technology, Atlanta, Georgia.

38. Auxier, J. A., Shinpaugh, W. H., Kerr, G. D., and Christian, D. J. (1974). Preliminary Studies of the Effects of Sealants on Radon Emanations from Concrete, Health Phys., Vol. 27, p. 390.

39. Franklin, J. C. and Nuzman, L. T. (1975). Polymeric Materials for Sealing Radon Gas into the Walls of Uranium Mines, Spokane Mining Research Center, Washington, U.S. Bureau of Mines, RI-8036, HO-220006 and HO-230007.

40. Kraner, Hobart W., Schroeder, Gerald L., and Evans, Robley D. (1964). Measurements of the Effects of Atmospheric Variables on Radon-222 Flux and Soil-Gas Concentrations, in The Natural Radiation Environment (Adams, J. A. S. and Lowder, W. M., Eds.), University of Chicago Press.

41. Pearson, John E. (1967). Natural Environmental Radioactivity from Radon-222, U.S. Department of Health, Education and Welfare, Public Health Service Publication No. 999-RH-26.

42. Pohl-Ruling, Johanna and Pohl, Egon (1969). The Radon-222 Concentration in the Atmosphere of Mines as a Function of Barometric Pressure, Health Phys., Vol. 16, p. 579.

43. Wilkening, M. H., Clements, W. E., and Stanley, D. (1975). Radon-222 Flux Measurements in Widely Separated Regions, proc. symp. on The Natural Radiation Environment III (Adams, Lowder, and Gesell, Eds.), p. 717.

44. Clements, William E. and Wilkening, Marvin H. (1974). Atmospheric Pressure Effects on Rn-222 Transport Across the Earth-Air Interface, J. Geophys. Res., Vol. 79, p. 5025.

45. Jonassen, Neils (1975). On the Effect of Atmospheric Pressure Variations on the Radon-222 Concentration in Unventilated Rooms, Health Phys., Vol. 29, p. 216.

46. Stranden, E., Berteig, L., and Ugletvett, F. (1979). A Study of Radon in Dwellings, Health Phys., Vol. 36, p. 413.

47. Jonassen, Neils and McLaughlin (1978). Exhalation of Radon-222 from Building Materials and Walls at Constant and Falling Pressures, proc. symp. on The Natural Radiation Environment III, Houston, Texas.

48. Giletti, Bruno J. and Kulp, J. Laurence (1955). Radon Leakage from Radioactive Minerals, American Mineralogist, Vol. 40, p. 481.

49. Megumi, Kazuko and Mamuro, Tetsuo (1972). A Method for Measuring Radon and Thoron Exhalation from the Ground, J. Geophys. Res., Vol. 77, p. 3052.

50. Megumi, Kazuko and Mamuro, Tetsuo (1973). Radon and Thoron Exhalation from the Ground, J. Geophys. Res., Vol. 78, p. 1804.

51. Gabrysh, A. F., McKee, N. D., and Eyring, H. (1962). Determination of Radon Emanation from Carbonate Rocks and Its Potential Hazard in Building Materials, Materials Res. and Standards, No. 2, p. 265.

52. Handley, T. H. and Barton, C. J. (1973). Home Ventilation Rates: A Literature Survey, Oak Ridge National Laboratory, ORNL/TM-4318.

53. Haywood, F. F. and Hawthorne, A. R. (1978). Measurements of Air Exchange Rates in Houses Using CO_2 Tracer Gas, Oak Ridge National Laboratory, to be published as an ORNL report.

54. Elkins, P. H. and Wensman, C. E. (1971). Natural Ventilation of Modern Tightly Constructed Homes, Conference on Natural Gas Research and Technology, American Gas Association and Institute of Gas Technology, Chicago, Illinois.

55. Spitz, Henry B. and Wrenn, McDonald E. (1978). Ventilation Rate of a Structure Inferred from Measurements of Indoor and Outdoor Radon and the Flux through the Foundation, proc. Health Phys. Soc. 23rd Annual Meeting, Minneapolis, Minnesota.

56. Wilkening, M. H. and Watkins, David E. (1976). Air Exchange and Rn-222 Concentrations in the Carlsbad Caverns, Health Phys., Vol. 31, p. 139.

57. Tsivoglou, E. C., Ayer, H. E., and Holaday, D. A. (1953). Occurrence of Nonequilibrium Atmospheric Mixtures of Radon and Its Daughters, Nucleonics, Vol. 11(9), p. 40.

58. Cliff, K. D. (1978). Measurements for Radon-222 Concentrations in Dwellings in Great Britain, proc. symp. on The Natural Radiation Environment III, Houston, Texas.

59. Fitzgerald, Joseph E., Jr., Guimond, Richard J., and Shaw, Roger A. (1976). A Preliminary Evaluation of the Control of Indoor Radon Daughter Levels in New Structures, U.S. Environmental Protection Agency Report EPA-520/4-76-018.

60. Porstendorfer, J., Wicke, A., and Schraub, A. (1978). The Influence of Exhalation, Ventilation and Deposition Processes Upon the Concentration of Radon-222, Thoron-220, and Their Decay Products in Room Air, Health Phys., Vol. 34, p. 465.

61. Pearson, John E. and Moses, Harry (1966). Atmospheric Radon-222 Concentration Variation with Height and Time, J. Appl. Meteorology, Vol. 5, p. 175.

62. Hosler, Charles R. (1965). Simultaneous Measurements of Rn-222, Pb-214, and Bi-214 in Air Near Ground, J. Geophys. Res., Vol. 70, p. 4537.

63. Israel, H. and Horbert, M. (1966). Results of Continuous Measurements of Radon and Its Decay Products in the Lower Atmosphere, Tellus, Vol. XVIII, p. 638.

64. Cox, William M., Blanchard, Richard L., and Kahn, Brend (1970). Relation of Radon Concentration in the Atmosphere to Total Moisture Detention in Soil and Atmospheric Thermal Stability, in Radionuclides in the Environment, Advances in Chemistry Series 93, p. 436.

65. Malakhov, S. G., Bakulin, V. N., Dmitrieva, G. V., Kirichenko, L. V., Ssissigina, T. I., and Starikov, B. G. (1966). Diurnal Variations of Radon and Thoron Decay Product Concentrations in the Surface Layer of the Atmosphere and Their Washout by Precipitations, Tellus, Vol. XVIII, No. 2, p. 643.

66. Travis, C. C. (1979). Environmental Sources of Radon, Trans. of the American Nuclear Society, Vol. 33, p. 144.

67. Kerr, G. D. (1975). Measurements of Radon Progeny Concentrations in Air by Alpha Spectrometry, Oak Ridge National Laboratory, ORNL/TM-4924.

68. Kerr, G. D., Ryan, M. T., and Perdue, P. T. (1978). Measurements of Airborne Concentrations of Radon-220 Daughter Products by Alpha Spectrometry. Paper presented at the 11th Midyear Topical Symposium of the Health Physics Society, San Diego, California.

69. Perdue, P. T., Dickson, H. W., and Haywood, F. F. (1980). Radon Monitoring Instrumentation, to be published in Health Phys.

70. Formerly Utilized MED/AEC Sites Remedial Action Program, Radiological Survey of the Former Vitro Rare Metals Plant, Canonsburg, Pennsylvania, Final Report (1979), DOE/EV-0005/3.

71. Hawthorne, A. R. (1979). New Developments in Monitoring Instruments, Symposium on Potential Health and Environmental Effects of Synthetic

72. Budnitz, Robert J. (1974). Radon-222 and Its Daughters--A Review of Instrumentation of Occupational and Environmental Monitoring, Health Phys., Vol. 26, p. 145.

73. Spitz, Henry and Wrenn, McDonald E. (1977). Design and Applications of a Continuous, Digital-Output, Environmental Radon Measuring Instrument, in Workshop on Radon, U.S. ERDA Report HASL-325, p. 48.

74. Countess, Richard J. and Thomas, Jess W. (1977). Radon Flask Monitor, U.S. ERDA Report HASL-330.

75. Countess, Richard J. (1977). Measurement of Rn-Flux with Charcoal Canisters, in Atomic Industrial Forum Program Reports, Vol. 13(9), p. 139.

76. Lucas, Henry F., Jr. (1977). Alpha Scintillation Radon Counting, in Atomic Industrial Forum Program Reports, Vol. 3.

77. George, Andreas C. (1976). Scintillation Flasks for the Determination of Low-Level Concentrations of Radon, proc. Ninth Midyear Topical Symposium of Health Physics Society, p. 112.

78. Kristan, Janez and Kobal, Ivan (1973). A Modified Scintillation Cell for the Determination of Radon in Uranium Mine Atmosphere, Health Phys., Vol. 24, p. 103.

79. Thomas, Jess W. (1974). Evaluation of Activated Carbon Canisters for Radon Protection in Uranium Mines, HASL-280, U.S. Atomic Energy Commission.

80. Present R. D. (1958). Kinetic Theory of Gases, McGraw-Hill Book Company, New York, New York.

81. Jacobi, W. (1972). Activity and Potential α-Energy of ^{222}Rn and ^{220}Rn Daughters in Different Air Atmospheres, Health Phys., Vol. 22, p. 441.

82. Krisiuk, E. M. (1980). Airborne Radioactivity in Buildings, Health Phys., Vol. 38, p. 199.

83. Raabe, Otto G. (1969). Concerning the Interactions that Occur Between Radon Decay Products and Aerosols, Health Phys., Vol. 17, p. 177.

84. Wilkening, M. H. (1952). Natural Radioactivity as a Tracer in the Sorting of Aerosols According to Mobility, Review of Scient. Instr., Vol. 23, p. 13.

85. Wrenn, McDonald E., Rosen, Jerry C., and Van Pelt, Wesley R. (1969). Steady State Solutions for the Diffusion Equations of Radon-222 Daughters, Health Phys., Vol. 16, p. 647.

86. Clough, W. S. (1973). Transport of Particles to Surfaces, J. Aerosol Science, Vol. 4, p. 227.

87. Möller, U. and Schumann, G. (1970). Mechanism of Transport from the Atmosphere to the Earth's Surface, J. Geophys. Research, Vol. 75, p. 3013.

88. Landau and Levich (1962). Cited in Reference 87.

VITA

Mahmoud M. Abdelrazek was born in Kom-Elnoor, Egypt, on November 4, 1938. He attended Kom-Elnoor public school and graduated in 1958. He entered the faculty of science, Cairo University, majoring in physics and geology and received his B.S. degree in July 1963. He started to work for the Egyptian Atomic Energy Commission in December 1963 as a scientific member of the Radiation Protection Division. In 1964, in addition to his job, Abdelrazek entered Cairo University for one year and received a diploma in radiation physics in 1965. In the same year he started his graduate studies at Cairo University and was awarded the M.S. degree in nuclear physics in August 1968. He continued to work for the Egyptian Atomic Energy Commission until 1972 when he was granted a 16-month fellowship from the International Atomic Energy Agency, Vienna. On February 15, 1972, he came to the United States and joined the Health Physics Division of Oak Ridge National Laboratory (ORNL) for the term of his fellowship. At the end of his fellowship in the fall 1973 he started his Ph.D. program at The University of Tennessee, Knoxville. During his graduate studies he worked as a Graduate Teaching Assistant in the Departments of Physics, Engineering Science and Mechanics, and Mathematics. Abdelrazek was employed several times (as consultant, part time or full time) by ORNL from 1973 to 1980. He is the author or co-author of 14 publications. He is married to Laila M. Emara, and has two daughters, Nermeen and Margie, and a son, Ahmad.