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The 1991 International Symposium on Radon and Radon Reduction Technology

Volume III: Measurement Methods

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Opening Presentation

Session III: Measurement Methods

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COMPARATIVE DOSIMETRY OF RADON IN MINES AND HOMES: AN OVERVIEW OF THE NAS REPORT

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ABSTRACT

The findings of the recent report by a National Academy of Sciences panel on radon dosimetry are reviewed. The committee was charged with comparing exposure-dose relations for the circumstances of exposures in mines and homes. The community first obtained data on the various parameters included in dosimetric lung models and then selected values that it judged to be best supported by the available evidence. Dosimetric modeling was used to calculate the ratio of exposure to radon progeny to dose of alpha energy delivered to target cells for various scenarios. The committee's modeling shows that exposure to radon progeny in homes delivers a somewhat lower dose to target cells than exposure in mines; this pattern was found for infants, children, men, and women.

The work described in this paper was not funded by the U.S. Environmental Protection Agency and therefore the contents do not necessarily reflect the views of the Agency and no official endorsement should be inferred.

INTRODUCTION

Radon, an inert gas, is a naturally occurring decay product of radium-226, the fifth daughter of uranium-238. Radon decays with a half-life of 3.82 days into a series of solid, short-lived progeny; two of these progeny, polonium-218 and polonium-214, emit alpha particles. When radon progeny are inhaled and these alpha emissions occur within the lungs, the cells lining the airways may be injured and damage to the genetic material of the cells may lead to the development of cancer.

Radon has been linked to excess cases of lung cancer in underground miners since the early decades of the twentieth century. Epidemiologic evidence on radon and lung cancer, as well as other diseases is now available from about 20 different groups of underground miners (1,2). Many of these studies include information on the miners' exposure to radon progeny and provide estimates of the quantitative relation between exposure to progeny and lung cancer risk (2,3); the range of excess relative risk coefficients, describing the increment in risk per unit of exposure is remarkably narrow in view of the differing methodologies of these studies (2).

As information on air quality in indoor environments was collected during the last 20 years, it quickly became evident that radon is ubiquitous indoors and that concentrations vary widely and may be as high as levels in underground mines in some homes. The well-documented and causal association of radon with lung cancer in underground miners appropriately raised concern that radon exposure might also cause lung cancer in the general population. The risk of indoor radon has been primarily assessed by using risk assessment approaches that extend the risks found in the studies of miners to the general population. Risk models that can be used for this purpose have been developed by committees of the National Council on Radiation Protection and Measurements (NCRP) (4), the International Commission on Radiological Protection (5) (1987), and the National Academy of Sciences (Biological Effects of Ionizing Radiation (BEIR) IV Alpha Committee) (1).

Extrapolation of the lung cancer risks in underground miners to the general population is subject to uncertainties related to the differences between the physical environments of homes and mines, the circumstances and temporal patterns of exposure in the two environments, and potentially significant biological differences between miners and the general population (Table 1). A number of these factors may affect the relation between exposure to radon progeny and the dose of alpha-particle energy delivered to target cells in the tracheobronchial epithelium; these factors include the activity-aerosol size distribution of the progeny, the ventilation pattern of the exposed person, the morphometry of the lung, the pattern of deposition and the rate of clearance of deposited progeny, and the thickness of the mucous layer lining the airways.

The activity-aerosol size distribution refers to the physical size distribution of the particles containing the alpha activity. The term "unattached fraction" has historically been applied to progeny existing

models that it judged to be best supported by the available evidence. The committee then utilized a dosimetric model, developed in part by the Task Group of the International Commission for Radiological Protection, to compare exposure-dose relations for exposure to radon progeny in homes and in mines. While the report provides the exposure-dose figures, the committee expressed its principal findings as a ratio, termed K in the BEIR IV report (1). K, a unitless measure, represents the quotient of the dose of alpha energy delivered per unit of exposure in a home to the dose per unit exposure for a male miner exposed in a mine. If the K factor exceeds unity, the delivered dose per unit exposure is greater indoors whereas if it is less than unity, the delivered dose per unit exposure is less indoors.

Factors other than lung dosimetry of radon progeny also introduce uncertainty in extrapolating risks from the studies of underground miners to the general population. The committee briefly reviewed the evidence on cigarette smoking, tissue damage, age at exposure, sex, and exposure pattern. These sources of uncertainty were considered in a qualitative rather than a quantitative fashion.

THE COMMITTEE'S FINDINGS

The committee selected several different sets of exposure conditions in homes and in mines (Table 2,3). The mining environment includes the areas of active mining, the haulage drifts, and less active and dusty areas such as lunch rooms. In some analyses, the values for active mining and haulage ways were averaged to represent typical conditions. Separate microenvironments considered in the home included the living room and the bedroom. Parameters for the living room and the bedroom were averaged to represent a typical scenario for the home. The effects of cooking and cigarette smoking on radon progeny aerosol characteristics were also considered. While the contrast between the home and mining environments was somewhat variable across the scenarios, homes were characterized as having greater unattached fractions and smaller particles. Higher average minute volumes were assumed for the mining environment (Table 2,3).

The committee also examined uncertainties associated with other assumptions in the dosimetric model. Doses to basal and secretory cells in the tracheobronchial epithelium were calculated separately, because all types of cells with the potential to divide were considered to be potential progenitor cells for lung cancer. The committee also compared the consequences of considering: lobar and segmental bronchi rather than all bronchi as the target; radon progeny as insoluble or partially soluble in the epithelium; of breathing through the oral or nasal route exclusively; of varying the thickness of the mucus lining the epithelium and the rate of mucociliary clearance; and cellular hyperplasia leading to thickening or injury causing thinning of the epithelium.

Across the wide range of exposure conditions and exposed persons considered by the committee, most values of K were below unity (Table 4). For both secretory and basal cells, K values indicated lesser doses of alpha energy per unit exposure, comparing exposures of infants,

as ions, molecules, or small clusters; the "attached fraction" designates progeny attached to ambient particles (6). Using newer methods for characterizing activity-aerosol size distributions, the unattached fraction has been identified as ultrafine particles in the size range of 0.5 to 3.0 nm (6). Typically, mines have higher aerosol concentrations than homes and the unattached fraction would be expected to be higher in homes than in mines. Because of differing sources of particles in the two environments, aerosol size distributions could also plausibly differ between homes and mines.

The physical work involved in underground mining would be expected to increase the amount of air inhaled in comparison with the generally sedentary activities of time spent at home. The greater minute ventilation of miners would result in a higher proportion of the inhaled air passing through the oral route, in comparison with ventilation during typical activities in residences. The physical characteristics of the lungs of underground miners, almost all adult males, differ significantly from those of infants, children and thickness of the epithelial layer could also plausibly differ, comparing miners with the general population, because of the chronic irritation by dust and fumes in the mines.

Methods are available for characterizing the effects of these factors on the relation between exposure to radon progeny and the dose of alpha energy delivered to target cells in the respiratory tract. Using models of the respiratory tract, the dose to target cells in the respiratory epithelium can be estimated for the circumstances of exposure in the mining and indoor environments. One of the recommendations of the 1988 BEIR IV Report (1) was that "Further studies of dosimetric modeling in the indoor environment and in mines are necessary to determine the comparability of risks per WLM [working level month] in domestic environments and underground mines". The BEIR IV Report had included a qualitative assessment of the dosimetry of progeny in homes and in mines, but formal modeling was not carried out.

Consequently, the U.S. Environmental Protection Agency asked the National Research Council to conduct a study addressing the comparative dosimetry of radon progeny in homes and in mines. This paper reviews the findings of the recently published report of the committee (Panel on Dosimetric Assumptions Affecting the Application of Radon Risk Estimates). The panel was constituted with the broad expertise, covering radon measurement and aerosol physics, dosimetry, lung biology, epidemiology, pathology, and risk assessment, needed for this task.

THE COMMITTEE'S APPROACH

To address the charge of undertaking further dosimetric modeling, the committee obtained data on the various parameters included in dosimetric lung models that contributed to uncertainty in assessing the risk of indoor radon. The committee not only reviewed the literature, but obtained recent and unpublished information from several investigators involved in relevant research. After completing this review, the committee selected values for parameters in dosimetric

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children, men and women in homes with exposures of male miners underground. While the highest values of K were calculated for children, the values for children did not exceed unity, suggesting that children exposed to radon progeny are not at greater risk for lung cancer on a dosimetric basis.

The committee explored the sensitivity of the K factors to underlying assumptions in the dosimetric model. The general pattern of the findings was comparable for secretory and basal cells. The K factors remained below unity regardless of whether the radon progeny were assumed to be insoluble or partially soluble in the epithelium. The K factor was also not changed substantially with the assumption that lobar and segmental bronchi, rather than all bronchi, are the target. Assumptions regarding breathing route also had little impact. After the committee had completed its principal analysis, new data became available suggesting that recent higher values for nasal deposition reported by Cheng et al. (7) might be preferable to lower values from the 1969 report of George and Breslin (8); other new evidence suggested that a value of 0.15 um should be used for aerosol size in the haulage drifts. Inclusion of these two modifications of the committee's preferred parameter values in the dosimetric model reduced the values of K by about 20 percent.

The committee did not attempt to reach quantitative conclusions concerning sources of uncertainty not directly addressed by the dosimetric modeling. It noted the paucity of data on such factors as cigarette smoking, age at exposure and particularly the effect of exposure during childhood, and exposure pattern. The evidence on these factors received detailed review in the BEIR IV report (1) and the present committee did not reach any new conclusions on these sources of uncertainty. The committee also commented on the potential effects of the miners' exposures to dust and fumes while underground. Increased cell turnover associated with these exposures may have increased the risk of radon exposure for the miners.

SUMMARY

The Panel on Dosimetric Assumptions Affecting the Application of Radon Risk Estimates comprehensively reviewed the comparative dosimetry of radon progeny in homes and in mines. The committee's modeling shows that exposure to radon progeny in homes delivers a somewhat lower dose to target cells than exposure in mines; this pattern was found for infants, children, men, and women. This finding was not sensitive to specific underlying assumptions in the committee's modeling. Assuming that cancer risk is proportional to dose of alpha energy delivered by radon progeny, the committee's analyses suggests that direct extrapolation of risks from the mining to the home environment may overestimate the numbers of radon-caused cancers.

TABLE 1. POTENTIALLY IMPORTANT DIFFERENCES BETWEEN EXPOSURE TO RADON IN THE MINING AND HOME ENVIRONMENTS*

Physical Factors	
Aerosol characteristics	Greater concentrations in mines; differing size distributions
Attached/unattached fractions	Greater unattached fraction in homes
Equilibrium of radon/decay products	Highly variable in homes and mines
Activity Factors	
Amount of ventilation	Probably greater for working miners than for persons indoors
Pattern of ventilation	Patterns of oral/nasal breathing not characterized, but mining possibly associated with greater oral breathing
Biological Factors	
Age	Miners have been exposed during adulthood; entire spectrum of ages exposed indoors
Gender	Miners studied have been exclusively male; both sexes exposed indoors
Exposure pattern	Miners exposed for variable intervals during adulthood; exposure is lifelong for the population
Cigarette smoking	The majority of the miners studied have been smokers; only a minority of U.S. adults are currently smokers

*Taken from Table 1-2 in reference (6).

TABLE 2. ASSUMPTIONS FOR EXPOSURE SCENARIOS ASSUMED FOR MINES AND HOMES*

SUMMARY OF RADON PROGENY AEROSOL CHARACTERISTICS ASSUMED TO REPRESENT EXPOSURE CONDITIONS IN MINES AND HOMES

Exposure Scenario	f_p	AMD of Room Aerosol (μm)	AMD of Aerosol in respiratory tract (μm)
Mine			
Mining	0.005	0.25	0.5
Haulage drifts	0.03	0.25	0.5
Lunch room	0.08	0.25	0.5
Living Room			
Normal	0.08	0.15	0.3
Smoker - average	0.03	0.25	0.5
- during smoking	0.01	0.25	0.5
Cooking/vacuuming	0.05	0.02/0.15+ (15%/80%)	0.02/0.3 (15%/80%)
Bedroom			
Normal	0.08	0.15	0.3
High	0.16	0.15	0.3

*Based on Tables 3-1 and 3-2 in reference 6.

+The radon progeny aerosol produced by cooking/vacuuming has three size modes; 5% of potential alpha energy is unattached, 15% has an AMD of 0.02 m, and 80% has an AMD of 0.15 μm. The 0.02 μm AMD mode is hydrophobic and does not increase in size within the respiratory tract.

TABLE 3. ASSUMPTIONS FOR EXPOSURE SCENARIOS ASSUMED FOR MINES AND HOMES*

LEVELS OF PHYSICAL EXERTION AND AVERAGE MINUTE VOLUMES ASSUMED FOR UNDERGROUND MINERS AND FOR ADULTS IN THE HOME

Exposure Scenario	Level of Exertion	Average $\dot{V}_E$ (liters/min) Man	Woman
Underground Mine			
Mining	25% heavy work/75% light work	31	--
Haulage way	100% light work	25	--
Lunch room	50% light work/50% rest	17	--
Home-Living Room			
Normal and smoker	50% light work/50% rest	17	14
Cooking/vacuuming	75% light work/25% rest	21	17
Home-Bedroom			
Normal and high	100% sleep	7.5	5.3

*Based on Tables 3-1 and 3-2 in reference 6.

TABLE 4. SUMMARY OF K FACTORS FOR BRONCHIAL DOSE CALCULATED FOR NORMAL PEOPLE IN THE GENERAL ENVIRONMENT RELATIVE TO HEALTHY UNDERGROUND MINERS*

Subject Category	K Factor for Target Cells	
	Secretory	Basal
Infant, age 1 month	0.74	0.64
Child, age 1 year	1.00	0.87
Child, age 5-10 years	0.83	0.72
Female	0.72	0.62
Male	0.76	0.66

*Taken from Table 5-1 in reference 6.

Session III:

Measurement Methods

CURRENT STATUS OF GLASS AS A RETROSPECTIVE RADON MONITOR

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ABSTRACT

Measurement of alpha activity on household glass surfaces has developed from an interesting idea into an attractive technique for (1) indoor radon screening and (2) improving estimates of long-term radon exposures for epidemiological studies. Early glass samples, which spanned a narrow range of radon exposures, displayed a positive correlation between exposure and surface activity. However, the age and exposure history of these early samples is uncertain. We now have calibration data from 33 pieces of glass with known exposures between 0 to 2000 kBq yrs m^{-3} and surface alpha activities as high as 2000 Bq m^{-2}. Glass has been exposed in four independent radon chambers, including the EPA Las Vegas laboratory. We also have samples from four houses, two from Minnesota, one from New York, and one from New Jersey.

There is a strong positive correlation between the exposure and surface activity, with a best-fit coefficient of variation (COV) of 70%. The calibration curve fits the house samples with a COV of 80%—somewhat larger owing to indeterminate exposure of the glass. Because indoor radon can have large spatial and temporal variability (COVs from 80% to 150% in Minnesota), we suggest that surface alpha activity is currently the best technique to acquire long-term averages of indoor radon.

Activity on glass can be measured rapidly using semiconductor or pulse ionization spectroscopy and inexpensively by track-registration detectors. These measurements provide reproducible techniques to screen homes for radon because they are insensitive to short-term radon fluctuations that confound present screening techniques. Glass surface activity is an ideal monitor for use in epidemiological studies since it can integrate radon daughter activity over decades. The method can properly average short-term radon fluctuations and radon changes due to structural alterations, and it has the ability to track exposures on glass that has been in more than one radon environment.

INTRODUCTION

A recurring dilemma for agencies that provide guidance on acceptable levels of indoor radon, and for epidemiologists trying to assess the effects of exposure to radon and radon daughter products, is that current measurement techniques collect data for periods of less than one year (1), and often for not more than two days (2). Current radon screening procedures produce relatively poor long-term radon estimates since they are subject to relatively large errors due to natural or induced variations within the home environment. These variations occur on the same time scales as the measurement protocols (3) and can be significantly greater than a factor of two (4). Additional error is introduced by assuming a fixed relationship between the measured radon and the radon daughter product concentrations. As a result, short-term screening measurements are not sufficiently accurate to assess the possible health impact of indoor radon exposure (5). Year-long measurements, although better, are still subject to long-term variability, can take a year to acquire, and may not accurately reflect lifetime exposures. The above mentioned problems illustrate the limited potential of the data to be used for epidemiological studies. Current measurement protocols also do not take into account possible exposure in the workplace or changes in housing environments over a life-time.

In 1987 Lively and Ney (6), reporting on a study of surface alpha radioactivity from radon daughter products, proposed that surfaces such as glass could be used to estimate an integrated ^{222}Rn concentration within a room. Samuelsson (7), citing the technique as a retrospective radon exposure meter, measured the activity on six pieces of household glass and reported a good correlation between surface alpha activity and radon exposure (0.5 to 9 kBq yrs m^{-3}). We report here on the correlation and reproducibility of glass surface activity with samples that were exposed in radon chambers for between 0.5 to 2,000 kBq yrs m^{-3}. We also compare these results with measurements from glass in several homes.

Surfaces can be used as radon monitors because they are passive collectors of radon daughter products. The long-lived radon decay product ^{210}Pb ($t_{1/2}$ = 22 y), which is followed by an alpha emitting isotope, ^{210}Po ($t_{1/2}$ = 138 d), will therefore display a relationship to the concentration of the radon source. In this paper we are attempting to understand and define the relationship between activity and exposure for simple environments in radon chambers with known radon concentrations and low aerosol contents. If the relationship is determinate for simple situations, it will then be possible to extend the monitoring to more complex environments, such as those in most homes, with some hope of understanding the results.

Glass is an appropriate surface for retrospective radon analysis because it is readily available, it can be recovered from a variety of environments and exposure histories, it tends to have a low intrinsic background, and it is nonporous and impermeable. The latter characteristics are important to accurate measurement of the alpha spectra. On porous, aged surfaces, such as gypsum board or plaster, a significant fraction of ^{210}Pb has diffused off the surface or beyond the range of alpha particle emission from ^{210}Po (6). In glass, however, ^{210}Pb has a very short diffusion length, estimated at 1 micron in twenty years (8). The tailing and FWHM of ^{210}Po alpha spectra from a piece of glass exposed for two weeks was the same as the spectra from a piece of glass exposed for more than thirty years.

Surfaces collect radon daughters at all stages in the decay chain between ^{222}Rn and ^{210}Pb. Some of the daughters are attached to aerosol particles and some exist as "unattached ions" or "ultrafine particles." Recoil of a parent atom by emission of an alpha particle can embed the daughter atom in the glass. Depths of recoil implantation, based upon studies of 100 keV ions, are estimated to be 0.04 ± 0.007 microns (9). Vanmarcke (10) presented a model for determining the fraction of daughters that embedded in a surface. He observed that the fraction embedded is in part controlled by the geometry of the decay/recoil couple and by the aerosol content of the room, which affects the attached-to-unattached ratio. We found early in our studies that if the glass was very dirty, up to half of the surface activity could be removed by washing. Analysis of glass from "clean" environments indicates that the removable fraction can be less than 10%.

METHODOLOGY

The calibration data in Figure 1 was obtained by exposing new pieces of glass in both static and dynamic radon atmospheres for known intervals. Multiple exposures from a single type of environment reduces the uncertainty associated with aerosols and exposure history. We used three of our own radon chambers: two were Radium Ore Revigators with internal volumes of about 5 liters and equilibrium radon activities between 3,700 to 9,000 kBq m^{-3}; the third was a large volume radon chamber (1 m^3) with higher humidity and air velocities. Several pieces of glass were also sent to the EPA Las Vegas radon laboratory for exposure in the radon chamber. The integrated exposure times from these chambers ranged from a few days to several months, simulating decades of exposure at lower radon levels.

The surface alpha activity was measured with surface-barrier alpha detectors and 10 cm^2 pieces of glass cut from 160 cm^2 exposed samples. The detectors have a high signal-to-noise ratio and good resolution and sensitivity for ^{210}Po. Count times range from three to twelve days. All of the measurements were on

washed surfaces after the short-lived daughters had decayed. The detectors were calibrated, and counting efficiencies were determined using NBS traceable ^{210}Po and ^{241}Am solid sources.

Low levels of surface alpha activity mean that background activities may become very important and should be evaluated. Uranium in the glass can result in ^{210}Po activity that is unrelated to the surface deposition of radon daughter products, and there is the possibility that glass may be exposed to radon prior to emplacement in the environment of interest. Our measurements to date, over an energy range of 4-8 MeV, have detected no activity other than that related to ^{210}Po. We also tested for ^{210}Po in the glass by remeasuring several low ^{210}Po activity samples after etching the surface with hydrofluoric acid. Polonium-210 count rates from these samples were indistinguishable from the detector background, indicating that there was no supported ^{210}Po activity in the glass. A nonexposed piece of glass from the group used for several low activity exposures was measured both with and without etching. The glass background activities did not differ from each other or from the detector background, indicating that there was no measurable preexposure activity on that surface. Although these results may not apply to all glass, they do indicate that for the samples tested, all of the measured alpha activity on the glass surface can be attributed to deposited ^{210}Po.

RESULTS AND DISCUSSION

We have measured the surface alpha activity from 37 pieces of glass that were exposed in four different radon chambers and four homes. The results have been corrected for disequilibrium between ^{210}Po and ^{210}Pb and for the decay of ^{210}Pb since the beginning of the exposure. The correction for ^{210}Pb decay is dominant for glass more than five to six years old. Activities were measured to better than 10% counting statistics; however, nonsystematic variations of activity on different areas of the same glass average 25%. Exposure times for the chamber samples were known, but the concentrations of radon in the chambers have a measurement error due to unsampled temporal variations. Exposure errors for the houses combined the best estimates of radon levels based on short-term radon measurements and the length of time the glass was in place.

The results (Figure 1 and Table 1) expand the range of exposures and measured activities to cover four orders of magnitude (up to 2,000 Bq m^{-2} activity and 2,000 kBq y m^{-3} exposure). We have presented the data in Figure 1 as a log/log plot to display the complete range of data.

Linear regression analysis (ordinary least squares—OLS) shows a strong positive linear correlation between exposure and surface activity: $R=0.97$, $p<0.001$. However, because we have estimates of uncertainties associated with both exposure and

surface activity, the best fit line shown in Figure 1 was calculated by using a weighted least squares fit (WLS) that more accurately reflects the influence of x-y errors at low activities and exposures. Although the scatter in the data exceeds our uncertainty estimates, the best WLS fit has a COV of 70%. This COV is similar in size to COVs for short-term Rn screening measurements and long-term Rn variations in houses(4).

That we can fit two different curves to data below exposures of 50 Bq m^{-2} results from glass samples within two environments, one static and one dynamic. Nine pieces of glass in Figure 1 were exposed for up to 15 kBq yr m^{-3} in a chamber where air velocities approach 2 m s^{-1} (clearly much breezier than the inside of most homes). In this environment, the slope of the best WLS calibration line is three times smaller than that obtained from the glass samples exposed in static air. The lower slope indicates that under dynamic air flow up to three times more radon daughter products were deposited on the surface during similar exposure intervals. In most homes, we expect that the daughter product deposition velocity would would be between static and dynamic conditions. A simple adjustment can therefore be made to the WLS calibration curve to account for indoor air flow estimates or measurements.

When the calibration equation is applied to the four different household glass samples, a COV of 80% is obtained, which is very similar to the glass calibration COV of 70%. This implies that aerosols and cleaning histories may not significantly affect the relationship between glass surface activity and exposure. When collecting data from homes, the primary limitation on the effectiveness of the technique is the uncertain surface exposure time, and that uncertainty is the major contributor to the COV.

Because of the large spatial and temporal variability of indoor radon (COVs from 80% to 150% in Minnesota), long-term estimates of radon and radon daughter concentrations based on short-term measurements are inadequate. Deposition of daughter products on surfaces represents an integrated measurement that can provide useful data over exposure intervals of approximately seventy years and a very wide range of radon concentrations. Our calibration data indicate that air flow does influence the deposition velocity of the daughter products. To obtain the most accurate long-term estimate of radon from the best-fit calibration, an evaluation of air movement during an exposure interval should be attempted.

Radioactivity on glass can be measured over a few hours or days using semiconductor (this paper) or pulse ionization (7) spectroscopy. In addition, inexpensive track-registration detectors can be used to measure the glass surface activity over a period of one year while at the same time measuring the radon concentration in the adjacent room. These types of measurements

provide rapid, reproducible analyses that can be used to screen homes for radon because they are insensitive to short-term radon fluctuations and tampering that can confound present screening techniques. Alpha activity on glass surfaces is an ideal monitor for use in epidemiology since it can integrate radon daughter activity over decades, and the variations from a best-fit line over a wide range of calibration data are less than a factor of two. The method properly averages short-term radon fluctuations, radon changes due to structural alterations, and it has the ability to track exposures on glass that has been in more than one radon environment. As a result, we believe that surface alpha activity is an attractive and probably the best available technique to assess the long-term indoor radon environment.

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TABLE 1. LINEAR REGRESSION ESTIMATES FOR SLOPE AND INTERCEPT

Method	Intercept estimate	Slope estimate
Ordinary least squares*	38 ± 12	1.4 ± 0.1
Weighted least squares*	-2.5 ± 0.3	1.7 ± 0.2
WLS — Static chambers†	-3.0 ± 0.1	1.9 ± 0.2
WLS — Dynamic chamber†	-0.1 ± 0.1	0.6 ± 0.2

*Full range of calibration activities.
†Activity range 0 — 50 Bq m^{-2}.

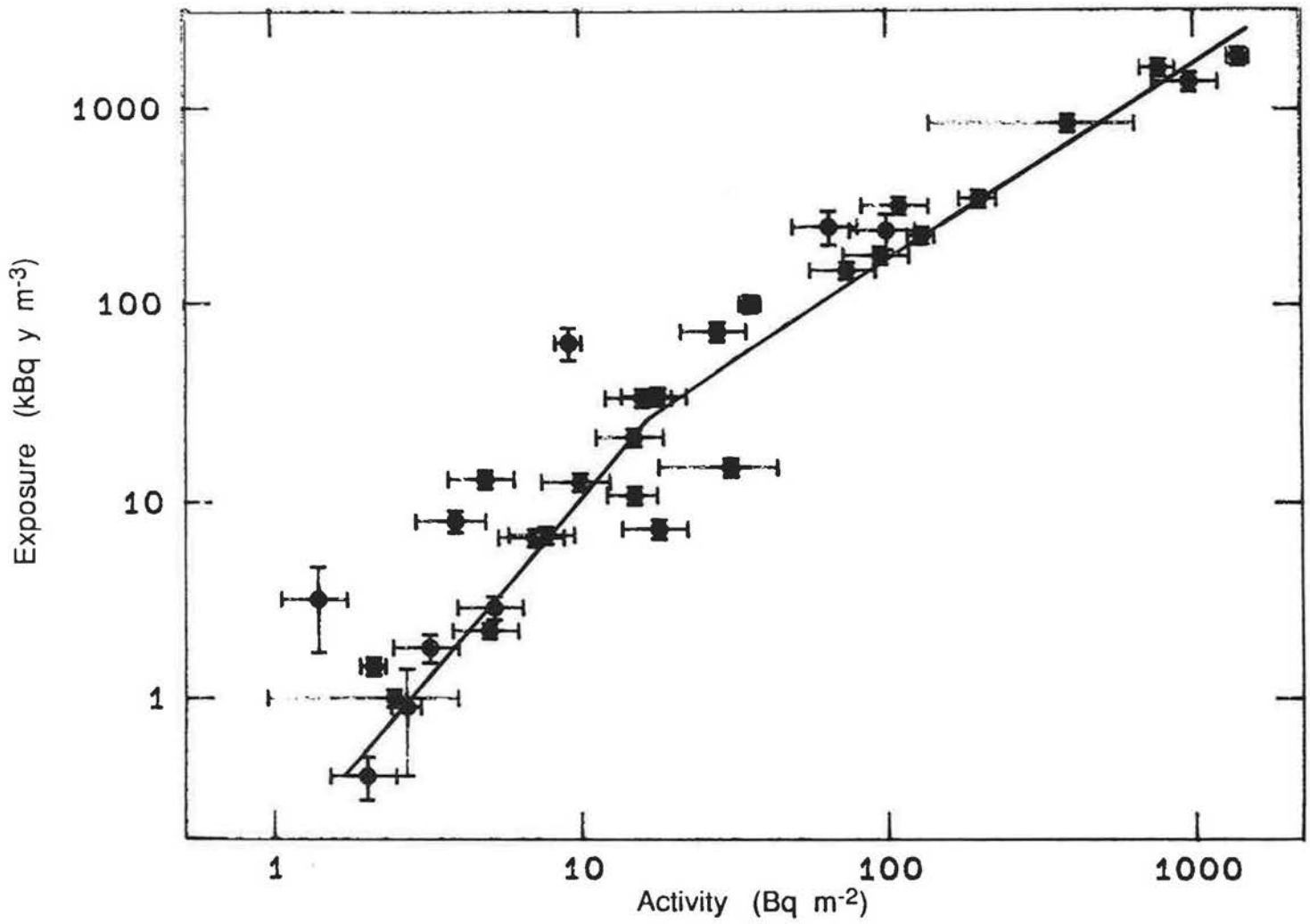

Figure 1. Log/log plot of exposure versus activity on calibration-glass surfaces. The curve is obtained from the weighted least squares equation in Table 1.

SOIL GAS MEASUREMENT TECHNOLOGIES

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ABSTRACT

A wide range of methods for characterizing the radon potential of land areas have evolved over the last decade through research programs in this country and abroad. Ideally, methods would provide information on radon production in the soil, as well as diffusion and permeability. This can be accomplished through various combinations of direct measurements and theoretical assumptions. Basic technologies concentrate on measuring: (1) radon in soil gas, (2) radon flux from the surface, or (3) radium content of the soil. Approaches may also include attendant measures of soil characteristics and other factors to support predefined indexes of radon potential.

Basic measurement approaches for radon potential are reviewed in terms of the following technical issues: measurement parameters, field/laboratory methodologies, quality assurance, and model concepts applied to data to estimate radon potential. Theoretical aspects of these factors are also considered. Radium-based measurements, for example, have the distinct advantage of being suited to testing water-saturated soils, but may require weeks to deliver results. Soil gas and flux measurements, on the other hand, generally fail to obtain samples from saturated soils because the gas volume is nearly zero, but approaches exist to deliver.prompt results on site. If time is of the essence, then, recognition factors to avoid generally saturated conditions are necessary to ensure sample validity.

This paper has been reviewed in accordance with the U.S. Environmental Protection Agency's peer and administrative review policies and approved for presentation and publication.

INTRODUCTION

Soil gas entry is generally regarded as a major source of indoor radon. Consequently, attention has been directed to developing the means to evaluate radon potential based on soil factors. National- and regional-scale maps consolidating radiometric, geologic, and soil information were developed to display broad trends (Wilson 1984; Duval et al. 1989). Studies to establish site-specific methods geared to finer detail through direct measurement have also been underway.

Early efforts in Sweden to devise site-specific methods recognized the importance of air permeability of the soil and moisture conditions in determining radon concentrations in soil gas and pressure-driven transport into buildings, leading to development of a series of in-situ measurements to complement exhalation tests in the laboratory (Akerblom et al. 1984; Lindmark and Rosen, 1985). Based on model assumptions, a regional classification could be considered to define class intervals of radon potential based on air permeability of the soil and soil gas (Swedjemark 1986).

It also became clear that the building itself exerts perhaps the strongest influence on radon entry by (1) creating the pressure forces to drive radon entry and (2) presenting the cracks, joints, and service penetrations that form radon entry pathways. Consequently, soil-based measurements for indoor potential must

encompass two levels of analysis: one to evaluate radon production and mobility in the soil, a second to evaluate transport into the building. Most indexes of radon potential, such as the various permutations of the Radon Index Number (RIN) and the Radon Availability Number (RAN), are formulated to define the radon that is available for transport, but do not explicitly consider radon entry into the building. Additional considerations would be required to account for differing entry routes and building pressures that occur with slab-on-grade, crawl space, and basement configurations.

Continuing work by Nazaroff and Sextro (1989) is headed in this direction. They have derived a generalizable framework that integrates soil factors with building factors. Although this latter approach requires specification of radon entry pathways, it provides a quantitative means to judge the radon potential of land areas in terms of the total effect. In the State of Florida, numerical modeling is being employed in this context (Rogers and Nielsen 1990).

This paper presents a general overview of the basic technologies applied to estimating radon potential, concentrating on measurement strategies for radium content, soil gas radon, and radon flux.

MEASUREMENT STRATEGIES

Quantitative estimates of radon potential are predicated from (1) a soil volume in flow communication to the building, (2) a supply of radon to the pore spaces of the soil, and (3) transport mechanisms to convey radon into the building. The situation is complicated by a number of factors arising from soil characteristics and environmental influences in addition to the effects of the building. Measurement strategies hinge on detecting radioactivity and other characteristics of a known sample volume (or mass) whose history has been controlled to represent one or more processes germane to the production and migration of radon in the soil.

As summarized in Figure 1, radium content is measured by isolating a defined volume of soil to retain the emanating fraction. At radioactive equilibrium, the activity concentration of radon and radon progeny is equated with the radium concentration. Soil gas measurements, on the other hand, seek to isolate radon in the pore spaces without affecting emanation or transport. Flux-based measurements rely on natural or induced transport through the soil column to deliver the radon to a sampling volume defined over a specified area of the soil.

Measurement strategies may involve in situ techniques wherein the sampling volume and detection volume coincide in the soil environment as well as extraction techniques wherein the sample is removed from the soil matrix for analysis. Procedures exist to provide point-in-time as well as time averaged and continuous measurements.

Analysis of soil-derived samples makes use of the same detectors that are commonly applied to air and water samples. The decay series from radon, through its short-lived decay products, releases three alphas (Rn-222, Po-218, Po-214) and two gammas (Pb-214, Bi-214). For samples that can be delivered to the detector in the gas phase, alpha detection is generally preferred. Samples that immobilize radon in a solid matrix (e.g., bulk soil samples, radon adsorbed onto activated carbon) are generally analyzed using gamma spectrometry.

As a rule, additional steps are required to estimate other factors such as permeability. These additional steps, however, do not necessarily entail new measurements. In developing county-scale maps of radon potential, Gundersen et al. (1988) and Otton et al. (1988) utilized water permeability data compiled by the Soil Conservation Service as a means to define classes of gas permeability. Yokel (1989) presents a detailed summary of theoretical and empirical relationships that can be used to estimate gas permeability from soil characteristics and water permeability.

In a similar vein, soil moisture could be characterized from climatological principles (Rose et al. 1990; Thornthwaite and Mather 1955). While such estimators are unlikely to approach the accuracy of standard determinations of soil moisture (e.g., ASTM D-2216), accuracy may be acceptable because underlying models are still required to extrapolate measurements to other conditions representative of seasonal or annual levels. Similarly, Rogers and Nielson (1990) cite ongoing work to estimate gas permeability and radon diffusion coefficients from standard data sources.

RADIUM CONTENT

Basic approaches for measuring the radium content of soils are summarized in Table 1. Conventional measurements of radium content involve sealing a dried soil sample in a leakproof container, storing the sealed sample for a long enough period of time to establish radioactive equilibrium, and analyzing for radionuclides of interest using gamma spectroscopy. Protocols frequently accommodate concurrent analysis of moisture content, laboratory estimates of radon emanation, and other analyses by subdividing field samples. This is the basic procedure adopted by the Florida Radon Research Program (Williamson and Finkel 1990) and utilized in other large-scale radon studies (e.g., Kunz 1989), as well as standardized radiological surveys (Myrick et al. 1983).

The general laboratory procedure is geared to a single analysis at radioactive equilibrium. An intermediate count is sometimes included a few hours after sealing the dried sample to measure the level of nonemanating radon in order to estimate the emanation fraction under standard conditions.

The prompt bismuth technique of Stief et al. (1987) features repeated analyses over shorter timeframes to evaluate the secular equilibrium between radium and radon. In this approach, the sample is sealed under field conditions and count-rate measurements are initiated very soon thereafter to establish the basis for extrapolating subsequent measurements back to the time of collection. The prompt bismuth method is directed primarily at evaluating soil gas radon; adaptations could be considered to estimate radium concentrations in timeframes of 24 hours or less. While the prompt bismuth method has shown promising results based on developmental tests and early applications, it has not entered widespread use, and warrants comparative testing against standard techniques to establish general reliability.

SOIL GAS MEASUREMENTS

Basic technologies for measuring radon concentrations in soil gas have evolved along three complementary pathways: (1) gas extraction from depth using hollow tubes, (2) analysis of bulk soil samples, and (3) in situ detectors.

Soil Gas Extraction

A number of field investigations have utilized soil gas extraction techniques to evaluate radon concentrations in soil gas. Leading approaches are summarized in Table 2. In many cases procedures incorporate measurement of permeability using an approach originally suggested by Scott (DSMA 1983). The basic approach involves driving a hollow probe to the desired depth, connecting the upper end of the probe to a vacuum source, and monitoring pressure and flow rate through the probe. Permeability is calculated from the ratio of flow to pressure with adjustments for a probe-specific calibration factor.

The reconnaissance probe described by Reimer (1990) is a relatively simple system consisting of a small-diameter (6- to 9-mm) thick-walled carbon-steel tube that is driven to sampling depth (75 cm, nominal) using a slide hammer. The relatively small probe volume (3 cm^3) allows for purging and sample collection using a hypodermic syringe for subsequent analysis using alpha scintillation. This method has been applied through a number of field surveys (Schumann and Owen 1988; Gundersen et al. 1990), and has an operating history that dates from the mid-1970s.

The permeameter probe described by Nielson et al. (1989) is also a small-diameter (13-mm) probe that is driven to sampling depth by hand. This system, however, is further equipped for controlled flow extraction to allow for estimates of soil permeability from pressure/flow relationships as well as radon concentration by alpha scintillation. This method constitutes basic procedures adopted by the Florida Radon Research Program (Williamson and Finkel 1990), and similar systems have been used in major field studies (e.g., Kunz 1989; Liu et al. 1990). A plastic foam whose permeability is verified through independent measurement serves as a test medium to establish the calibration constant (Nielson et al. 1989). This probe is also compatible with the developmental work reported by Nazaroff and Sextro (1989).

The packer probe described by Tanner (1988a, 1988b) is a more complex apparatus that also provides simultaneous measures of radon and per- meability. This system features inflatable packers to create a "waste space" in the augered hole between the packers and a "sample space" below the lower packer. The system is devised so that the waste pump draws at a slightly greater suction on the waste space than on the sample space, intercepting any surface air so that the sample fully represents the subsoil environment.

Design elements of the packer probe originate from the invention of Hassler (1940). The inflatable packers provide the means to more firmly shape the collection geometry for drawing air from the soil pores. Preliminary experiments, however, have indicated that the sample flow is little affected by the flow level from the waste space, reinforcing the concept that permeability is greater in the horizontal than in the vertical (Tanner 1988a, 1988b). It would seem, therefore, that the main benefits of the inflatable packers is to guarantee sealing against the augered hole, and the soil ultimately shapes the transport field.

Analysis of Bulk Samples

Basic approaches for determining soil gas concentrations from bulk samples of soil generally involve sealing the sample under known conditions and measuring the evolution of radon in the sample with time. As shown in Table 3, three basic patterns can be recognized: (1) emanation, a variation of the standard laboratory test for radium that infers pore gas radon from time-related changes in a sample that has been baked to controlled dryness; (2) prompt bismuth, a second variation of the radium test that monitors time evolution of a sample that is sealed under field conditions; and (3) exhalation, involving analysis of radon escaping from the sample to a headspace.

Both the emanation technique and the prompt bismuth technique monitor the ingrowth of radon in the soil sample, producing data that readily estimate the undepleted soil gas concentration. The exhalation technique, on the other hand, is used primarily to determine the time rate of release of radon from the sample (hence the term exhalation), and requires additional information to estimate the undepleted soil gas concentration of the sample.

Exhalation tests in the laboratory involve placing a soil sample in a sealed container and monitoring the ingrowth of radon in the headspace. The radon concentration in the headspace is related to the exhalation rate through a simple mass-balance model. Colle et al. (1981) observed that, while many investigations of radon exhalation have followed the same broad principles, great differences occurred with regard to the size and composition of containers and the length of time allowed for radon to build up. Nonetheless, tests of this type have provided valuable insight with regard to the role of moisture (Stranden et al. 1984; Stranden 1983).

Back-diffusion into the sample is a concern in exhalation tests because the buildup of radon is secondarily affected, masking interpretation of the free or unattenuated exhalation rate (Jonasson 1983). Based on theoretical considerations, Samuelsson (1990) suggests that the outer volume should be at least 10 times larger than the pore volume of the sample.

In Situ Detection

Direct burial of detectors to estimate radon concentrations in the soil has been in use for some time. The main avenue of development entails forming a suitable detection volume in the soil and detecting alpha activity from radon diffusing into the cavity and subsequent decays of the short-lived progeny. As shown in Table 4, two basic techniques are evident: (1) passive detection and (2) active detection.

Passive in situ detection is probably the most widely used approach. The alpha track detector, originally developed to support uranium exploration (Fleischer at al. 1980), was soon adapted to support studies of radon in buildings (Akerblom et al. 1984). While the alpha track detector is still the system most closely identified with in situ passive measurements in the soil, the basis can be extended to other technologies. Although definitive studies remain to be done, the feasibility of the passive electret technology has been demonstrated for measuring soil gas (Kotrappa et al. 1987; Dempsey and Kotrappa 1989). Burial of activated carbon canisters to collect radon in the subsoil has been used as well (Akerblom et al. 1984). Both the alpha track technology and the electret technology require a permeable membrane to prevent thoron entry into the detection volume. Thoron does not significantly interfere with the activated carbon canister approach.

The second approach, involving placement of an active detection system (Warren 1977), presents an opportunity to study short-term effects. This latter approach has not entered widespread use. Cotter and Thomas (1989) have used this technique for continuous in situ detection of soil gas in Hawaii.

RADON FLUX

Measurement systems for radon flux seek to determine the net transfer from the soil to the atmosphere. General summaries of measurement technologies appear in publications by Colle et al. (1981), Freeman and Hartley (1986), and NCRP (1988). Basic approaches have focused on capturing radon using (1) closed accumulators, (2) flow-through accumulators, and (3) adsorption. Each of these approaches, summarized in Table 5, involves isolating an area of soil and measuring the amount of radon captured over a defined period of time.

Each of these basic methods is predicated on using the naturally prevailing convective/diffusive transport to deliver radon to the measurement system. It is logical, then, to conceive a fourth category, induced flux, to transport radon under controlled conditions. Additional methods that are frequently mentioned include the vertical profile method, which utilizes patterns of atmospheric radon and meteorology to estimate flux over large areas, and the soil concentration gradient method, which involves model estimates of surface flux from soil gas concentration data. Neither of these methods has entered widespread use.

Closed Accumulation

This approach involves direct accumulation of radon into a volume defined by the soil surface and a vessel whose open face is affixed to the soil. The radon concentration in the accumulator begins to increase as soon as the vessel is emplaced because dispersion to the atmosphere is negated. Initially, the concentration grows rapidly in direct proportion to flux. Soon, however, the rate of growth in the accumulator slows due to back-diffusion into the soil. Consequently, methodologies generally focus on acquiring grab samples during the very early stages of accumulation where linear relationships apply. Currently, grab sampling is generally accomplished by direct transfer to evacuated scintillation cells (Freeman and Hartley 1986). Of course, samples can also be drawn using intermediary containers (e.g., syringes, Tedlar bags) for subsequent transfer to scintillation cells.

Flow-through Accumulation

In an effort to more closely simulate natural conditions in the collection volume, the radon can be swept out of the accumulator and replaced with ambient air. If radon concentrations in the accumulator are maintained low enough to suppress back-diffusion, radon flux into the accumulator is proportional to the radon content of the exiting air stream. Early implementations of this method employed a closed-flow loop with a charcoal trap (chilled with dry ice) to collect radon for analysis (Pearson and Jones 1965). Subsequent designs directed the air stream to a continuous monitoring system with flow compensation drawn from ambient air (Scherry et al. 1984; Freeman and Hartley 1986). The flow-through accumulation method permits the use of larger collection areas and longer measurement periods.

Adsorption

Current applications of the adsorption method are generally drawn from the work of Countess (1976, 1977). The basic method involves placing a charcoal canister in contact with the surface for a period that may range from a few hours to a few days. Radon adsorbed on the charcoal is determined by measuring the gamma activity of the radon decay products in equilibrium with the adsorbed radon. The size and construction of charcoal canisters range from prepackaged cartridges designed for respirators to large-diameter (25 cm) canisters especially designed for flux measurements (Freeman and Hartley 1986).

Induced Flux

Principal references discussing the traditional flux measurement technologies dwell, to varying degrees, on short-term and long-term fluctuations caused by meteorological conditions, and soil state, and how these factors can influence the estimation of representative flux rates from limited duration data. This treatment is analogous to concerns for interpreting soil gas data that are unsupported by information on porosity.

Alternative approaches, then, could be considered wherein the natural transport is simply overpowered. Hassler (1940), in the patent that inspired the packer soil gas probe discussed earlier, presented the basis for a flow hood to provide for controlled transport of soil gas from the surface. Basic components involve an annular guard (to discourage re-entrainment of surface air) and an inner hood to capture soil gases.

TECHNICAL CONSIDERATIONS

Currently, there are no hard and fast criteria to provide an unambiguous reference for judging the performance of measurement technologies for radon potential. While there is little doubt that site-specific measure- ments can be used to determine the radon potential of land areas, interpretations are driven by empirical correlations and theoretical considerations. A broad consensus, however, highlights the importance of examining (1) the abundance of radon in the soil, (2) its propensity to migrate in the soil, and (3) explicit building effects.

Technologies geared to measuring (1) radium concentrations in bulk soil samples or (2) soil gas concentrations are readily applied to the problem of estimating the undepleted radon concentration in soil gas. Measurements of unattenuated flux provide estimates of diffusive transport which, in turn, could be used to estimate soil gas concentrations at depth. The induced flux method, although untested, may provide the means to directly simulate radon entry for slab-on-grade and crawl space construction. Laboratory measurements of exhalation, on the other hand, while not readily extrapolated to the soil environment, may provide clues to the relative strength of radon sources through comparative tests.

Radium-based measurements have the distinct advantage of being suited to testing water-saturated soils. Soil-gas-based measurements (extraction probes, in situ detection, flux), on the other hand, generally fail to obtain samples from saturated soils because the gas volume is nearly zero. Recognition factors to avoid generally saturated conditions can be built into protocols, as can rules to invalidate samples attempted from saturated layers encountered at depth.

Material that is permanently saturated in the native state but likely to reach varying degrees of dryness after construction, therefore, is best characterized through radium-based measurements. These circumstances are likely to occur with fill material and may occur in areas with a shallow water table that could recede as property development alters drainage patterns.

Quality assurance is a vexing question for soil gas measurements. Although analytical proficiency can be deemed acceptable, there is little information at hand to evaluate system-level performance because relatively few studies have explicitly compared technologies. A number of studies have included more than one soil measurement technique, but additional analysis would be required to formally compare methods.

PRACTICAL CONSIDERATIONS

Practical decisions are likely to be guided by two absolutes: (1) avoidance of clearly inappropriate technologies, and (2) meeting the schedule demands of the situation. For the radium-based measurements, the all-weather capability must be judged against the lengthy time period necessary to achieve radioactive equilibrium. Delays could be shortened by taking more counts during the ingrowth period to extrapolate data to equilibrium levels. For soils with a low emanation fraction, a number of days may need to elapse to resolve the trend, but turnaround time could, in concept, be reduced to a matter of days. Further, initial count data offer information to provide a rough estimate without extended waits.

While the soil gas extraction techniques are not suited to testing under saturated conditions, the simplicity of equipment and field operations for the hand-driven probes can deliver prompt results, making the reconnaissance probe and the permeameter probe likely candidates for widespread use. The packer probe is a bit more complex and requires an augered hole, but delivers data in a short timeframe.

In situ detectors offer possibly the least expensive approach. Emplacing detectors at a satisfactory depth (1 m) and retrieving them may present a problem. The main disadvantages, however, could arise from the need to sample for relatively long periods of time and from unreliable results in the presence of high moisture levels.

As noted earlier, measurements of unattenuated flux can be converted to estimates of soil gas radon at depth. This conversion, however, is predicated on model assumptions that may go unverified in the field. Similarly, laboratory exhalation cannot be readily extrapolated to quan- tatative estimators of radon potential. The induced flux technique may prove to be a useful test apparatus for soils receiving slab-on-grade or crawl space construction. At the present time, however, it is an untested technology.

CONCLUSIONS

At the present time, the principal means to confirm indoor radon levels involves testing buildings, not land. Although this is likely to continue to be the main verification, soil-based measurements can help to identify land areas warranting special attention for risk communication programs as well as site-specific decisions for varying degrees of radon-resistant construction.

Each of the technologies summarized here is capable of providing useful information to evaluate radon potential. With the exception of the induced flux

technique, all of the measurement techniques discussed here are supported by documented field experience and, in many cases, by published protocols. However, the means to estimate radon potential from field data is still evolving. How this evolution affects the definition of consensus protocols remains to be seen.

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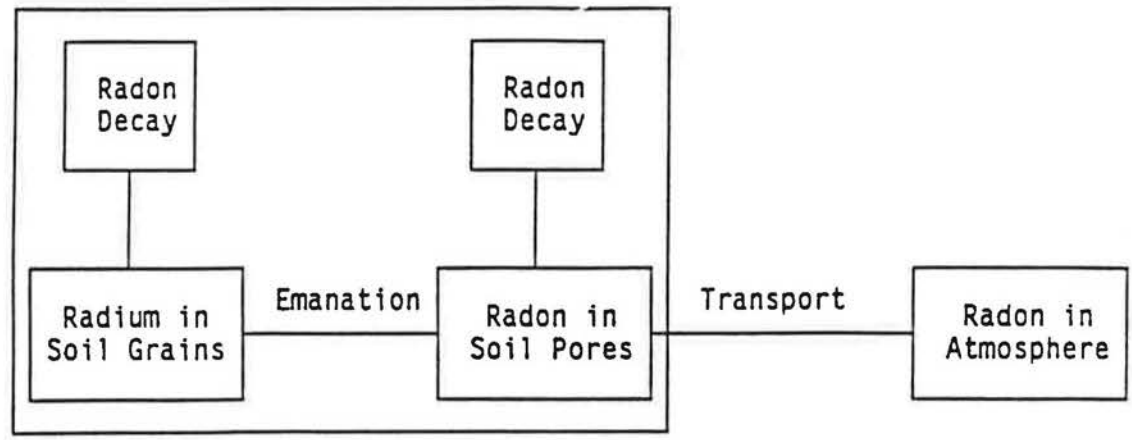
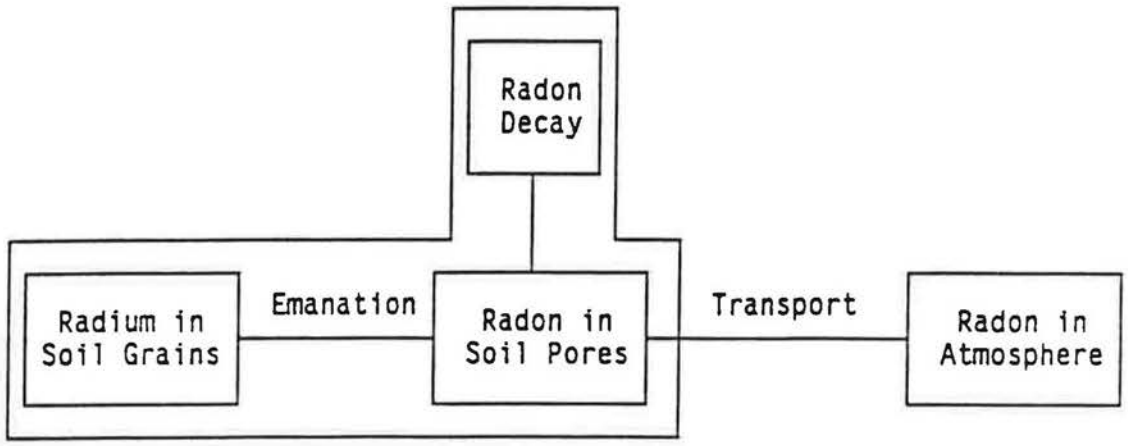
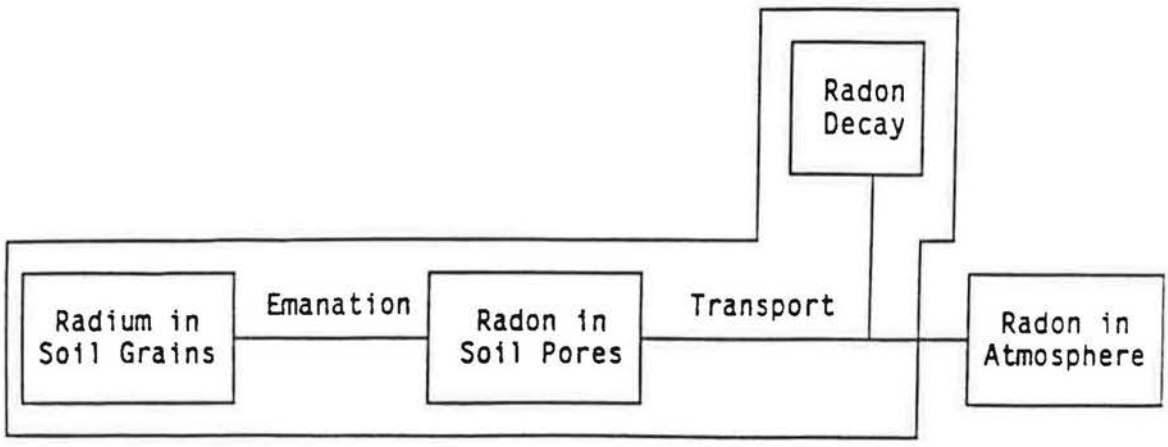


Radium Content



Radon in Soil Gas



Radon Flux

Figure 1. Sampling strategies for radon in the soil.

TABLE 1. SUMMARY OF APPROACHES TO MEASURING RADIUM CONTENT OF SOILS

Approach	Procedures/Equipment	References
Laboratory Analysis	Weigh sample, heat to dryness, store in sealed container to achieve radioactive equilibrium, analyze by gamma spectroscopy.	Williamson and Finkel (1990)
Prompt Bismuth	Seal sample and weigh at time of collection, analyze by gamma spectroscopy within 2 hours after collection, repeat analysis at 4 to 12 hours and at radioactive equilibrium.	Stieff et al. (1987)

TABLE 2. SUMMARY OF APPROACHES FOR MEASURING RADON IN SOIL GASES WITH GAS EXTRACTION PROBES

Approach	Procedures/Equipment	References
Reconnaissance Probe	Small-diameter (9-mm) probe is driven to 75-cm depth, gas sample is extracted by syringe, analysis is by scintillation.	Reimer (1990)
Permeameter Probe	Small-diameter (13-mm) probe is driven to depths of 46, 61, 76, and 122 cm; pressure/flow relationships are used to estimate permeability; soil gas samples are drawn from the 122 cm depth to flow-through scintillation cells for subsequent analysis.	Nielsen et al. (1989)
Packer Probe	Moderate diameter (27-mm) probe is inserted into 3.5-cm diameter hole to augered depth of 1 m; inflatable packers isolate sample space, and sample air is drawn to flow-through scintillation cell for analysis; permeability is estimated from pressure/flow relationships.	Tanner (1988a,b) Hassler (1940)

TABLE 3. SUMMARY Of APPROACHES FOR MEASURING RADON IN SOIL GAS FROM BULK SAMPLES

Approach	Procedures/Equipment	References
Emanation	Weigh sample, heat to dryness, reweigh, store in sealed container; analyze by gamma spectroscopy within 4 to 36 hours of sealing and again after radioactive equilibrium is achieved.	Williamson and Finkel (1990)
Prompt Bismuth	Seal and weigh sample at time of collection, analyze by gamma spectroscopy within 2 hours after collection; reanalyze at 4 to 12 hours, and at radioactive equilibrium.	Stieff at al. (1987)
Exhalation	Place soil sample in sealed container, measure outgassed radon in headspace.	Jonasson (1983) Samuelsson (1990)

TABLE 4. SUMMARY OF APPROACHES FOR IN SITU DETECTION OF RADON IN SOIL GAS

Approach	Procedures/Equipment	References
Passive Dosimeter	Passive dosimeter is buried in soil; decays of radon diffusing into detection volume and subsequently formed radon progeny are registered.	Fleischer et al. (1980)
Active	Electronic detector is buried in soil; decays of radon diffusing into detection volume and subsequently formed radon progeny are recorded by the detector.	Warren (1977)

TABLE 5. BASIC APPROACHES FOR MEASURING SOIL FLUX

Approach	Procedures/Equipment	References
Closed Accumulation	An open-ended vessel is sealed to the surface; ingrowth of radon is measured over time.	Wilkening et al. (1972)
Flow-through Accumulation	An open-ended vessel is sealed to the surface; radon entering the vessel is swept to a collector or monitor for measurement.	Freeman and Hartley (1986)
Adsorption	Exhaled radon is adsorbed onto granular charcoal; the charcoal bed is removed to laboratory for analysis.	Countess (1976, 1977)
Induced Flux	A hood is attached to the surface and radon is transported under controlled evacuation.	Hassler (1940)