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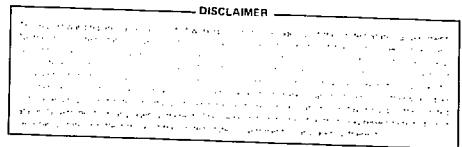
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**RADON AND ITS DAUGHTERS IN ENERGY-EFFICIENT BUILDINGS**

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ABSTRACT

Our group has been carrying out work on several aspects of radon and its daughters indoors. We have measured radon emanation rates and radionuclide concentrations in building materials, performed surveys of radon and daughter concentrations in residences, begun to examine control techniques and strategies, and devoted significant efforts to instrumentation developments. To more completely characterize radon and its daughters indoors, more substantial efforts are needed on the questions of geologic distribution of radon, transport into structures, daughter behavior indoors, and instrument response under various conditions.

## INTRODUCTION

Indoor exposures to daughters of radon 222 (hereafter referred to as "radon") contribute significantly to the overall radiation dose to which the general population is exposed, but the nature and extent of indoor exposures is not known precisely. Interest in this subject has risen in recent years because of increasing use of measures that would decrease mechanical ventilation or infiltration rates in buildings, possibly raising daughter concentrations. We are investigating sources and indoor concentrations of radon and its daughters and evaluating the effectiveness of selected control measures. In our efforts we have studied: 1) radon emanation from building materials; 2) radon and radon-daughter concentrations in conventional and energy-efficient buildings; and 3) the effectiveness of mechanical ventilation with air-to-air heat exchangers as a control measure. Instrumentation development has been associated with each of these studies. However, to characterize indoor radon and its daughters adequately, more intensive research efforts are required on geographic distribution of radon sources, mechanisms for radon transport, the behavior of radon daughters, and the sensitivity of monitoring instruments.

## EMANATION RATES AND RADIONUCLIDE CONCENTRATIONS IN BUILDING MATERIALS

We have examined samples of building materials commonly used in the United States, emphasizing concrete, to determine their contribution to radon concentrations observed indoors. Our principal measurement technique employs gamma detection using a heavily shielded sodium iodide system. Measurement of gamma rays from  $^{214}\text{Bi}$  and  $^{208}\text{Tl}$  from a sample in a sealed container indicates concentrations of the  $^{238}\text{U}$  and  $^{232}\text{Th}$  decay chains, respectively. Subsequent measurement with the container open, allowing radon (the parent of  $^{214}\text{Bi}$ ) to escape, then yields the radon emanation rate. (In both cases  $^{40}\text{K}$  is measured from its own gamma ray.) An alternative technique measures radon emanation directly by sealing the sample in a container for one to three days and then transferring the radon collected to a scintillation cell for counting.

Emanation rates (per unit mass) and elemental concentration for samples of ordinary concrete from ten U.S. metropolitan areas are shown in Table 1. The mean rate for the ten sample groups ranges from 0.4 to 1.2 pCi kg<sup>-1</sup> h<sup>-1</sup>. Although not shown, a group of concretes containing fly ash also fell within this range, and samples containing phosphate slag fell below. Gypsum, brick, and wood were toward the bottom of this range or lower, as were solar rock bed samples with high uranium content and a radon escape-to-production ratio of 4% (ref. 1).

The materials examined appear to contribute only a small portion of the radon levels ordinarily found in U.S. homes. For 0.2 m thick concrete, 1 pCi kg<sup>-1</sup> h<sup>-1</sup> corresponds to an emanation rate per unit area of approximately 0.05 pCi m<sup>-2</sup> s<sup>-1</sup>, which would contribute 0.1 pCi/l to the indoor radon concentration of a house having an air exchange rate of 1

per hour and an emanating-surface-to interior-volume ratio of  $0.5 \text{ m}^2$  per  $\text{m}^3$ . We are now examining emanation rates of individual concrete components surveying more fly-ash concretes and measuring the radon diffusion length in concrete samples.

#### INDOOR CONCENTRATIONS OF RADON AND ITS DAUGHTERS

We have surveyed three housing groups, one consisting of energy-efficient houses and two of conventional, in each case measuring radon (and usually radon daughters) in grab samples and measuring the infiltration rate at the same time. Occupants were asked to close windows and doors for the night prior to measurements to assure a degree of correspondence between the observed radon concentrations and infiltration rates and thus to yield the radon source strength. These surveys indicate the range of source strengths and concentrations in typical houses, providing an initial basis for more intensive research characterizing indoor exposures and the influence of energy-conservation and control measures.

Most of the 17 "energy-efficient" houses surveyed in the United States and Canada (ref. 2) were demonstration or research houses, privately-owned residences built to assure low infiltration rates, or passive solar houses with rock-bed heat storage. Air samples were taken in Tedlar bags, from which radon concentrations were determined using scintillation cells. Infiltration was measured by releasing a tracer gas in the house and measuring its concentration as a function of time using an infrared analyzer. Results are shown in Figure 1. If the radon source strength were the same for all these measurements, and if the outdoor radon concentrations were small, the points would be on a straight line. Source strengths were found to differ by more than a factor of 10, however, even among this small number of houses. (The line in the figure is the result of detailed measurements in a single house, discussed below.) An examination of U.S. data from conventional housing indicates that indoor radon concentrations vary at least two orders of magnitude (ref.3), and most of this variability appears to be due to differences in source strength.

The second group, 26 conventional houses in the San Francisco Bay Area, was surveyed (ref. 4) in a similar manner. Radon concentrations in these houses ranged from 0.4 to 0.8 pCi/l, typically much lower than those obtained in our energy-efficient sample. Measured infiltration rates ranged from 0.02 to 1.2/hr, with 80% in the range 0.1-0.6; these infiltration rates are lower than typical for these houses because of mild weather conditions at the time of measurement.

The third group of houses surveyed were conventional houses in the vicinity of the single energy-efficient house in Maryland that showed the highest radon concentration (See Figure 1). Our preliminary results indicate that indoor radon concentrations in this particular area are atypically high. The data from these two surveys of conventional houses indicate differences in indoor source strengths of more than a factor of

100.

## CONTROL TECHNIQUES AND STRATEGIES

Of available techniques for controlling indoor radon and daughter concentrations, our research program has investigated those that directly affect the airborne concentration rather than those that reduce the source strength. In particular, we have investigated the effect of mechanical ventilation systems incorporating air-to-air heat exchangers on indoor concentrations. Such systems avoid much of the energy loss ordinarily associated with mechanical ventilation in heating and cooling seasons, can control indoor pollutants other than radon and its daughters, and are applicable to existing as well as new structures. For our initial tests, we selected the Maryland house mentioned above. Using a mechanical ventilation system with air-to-air heat exchange to vary the air exchange rate, we measured both radon and radon daughter concentrations for a two-week period. The line shown in Figure 1 represents the equilibrium radon concentration. Individual daughter concentrations yielded the potential alpha energy concentrations (PAEC) shown in Figure 2. The daughters have a much more complex dynamic behavior than radon.

We have also begun to study the effectiveness of these ventilation systems for radon control in a number of recently built conventional houses with relatively low infiltration rates (0.6/h or less). An automatic system is monitoring radon and infiltration rate in these houses for week-long periods with and without mechanical ventilation. We will shortly begin laboratory studies of air cleaning systems in a highly instrumented "radon research house."

We have begun to evaluate the role of such techniques in programs to control exposures in conventional or energy-efficient houses (ref. 6). Present exposure rates vary substantially among geographic areas and from house to house in the same area, with a significant incidence of public exposures in the "occupational" range, i.e., in the vicinity of 1 WLM/yr or higher. Ordinary tightening measures (i.e., those that reduce infiltration rates, typically 0.5-1/h, by about 25%) will not affect this incidence greatly, but elevated exposures appear to require remedial action in any case. On the other hand, construction of very tight houses (infiltration rates as low as 0.1/hr) appears to require incorporation of ventilation or air cleaning systems. Strategies to control public exposures could have two complementary goals: to assure that virtually no exposures exceed some designated limit, as yet undetermined; and to design buildings to assure an acceptably low average exposure, also to be determined. Development or implementation of either limit depends on future work of the kind discussed below.

## INSTRUMENTATION DEVELOPMENT

We have devoted substantial efforts to developing and evaluating instrumentation, primarily for measuring airborne radon and daughter concentrations, but also for source monitoring.

Alpha spectroscopic methods for field monitoring of radon daughter concentrations are examined in detail in a separate paper (Nazaroff et al, "Alpha Spectroscopic Techniques for Field Measurement of Radon Daughters"). That analysis adjusts timing intervals for two-count procedures to maximize precision in measuring individual daughters and compares the precision with that of other techniques. In addition, a single-count procedure taking a total of 11 minutes yields the potential alpha energy concentration (with less than 20% uncertainty) and the polonium 218 concentration.

We have also designed and fabricated an automatic system (the "Aardvark") for field measurement of: indoor radon concentration, following Thomas (ref. 7); infiltration rate, by tracer gas techniques using infrared analysis (ref. 8); indoor and outdoor temperatures; and furnace activity. A more complete system, incorporating an automatic daughter monitor of our own design, is under development for a "radon research house".

We routinely engage in testing and refining integrating radon monitors for our field program, including those based on thermoluminescent and track etch detection. A new development has been the design and fabrication of a prototype radon "sniffer" for identifying places at which radon enters the houses. Laboratory and field testing of this instrument is not complete. Such a device may also be effective in determining correlations between source behavior and observed indoor concentrations.

#### FUTURE EFFORTS REQUIRED TO CHARACTERIZE INDOOR RADON

Successfully characterizing indoor radon will require intensive investigation of several fundamental questions: 1) geographic characterization of radon source strength; 2) radon transport into structures; 3) behavior of daughters indoors (including the effect of removal processes); and 4) response of measurement techniques in various conditions.

The rate at which radon emanates from the ground or from samples of building materials varies strongly from one site or sample to another. Investigating how emanation rates are related to the geologic (and radiologic) character of an area, or how various monitoring strategies might yield such information, could considerably ease the problem of characterizing indoor radon concentrations on a large scale. One U.S. information resource not yet utilized is the aerial uranium survey program. This survey measures  $^{214}\text{Bi}$  gamma emissions and hence gives a direct indication of radon daughter concentrations. Related ground measurements are required to determine the correspondence between aerial data and radionuclide concentrations or emanation rates.

Investigating the transport of radon into structures, while difficult, deserves a significant effort and can profit from the substantial understanding of underground radon transport acquired in uranium mines and elsewhere. We plan a modest program at the sites of houses in which we measure indoor radon concentrations. Initially, we will use the

equivalent of grab samples to indicate source strength i.e., measurements of radionuclide concentration, radon concentration in soil gas, emanation rate, and gamma spectra. More sophisticated measurements could correlate source strengths with indoor concentrations on a real-time basis.

Fundamental to our understanding of indoor exposures and control methods is study of the dynamic behavior of radon daughters in structures. Such studies require sophisticated instrumentation and test facilities simulating structures among the building stock. Our major new effort for the near future will use such a facility to study the influence of various factors on the airborne concentration of radon daughters and on their attachment to particles. An automatic system based on the Aardvark will monitor air exchange rate, radon and radon daughter concentrations, indoor and outdoor temperature, wind, and state of the mechanical ventilation system. Particulate mass concentration and size distribution will also be monitored.

These facilities, together with complementary environmental chambers, are necessary for studying the response of instruments, particularly daughter monitors, in a range of conditions. This testing program must precede any comprehensive survey of the building stock or formulation of concentration standards.

Significant efforts are already beginning on the fundamental questions of geologic distribution, radon transport, daughter behavior, and instrument response. But these efforts need to be focussed and intensified to serve as an adequate basis for large-scale monitoring programs, exposure characterization, estimation of the effect of energy-conserving measures, and formulation of control strategies.

#### ACKNOWLEDGEMENT

This work was supported by the Assistant Secretary for Conservation and Renewable Energy, Office of Building Energy Research and Development, Building Systems Division, and by the Director, Office of Energy Research, Office of Health and Environmental Research, Human Health and Assessments Division and Pollutant Characterization and Safety Research Division of the U.S. Department of Energy under contract no. DE-AC03-76SF00098.

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TABLE 1. EMANATION RATES AND RADIONUCLIDE CONCENTRATIONS IN ORDINARY CONCRETE

Origin	No. of Samples	Emanation Rate per Mass ( $\mu\text{Ci kg}^{-1} \text{h}^{-1}$ )			Elemental Concentrations (mean)			Average Escape-to-Production Ratio (%)
		Range	Mean	S.D.	U (ppm)	Th (ppm)	K (%)	
Albuquerque, N. Mex.	12	0.70-1.95	1.22	0.35	2.5	6.0	1.5	22
Kansas City, Mo.	12	0.40-0.65	0.53	0.07	1.0	2.0	0.7	25
Philadelphia, Penn.	7	0.35-0.55	0.42	0.08	0.6	1.5	0.7	17
Salt Lake City, Utah	9	0.50-0.75	0.64	0.08	2.0	4.0	0.6	14
San Francisco - Oakland, Cal.	21	0.65-1.10	0.83	0.12	1.5	3.0	0.6	24
Austin, Texas	8	0.60-0.92	0.73	0.12	1.3	1.5	0.8	24
San Antonio, Texas	8	0.27-0.72	0.46	0.14	3.0	7.5	1.5	8
Chicago, Ill.	12	0.25-1.39	0.66	0.36	1.5	2.0	0.5	25
St. Paul - Minneapolis, Minn.	5	0.54-0.75	0.62	0.09	1.5	4.0	1.5	19
Knoxville, Tenn.	6	0.46-0.78	0.59	0.12	1.0	1.2	0.5	23

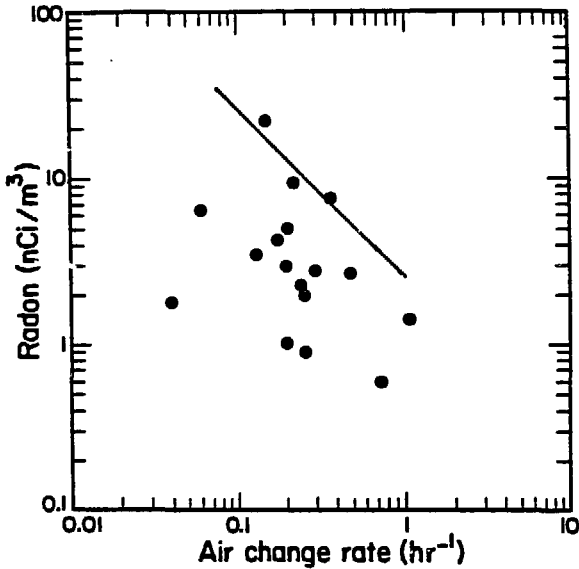


Figure 1. RADON CONCENTRATIONS AND VENTILATION RATES IN ENERGY-EFFICIENT HOUSES. Results are from grab radon samples and concurrent ventilation rate measurements. The line indicates results from a house in which the ventilation rate was varied using mechanical ventilation with air-to-air heat exchange.

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Figure 2. VARIATION OF POTENTIAL ALPHA ENERGY CONCENTRATION WITH TIME. Five ventilation rates were examined, using mechanical ventilation with air-to-air heat exchange during a two-week test period. (The indicated EPA guideline for average PAEC applies to houses on land reclaimed from phosphate mining.)

