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CHARACTERIZING THE SOURCE OF RADON INDOORS

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CHARACTERIZING THE SOURCE OF RADON INDOORS

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ABSTRACT

Average indoor radon concentrations range over more than two orders of magnitude, largely because of variability in the rate at which radon enters from building materials, soil, and water supplies. Determining the indoor source magnitude requires knowledge of the generation of radon in source materials, its movement within materials by diffusion and convection, and the means of its entry into buildings.

This paper reviews the state of understanding of indoor radon sources and transport. Our understanding of generation rates in and movement through building materials is relatively complete and indicates that, except for materials with unusually high radionuclide contents, these sources can account for observed indoor radon concentrations only at the low end of the range observed. Our understanding of how radon enters buildings from surrounding soil is poorer, however recent experimental and theoretical studies suggest that soil may be the predominant source in many cases where the indoor radon concentration is high.

keywords: indoor air quality, pollutant sources, radon, residential buildings.

INTRODUCTION

It has long been known that the dose equivalent to the lung caused by alpha radiation from deposited ^{222}Rn and ^{220}Rn daughters substantially exceeds the whole body dose equivalent attributed to naturally occurring radionuclides, i.e., about 1 mSv (0.1 rem) per year. With the recent utilization of an "effective dose equivalent", it is now possible to say more precisely that radon-daughter exposures of the lung contribute approximately half of the average total effective dose equivalent of 2 mSv per year for the general population[1]. Most of the radon-daughter exposure occurs indoors.

What is of most interest from the point of view of sources is the substantial variability in time-averaged exposures to the daughters, ranging from rates more than a factor of 10 lower to more than a factor of 10 higher than average, even among the general population. To some extent, this variability is connected with different equilibrium factors, but it is caused mainly by the difference in indoor radon concentration from one building to another. Furthermore, although the indoor radon concentration is definitely affected by ventilation rate, it now appears that the major cause of the variability in radon concentration from one building to another is differences in the rate at which radon enters the indoor atmosphere from its various sources. This conclusion is supported by various types of work, but none makes it clearer than direct measurement of both indoor radon concentration C_R and air-exchange rate λ_v in samples of housing. For example, in grab-sample measurements for 101 houses in the United States, any presumed correlation between C_R and λ_v was imperceptible; in fact, the variance in radon entry rate, calculated as the product of C_R and λ_v, far exceeded the variance in λ_v[2]. This observation also appears to be true of time-averaged measurements taken over periods of several months[3].

In this respect, the case of radon and its daughters is similar to other classes of indoor air pollutants, e.g., gases or particles arising from indoor combustion processes and formaldehyde and other organics arising from building materials and furnishings. Indoor concentrations are affected by ventilation rates and at least potentially by various interactive processes, involving other airborne pollutants and the building structure and equipment. But average indoor concentrations for each pollutant class are found to vary over one or two orders of magnitude or more, and the major cause of this variability is differences in rates of emission into the indoor atmosphere[4].

A major reason for the rising interest in studying these pollutant classes has been the possibility that energy-saving programs may raise concentrations by lowering ventilation rates. It is therefore ironic to find that the major cause of variability in concentrations is variability in source strengths rather than ventilation rates. On the other hand, this presents a substantial opportunity to lower collective and individual exposures by systematically identifying the portion of the building stock with high source strengths and by applying appropriate control measures, either remedially or in new buildings. A further irony in this context is that the same forces that drive air-exchange rates, at least in single-family residences, may also drive radon entry to a substantial degree.

The wide variability of radon source strengths and the associated potential for reducing indoor concentrations are ample reason for detailed consideration of our ability to characterize the origin of radon indoors. Radon enters buildings from several major sources, principally building materials and the soil or rock that underlie or surround building foundations; in some circumstances radon can also enter via water supplies (or even natural gas) at substantial rates. Characterizing the source of radon indoors requires attention to the rate at which radon is generated in source materials, to the modes of radon transport through various materials, and finally to the manner in which radon actually enters indoor atmospheres.

Previous reviews have focused on characterization of the source materials themselves and, to a lesser extent, on how radon moves through or out of materials[1,5-9]; they have not examined the question of radon entry routes in detail, although substantial work on this problem has been undertaken in recent years (e.g., refs. 10-12). The purpose of this paper is to review the state of understanding of radon sources and transport, with particular attention to characterizing the factors that affect the rate at which radon enters buildings.

The sources emphasized in this review are building materials and soil or rock, although we give some attention to entry via water. The source of ^{222}Rn is the main topic, although ^{220}Rn is included to some degree. We first examine source materials focusing on radium contents and associated radon emanation rates, then consider transport through earth and building materials. We next consider mechanisms for entry into buildings. Finally, we indicate potential means for identifying high source strengths systematically either on a regional scale or in individual buildings, and for reducing the higher entry rates.

Before proceeding, it is useful to provide a rough scale for rates of radon entry into buildings and for the approximate range of contributions from various sources. For a building at steady state with a spatial-average concentration of 30 Bq m^{-3} (0.8 pCi l^{-1}) and an air-exchange rate of 0.5 h^{-1}, the radon entry rate (given per unit volume) must be 15 Bq m^{-3} h^{-1} (0.4 pCi l^{-1} h^{-1}), assuming ventilation is the only removal mechanism. Entry rates for the U.S. housing sample mentioned above[2,11] had a geometric mean of 14 Bq m^{-3} h^{-1} (0.37 pCi l^{-1} h^{-1}), with geometric standard deviation (GSD) of 4.0, comparable to results for housing in Great Britain[13]. As discussed below, the contribution expected from most building materials, such as ordinary U.S. concretes, is much less than the observed geometric mean; it would be particularly difficult to account for the higher entry rates that are observed (hundreds of Bq m^{-3} h^{-1}) on the basis of ordinary building materials. On the other hand, the flux from soils can account for the observed range of entry rates.

In some cases building materials can constitute the predominant source. Materials with high radionuclide contents have been found that can constitute large source terms, an example being the alum-shale concretes previously used in Swedish housing[14]. Moreover, multi-story buildings typically have source strengths that are lower than those for one-story structures; these smaller rates can be accounted for by radon emanating from building materials, and the incorporation of certain industrial byproduct materials has the potential for raising rates. In this paper, we will review work pertaining in general to characterization of indoor radon source strengths, but give special attention to the causes of higher-than-average rates.

EMANATION CHARACTERISTICS OF SOURCE MATERIALS

Characterizing the source of radon indoors requires attention to much more than the distribution of radium, the parent of radon. As a first step, it is useful to consider the factors that affect the generation of radon atoms that are able to move a significant distance from their site of formation. We also summarize what is known about exhalation rates from building materials and soil, leaving until the next section explicit consideration of how radon moves through various materials.

The total rate at which radon atoms are generated is given simply by the radium activity at the point in question: $dN_{Rn}/dt = I_{Ra}$. Since the corresponding radon activity is $I_{Rn} = \lambda_{Rn} N_{Rn}$, where λ_{Rn} is the radon decay constant (2.1×10^{-6} s^{-1} for ^{222}Rn and 1.3×10^{-2} s^{-1} for ^{220}Rn), the total rate at which radon activity is generated is $G = dA_{Rn}/dt = \lambda_{Rn} I_{Ra}$. However, only a fraction of the radon generated is free to move through the material, so that we define an effective radon-generation rate (or emanation rate) to be the product of an "emanation ratio", r, and the total generation rate:

$$G_e = r\ \lambda_{Rn} I_{Ra}. \qquad (1)$$

(Each of these quantities, except λ_{Rn} and r, can be considered as concentration per unit volume or mass.)

The emanation ratio (or fraction) has also been used by some to designate the fraction of radon generated that actually escapes the source material, in which case it is dependent on the size and shape of the material, as well as other factors affecting transport. For materials whose dimensions are small compared with macroscopic transport lengths, these two ratios are approximately equal and the ambiguity causes no difficulty.

Tanner[5,6] and others review mechanisms for generation of radon that is free to move. Basically, although a parent radium atom is ordinarily in or on a solid grain, the radon atom produced upon alpha decay has a significant recoil energy and can come to a stop in the fluid (air or water) in the space between the grains, i.e., the pore space; it can then migrate through the porous medium by various mechanisms. As the recoiling radon atom can move through only a small amount of material before stopping, two conditions must ordinarily be met for its recoil to end in a pore space: it must be sufficiently close to the pore, and the fluid in the pore must succeed in stopping it. A reasonable scale for recoil considerations is 0.1 mm: the recoil range in solid materials is somewhat less than this value; the range in water is comparable; and the range in air is three orders of magnitude greater (stopping power being roughly proportional to density). A radon atom that does not reach a pore on recoil will not do so by diffusion, since diffusion lengths in solids are extremely small, and one that recoils across a pore and is embedded in solid material can return to the pore only if the stopping process effectively forms a pathway for return.

Detailed accounting of the emanation ratio would therefore require consideration of such factors as the size of the grains or microscopic structure of the material in question (characterized, for example, by a diameter or other scale length), the distribution of radium in the grains (perhaps characterized by the fraction contained in a surface layer of specified thickness), the fraction of space occupied by pores that are

connected with each other on a macroscopic scale (given by the porosity), and the amount of water in the pores (specified, to some degree, by the moisture content). Not all this information is easily accessible. As a result, source materials are often characterized merely by the effective radon-generation rate or by the radium content and emanation ratio.

Certain of the parameters mentioned above have significance beyond the context of radon emanation or generation rates per se. Porosity and moisture content, as well as the microscopic structure of the material, affect the diffusion of radon or the flow of radon-bearing air. Furthermore, the radium content is a parameter that is easily measured and - together with other information - may serve as an index to the radon emanation rate. These questions will be discussed after considering what is known about radium contents, emanation ratios, and emanation rates of building materials and soils.

Building materials are more easily characterized as radon sources than are the soil or rock that constitute the building site. The observed ^{226}Ra concentrations for materials derived from components of the earth's crust are comparable to the values typical for major rock types and soils (see below). An earlier review[8] cites data on ^{226}Ra contents of European building materials: average values for the concrete sample groups examined ranged from 33 to 74 Bq kg^{-1} (0.9 - 2.0 pCi g^{-1}); the range for cement was similar, but that for brick was about 50 percent higher, and that for plaster was lower by about a factor of five. Except for materials recognized to have unusually high radionuclide contents, later reviews cite results with only a moderately wider range[1,7]. Examinations of U.S. concretes have yielded sample averages ranging from 9 to 44 Bq kg^{-1} (0.2 - 1.2 pCi g^{-1}), with values for brick and tile somewhat higher[15-17]. Concentrations of ^{226}Ra for building materials not derived from crustal components, e.g., wood, are much lower.

Considerably greater ^{226}Ra concentrations may be found in specific components of natural building materials. Granites exceed the range given above to some degree, and Italian tuffs and other materials used in building have concentrations of 200 Bq kg^{-1} (5 pCi g^{-1}) or more[18]. Probably

the most significant example in this class is the use of alum shale in Swedish aerated concretes during the period 1930-1975, resulting in concentrations averaging 1300 Bq kg^{-1} (35 pCi g^{-1}) [14].

Building materials incorporating residues from industrial processes may also have elevated ^{226}Ra contents, as discussed in an earlier review[7]. An example is the Federal Republic of Germany's use of "red mud" (from bauxite processing) for bricks and of blast-furnace slag for blocks, each case yielding materials with average ^{226}Ra concentrations of about 280 Bq kg^{-1} (7.6 pCi g^{-1}). A possibility with potentially wider implications is the use of wastes from processing sedimentary phosphate ore, which contains substantial concentrations of the ^{238}U series. One such byproduct, phosphogypsum, may be used for building materials such as wallboard, particularly in countries with little natural gypsum, and may have ^{226}Ra concentrations as high as 600 Bq kg^{-1} (16 pCi g^{-1})[7]. Another byproduct, phosphate slag, can be incorporated into concrete; this has led, in the United States, to the use of concretes with ^{226}Ra concentrations of about 740 Bq kg^{-1} (20 pCi g^{-1}) in an estimated 74,000 homes[19]. A final example of potential importance is the disposition of fly ash from coal-burning power plants by incorporating it into concrete; since fly ash contains ^{226}Ra at concentrations ranging up to several hundred Bq kg^{-1} [1,20,21], some increase in the ^{226}Ra content must be expected for concretes containing a few percent fly ash by weight.

In many of the materials mentioned above, the activity concentrations of the ^{232}Th series, including ^{224}Ra, the parent of ^{220}Rn, are similar to those of the ^{238}U series. A significant exception is that the alum shale used in Swedish aerated concrete has unusually high concentrations of the ^{238}U series, but not of the ^{232}Th series; the same is true of byproducts of phosphate production.

Measured concentrations of these two series are, of course, direct indicators of associated electromagnetic radiation fluxes, not discussed in this paper. However, since emanation ratios can vary considerably, ordinarily in the range of 1 to 30 percent, radionuclide concentrations are not a direct indicator of effective radon-generation rates. Relatively few

measurements of emanation rates or ratios have been performed for building materials, even those known to contain elevated radium concentrations. Selected results from a number of such measurements are summarized in Table 1, where, depending on the type of measurements performed, we give activity concentrations of ^{226}Ra, emanation rates (per unit mass), or emanation ratios.

One or more measurements have been performed for each of the major classes of building materials, or of components of such materials. In contrast to ordinary red brick, which had emanation ratios of at most several percent, the ordinary concretes examined had average emanation ratios between 3.5% and 28%[16,22,23]. Thus the observation that emanation ratios for ordinary materials are typically in the range 1-10%[1] does not apply to the important case of concrete. The corresponding radon emanation rates were found to be $(0.3 - 1.2) \times 10^{-5}$ Bq $kg^{-1}s^{-1}$ (0.3 - 1.2 pCi kg^{-1} h^{-1}), which are in reasonable agreement with other results for ordinary concrete[24,25]. In contrast, the emanation rate for a sample of Swedish alum-shale aerated concrete was found to be much higher, 44×10^{-5} Bq kg^{-1} s^{-1} (43 pCi kg^{-1} h^{-1}) [24]. Two studies of radon emanation from concrete containing fly-ash indicate that its radiological impact may be small: Stranden found the radon emanation rate from fly-ash concrete to be significantly lower than from otherwise identical concretes without fly-ash[20]; on the other hand, Smith et al.[21] found an average emanation rate of 0.41×10^{-5} Bq kg^{-1} s^{-1} (0.40 pCi $kg^{-1}h^{-1}$) for 20 samples of fly ash-concrete, 39% higher than that for control samples, but still at the low end of the range of emanation rates measured by Ingersoll for ordinary U.S. concretes. (In Smith et al., concrete components other than fly ash were selected to have low radionuclide contents.) Because of their low emanation ratios, emanation rates for brick are generally less than for ordinary concrete.

Radon emanation from components of building materials has been examined to some extent. Table 1 gives a few results for special materials; work referred to in the table also gives results for constituents of ordinary concrete. As is the case for brick, cement and

fly ash often exhibit very low emanation ratios; these low ratios may be associated with the fact that such materials ordinarily have very low moisture contents, affording little opportunity for recoiling radon atoms to come to a stop in pore spaces. Differing moisture contents may also account in part for the wide disparity in results from emanation measurements from different countries, e.g., the relatively low average concrete emanation ratio from the U.S.S.R. as compared with other countries, as seen in Table 1. These differences may also be due to differences in the microscopic structure of the materials studied.

In some cases, the radon source strength from building materials is given in terms of the exhalation rate (or flux) from the surface of the material, or even in flux per unit activity concentration (e.g., ref. 8). The flux depends on the macroscopic configuration of the material, but can be estimated from the emanation rate (given per unit mass) with knowledge of the material density and thickness; as discussed in the next section, if the material is relatively thick, the diffusion length is also required for accurate estimation. The ^{222}Rn flux from walls of ordinary concrete was measured to be 0.0046 Bq $m^{-2}s^{-1}$ (0.12 pCi $m^{-2}s^{-1}$) for a wall 0.2-m thick[25], and 0.0008 Bq $m^{-2}s^{-1}$ (0.02 pCi $m^{-2}s^{-1}$) for a 0.3-m-thick wall[24]. The U.S. data in Table 1 imply that flux from a 0.2-m thickness is in the range 0.0007 - 0.0021 Bq $m^{-2}s^{-1}$ (0.019 - 0.057 pCi $m^{-2}s^{-1}$), assuming a diffusion length of 0.13 m. Flux measurements in 15 paved basements in the U.S. gave a range of 0.0009 - 0.0067 Bq $m^{-2}s^{-1}$ (0.025 - 0.181 pCi $m^{-2}s^{-1}$), with a mean of 0.0028 Bq $m^{-2}s^{-1}$ (0.076 pCi $m^{-2}s^{-1}$)[26], suggesting the possibility of transmission of radon from the soil.

The flux can be compared relatively directly with the radon entry rate observed for a given indoor space. Flux is also the natural measure of radon emanating from soil, which is the major source of indoor radon in many cases. As in the case of soil, the flux of radon from building materials can vary considerably, not only with moisture content, but also with environmental parameters such as pressure and temperature, since these can directly affect the transport of radon.

Soil and rock have concentrations of elements in the ^{238}U and ^{232}Th decay series that vary over at least as great a range as the values in the building materials derived from them. In addition to the variation in the concentrations of the precursor radionuclides, the emanating fraction and transport-related parameters vary both with mechanical composition of the material and with environmental conditions, especially moisture content. Thus, to the extent that these source materials contribute to indoor radon, potential entry rates can be expected to vary over a wide range. Although our principal interest is the source of radon indoors, we first examine radium concentration and radon emanation rate for soil and rock itself and flux from ground that is not covered by a structure.

Average concentrations in soil of elements of the ^{238}U and ^{232}Th series are approximately 25 Bq kg^{-1} (0.68 pCi g^{-1})[1]. Reference 1 also cites typical ranges to be 10 - 50 and 7 - 50 Bq kg^{-1} (0.3 - 1.4 and 0.2 - 1.4 pCi g^{-1}) respectively; this contrasts with a less authoritative report citing a range of 0.7 - 66 Bq kg^{-1} (0.02 - 1.8 pCi g^{-1})[27]. Analysis of ^{226}Ra and ^{232}Th concentration in approximately 330 samples collected from 33 states in the U.S. showed ranges of 8.5 - 160 and 3.7 - 130 Bq kg^{-1} (0.23 - 4.2 and 0.10 - 3.4 pCi g^{-1}), respectively, with corresponding arithmetic mean values of 41 and 37 Bq kg^{-1} (1.1 and 1.0 pCi g^{-1}) [28]. Isolated reports of other measurements also indicate a range of 1-2 orders of magnitude for ordinary soils. For example, Pensko et al.[29] found a range of ^{226}Ra concentrations of 4 - 20 Bq kg^{-1} (0.1 - 0.5 pCi g^{-1}) for five samples from near Warsaw; nineteen measurements reported in Barretto et al.[30] indicate at minimum a range of 8 - 85 Bq kg^{-1} (0.2 - 2.3 pCi g^{-1}).

Values much higher than the ranges indicated above are associated with soils near uranium mining areas and mill tailings piles. Powers et al.[31] found a range of 15 - 1700 Bq kg^{-1} (0.4 - 46 pCi g^{-1}) for 28 samples collected near uranium mining and milling areas in Wyoming, New Mexico and South Dakota. The ^{226}Ra concentration reported for Jaduguda, Bihar, India, an area with known deposits of uraniferous minerals, vary from 40 - 200 Bq kg^{-1} (1-5 pCi g^{-1})[32]. Kalin and Sharma[33] measured ^{226}Ra concentrations in 63 samples from Canada collected at the surface and at a

depth of 20 - 25 cm at two mill tailings piles and found a mean of 1760 Bq kg^{-1} (48 pCi g^{-1}). Finally, Strong et al.[34], analyzing samples from two Australian tailings piles, measured ^{226}Ra concentrations of 11,000 and 200,000 Bq kg^{-1} (300 and 5400 pCi g^{-1}).

The range of values for rocks is expected to be similar to that for soils, with typical activity concentrations ranging from 0.4 - 60 Bq kg^{-1} (0.01 - 1.6 pCi g^{-1}) for ^{238}U series and 7-80 Bq kg^{-1} (0.19 - 2.2 pCi g^{-1}) for ^{232}Th series elements[8]. Results reported in Barretto et al.[30] indicate a minimum range of 6 - 740 Bq kg^{-1} (0.2 - 20 pCi g^{-1}) for the 50 samples they studied.

Measurements comparable to those for building materials have been performed of emanation rates or ratios for soil and rock, both in connection with interests in uranium exploration and for the use of ^{222}Rn as an atmospheric tracer; these measurements are reviewed by Tanner[5,6]. Pearson and Jones[35] measured radon emanation rates from 70 dried soil samples (excluding some from mining operations), mostly from Illinois, and found a mean value of 4.4×10^{-5} Bq kg^{-1} s^{-1} (4.3 pCi kg^{-1} h^{-1}) with a range of $(1.3 - 6.2) \times 10^{-5}$ Bq kg^{-1} s^{-1} (1.3 - 6.0 pCi kg^{-1} h^{-1}). Barretto et al.[30] report on both emanation rate and emanation ratio: fifty rock and nineteen soil samples yielded emanation rates in the ranges of less than 0.05×10^{-5} to 8×10^{-5} Bq kg^{-1} s^{-1} (0.05 - 8 pCi kg^{-1} h^{-1}) and $(0.3-9.2) \times 10^{-5}$ Bq kg^{-1} s^{-1} (0.2 - 9.5 pCi kg^{-1} h^{-1}), respectively. Corresponding ranges of emanation ratio were 1-26% for rock and 10-55% for soil. Consistent with these data are measurements of five samples of sandy soil by Pensko et al.[29] which yielded emanation ratios in the range of 12-50%.

Several measurements of source characteristics have been performed on soils near or under buildings in which indoor radon concentrations were being investigated, but systematic examinations have ordinarily not been performed. As examples, emanation rates of 2.2×10^{-5} Bq kg^{-1} s^{-1} (2.1 pCi kg^{-1} h^{-1}) were found as the average for two soil samples each from Maryland and California, each pair having average ^{226}Ra concentrations of approximately 40 Bq kg^{-1} (1.1 pCi g^{-1}), and therefore emanation ratios of roughly

25%[16]. A mean ^{226}Ra concentration of 37 Bq kg^{-1} (1.0 pCi g^{-1}) was found in soils adjacent to 19 houses in New York and New Jersey, with all but one value in the range 30 - 41 Bq kg^{-1} (0.8 - 1.1 pCi g^{-1})[26]. Emanation characteristics were not measured in this study, but fluxes into basements were, as mentioned above. Soil analysis for samples collected at two solar-heated apartment buildings in New England showed ^{226}Ra concentrations of 32 and 78 Bq kg^{-1} (0.86 and 2.1 pCi g^{-1})[36]. Soil samples collected in the crawl space of eight houses in Northern California and near Portland, Oregon, had radon emanation rates in the range (1.2 - 1.8) x 10^{-5} Bq kg^{-1} s^{-1} (1.2 - 1.8 pCi kg^{-1} h^{-1})[37]. Soil adjacent to an extensively studied house near Chicago was found to have 70 Bq kg^{-1} (1.9 pCi g^{-1}) of ^{226}Ra and an emanating fraction of 41%[38].

Because of the very small diffusion length of radon in crystalline materials, if ^{226}Ra were uniformly distributed in structurally-intact soil grains, the emanating fraction would be much smaller than the observed values. The inconsistency of this expectation with observations has been explained by suggesting first that the radium in soil grains may exist, in part, in crusts or films on the grain surface and, second, that chemical corrosion, weathering, and small-scale fracturing may lead to large internal surface areas and substantial internal voids[5,6]. These hypotheses are supported by various data. For example, Megumi and Mamuro[39] measured the radium contents of two size-segregated samples of soil from weathered granite and found, in one, monotonically increasing concentrations of ^{226}Ra, ^{224}Ra and ^{228}Ac with decreasing particle size (and therefore increasing surface area). They found that particles smaller than 200 mesh (i.e., those passing through a screen having 200 wires per inch), with a mean diameter of 20 μm, had 7-10 times the activity concentrations of particles larger than 5 mesh, having a mean diameter greater than 2800 μm. They also found the emanation ratio of ^{220}Rn to be independent of particle size, and about 10% for that sample. In contrast the ^{222}Rn emanation ratio for the small size fraction was 25%. Because the ratio of the decay constant of ^{222}Rn to that of ^{220}Rn is approximately 6100, the ratio of the diffusion lengths is roughly 80, so this result too suggests that the emanating atoms are produced principally in a surface layer. In a later study by the same authors, the activity ratio of ^{226}Ra to ^{238}U was

found to be approximately 2.0 for two soil samples, in contrast to values of 0.3 and 0.1 for samples of river and sea water, respectively, suggesting that ^{238}U may be dissolved and that its progeny may later be absorbed onto grain surfaces[40]. Jasinska et al.[41] found corroborating results: for 70 soil samples collected in the Krakow province, the ^{238}U and ^{232}Th concentrations (assuming radioactive equilibrium) increased linearly with the fraction of sample mass contained in particles with diameters less than 0.02 μm. They concluded that mechanical composition, rather than soil type, determined ^{232}Th and ^{238}U contents. On the other hand, Myrick et al.[28] concluded that for most of the 356 samples of surface soil they analyzed, radioactive equilibrium existed between ^{238}U and ^{226}Ra. And while Andrews and Wood[42] found increasing radon emanation with decreasing particle size in two of the three samples they studied, they suggest that the difference was primarily due to increasing emanation ratios rather than increasing radionuclide content.

The emanation ratio of soils has been observed to vary with moisture content: more radon emanates from soil when it is moist or saturated then when it is dry. Megumi and Mamuro[39] found ^{220}Rn emanation to vary from 10% for a dry sample to 12-13% for a sample having 2-8% water content. Strong and Levins[43] found the ratio of saturated to dry emanation ratios to be 3.9 ± 0.6 for ^{222}Rn released from eight samples of uranium ore and mill tailings. This phenomenon results from the increased stopping power of water compared to air for the recoiling radon atom: whereas the atom may traverse an air-filled pore and become embedded in another grain, the water-filled pore is likely to stop it, permitting radon transport out of the material.

Flux from the soil surface, combining emanation rate and diffusive transport properties, is a practical indicator of soil as a ^{222}Rn source. The world-wide average flux of ^{222}Rn from dry land has been estimated to be 0.016 Bq m^{-2} s^{-1} (0.43 pCi $m^{-2}s^{-1}$), with a range of 0.0002 - 0.053 Bq m^{-2} s^{-1} (0.005 - 1.4 pCi $m^{-2}s^{-1}$), the lowest values having been measured over lava[44]. Flux of ^{220}Rn from dry, uncovered soil, measured at six sites in New Mexico, was found to be 1.6 ± 0.3 Bq m^{-2} s^{-1} (42 ± 9 pCi $m^{-2}s^{-1}$) [45]. (The higher activity flux for ^{220}Rn than for ^{222}Rn reflects its shorter

half-life.) Some recent measurements of ^{222}Rn flux have been performed in connection with studies of radon indoors: the flux in the unpaved crawl space of a house with high indoor concentrations was found to be 0.27 Bq m^{-2} s^{-1} (7.3 pCi $m^{-2}s^{-1}$)[46], considerably higher than Wilkening's range; the basement fluxes mentioned above[26] may include a significant contribution from soil.

Clearly the flux from the soil surface depends on how radon moves through porous media. Radon transport and entry into buildings will be discussed in succeeding sections. However, the information above is sufficient to make a useful comparison of the measured overall entry rates into buildings and the strength of sources that contribute to these entry rates. The nominal entry rate of 15 Bq $m^{-3}h^{-1}$ (0.4 pCi $1^{-1}h^{1}$) mentioned earlier can be entirely accounted for by emanation from building materials in a room with floor, ceiling, and walls of 0.2-m-thick concrete with a flux of 0.002 Bq $m^{-2}s^{-1}$ (0.054 pCi $m^{-2}s^{-1}$), about 30% greater than the mean for U.S. concretes[16], and a surface-to-volume ratio of approximately 2 m^{-1}. However, the larger range of entry rates for single family dwellings cannot easily be attributed to building materials. Figure 1 shows a frequency distribution of entry rates for U.S. housing, on which is indicated the range of emanation rates for U.S. concretes[16], converted to an entry rate on the assumption that houses are one-story structures with a 0.2-m-thick concrete floor and that walls and ceiling do not contribute. The range of contributions from measured fluxes from soil[44] is also indicated. It is clear that the emanation rates from concrete cannot account for the observed entry rates, but that the fluxes from soil can, provided that in a substantial number of cases the effective flux from soil under the house is comparable to the flux from uncovered soil. Understanding the movement of radon through source materials and into buildings, particularly those in close contact with the soil, is one of the principal challenges in characterizing indoor radon.

TRANSPORT IN BUILDING MATERIALS AND SOIL

The characterization of radon sources in terms of radium concentrations, emanation ratios, emanation rates, or fluxes, while useful, is incomplete, both because of the dependence of these parameters on environmental parameters and because actual transport or entry mechanisms are more complex than indicated by emanation rates or diffusive fluxes from, for example, uncovered soil. Transport of radon from its generation site may occur via several mechanisms, described by many authors (e.g., refs. 5,6,22,24,47). Two of these cause movement on a local scale, i.e., molecular diffusion and flow of pore gas, while movement of radon over larger distances may occur along with water or through cracks and fissures. Careful consideration of diffusion and flow is necessary to understand the entry of radon into buildings, as is the influence of environmental parameters such as barometric pressure, temperature, windspeed, and moisture.

Diffusion can be described by a flux, J, that is proportional to the gradient of the concentration, C:

$$\vec{J} = -D\,\vec{\nabla}C \qquad (2)$$

For a porous medium, if C is taken to be the concentration in the pore volume, $\vec{J}$ designates the transport rate per cross-sectional area of either the bulk material, so that D is the effective bulk diffusion coefficient k_e, or the pore volume, in which case D is an effective interstitial diffusion coefficient k_e^*. As usually interpreted $k_e = \varepsilon k_e^*$, where ε is the soil porosity [48]. Because of conservation considerations, the concentration obeys the diffusion equation with decay and production terms:

$$\frac{\partial C}{\partial t} = k_e^* \vec{\nabla}^2 C - \lambda_{Rn} C + G_v \,, \qquad (3)$$

where G_v is the radon emanation rate per unit pore volume, $G_e\rho/\varepsilon$, ρ being the bulk density.

This equation has a characteristic diffusion length, $\ell = \sqrt{k_e^* / \lambda_{Rn}}$, and the bulk flux (e.g. at a surface) is then

$$\vec{J} = k_e \vec{\nabla} C = \varepsilon k_e^* \vec{\nabla} C = \varepsilon \lambda_{Rn} \ell^2 \vec{\nabla} C, \tag{4}$$

where C depends on ℓ, λ_{Rn}, and G_v.

We note the steady-state one-dimensional solutions for soil of infinite depth, for a slab of building material with thickness L, and for a combination of the two. The radon concentration in soil gas at a depth z below the surface, assuming the concentration at the surface is approximately zero, is

$$C(z) = C_\infty \left(1 - e^{-z/\ell} \right), \tag{5}$$

where $C_\infty = G_v/\lambda_{Rn}$ is, as expected, the pore radon concentration at large depths. The flux at the surface is therefore

$$J = \varepsilon \lambda_{Rn} \ell C_\infty = \varepsilon \ell G_v = \ell (G_e \rho), \tag{6}$$

i.e., simply the product of the diffusion length and the emanation rate per unit volume of the bulk material. Similarly, the exhalation rate from each surface of a slab is easily shown to be[24]

$$J = \ell \, (G_e \rho) \tanh\left(\frac{L}{2\ell}\right), \tag{7}$$

which reduces to equation (6) for large L, and to $\frac{L}{2}(G_e\rho)$ for $L \ll 2\ell$. Even for $L = 2\ell$, $J = 0.76\,\frac{L}{2}(G_e\rho)$, so that for typical diffusion lengths the diffusive flux depends primarily on the thickness of the slab and to a lesser degree on the diffusion length. Finally, for an uncracked slab lying on the ground, the radon flux transmitted by the slab[47] is

$$J_T = J_{Free}\left[\cosh\left(\frac{L}{\ell_s}\right) + \frac{\varepsilon_g \ell_g}{\varepsilon_s \ell_s}\sinh\left(\frac{L}{\ell_s}\right)\right]^{-1}, \qquad (8)$$

where J_{free} is the flux from soil without the slab (equation 6) and subscripts s and g refer to slab and ground, respectively. The larger term in the brackets is usually the second, which may often be in the range 10-100.

A number of workers have measured the diffusion characteristics of radon in concrete, as summarized in Table 2. Measured porosities range from less than 0.1 to greater than 0.3, consistent with the current understanding of the porosity of concrete and its components[47]. Diffusion lengths for ^{222}Rn range from less than 0.1 m to approximately 0.3 m (for some lightweight concretes), corresponding to interstitial diffusion coefficients in the range (2-20) x 10^{-8} $m^2 s^{-1}$. The variability of these parameters does not appear to be connected with the porosity or the type of concrete. Combined with the radon generation rates discussed above, these diffusion characteristics for concrete give exhalation rates that are consistent with the fluxes actually observed.

Soil is significantly different from building materials with respect to diffusion lengths and their dependence on environmental parameters, specifically moisture. A typical diffusion length for ^{222}Rn in soil is approximately 1 m (corresponding to an interstitial diffusion coefficient of 2 x 10^{-6} m^2 s^{-1}), but can decrease to the order of 0.01 m if the soil becomes saturated[5,43]; the diffusion length can exceed 1.5 m, e.g. for dry sand, but ought always to be substantially less than that for air, 2.2-2.4 m[5]. (In general, diffusion lengths for ^{220}Rn are approximately 1/80 those of ^{222}Rn because of their different decay rates.)

The porosity of soil is a factor in determining diffusion characteristics and permeability. For dry soil, without organic matter, porosity ranges from as low as 37% for some sandy loams to as high as 63% for some silt loams[49], assuming the average soil grain density to be 2.7×10^3 kg m^{-3} [50].

Sample calculations illustrate the internal consistency of the soil data and demonstrate the inadequacy of diffusive transport in accounting for many cases of high radon entry rate. A soil with a ^{226}Ra concentration of 25 Bq kg^{-1} (0.68 pCi g^{-1}), a bulk density of 1.5×10^3 kg m^{-3}, a porosity of 0.45, and an emanation ratio of 0.25 would have a ^{222}Rn concentration in soil gas well below the surface of 2.1×10^4 Bq m^{-3} (560 pCi l^{-1}), within the range of $(0.7-22) \times 10^4$ Bq m^{-3} (200 - 6000 pCi l^{-1}) observed by various workers (e.g., refs. 51-53). For a diffusion length of 1.0 m, the surface flux would be 0.021 Bq $m^{-2}s^{-1}$ (0.56 pCi $m^{-2}s^{-1}$), close to the world-wide average flux cited by Wilkening et al.[44].

A structurally-intact concrete slab, having a thickness of 0.2 m, a porosity of 0.25 and a diffusion length of 0.15 m, if placed atop this soil would transmit 4.3% of this flux, i.e. about 0.0009 Bq $m^{-2}s^{-1}$ (0.02 pCi $m^{-2}s^{-1}$). This flux is comparable to the low end of the range of fluxes expected from the concrete itself and while together they constitute a large portion of the flux observed through basement slabs[26], they are significantly less than the flux required to explain U.S. source strengths in general. The transmission factor may also be less than 4.3%: Colle et al.[47] cite 2-4% for a 0.1-m-thick slab. The presence of cracks in the slab may increase the transmission of the diffusive flux from the soil considerably. Using a mathematical model of a cracked slab, Landman[54] determined that 25% of the flux from uncovered soil would penetrate the slab if a 1-cm gap existed for every 1 m of slab. Even for such relatively large penetrations, however, the resulting diffusive flux is still very much smaller than the observed entry rates in some houses.

Flow of radon-bearing air through soil and building materials is an important mechanism for radon entry. Pressure differences induce a fluid flow per unit cross-sectional area that, according to Darcy's law, is proportional to the pressure gradient:

$$\vec{V} = -\frac{K}{\mu}\vec{\nabla}P \ , \qquad (9)$$

where K is the permeability of the medium, μ is the dynamic viscosity (18 $\times 10^{-6}$ nt s m^{-2} for air at 10°C), and P is the pressure. The effect of small variations in the pressure (relative to average atmospheric pressure) can be described by a second order differential equation analogous to the diffusion equation; however, for most purposes flows stabilize relatively quickly after a change in pressure, so that steady-state solutions are adequate for describing radon transport[47].

The significance of flow as a transport mechanism is the possibility that a greater volume of radon-generating material in and around a structure may contribute to the entry rate than is possible due to diffusion alone. One possibility is that air may flow through the bulk of the building materials, i.e., the walls or floor (e.g., in a basement) of the structure, in which case the permeability of the structural material is of direct interest. A more likely possibility is that pressures in the surrounding soil may drive radon toward the understructure and through openings in structural elements.

The limited data available on the permeability of concrete to air flow indicate that bulk air flow through structurally-intact concrete is unlikely to be an important contributor to radon entry rates. Flow through one concrete, perhaps fabricated to have low permeability, was 1.45×10^{-8} $m^3 s^{-1} m^{-2}$ for a pressure difference of 3500 Pa applied across 0.1 m, indicating a permeability of 8×10^{-18} m^2 [55]. McLaughlin and Jonassen[56] measured the radon exhalation from a basement wall into a can that was held at a reduced pressure. They found a linear dependence of exhalation rate

on pressure for underpressures in the range 700 - 3300 Pa, and observed a flux of 0.0051 Bq $m^{-2}s^{-1}$ (0.14 pCi $m^{-2}s^{-1}$), approximately 6 times the flux at atmospheric pressure, for an underpressure of 3200 Pa. Assuming their wall to have a thickness of 0.3 m, and that the flows of (3.0 - 20) x 10^{-7} $m^3s^{-1}m^{-2}$ required to maintain the can at the desired underpressure passed through the concrete, the permeability of that wall is on the order of 3 x 10^{-15} m^2.

It is unclear whether pressure differences on the order of 1000 Pa, such as those induced by barometric pressure variations, can be maintained across a wall even for as long as a few hours. A much smaller pressure difference, commonly on the order of 5 Pa, may persist, induced by buoyancy resulting from a higher temperature indoors than outside. In this case the contribution to radon entry of flow through the concrete is certain to be small: for a basement having 0.2-m-thick walls and floors of concrete having a permeability of 3 x 10^{-15} m^2, and a below grade surface area of 200 m^2, a pressure difference of 5 Pa applied uniformly across the walls and floor would lead to a flow of 8 x 10^{-7} m^3s^{-1}. Taking the concentration of radon in the soil gas surrounding the basement to be 2.1 x 10^4 Bq m^{-3} (570 pCi l^{-1}), and ignoring the decay of radon during transit through the concrete, this component of the flux would be approximately 9 x 10^{-5} Bq $m^{-2}s^{-1}$ (0.003 pCi $m^{-2}s^{-1}$), or 6% of the diffusive flux of radon from the average U.S. concrete[26].

Regardless of the magnitude of pressure difference across a concrete slab, flow through cracks, holes, and other penetrations is likely to dominate flow through the intact slab. For the example in the preceding paragraph, the flow through a hole of 2.3-mm diameter would equal the flow through the floor and walls. Penetrations through concrete in houses probably have a total area several orders of magnitude larger, so for practical purposes flow through intact concrete can be ignored.

The limited number of measurements of the air permeability of surface soil yield values in the range (0.007 - 3) x 10^{-10} m^2 [57-59], with the permeability of a given soil depending highly on the degree of compaction.

Even though these data show a broad range, much lower values are to be expected for packed clay and much higher for gravel. Little has been done to correlate this information with experiments on the transport of radon through or from soil under various meteorological conditions. The ^{222}Rn flux from the soil surface[60] and the concentration in soil gas[51] have been observed to change inversely with changes in barometric pressure. Kraner et al.[51] interpreted these changes to be due, possibly, to a piston-like displacement of radon-laden soil gas, after which a new equilibrium might be established over a period of days due to diffusion and decay. Clements and Wilkening[60] found good agreement between a theoretical model, which combined flow and diffusion effects during barometric pressure changes, and experimental results. Additionally, it has been observed that radon concentrations at moderate depths are depleted during periods of high wind speed[51,52], and it has been suggested that this is due to turbulent pumping, i.e., local pressure changes in the vicinity of the soil surface. Due to the low velocities very near the soil surface, the possibility of a classical Bernoulli effect is discounted; however, possible correlations between wind speed and barometric pressure were not mentioned.

Water-borne transport can be an important means by which radon enters residences. Radon concentrations in water have been observed to vary over an extremely wide range - at least four orders of magnitude - and to reach very high levels, often exceeding 10^6 Bq m^{-3}. Generally the highest concentrations of ^{222}Rn in water are observed in drilled wells, especially in granitic areas. Lower concentrations are found in water from dug wells, and surface water sources typically have the lowest concentrations. Hess et al.[61] measured ^{222}Rn concentrations in water from 2000 wells in Maine and found a range of 7×10^2 - 6.7×10^6 Bq m^{-3} (20 - 180,000 pCi l^{-1}); they observed an average concentration of 8.2×10^5 Bq m^{-3} (22,100 pCi l^{-1}) for wells in granite areas, an order of magnitude greater than the average for wells in chlorite areas. Results consistent with the lower mean value were reported in a review of radon measurements in well and spring water in the United States which showed that, excluding New England, 74% of 438 samples analyzed had ^{222}Rn concentrations of less than 7.4×10^4 Bq m^{-3} (2000 pCi l^{-1}), and only 5% exceeded 3.7×10^5 Bq m^{-3}

(10,000 pCi l^{-1}) [62]. Likewise, the average concentration provided to the 40% of the Swedish population served by public water works derived from ground water sources is 2.2×10^4 Bq m^{-3} (600 pCi l^{-1}); however, concentrations as high as 10^6 Bq m^{-3} (2.7×10^4 pCi l^{-1}) have been measured[63]. On the other hand, 45% of the Swedish population is served by water from surface sources having an average ^{222}Rn concentration less than 2000 Bq m^{-3} (50 pCi l^{-1}). (The remaining 15% of the population is served by private wells, not yet surveyed.) A survey of ^{222}Rn in drinking water in Finland showed a mean concentration of 2.5×10^4 Bq m^{-3} (670 pCi l^{-1}) in municipal supplies, in contrast to a corresponding value for drilled wells of 6.3×10^5 Bq m^{-3} (1.7×10^4 pCi l^{-1}) [64].

Studies of radon transfer from tap water to indoor air cite average per-person water use of 0.2 - 0.4 m^3 per day, and a use-weighted transfer efficiency of 0.5 - 0.6 [65,66]. Thus for four persons living in a house with a volume of 250 m^3 the radon entry rate via domestic water having a concentration of 10^5 Bq m^{-3} (2700 pCi l^{-1}) is expected to be in the range 7 - 16 Bq m^{-3} h^{-1} (0.2 - 0.4 pCi l^{-1} h^{-1}), comparable to the geometric mean of 14 Bq $m^{-3}h^{-1}$ (0.37 pCi l^{-1} h^{-1}) previously cited for a sample of U.S. housing. Because the concentration of ^{222}Rn in water can exceed this value by more than an order of magnitude, water-borne transport can in some cases contribute to radon entry at a rate comparable to the higher rates observed. On the other hand, surface water supplies, containing ^{222}Rn at a typical concentration of 2000 Bq m^{-3} or less, will contribute no more to the indoor concentration than a concrete slab floor.

Discounting the effect that water has on the emanation ratio, the equilibrium radon concentration in soil and rock pores should be independent of the pore fluid. It is noteworthy, then, that the highest concentrations observed for radon in water exceed the corresponding values reported for soil gas by almost two orders of magnitude, suggesting, perhaps, that the range reported for the latter may not reflect the higher values that may occur in some circumstances.

ENTRY OF RADON INTO BUILDINGS

The basic transport mechanisms discussed above afford ample opportunity for movement of radon within and from building materials or the soil surrounding buildings. The extent to which they account for actual entry rates depends on the specific structural characteristics of buildings, the diffusion of radon through structural elements, and the flow of radon-bearing air due to driving forces induced by winds, temperature differences or changes in barometric pressure. The potential effect of barometric pressure has been discussed above. It is the potential importance of winds and temperature differences, the same factors that affect the overall infiltration rate for buildings, that has been the focus of recent investigations, the implications of which are by no means fully understood. The premise for this attention is the desire to account for entry rates into buildings (even those having concrete understructures) that are comparable to the free flux expected from the underlying soil.

Before discussing some recent lines of investigation, it is useful to set forth the essentials that describe how air enters buildings through their shells, as opposed to entry through windows or ventilation systems. Infiltration rates are presently understood in terms of pressure across the shell that arise from two sources: winds and temperature differences. The first cause is superficially obvious, although the details have substantial subtlety and can be represented on a fundamental basis only with difficulty. The dependence of infiltration on wind speed has been parameterized as part of infiltration models as

$$Q_{wind} = A_o \, f_w \, V \, , \qquad (10)$$

where V is wind velocity, A_o is the effective leakage area of the building and f_w is the wind parameter, which accounts for local and terrain shielding effects, the distribution of leakage area in the building envelope, and the height of the building relative to the height at which wind speed is

measured[67]. As noted below, the details of how wind speed varies with height near the ground may be very important in characterizing how radon entry depends on this factor. Moreover, wind-induced pressures might be expected to drive radon-rich soil-gas into a structure on one side, while inducing indoor air to be drawn into the soil from the other.

The dependence of infiltration on temperature differences, particularly during cold seasons, is essentially due to a "stack" effect, where the higher temperature indoors implies a pressure across the building shell that varies (in sign as well as in magnitude) with height, inducing a convective loop that carries air into the building near the ground and out of the building toward the top of the structure. This driving force has also been parameterized in a simple way as part of an infiltration model:

$$Q_{stack} = A_o \, f_s \, \Delta T^{\frac{1}{2}} \qquad (11)$$

where ΔT is the absolute value of the temperature difference between indoors and outdoors and f_s is the stack parameter, which accounts for the building height and the distribution of leakage area[67].

Infiltration models are useful not only in considering the overall air-exchange rates induced by meteorological conditions, which thereby affect indoor concentrations, but also because they give an indication of the potential for comparable forces to drive radon into buildings via the understructure. The total rate at which soil gas would have to enter buildings to account for the observed entry rates is a small percentage of the total air exchange rate[11,12,68]: the geometric mean entry rate from the U.S. data, 14 Bq $m^{-3}h^{-1}$ (0.4 pCi $l^{-1}h^{-1}$), implies a soil-gas entry rate of only 0.0007 h^{-1} (assuming a soil-gas concentration of 2.1 x 10^4 Bq m^{-3}), only 0.1% of the total air-exchange rate typical of houses.

Infiltration models, and experimental data such as that discussed below, show that air exchange is driven by pressure differences of only a few Pa, far less than changes in barometric pressure; these larger pressure

differences in any case, cannot persist across the above-ground structural shell and probably persist only for short times (i.e., hours or less) across the understructure components and the surrounding soil as suggested in preceding sections. Given typical air-exchange rates for residences of 0.5 - 1.0 h^{-1} [69] any radon entering due to a short-lived pressure difference will be removed within several hours. A key question then is whether small but enduring pressure differences can drive significant amounts of radon through a substantial thickness of soil and into a building. Indications are that it can.

Significant attention has recently been given to the question of radon entry via soil gas in countries such as Sweden, the United States, Canada, and Great Britain, where portions of the housing stock have been found to have unusually high radon concentrations. The remainder of this section will indicate tentative results of ongoing research efforts of our group and others of which we are aware.

Radon entry into basements, especially in residences, has been a subject of extensive investigation over the last several years (e.g. refs. 10,26). The element that has recently been added to these studies is the detailed investigation of entry modes experimentally and to some extent, theoretically. These investigations seek to examine both radon source parameters and putative driving forces at a level of detail that is adequate to form a consistent picture of actual entry modes and directly observed indoor concentrations, a picture that does not exist as yet. The results of such investigations may then serve as a basis for reducing entry rates in circumstances where they are deemed excessive.

An example of such a study is an experiment mounted in a single-family house with a basement near Chicago, in which - for a period of five months - real-time measurements were performed of the indoor radon concentration and air-exchange rate, source-related parameters, and meteorological factors[38], using a monitoring system based on one described earlier[70]. A time-varying entry rate can be calculated from the measured radon concentration and air-exchange rate; the effect of driving forces on this entry rate and on radon concentrations in soil gas and at entry points can then

be examined. Figure 2 gives an example of the radon data for a week-long portion of the experiment. Here, in spite of the fact that the drain tile entrance into the sump was occluded by water throughout the monitoring period, the radon level at the sump is seen to vary substantially and to reach quite high values. The indoor radon concentration changes in correspondence with sump activity, implying that the same factors that affect the latter also influence radon entry, and suggesting that the sump may constitute an important pathway for radon entry into the house. The cause of the rapid increases in sump activity has not been identified, neither have the dips in soil gas activity, which seem to occur regularly during this period just prior to increases in sump activity, been explained.

Thus, detailed interpretation of the results of such an experiment is not straightforward, especially when the source distribution has a "singularity" - in this case a sump and drain-tile system - that is not well characterized. Canadian workers have had some success in dealing with such systems in a very practical way by, for example, adding a water trap where one didn't exist or providing sub-floor ventilation[71]. The main conclusion to be cited from the Chicago study is the inference that the radon entry rate appears to have two components, one that is independent of air-exchange rate (and, presumably, of the forces that drive it) and the other that is proportional to the air-exchange rate. The first component therefore acts like the entry of radon by diffusion, while the second behaves like pressure-driven entry. Estimation of the expected diffusion rate through the concrete basement, using parameters measured for concrete from the Chicago area[16,72], yielded an entry rate of approximately 2 Bq m^{-3} h^{-1} (0.06 pCi $l^{-1}h^{-1}$), which - as it happened - equalled the "diffusive" entry rate extracted from the experimental data. Accounting for the component that was dependent on air-exchange rate required that a pressure difference of about 3.5 Pa be able to draw 0.5 x 10^{-3} m^3s^{-1} of soil gas from of the soil surrounding the basement, which appeared possible if the permeability was relatively high or if a comparable flow of air was available along the exterior basement walls to act as a carrier of radon diffusing to the walls. In this way an entry rate of 63 Bq $m^{-3}h^{-1}$ (1.7 pCi $l^{-1}h^{-1}$) could be accounted for at this site.

It therefore appears possible that observed radon entry rates for buildings with basements are within the range that could be expected based on emanation and transport characteristics of the building materials and surrounding soil, provided pressure-driven flow is considered. Fundamental modeling of pressure differences and radon concentrations within the soil, of the type that has recently begun[73], will be required to verify this hypothesis. Even for the specific case of basements, considerably more experimental work is required to determine entry routes and transport mechanisms with some degree of certainty. An intriguing view of the processes proposed here, involving pressure differences that are wind or stack driven, is that a building effectively draws radon from the ground in a manner that is considerably more subtle than the "piston" action associated with changes in barometric pressure.

Radon entry through crawl spaces is known to be significant when the crawl space is unpaved and unvented[46]. However, a large number of U.S. homes have crawl spaces that are vented; in some of these, vents are being sealed to save energy, and plastic sheeting is placed over the soil in crawl space to prevent build-up of excessive moisture in the understructure. Investigations have recently been carried out in a few homes in California and Oregon, not only to ascertain the effect of such measures on radon entry, but also to determine the transmission of radon across crawl spaces that are not altered in this way[37]. These studies have monitored an array of parameters that is comparable to the basement study just described, except that air-exchange rate is not measured continuously, but rather is calculated based on an infiltration model[67] and real-time measurement of meteorological parameters.

Preliminary results show that a fairly large fraction of the radon flux from soil is transmitted by the crawl space (even when vents are left open), that a stack effect may contribute in some cases to raising the flux above the level expected from open soil, and that even unusual transport processes may at times occur. For example, Figure 3, a plot of data taken at a house in Oregon, shows that during a period of substantial rainfall and decreasing barometric pressure around March 30, 1983 the radon entry

rate rose markedly, suggesting that with reduced permeability in the surrounding soil the house became the route for soil gas to escape at a time of falling pressure. No such effect is observed on March 13, another day of heavy rainfall, probably because the barometric pressure was rising during that day.

This work has also entailed use of a tracer gas (sulfur hexafluoride) to study the degree of transmission between the crawl space and the interior of the house, one more type of measurement that is useful in characterizing radon entry into houses of this type. Indications are that during the heating season a primary mechanism by which a crawl space is ventilated is stack-effect driven flow into the house through the floor, the air being replaced by flow into the crawl space through the vents. Cross ventilation, driven by wind and by temperature differences between the crawl space and outdoor air, appears only comparable despite the large openings that the vents constitute, presumably because the winds speeds are low at the height of the vents and the temperature difference is small.

Other types of buildings have radon entry routes that differ significantly from those just indicated. The case that is most similar is that of a slab-on-grade building, in which case many of the same considerations applying to basements may be pertinent, albeit in a simpler geometry. Setting aside the question of pressure-driven transport for a moment, diffusive movement can of course be understood in terms of the more straightforward considerations set forth in earlier sections. However, as in the cases above, an essential element is the availability of experimental and theoretical results that are adequate to determine the relative importance of the different transport mechanisms in the circumstances of interest. As the number of stories in the building under study increases, the problem simplifies (at least in the absence of complicated ventilation systems) to the case where simple diffusion from the materials in the structure is the predominant radon entry mechanism, with the possibility of very small departures due to pressures across the above-ground structural components.

Certain building designs, not in common use now but attractive because of their energy efficiency, have the potential for high radon entry rates. Two examples are earth-bermed houses and solar houses; in either case high entry rates may arise because of the large amounts of crustal materials associated with the building structure. In such cases specific attention must be given to limiting the pathways for radon entry.

IDENTIFICATION AND CONTROL OF HIGH ENTRY RATES

Encouraging evidence that very high radon entry rates can be practically reduced or avoided is contained in the results of radon studies in areas where high indoor concentrations are observed. In such areas only a moderate fraction of the houses are usually observed to have very high concentrations; others have concentrations that are in the normal range. It is therefore possible to build houses with typical concentrations in areas having potential for high indoor levels. Thus, drawing on our current understanding of radon emanation, transport, and entry, we must address two questions: 1) what circumstances lead to high entry rates and 2) what can be done to control them. As a basis for considering the prospects, it is useful to reiterate the main conclusions from the previous sections.

The relative importance of various sources clearly depends on the circumstances. As an important example, emanation rates for ordinary U.S. concretes are in the range $(0.4 - 1.3) \times 10^{-5}$ Bq $kg^{-1}s^{-1}$ (0.4 - 1.3 pCi $kg^{-1}h^{-1}$), leading to fluxes from 0.2-m-thick concrete of about 0.0007 - 0.0021 Bq $m^{-2}s^{-1}$ (0.02 - 0.06 pCi $m^{-2}s^{-1}$). In many buildings, such fluxes can account for the observed indoor radon concentrations, but these cases tend to have concentrations at the low end of the range observed. Building materials with elevated ^{226}Ra concentrations, and correspondingly high emanation rates or fluxes, can contribute much more substantial indoor concentrations, even to the level of one or two orders of magnitude higher than the average. It is therefore clear that continuing attention ought to be given to the possibility of elevated emanation rates due to use of specific natural materials or industrial byproducts. The latter materials

are easily subjected to the scrutiny of routine monitoring programs prior to decisions about incorporating them into building materials, for example, in the manner of investigations being carried out in Europe and the United States on the use of fly ash in concrete. Delineating potentially high contributions from natural materials is more difficult unless there is some reason a priori to suspect that they may have high emanation rates. This difficulty may be solved partially by associating this question with the characterization of soil as a potential source of radon.

Similarly, most water supplies do not contribute substantially to the radon entry rate. However, the occurrence of large contributions in some circumstances, e.g., ^{222}Rn concentrations in water of 10^6 Bq m^{-3} (30,000 pCi l^{-1}) which can contribute 100 Bq $m^{-3}h^{-1}$ (3 pCi $l^{-1}h^{-1}$) to the radon entry rate, requires that one ask how to identify such problems systematically. Some countries are approaching this question directly by performing measurements on community water supplies derived from underground sources. Generalizing from results of specific measurements or even such wide-scale programs may be facilitated by considering some of the same factors that apply to the question of characterizing soil as a radon source.

The most substantial issue for many smaller buildings, including but not restricted to single-family residences, is the contribution from soil. As indicated in data already available, in many cases the contribution to indoor concentrations from the soil must approximate the flux from open soil, which averages about 0.016 Bq $m^{-2}s^{-1}$ (0.43 pCi $m^{-2}s^{-1}$). As suggested in the last section, there appear to be entry routes and transport mechanisms that can permit the bulk of the flux from soil to appear inside buildings. It would be useful to be able to predict the contribution to be expected from soil in specific circumstances and to indicate methods for controlling excessive rates.

Characterizing geographic areas in respect to radon source strength from the ground would be the optimum solution, although even if possible it would be hard to achieve. One approach would be to perform flux measurements on a wide scale. However, these depend on meteorological conditions at the time of the measurement and do not indicate a great deal about the

soil characteristics in respect to pressure-driven transport. An alternative approach is to investigate radionuclide content and soil characteristics on a wide scale. This approach is attractive particularly in countries such as the United States where the entire country has been mapped for potential uranium sources by aerial radiometric measurement of the ^{214}Bi gamma ray at 1.76 MeV. This is of course not a measure of ^{222}Rn emanation rates, but it is a fundamental parameter that, together with other information on soil characteristics (particularly those related to diffusion and flow), could serve as a useful indicator to areas with potentially high source strengths. Because of the difficulty of characterizing as large an area as the United States in any other way, the potential for this approach is being investigated. It is useful to note that this approach may correlate well with a systematic effort to characterize the radon content of ordinary building materials or of domestic water supplies, since both of these are usually derived from local natural sources. However, the efficacy of this approach depends critically on a better understanding of how radon actually enters buildings from the ground and how measurement of particular source-related parameters can serve as a guide to the entry rate. These same considerations relate to the question of how one might reduce the entry rate.

Control of high entry rates from the soil is substantially more difficult than control of radon entry from building materials or water. The ultimate control measure for building materials is attention to the constituents; and radon can be removed from water by aeration or storage. Control of radon entering from soil requires attention to the questions indicated above and, in addition, formulation of means to inhibit entry. Specific control measures, such as sealants and sump or sub-floor ventilation, are beyond the scope of this paper. However, the kind of experimental and theoretical work summarized in the last section is intended to lead to practical and inexpensive means of controlling radon entry in new buildings as well as those already existing. In the latter case, efforts can be directed to blocking specific points of entry, provided the scientific basis for identifying them has been established and the monitoring methods for localizing them have been devised.

It thus appears that we have an understanding of radon sources that is sufficient to indicate the nature of the problem and, in particular, the circumstances in which high radon entry rates may occur. Substantial experimental and theoretical work remains to be done before a capability for identifying and controlling the cases of interest can be established. Only then will it be possible, on a systematic basis, to implement standards that have as one goal the limitation of radon entry rates to specific levels in new and existing buildings.

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Table 1. ^{222}Rn source characteristics of building materials.

Country / Material	No. Samples	^{226}Ra (Bq kg^{-1})	^{222}Rn Eman. Rate (10^{-5} Bq kg^{-1} s^{-1})	Eman. Ratio	Comments	Ref.
USSR						(22)
heavy concrete	18	66	0.32	0.035		
lightweight concrete	19	141	0.41	0.021		
red brick	12	50	0.16	0.017		
Hungary					Samples weighted to yield country-wide average	(23)
concrete	~100	13	0.78	0.28		
red brick	~200	55	0.39	0.036		
Poland						(29)
fly ash	33	96		0.0054		
slag	11	67		0.0070		
phosphorite	6	705		0.20		
red brick	3	18		0.023		
Denmark						(24)
concrete	4		0.47			
alum-shale concrete	1		44		Swedish, old-type	
brick	2		0.017			
gypsum board	1		0.023			
Norway					Emanation ratio determined from subset of samples	(25)
concrete	137	28		0.01-0.20		
brick	18	63		0.01		
Leca[a]	12	52		0.01-0.20		
United States						(16)
concrete	106/21[b]	9-32	0.43-1.26	0.13-0.25	10 metropolitan areas	
fly-ash concrete	8	19	1.04	0.26	Knoxville, Tennessee	
red brick	6	45	0.10	0.01	California	
gypsum	12	12	0.63	0.28	from 5 areas	

[a] lightweight expanded clay aggregate

[b] ^{222}Rn emanation rate measured for 106 samples from 10 cities; ^{226}Ra concentration and emanation ratio measured for 21 samples from seven cities.

Table 2. Diffusion characteristics of building materials.

Material	Bulk diff. coef. (k_e^*) (10^{-8} m^2 s^{-1})	Interstitial diff. coef. (k_e) (10^{-8} m^2 s^{-1})	Diffusion length (ℓ) (m)	Porosity (ε)	Comments	Ref.
heavy concrete			0.13			(22)
lightweight concrete			0.26			
red brick			0.15			
concrete	~0.2		~0.1		assuming ≈0.1	(48)
alum-shale lightweight concrete		1.16	0.074	0.265		(24)
ordinary concrete		0.34	0.04	0.45		
concrete		3.6	0.13			(25)
brick		4.7	0.15			
Leca[a]		8.4	0.20			
concrete		3.3	0.126	0.068		(72)
concrete		6.0	0.169	0.32		

[a] lightweight expanded clay aggregate

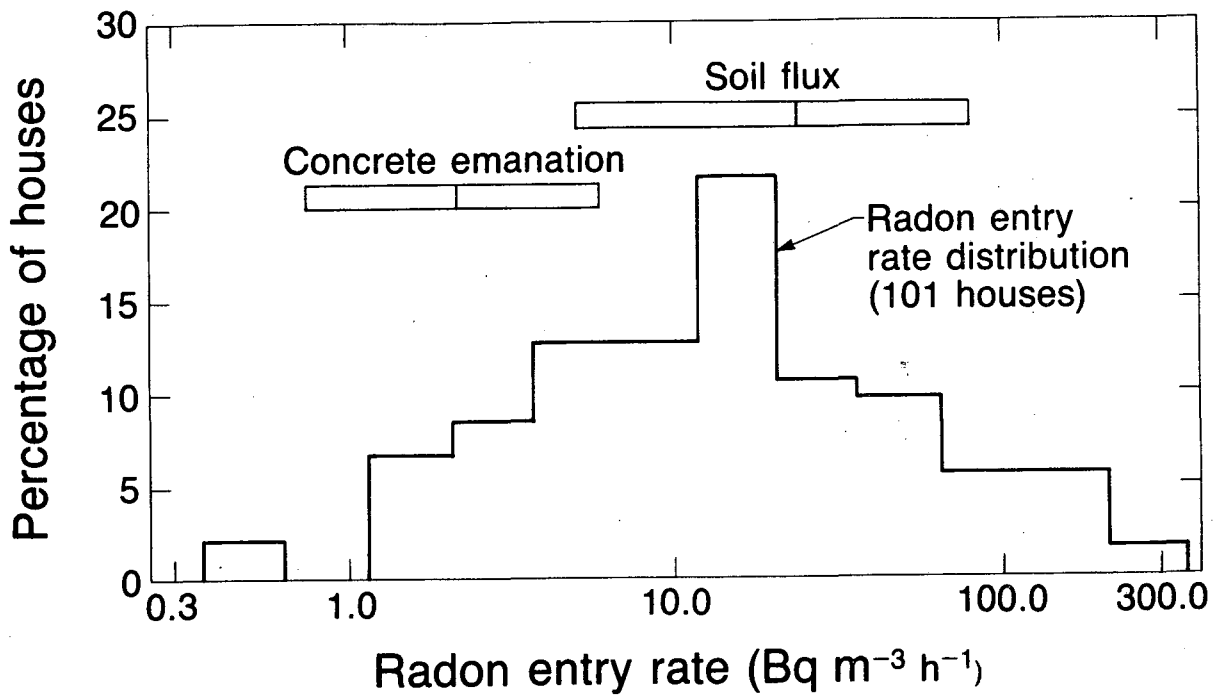

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Figure 1. Histogram of radon entry rates based on a grab-sample measurement of radon concentration and a tracer gas decay measurement of air-exchange rate[2]. The bar labelled "concrete emanation" indicates the range of contributions of a sample of 106 U.S. concretes assuming half the radon emanating from a 0.2-m floor enters a single-story house having a 2.4-m ceiling height[16]. The bar labelled "soil flux" indicates the range of mean contributions of 971 measurements at 62 sites of flux from uncovered soil (excluding measurements over lava), assuming all the radon that would emanate from uncovered soil enters the house, again with a 2.4-m ceiling height[44]. For each bar the center line indicates the mean value.

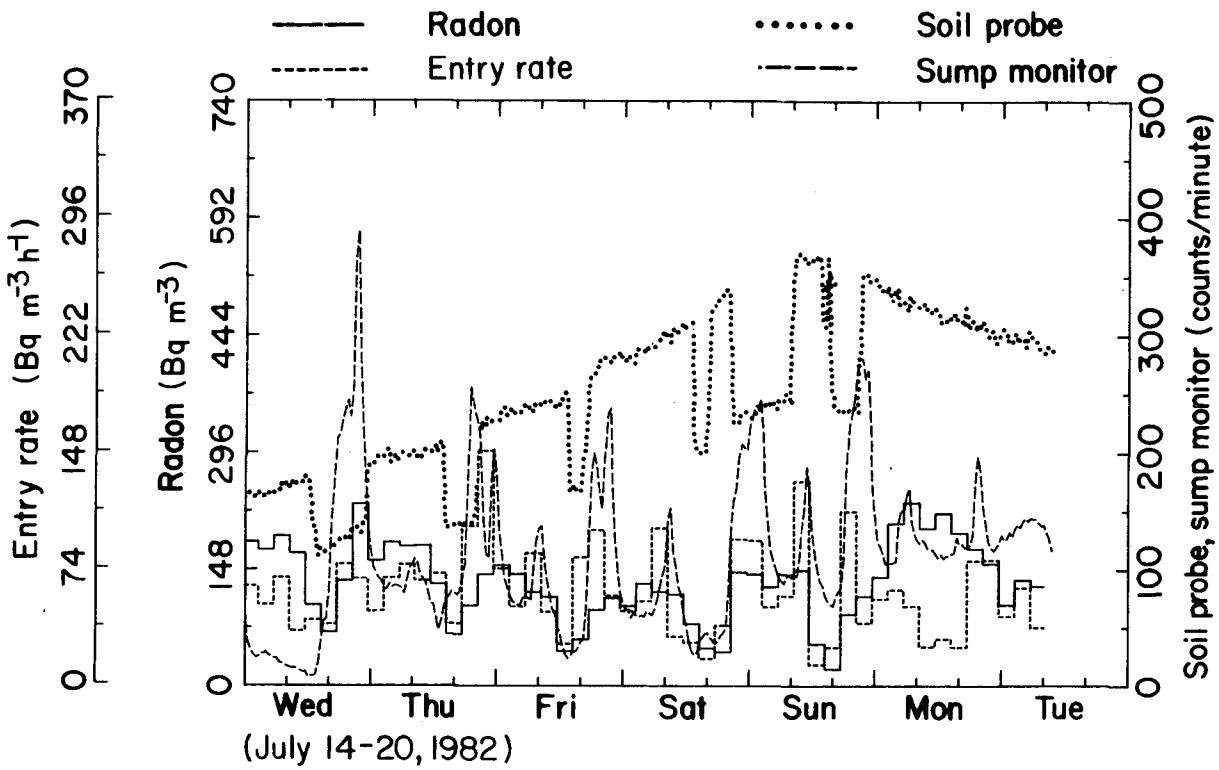

Figure 2. Radon measurements and source-related parameters for a week-long period in a single-family house near Chicago. During this period the water level in the basement sump was maintained above the entrance from the drain-tile system that extended around the house perimeter. The soil probe monitored alpha activity in the soil air adjacent to the basement, about 0.5 m below the soil surface. The sump monitor sampled air at a hole in the sump cover. For each device, the steady state response to ^{222}Rn in room air is 1 count min^{-1} = 17 Bq m^{-3} (38).

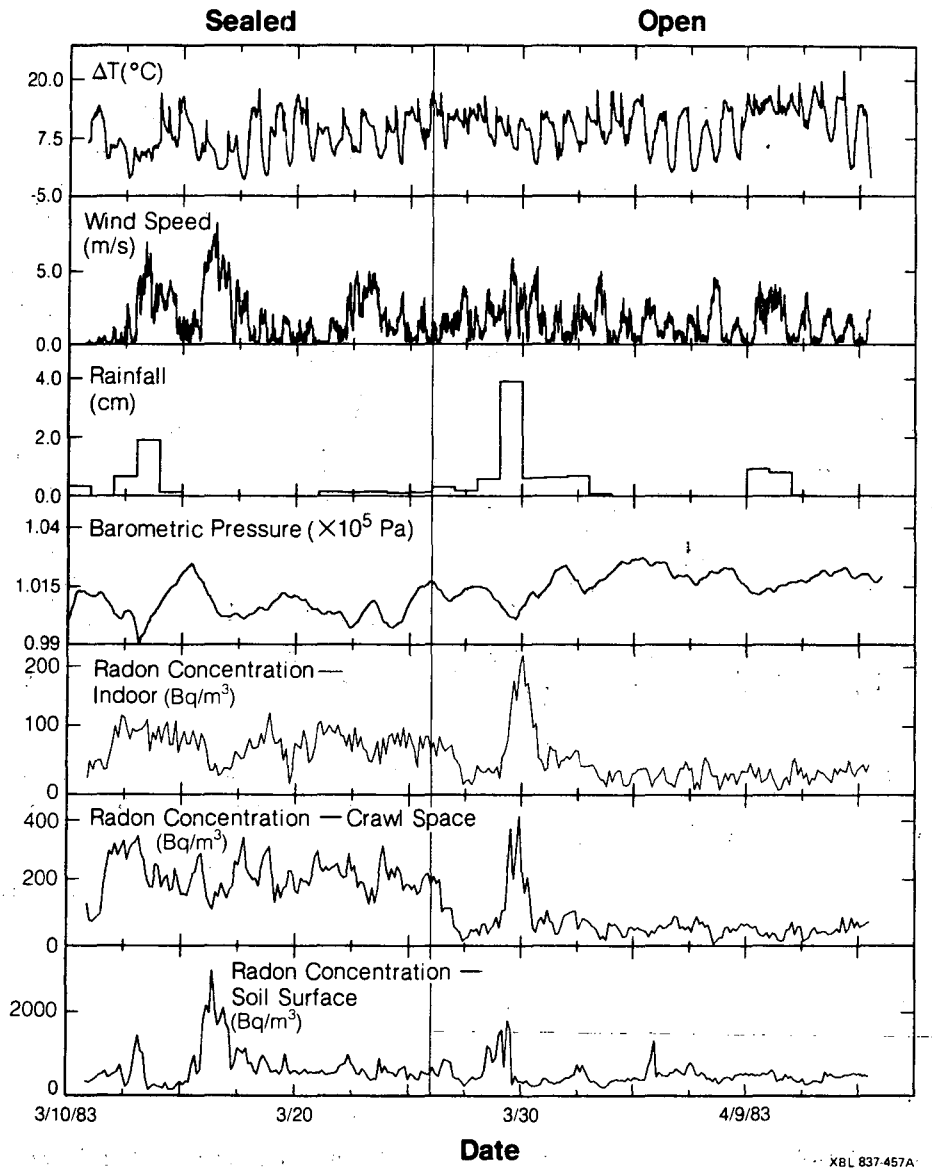

Figure 3. Radon concentrations and meteorological parameters measured over a five-week period in a house in Portland, Oregon. The temperature difference is positive when the indoor temperature is greater than that outdoors. Wind speed was measured on site at 9 m. Rainfall and barometric pressure data were obtained from the National Weather Service. The radon concentration at the soil surface was measured by sampling air at 15×10^{-6} m^3s^{-1} from a five-sided aluminum box whose open side covered 0.95 m^2 of soil adjacent to the house. During the first two weeks of sampling the crawl space vents were sealed with polyethylene sheets and tape; during the last three weeks they were open. The soil in the crawl space was covered by 0.15-mm (6 mil) black polyethylene sheet[37].

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