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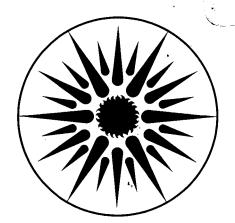
UNDERSTANDING THE ORIGIN OF RADON INDOORS -- BUILDING A PREDICTIVE CAPABILITY

R.G. Sextro

December 1985

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UNDERSTANDING THE ORIGIN OF RADON INDOORS--BUILDING A PREDICTIVE CAPABILITY

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December 1985

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Abstract--Indoor radon concentrations one to two orders of magnitude higher than the U.S. average of ~60 Bq m⁻³ (~1.5 pCi L⁻¹) are not uncommon, and concentrations greater than 4000 Bq m⁻³ have been observed in houses in areas with no known artificially-enhanced radon sources. In general, source categories for indoor radon are well known: soil, domestic water, building materials, outdoor air, and natural gas. Soil is thought to be a major source of indoor radon, either through molecular diffusion (usually a minor component) or convective flow of soil gas. While soil gas flow into residences has been demonstrated, no detailed understanding of the important factors affecting the source strength of radon from soil has yet emerged. Preliminary work in this area has identified a number of likely issues, including the concentration of radium in the soil, the emanating fraction, soil type, soil moisture content, and other factors that would influence soil permeability and soil gas transport. Because a significant number of dwellings are expected to have indoor radon concentrations above guideline levels, a predictive capability is needed that would help identify geographical areas having the potential for high indoor concentrations. This paper reviews the preliminary work that has been done to identify important soil and building characteristics that influence the migration of radon and outlines the areas of further research necessary for development of a predictive method.

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Key word index: convective air flow, diffusion, indoor air quality, lognormal distribution, radon, residential buildings, soil characteristics, source strength.

INTRODUCTION

Radon-222, an inert gas, is part of the ²³⁸U decay series. Its immediate parent, ²²⁶Ra, is a ubiquitous constituent of all crustal material, and is found in soils and in building materials having rock- and soil-based components. The radon gas formed by the alpha decay of radium can be transported through and from these materials by molecular diffusion or convective (bulk) flow and into the interior of houses. In the following discussion, this paper refers to ²²²Rn. Radon-220 may also be present in indoor air, but in general its concentrations are limited by its short (55 s) half-life.

Radon is also radioactive and its alpha decay forms a sequence of radionuclides, commonly termed radon progeny. The radium and radon decay chain is shown in Fig. 1. These radon decay products are chemically active and can attach to surfaces, such as room walls, airborne particles, or upon inhalation, lung tissue. This latter process is primarily responsible for the health effects associated with radon, where the alpha decays of the progeny damage nearby lung tissue and result in an increased risk of lung cancer (UNSCEAR 1982).

The distribution of indoor radon concentrations in homes across the U.S., as for many other indoor pollutants, is not well characterized. However, a recent study (Nero et al., 1984) systematically assessed the results of various investigations of indoor radon concentrations in locations across the country and compiled an aggregate data set that explicitly accounts for differences in selection and measurement procedures. The resulting distribution, shown in Fig. 2, appears to be lognormal, with a geometric mean (GM) of 35 Bq m⁻³, a GSD of 2.84, and an arithmetic mean (AM) of 61 Bq m⁻³. The lifetime lung cancer risk associated with this average concentration is approximately 0.4% (NCRP, 1984a), which is one to two orders of magnitude larger than that usually associated with environmental risks.

The National Council on Radiation Protection and Measurements (NCRP) has proposed an 'action' guideline equivalent to approximately 300 Bq m⁻³ (NCRP, 1984b). One to two percent of the houses in the distribution shown in Fig. 2 have average indoor radon concentrations greater than or equal to this guideline. If this distribution is representative of radon concentrations in the U.S. housing stock, 1 to 2 million homes could equal or exceed this action level. Exposures to high concentrations of radon pose a potentially serious public health problem, and efforts to reduce public exposures may depend upon identifying those factors that significantly influence indoor concentrations.

The focus of this paper, after discussing briefly the sources of indoor radon, is to review preliminary efforts to develop a predictive capability for identifying areas with the potential of having high indoor radon concentrations.

SOURCES OF INDOOR RADON

There are five potential sources of indoor radon: the soil surrounding the building shell, building materials, natural gas, potable water, and outdoor air. These are illustrated schematically in Fig. 3 for two of the three basic types of house substructure (the third basic substructure, slab-on-grade, can have cracks and penetrations similar to those found in basement floors). As shown in Fig. 3, radon can enter the building shell through cracks and other openings in the building substructure, which may be due to the settling or aging of the structure or due to the design and construction practices used. Penetrations through basement walls or floor slabs for utility services, joints between the basement walls and floor, or an untrapped weeping-tile and sump system are all examples of openings through which radon may pass.

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Earth-based building materials also contain trace quantities of radium, and concrete and gypsum board are sources of radon emanation, although the source strength from these materials is usually quite small. Groundwater and natural gas accumulate radon produced by radium in the subsurface rock formations which surround the fluid reservoir or through which the gas or water flow. Use of these fluids indoors will release the incorporated radon into the indoor air. Finally, infiltrating outdoor air will carry with it radon that is present in the ambient air.

The range of expected radon source strengths from each of these sources can be compared to the frequency distribution of radon entry rates that have been observed in single-family residences, as illustrated in Fig. 4. The steady-state radon entry rate is the product of the indoor radon concentration, C_i , and the air exchange rate, λ_V . The solid line in Fig. 4 summarizes time-averaged data from 73 homes in several cities in which C_i and λ_V were measured simultaneously (Nazaroff and Nero, 1984). The dashed line in the figure is the radon entry rate distribution derived from the observed distribution for indoor radon concentrations, shown in Fig. 2, and the distribution of measured air exchange rates from 578 houses, based on an analysis in Nazaroff et al. (1985b). Since the distributions for C_i and λ_V can be approximated by a lognormal distribution, the product distribution is lognormal as well.

As can be seen, with the exception of the unattenuated soil flux density, the other sources contribute only to the lower end of the entry rate distribution. The equivalent radon entry rate for natural gas use is quite small (Johnson et al., 1973) and is not illustrated in Fig. 4. Both building materials and infiltrating outdoor air make small contributions to indoor concentrations. In the case of high-rise buildings where indoor radon concentrations are usually small, radon from the soil may not be significant, hence outdoor air and building materials may be the major sources of radon (see for example

Abu-Jarad and Fremlin, 1982). The importance of water as a potential source of radon varies since the average concentration of radon dissolved in water depends upon the source of the water. In some areas of the country, high concentrations of radon dissolved in groundwater have been reported, and the resulting contributions to indoor radon levels are substantial (Hess et al., 1983). Since the water-to-air transfer factor is ~10⁻⁴, radon concentrations in water must be on the order of 100,000 Bq m⁻³ to contribute radon to indoor air equal to that contributed by outdoor air. For public water supplies, derived either from surface water or from groundwater sources, typical radon concentrations are low, averaging 1 x 10³ and 1 x 10⁴ Bq m⁻³, respectively. For small public systems using groundwater - serving less than 1000 people - or for private groundwater sources, radon concentrations in water vary widely. In some of these water supplies, the levels of radon in water can be significant, and will contribute measurably to indoor air concentrations (Nazaroff, et al., 1985b). These situations appear to be very isolated, and are highly dependent upon local geological conditions.

The diffusive radon flux density from soil is, potentially, an important source of indoor radon. The range of unattenuated soil flux densities shown in Fig. 4 is based on measurements for a variety of soils (from which the very low flux densities for lava have been excluded) (Wilkening et al., 1972). This estimate assumes that the area of soil is determined by the area of the house, that the building shell simply accumulates the. radon, and the resulting indoor concentration is determined by the ventilation rate of the shell. This estimate is also based on the exhalation of radon from uncovered soil, hence any covering, such as a concrete slab floor will significantly attenuate the flux density (Colle et al., 1981). As shown in Fig. 4, however, this source does not account for the radon entry rates observed in the upper end of the frequency distribution of source strengths. On the other hand, it appears that convective flow of radon-bearing soil gas into residences can account for high radon source strengths. In this case, the building shell is not simply a passive accumulator of radon diffusing from the soil, but has an active role in drawing radon from the soil into the building interior. This has been discussed by a number of authors (Akerblom et al., 1984; DSMA, 1983; Scott and Findlay, 1983; Nero and Nazaroff, 1984; Nazaroff et al., 1985a; Nazaroff et al., 1985c), and is the basis for the discussion here of a predictive approach.

BUILDING A PREDICTIVE CAPABILITY

As discussed previously, the number of houses that might be expected to have indoor radon concentrations above the NCRP action guideline is quite large in absolute terms - between one and two million structures - but at one to two percent of the total

housing stock, is fairly diffuse. It appears, based on the observation that the GM's of the 22 individual data sets included in the compilation discussed earlier vary lognormally, with a GSD of 2.0, that there is also some geographic variability in the observed mean concentrations (Nero et al., 1984). While individual homes with high indoor Rn concentrations may be found either in random surveys with a broad geographic coverage or in surveys of areas already known to have homes with significant indoor levels, it is clear that such surveys are likely to miss a large number of homes and a more systematic approach is needed.

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If there are factors common to houses with high indoor levels, these might be used to identify other houses with high indoor radon levels. Since convective flow from soil appears to be the principal source of radon in houses, at least for those homes with high concentrations, soil-related factors should be useful predictors. It is also apparent that as a screening method, a geographically-based approach would be preferable, since it would serve to locate areas where the potential for high indoor radon concentrations exists, and where further detailed field survey work would be warranted and more cost-effective.

A Discussion of the Major Components:

In general, there are two elements that determine radon entry rates: radon availability and the subsequent transport of radon into buildings. Radon migration in soils has been reviewed by Tanner (1964 and 1980), and radon entry into buildings from various sources has been discussed by Nero and Nazaroff (1984). Understanding the factors that influence these elements, such as radium concentration in the soil and the soil permeability, will provide a basis for developing a predictive capability.

Radon in the Soil -- Radium concentrations in samples of ordinary, non-uraniferous soil from 33 states in the U.S. range from 8.5 to 160 Bq kg⁻¹, with an average of 41 Bq kg⁻¹ (Myrick et al., 1983). In some cases, the radium is incorporated within the crystal-line material, while in others, it may be deposited on the surface of soil particles. The alpha decay of radium imparts momentum to the recoiling radon nucleus, which can travel 60 μ m in air, about 75 nm in water and approximately 35 nm in crystalline material. A fraction of the recoiling radon nuclei will travel across air spaces between soil grains and become embedded in adjacent grains. Typically, the molecular diffusion of radon out of crystalline matter is much slower than the radon half-life, thus these embedded recoils contribute little to the availability of radon in the soil gas. Since the recoil range in water is about three orders of magnitude smaller than that for air, the recoiling nuclei can be slowed or stopped by the presence of water in the soil pore spaces. Radon that stops in the pore spaces is available for transport, although, too much water

in the soil will reduce both the diffusive flux and the flow of soil gas in the soil.

The ratio of the amount of radon released from the soil materials (i.e. available for transport) to the total amount of radon produced by the radioactive decay of radium in the soil is the emanating fraction, r. This ratio has been observed to range from 10 to 55 percent (Nero and Nazaroff, 1984), depending upon the soil type, moisture content and temperature (Stranden et al., 1984). The relationship between these various parameters can be seen from the equation for the maximum soil gas radon concentration, C_{∞} , in undisturbed soil, which is given by

$$C_{\infty} = \frac{\rho_{\rm s} \ r \ A_{\rm Ra}}{\epsilon}. \tag{1}$$

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This equation assumes that radioactive decay is the only removal process. The parameter ρ_8 is the bulk soil density (which is typically on the order of 10^3 kg m⁻³), A_{Ra} is the radium activity in the soil, and ϵ is the soil porosity.

Radon Transport -- While the unattenuated diffusive radon flux from the soil does not provide a sufficient radon entry rate to account for radon entry rates in the upper portion of the frequency distribution in Fig. 4, the observed concentration of radon in soil gas is between (0.7 and 22) x 10⁴, with a typical concentration of 2 x 10⁴ Bq m⁻³ (Nero and Nazaroff, 1984). This source can supply radon in sufficient quantity to account for high radon concentrations in indoor air provided that the gas is transported into the building shell.

There are two basic mechanisms for transport of radon into buildings, diffusion and convective (bulk) flow. Diffusion, described by Fick's law, results in a flux density, J, that is proportional to the gradient of the concentration, C:

$$\vec{J} = -D\vec{\nabla}C \tag{2}$$

where D is the diffusion coefficient. Diffusion of radon into buildings can occur via two general routes, directly from uncovered soil and through building materials (such as through a cement slab floor lying directly on top of soil materials). As noted earlier in the discussion of Fig. 4, the diffusive flux density from uncovered soil cannot account for the high radon source strengths that have been observed. This is especially true when the factor of 25 to 50 reduction in the diffusive flux density by the presence of an unbroken concrete slab is taken into account (Colle et al., 1981).

Pressure-driven convective flow can be characterized by Darcy's law, which relates the flow per unit cross-sectional area, V, to the gradient of the pressure, P:

$$\vec{\nabla} = -\frac{K}{\mu} \vec{\nabla} P \tag{3}$$

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where K is the permeability of the medium and μ is the dynamic viscosity (which for air is 18×10^{-6} nt s m⁻²). The route of entry for such flows, such as openings in the basement walls or floors, were illustrated schematically in Fig. 3.

Pressure differences can occur in several ways. Meteorological conditions may produce changes in atmospheric pressure. Pressure gradients across a building shell (which is the interface between the soil and the gas contained within it and the building interior) can develop due to wind loading on the building exterior, temperature differentials between the interior and exterior, and any building ventilation system that might pressurize or depressurize the building shell with respect to atmospheric pressure.

The first of these, changes in barometric pressure, can be quite large in magnitude, several percent of atmospheric pressure or several thousand pascals. Changes in barometric pressure have been shown to influence radon flux from the surface of uncovered soil (Clements and Wilkening, 1974; Schery et al., 1984). It is not clear, however, whether barometric pressure changes lead to persistent pressure differentials across the building shell. To date, the experimental evidence is mixed. Based on experiments conducted in a single-storey house with a basement in which environmental parameters and indoor radon concentrations were extensively monitored (Nazaroff et al., 1985a), no relationship between barometric pressure changes and indoor radon concentrations was observed. On the other hand, the influence of the rate of change of the barometric pressure on the observed indoor radon concentration has been reported (Hernandez et al., 1984).

The effect of wind on the building shell and the stack effect, caused by the indooroutdoor temperature difference are, in general, responsible for infiltration into the building shell. These two effects are usually parameterized in infiltration models (Grimsrud et al., 1982) as

$$Q_{wind} = A_o f_w V (4)$$

and

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where A_0 is the effective leakage area of the building shell, f_w is the wind parameter, which accounts for local and terrain shielding effects, V is the wind velocity, fs is the stack parameter, accounting for the building height and the distribution of the leakage area, and ΔT is the indoor-outdoor temperature difference. These two effects can lead to pressure differentials of the order of a few pascals at the bottom of the building shell. While much smaller than the changes in barometric pressure, these wind and thermal stack effects generally produce a persistent ΔP across the shell, one that appears to be sufficient to provide a radon entry rate comparable to the higher entry rates observed, as can be illustrated by the following example. Assuming an entry rate of 100 Bq m⁻³ h⁻¹. which is twice the average radon entry rate based on data shown in Fig. 4, and a typical radon concentration in soil gas of $\sim 2 \times 10^4$ Bq m⁻³, the resulting soil gas entry rate is 0.005 h⁻¹. This implies that about 0.6% of the infiltration flow enters the house through the soil. Volumetrically, this flow is around 3 x 10⁻⁴ m³ s⁻¹, which is the equivalent to the flow through an opening 0.8 cm in diameter and 10 cm long and driven by a pressure differential of 5 pascals. Additional evidence for the influence of the stack effect is the observation of seasonal differences in indoor radon concentration (Turk, 1985). These differences are too large to be accounted for only by increased ventilation rates in the summer months compared with the minimum ventilation rates in the winter. The indoor-outdoor temperature differences are small or non-existent in summer, while in winter, the differences are at a maximum.

Coupling between the building shell and the soil induces a pressure differential in the soil near the house, and provides for the bulk flow of soil gas. There is experimental evidence for the existence of a negative pressure field in the soil surrounding a house established by the depressurization of the building basement (Nazaroff et al., 1985c). Comparison between the observed pressure field and a model of the spatial dependence of the pressure in the soil suggested that anisotropies in the soil permeability significantly influenced the pressure field. This is also suggested by the observation that at some sampling locations close to the house, no steady depressurization reading was made, while at some points up to 5 m away from the basement wall, a measurable depressurization effect was detected. Such anisotropies should not be too surprising since soil is often not a homogeneous mixture.

Tracer gas experiments, conducted at the same time, showed a substantial effect of the pressure gradient on movement of the tracer gas through the soil; in a case where the basement was depressurized by 30 Pa, effective transport velocities exceeded 1 m h⁻¹.

Bulk flow of soil gas through the soil has important implications. Since the diffusion length of ²²²Rn in soil is about 1 m, diffusive transport only through the soil limits the volume of soil from which radon can enter the building shell. Bulk flow of soil gas, on the other hand, can extend this source volume significantly, depending of course, upon the permeability and other characteristics of the soil.

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The air permeability of soil ranges over twelve orders of magnitude, from 10^{-7} m² for coarse gravel to 10^{-19} m² for glacial till and marine clay (Freeze and Cherry, 1979). At the low end of this range, the permeabilities are of the same order as that of concrete, ca. 10^{-15} m² (Nero and Nazaroff, 1984), in which case pressure-driven flows through these soils are quite small and contribute little to indoor radon concentrations. The air permeability of soil is influenced by both the porosity and the moisture content of the soil, and the distribution of pore sizes is an important determinant of permeability. In most well-drained soils, water is present in the small pores while the larger open volumes are air filled. Air flow occurs principally through these large pores, and only when water begins to fill these larger voids does soil moisture content significantly affect air permeability.

Assembling the Major Elements

Thus far we have discussed the important characteristics of radon accumulation in and transport through soil and into houses. An elementary approach to developing a predictive capability would be to combine information bearing on radon production with data regarding gas transport -- both diffusive and convective flow -- through the soil. Information on building structure and operation would help characterize the driving force(s) for bringing radon into the building interior. The coupling of the building shell with the surrounding soil will determine how much of the available radon is then brought into the indoor air.

As noted earlier, given the nature of the problem, i.e. how to go about locating one to two million homes with high indoor radon levels in a systematic fashion, a geographically-based method would be preferred. This approach would help locate areas with the potential for high indoor radon concentrations so that more detailed follow-up surveys could be conducted. However, such a method will depend upon the availability of relevant geological information at a sufficient geographical scale.

Data on radium concentrations in the surficial soil are available for much of the United States, compiled by the National Airborne Radiometric Reconnaissance program. These aerial measurements of gamma radiation from radionuclides in the earth's surface, including ²¹⁴Bi, were part of a program to survey the uranium resource potential in the U.S. A preliminary analysis of these data has been conducted for several western states

and variations in radium content were seen at a regional scale (Moed et al., 1984). In addition, field measurements in two specific areas confirmed the spatial variability seen in the detailed aerial data. Additional investigations of the data for the remainder of the U.S. are currently in progress, including an approach to utilizing the data at a geographical scale of approximately a kilometer.

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As discussed earlier, two important pieces of data on soils are air permeability and moisture content. In general, air permeability data for soils do not exist at a geographical scale, although data on soils do cover a number of other parameters, including general soil classifications and, in many cases, water permeability. One source of soil information is the extensive soil data collected by the Soil Conservation Service (SCS) of the U.S. Department of Agriculture. Soils are typically characterized by the distribution of particle sizes, from which porosities can be obtained (Day, 1965). The particle size data may also be an indicator for air permeability, and additional work is needed to define the ranges of porosities and/or permeabilities associated with each particle size class. A second approach for obtaining soil air permeabilities would be to utilize existing soil water permeability information. If the relationship between water permeability and air permeability for most soils can be developed, wider geographic estimates of soil air permeabilities may then be available. Finally, in as much as air permeabilities and radon diffusion coefficients are influenced by the water content of the soil, estimates of soil moisture content are needed. Again, SCS data, used in conjunction with climatic data for a specific region, may be useful.

Assembling a predictive capability will require additional research on both the individual physical components and their mathematical relationship. A Radon Index Number (RIN) has been proposed (DSMA, 1983) which relates some of the soil parameters just discussed to an index of expected indoor radon concentrations,

$$RIN = \log A_{Ra} + n \log K + \sum B_i$$
 (6)

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where A_{Ra} is the radium concentration in the soil, K is the permeability, n is an index less than 1, and the parameters B_i incorporate meteorological effects (wind speed, temperatures, etc.), housing characteristics (substructure type, ventilation rate, etc.) and other variables. The form of this equation as suggested here is only a first step in evaluating the factors influencing indoor radon concentrations. At present few data exist for testing this approach. Eaton and Scott (1984) have evaluated a slightly different version of the RIN equation and have seen some correlation between RIN and observed

indoor radon concentrations, based on a very limited data set. However, there is considerable scatter in the data.

Among the questions such an approach raises are which of the B_i terms are significant, how should they be specified, and at what geographic scale is the RIN appropriate. Resolution of these questions is likely to be based on a semi-empirical approach, as additional soil data are gathered and correlated with observed indoor radon concentrations. Building factors that might be utilized in the B_i terms are likely to include the substructure designation, possibly by incorporating a flow resistance term. While the data aren't yet sufficient to designate which substructure type is more resistant to pressure-driven flow of soil gas, houses with ventilated crawl spaces would appear to be least well-coupled to the soil. Because both wind and indoor-outdoor temperatures affect infiltration flows into dwellings, climatic variables such as heating degree days and average wind speeds might be included as B_i terms in the RIN equation.

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The geographic scale for application of the RIN approach will depend, in part, upon the geographic scale of the data. In some cases, major variations in soil characteristics can be seen at the sub-county level as shown by the SCS soil survey reports, which are done on a county basis. Local meteorological information can also vary within counties, depending upon local terrain effects, although whether the RIN equation will be sensitive to local variations isn't clear. However, a predictive approach is not intended to yield predictions on a house-by-house basis; rather its development and fine-tuning will be done based on aggregated samples of sufficient size to be statistically valid.

CONCLUSIONS

Soil is the major source of radon in houses with higher-than-average indoor concentrations. Identifying the estimated one to two million homes with indoor concentrations exceeding the NCRP guideline of ~300 Bq m⁻³ is not an easy task, however. Even in areas where indoor levels in some houses have been observed to exceed these guidelines, most of the homes surveyed in these areas have indoor radon concentrations close to the average of 60 Bq m⁻³ (DSMA, 1983). A geographically-based predictive approach would, ideally, locate areas with the potential for having high indoor radon levels; such areas would then be investigated in more detail. The efficacy of such an approach will depend upon the availability and quality of data for the characterization of soils in terms of both radon availability and radon transport.

This paper has discussed some of the factors that appear to be important constituents of a predictive capability. Although exploration of existing data bases with wide

geographic coverage has begun, further research is necessary both to investigate the validity of the approach and to define a suitable mathematical relationship among the important parameters.

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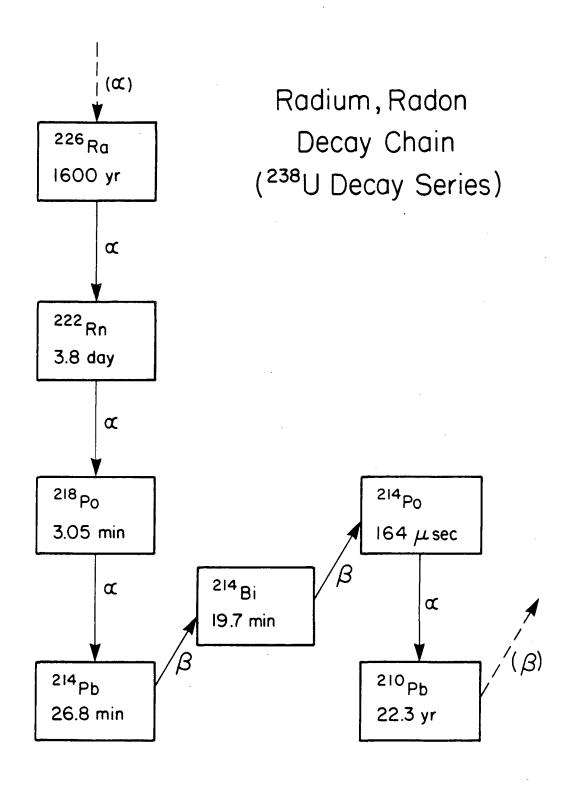
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XBL 853-10395

Figure 1: Radium and radon decay chain. The nuclides ²¹⁸Po, ²¹⁴Pb, and ²¹⁴Bi are of primary radiological concern due to inhalation and subsequent alpha decay.

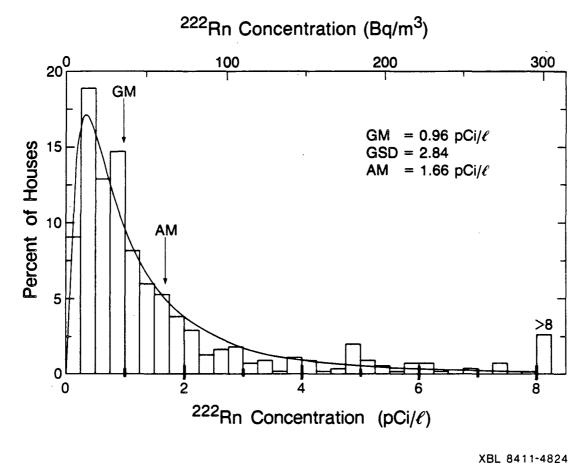
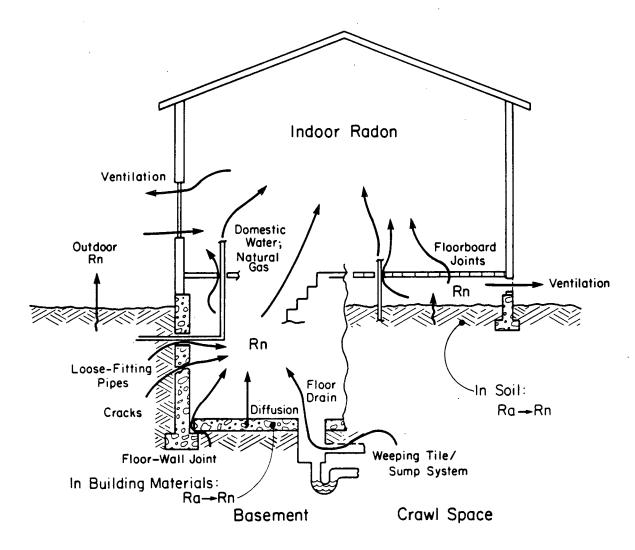


Figure 2: Probability distribution of ²²²Rn concentrations in 552 U.S. houses. The smooth curve is a lognormal functional form with a GM of 35.5 Bq m⁻³ and a GSD of 2.84 (figure from Nero *et al.*, 1984).



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Figure 3: Schematic diagram of radon entry pathways for typical residential building substructures.

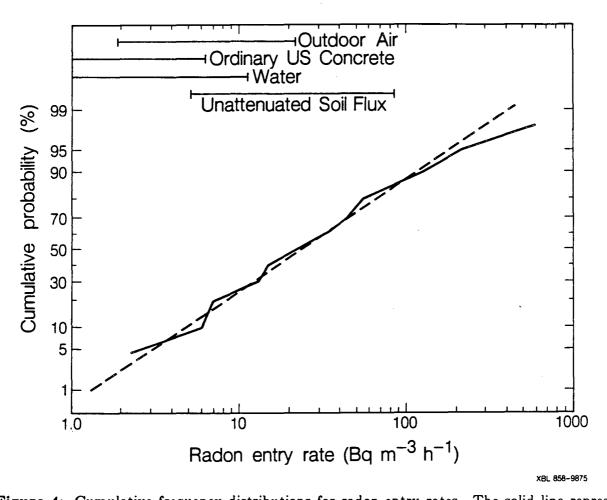


Figure 4: Cumulative frequency distributions for radon entry rates. The solid line represents the distribution for 73 simultaneous measurements of indoor radon concentration and building ventilation rate, while the dashed line is the distribution derived from separate data on radon concentrations and air exchange rates (see text). The source strengths shown at the top the figure are estimated based on an assumed single-storey house with 100 m² floor area, a 2.4 m ceiling height, and a 0.2 m thick concrete slab floor. Assumptions regarding the contributions of individual sources are as follows. The average outdoor air concentration, 9 Bq m⁻³, is taken from Gesell (1983), with the range in ventilation rates represented by the 5 and 95 percent probability intercepts on the distribution of λ_V from Nazaroff et al. (1985b). The range of concrete emanation rates is from measurements reported by Ingersoll (1983), with the further assumption that half of the radon released from the concrete goes indoors. The range for radon concentrations in water, 40 to 1 x 10⁵ Bq m⁻³, is based on the 5 and 95 percent probability intercepts from the population-weighted aggregate distribution of radon concentrations in water and the average water-to-air transfer factor of 1.1 x 10⁻⁴, from Nazaroff et al. (1985b). The unattenuated soil flux is based on data from Wilkening et al. (1972).

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