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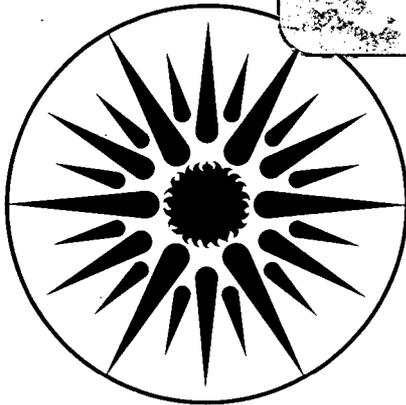
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CHARACTERIZING THE SOURCES, RANGE, AND ENVIRONMENTAL INFLUENCES OF RADON 222 AND ITS DECAY PRODUCTS

A.V. Nero, R.G. Sextro, S.M. Doyle, B.A. Moed, W.W. Nazaroff, K.L. Revzan and M.B. Schwehr

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ABSTRACT

Recent results from our group directly assist efforts to identify and control excessive concentrations of radon 222 and its decay products in residential environments. We have demonstrated directly the importance of pressure-induced flow of soil gas for transport of radon from the ground into houses. Analysis of available information from measurements of concentrations in U.S. homes has resulted in a quantitative appreciation of the distribution of indoor levels, including the degree of dependence on geographic location. Experiments on the effectiveness of air cleaning devices for removal of particles and radon decay products indicate the potential and limitations of this approach to control.

## INTRODUCTION

A variety of measurements from the United States suggests that radon 222 concentrations in residences average about  $40 \text{ Bq/m}^3$  ( $1.1 \text{ pCi/l}$ ). Estimation of the incidence of lung cancer due to the decay products associated with this much radon 222 yields thousands of cases per year among the U.S. population. Measurements to date also indicate that indoor levels in the range of  $400\text{-}4000 \text{ Bq/m}^3$  occur with startling frequency. And, whereas the risk associated with even  $40 \text{ Bq/m}^3$  is very large compared with many environmental insults of concern, living for prolonged periods at the higher concentrations observed leads to estimated individual lifetime risks of lung cancer that exceed 1% and that sometimes approximate the risk associated with cigarette smoking. Considering these levels and the associated risks, we - among others - have devoted substantial efforts to characterizing the factors affecting indoor concentrations of radon 222 and its decay products. We here report recent results from experimental studies on the mechanisms for entry of radon 222 into homes, from our analysis of most of the concentration measurements reported for U.S. homes, and from experimental studies of the behavior of radon-222 decay products.

The initial observation of significant concentrations of radon (taken to be radon 222 unless otherwise stated) in ordinary U.S. homes (ref.1) occurred at about the same time that programs to increase the efficiency of energy use were hitting their stride. Reducing ventilation rates in buildings can be a cost-effective, and hence attractive, component of such programs. However, given the expectation in first order that indoor pollutant concentrations equal the ratio of the entry rate per unit volume to the ventilation rate, energy-conservation programs were thought to have the potential to exacerbate the indoor air quality problem significantly.

The expectation that, with supply of differing amounts of mechanical ventilation in a given house, the indoor radon concentration varied as the inverse of the ventilation rate, was observed directly in an energy-efficient house (ref. 2). However, paired measurements of radon concentration and ventilation rate in a number of housing samples showed no apparent correlation between these two parameters (ref. 3). And, for the aggregate sample, radon concentrations showed a significantly larger variability than the ventilation rate, suggesting that the source strength was the dominant determinant of the wide range of concentrations observed in U.S. housing.

## SOURCES OF INDOOR RADON

Indications of the importance of soil as the source of radon in U.S. homes had arisen in early studies, when it was found, for example, that measurements of radon emanating from structural materials could not account for observed indoor concentrations, based on estimates of the air exchange rate (ref. 1). A clearer picture emerged from the distribution of entry rates inferred from the direct measurements of radon concentration and ventilation rate alluded to above (ref. 3). Figure 1 shows such entry-rate distributions from various countries, as well as indicating the potential contribution of various sources. As indicated there, a median (or geometric mean) entry rate for U.S. single-family homes is in the vicinity of  $20 \text{ Bq m}^{-3} \text{ h}^{-1}$  ( $0.5 \text{ pCi l}^{-1} \text{ h}^{-1}$ ). Based on emanation rate measurements

from concretes (see Fig. 1, caption), one might expect emissions from this source to account for a median of about  $2-3 \text{ Bq m}^{-3} \text{ h}^{-1}$  (assuming all houses are one-story with a concrete slab-on-grade), far below the total observed. On the other hand, the potential contribution from unattenuated soil flux, a median of  $25 \text{ Bq m}^{-3} \text{ h}^{-1}$  (based on ref. 4), corresponds well with the median indoor observations. However, houses have understructures that might be expected to impede substantially the ingress of radon, at least by diffusion. As a result, a major focus of research on radon entry has been to understand how the rate of radon entry could be approximately equal to the unimpeded flux from the ground (i.e., in the absence of the house) and, sometimes, even higher.

Recent work of our group has been devoted to direct characterization of mechanisms that cause entry of radon-bearing soil gas. A study of radon entry in a single-family house with a basement analyzed indoor concentration versus the ventilation rate, measured continuously over a period of months, and concluded that entry could usefully be represented by a sum of two components: one - the smaller - independent of ventilation rate, much as diffusion would be, and a larger term that is proportional to ventilation rate, as pressure-driven flow might be (ref. 5). Moreover, the observed pressure and soil parameters were consistent with the soil-gas infiltration rate that was implied by the measured concentrations and ventilation rates. These results, together with recent theoretical work (ref. 6) are helping to constitute a fundamental picture of the pressure and velocity fields in the soil surrounding homes with basements. Even more recently, we have directly observed, in two houses with basements, the coupling between depressurization of the building shell and a negative pressure field induced in the surrounding soil; we have also measured underground soil-gas movement by injecting  $\text{SF}_6$  tracer gas and monitoring concentrations in the soil and in the basements (ref. 7). These results provide a basis for optimization of control techniques, as well as for strategies to identify houses having excessive concentrations. They may also have significant implications for entry of other pollutants from the soil.

The small pressure differences that occur between the lower part of the house interior and the outdoors, and that appear to drive radon entry, are the same pressure differences that cause infiltration during seasons when the windows are closed. These pressure differences arise from two environmental factors: the thermal stack effect and wind loading on the building shell. The stack effect, established by the difference in temperature between indoors and outdoors, causes a convection pattern that exchanges indoor air with outdoor; the outdoor air is drawn from the vicinity of the understructure during the heating season. The second factor of importance is wind, which causes differences in pressure across the different walls, depending upon the wind direction. The pressure differences caused by temperature differences and winds are roughly comparable in size, averaging on the order of a few pascal (with higher values in relatively severe climates). These extremely small pressure differences account for air infiltration through openings in walls (and other components of the house), the dominant contributor to home air exchange during the heating season. These same pressure differences can, in principle, drive small flows of soil gas: soil gas contains enough radon that only 0.1% of infiltrating air would have to be drawn from the soil (ref. 30) to account for the observed rate of radon entry into homes. We note in passing that models are available to parameterize infiltration rates in terms of the temperatures differences and wind loads (ref. 21; ref. 9).

These results for structures with basements might also be expected to apply with some modification to slab-on-grade structures; however, direct measurements in such structures have not been performed. For houses with a crawl space, there is some isolation of the interior from the soil, but limited measurements of the transport efficiency of radon through crawl spaces have given the result that a substantial portion of the radon leaving uncovered soil manages to enter the interior, even if vents are open to permit natural ventilation of the space (ref. 10). In retrospect, this is not entirely surprising, since the stack effect will still tend to draw infiltrating air into the home from the crawl space. Another result of this study is that energy conservation efforts that focus on tightening the floor above a crawl space can significantly reduce infiltration rates, while reducing radon entry a corresponding amount, so that indoor radon concentrations are little affected.

Of the radon sources other than soil, domestic water drawn from underground sources is probably more important than building materials as a source of radon, particularly in certain parts of the U.S. Surface waters have such low radon concentrations that its use indoors cannot affect indoor concentrations. On the other hand, ground water accumulates radon generated within the earth's crust. As a result, very high radon concentrations have been found in certain potable water supplies, particularly those supplies - such as wells supplying individual households or small public systems - where the storage or hold-up time is too short to allow time for radon decay. As an example, concentrations exceeding  $3.7 \times 10^6$  Bq/m<sup>3</sup> (100,000 pCi/l) have been found in wells in Maine (ref. 11). With normal water use, if the radon from such (admittedly rare) water enters the air of a typical house, an indoor concentration of about 400 Bq/m<sup>3</sup> would result, which would be in the range of concern.

Past examinations of the overall potential contribution from water supplies have been little more sophisticated than the estimate just given, which corresponds to a ratio of radon in air to radon in water of  $10^{-4}$ , comparable to estimates and direct observations made by a number of authors (e.g., ref. 11). However, substantial data have recently become available on concentrations of radon in public water supplies drawn from ground water (ref. 12), indicating that the majority of such supplies have concentrations below  $4 \times 10^4$  Bq/m<sup>3</sup>, but that a very small percentage have concentrations exceeding  $4 \times 10^6$  Bq/m<sup>3</sup>. Moreover, data are available to assess the effect of radon release to indoor air in a more comprehensive way: a recent analysis by our group has combined information on water use rate, efficiency of radon release from domestic water used in various ways, house volumes, and ventilation rates, to yield a frequency distribution of air-to-water ratios that is approximately lognormal, with a geometric mean (GM) of  $0.65 \times 10^{-4}$  and a geometric standard deviation (GSD) of 2.88 (ref. 13). The importance of these developments is that they permit quantitative assessment of the contribution of water to indoor radon concentrations. Such an assessment (ref. 13) indicates that public groundwater supplies contribute an average of approximately 1 Bq/m<sup>3</sup> in the 30% of U.S. homes served by these supplies, a contribution equalling about 3% of the average indoor radon concentrations in U.S. homes (see below).

However, the very high water-borne concentrations that are sometimes found - particularly from private wells - will contribute much larger airborne concentrations in the homes affected. Using the few data that are

available for the approximately 18% of the population using private wells, we estimate the indoor radon concentration from water for this segment of the housing stock to average about 20 Bq/m<sup>3</sup>. About 10% of the houses served by private wells are estimated to have indoor concentrations from water of 40 Bq/m<sup>3</sup> or more, which translates into about 2% of the total U.S. housing stock. Although these estimates for private-well contributions cannot be regarded to be reliable, they suggest that the portion of the population using private wells may be experiencing significantly higher radon exposures than average, particularly in areas with high radon activity in water.

Overall, the approximate contribution of various sources to U.S. residences may be summarized as in Table 1, where the observed average indoor concentration (55 Bq/m<sup>3</sup>) is taken from the work described in the next section. As indicated in the Table, the dominant contributor to indoor concentrations in single-family houses is the soil, with water and building materials contributing only a few percent. In contrast, for large buildings, the main contributors appear to be building materials and outdoor air, the latter also contributing significantly to concentrations in single-family housing. However, for the portion of residences served by private wells in high activity areas, the contribution from water may be much larger than in ordinary circumstances.

TABLE 1

Approximate Contributions of Various Sources to  
Observed Average Indoor Radon Concentrations

	Single-Family Houses (Bq/m <sup>3</sup> )	High-Rise Apartments (Bq/m <sup>3</sup> )
Soil potential (based on flux measurements)	55	<40
Water (public supplies)	0.4*	0.4*
Building materials	2	4 <sup>+</sup>
Outdoor air	10	10
Observed indoor concentrations	55	(12)

\* Applies to 80% of population served by such supplies; contribution from water may average about 20 Bq/m<sup>3</sup> in homes using private wells, with even higher contributions in high-activity areas.

<sup>+</sup> The contribution to single-family houses is less than that estimated in the text because not all houses are slab-on-grade or one-story. A higher contribution to apartment air is suggested on the presumption that, on the average, high-rise apartments have a higher amount of radon-bearing building materials per unit volume than do single-family houses.

## FREQUENCY DISTRIBUTION OF INDOOR CONCENTRATIONS IN U.S. RESIDENCES

Despite a broad range of efforts to characterize indoor radon, and a significant number of studies that have included measurements in existing U.S. homes, no unequivocal estimates have been made of the concentrations to which the U.S. population is exposed. Studies have varied significantly in incentives, scientific objectives, selection of homes, and measurement procedures, and the results, not surprisingly, vary significantly, as may the conclusions that can be drawn from them. No useful quantitative appreciation of the actual distribution in U.S. homes has been available, although knowledge of this distribution would be extremely valuable in formulating a strategy for controlling excessive concentrations, as well as for making a reliable estimate of even the average population risk.

However, the data already available are quite substantial and deserve careful evaluation, if only as a basis for proper design of subsequent larger-scale, monitoring efforts. We have undertaken a systematic analysis of U.S. results, explicitly considering the differences between studies and using lognormal representations as a tool for aggregating the various data sets to yield a nominal distribution for the United States. The results are quite robust, i.e., they have little dependence on selection of data sets, on normalizations having to do with season of measurements, and on weighting of the data (ref. 14). Figure 2 shows the result of direct aggregation of 19 of the data sets that are available as individual data, totalling 552 houses. Because of the lack of proper normalization and weighting, no general conclusion may be drawn from this specific aggregation, aside from the substantial conformance to a lognormal representation, a result that has been observed in many individual studies.

The more complete analysis (ref. 14) utilized from 22 to 38 sets, corresponding to different areas (usually a state or urban area) of the United States, with the larger number including monitoring efforts that were initiated because of some prior knowledge of a potential for elevated concentrations. Characterizing the results of each set in terms of a lognormal function, the geometric mean radon concentrations from these sets range from 11 - 210 Bq/m<sup>3</sup>, with geometric standard deviations ranging from 1.3 to 4. As just noted, the results of aggregating these sets are quite robust, the main differentiation among different aggregations being that including the full 38 sets yields somewhat higher results than including only the 22 "unbiased" samples. The overall result, relying primarily on the 22-set aggregations and including a renormalization of data taken only during heating season, is a geometric mean of about 33 Bq/m<sup>3</sup> (0.9 pCi/l) and a geometric standard deviation of 2.8, implying an average concentration of 55 Bq/m<sup>3</sup> and 1 to 3% of houses exceeding 300 Bq/m<sup>3</sup>. This result can only be associated with the portion of the housing stock consisting of single-family houses, since 99% of the data are drawn from such houses. However, this is the dominant element in the U.S. housing stock, and the results of this analysis suggest that of the order of a million single-family houses have annual-average concentrations of 300 Bq/m<sup>3</sup> or more. This corresponds to exposures approaching the 2 WLM/yr remedial-action limit recently recommended by the National Council on Radiation Protection and Measurements (NCRP) (ref. 15), a level that is similar to exposures now received by uranium miners. Another interesting observation from this analysis is that the geometric means of the 22 sets are themselves lognormally distributed, with a geometric standard deviation of 2.0. This demonstrates the substantial variability in mean radon concentration from one area to another

and indicates the potential value of strategies that locate homes with high concentrations by first trying to identify areas that have, or are likely to have, unusually high mean concentrations.

#### BEHAVIOR OF RADON DECAY PRODUCTS

The chemically active radioactive decay products of radon can attach to surfaces, such as room walls, airborne particles, or - upon inhalation - lung tissue. The health effects associated with radon are due to the alpha decay of two members of the radon decay series, polonium 218 and 214. Decay products not attached to airborne particles are thought to have a greater lung dose than those attached to particles before inhalation (ref. 16). Thus the interaction between airborne particles and radon decay products is an important element in determining decay-product behavior and in estimating the associated health risks.

Recent research efforts in our group have been directed toward evaluating techniques for control of indoor particles and radon decay products and a general experimental examination of decay-product behavior. A variety of stand-alone (unducted) and in-duct air cleaning devices have been examined using tobacco smoke as a test aerosol (ref. 17, 18). Particle concentrations, size distributions, and radon and decay product concentrations were monitored continuously during the experiments.

In general, the results for particle removal were similar for ducted and unducted devices, with high efficiency particle air (HEPA) filters and electrostatic precipitators having the greatest particle and decay-product removal capabilities. Simple panel filters, either incorporated into small, unducted fan-filter devices or used in ducted systems (such as a furnace filter in a forced-air space heating system) showed little reduction in particle or decay-product concentrations. Air circulation alone was also ineffective in reducing particle or decay-product concentrations.

The interaction between particle and decay-product concentrations is summarized in Figure 3, where the normalized radon decay-product concentration (equivalent to the equilibrium factor, F) is shown as a function of particle concentration. The data points are from studies of the unducted air cleaning devices. The solid line labeled 'Total' is the calculated equilibrium factor based on inferred values for decay-product deposition rates and measured ventilation rates in the experimental chamber. The dashed curve indicates the calculated relative concentration of unattached radon decay products. At low particle concentrations, almost all the airborne decay products are unattached; as particle concentrations increase a greater fraction attach to these particles. The total airborne decay-product concentration declines with decreasing particle concentration. This effect is due principally to the much higher deposition rates for the unattached decay products (on the order of  $15 \text{ hr}^{-1}$ ) compared with those that are attached (ca.  $0.15 \text{ hr}^{-1}$ ). Thus air cleaning contributes to the reduction in radon decay-product concentration due to their direct removal and due to the increased deposition rate of unattached decay products, which predominate at lower particle concentrations.

Another important aspect of the use of air cleaning for control of radon decay-product concentrations is also illustrated in Figure 3, where the relative radiological dose to the lungs has been calculated for various

relative decay-product and particle concentrations. These calculations, based on similar estimates in ref. 19, are indicated by the two solid lines referring to the right hand scale; two cases are shown, the upper curve is for children undergoing light activity, the lower curve is for adults at rest. These cases are thought to represent reasonable upper and lower bounds. The calculations assume that at a particle concentration of  $10^5 \text{ cm}^{-3}$  all the decay products are attached to particles, therefore having a relative dose of 1. As particle concentrations decrease, the increasing fraction of the unattached decay products becomes more important, even though the total radon decay-product concentration declines. Thus, the overall health risk does not appear to decrease significantly as particle concentrations are reduced.

#### PROSPECTS FOR CONTROLLING EXCESSIVE CONCENTRATIONS

These and related studies are helping to shape an effective strategy for finding and reducing excessive radon concentrations in U.S. residences. A very wide range of concentrations is found in U.S. homes, and it appears that this wide variability is determined, in order of importance, by source strengths, ventilation rates, and reactions of the radon decay products. The factor of greatest importance appears to be the ease with which radon-bearing soil gas is drawn through the surrounding soil into the lower part of the structure. Entry is affected critically by convective air flow, which - interestingly - also plays an important role in ventilation rates and in decay-product behavior.

Corresponding to the relative influence of these factors on indoor concentrations, we have available to us an array of techniques for controlling excessive concentrations, including reduction of the source strength, provision of more ventilation (including use of energy-efficient techniques), and removal of radon decay products using systems that clear particles from the air. It appears that source reduction, followed by increased ventilation, has the greatest potential effectiveness. But even more challenging than the development of specific control techniques may be the formulation of an overall strategy within which they may effectively be employed.

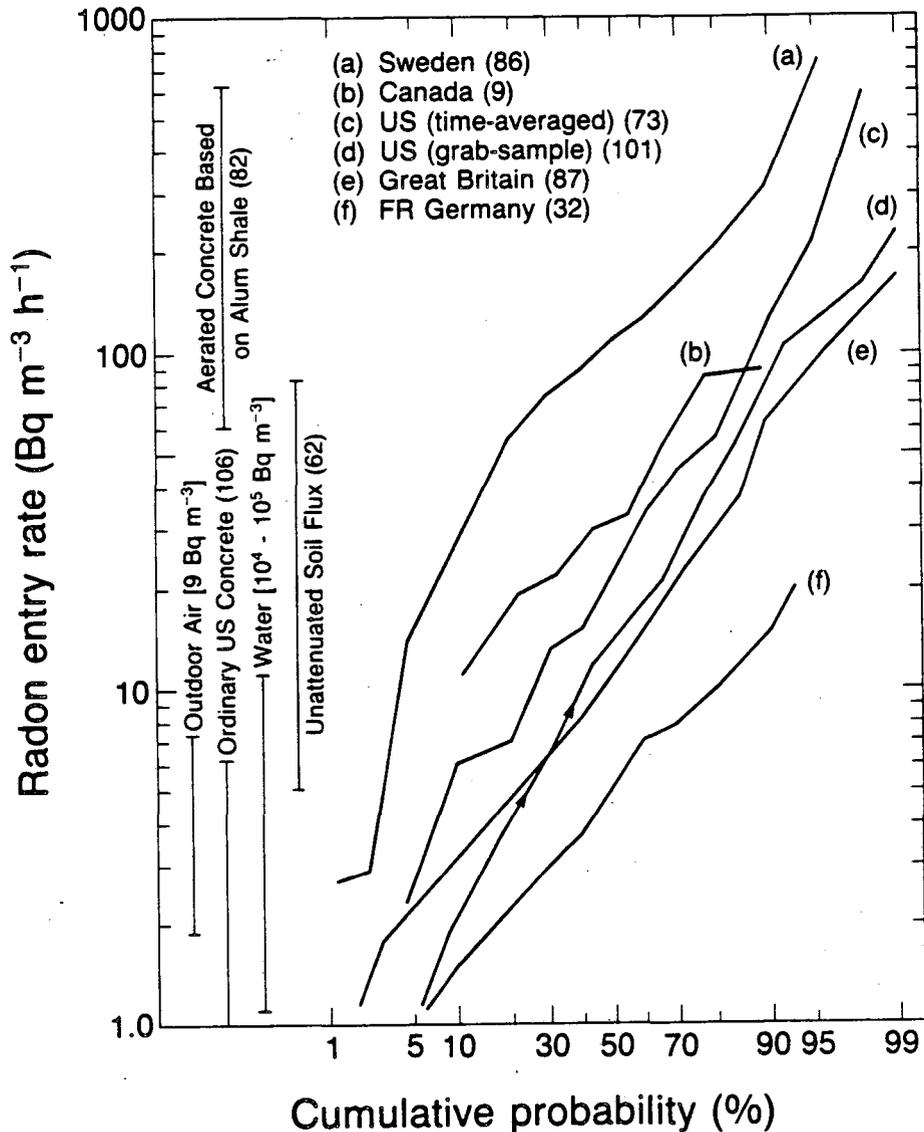
This work was supported by the Director, Office of Energy Research, Office of Health and Environmental Research, Human Health and Assessments Division and Pollutant Characterization and Safety Research Division, and by the Assistant Secretary for Conservation and Renewable Energy, Office of Building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

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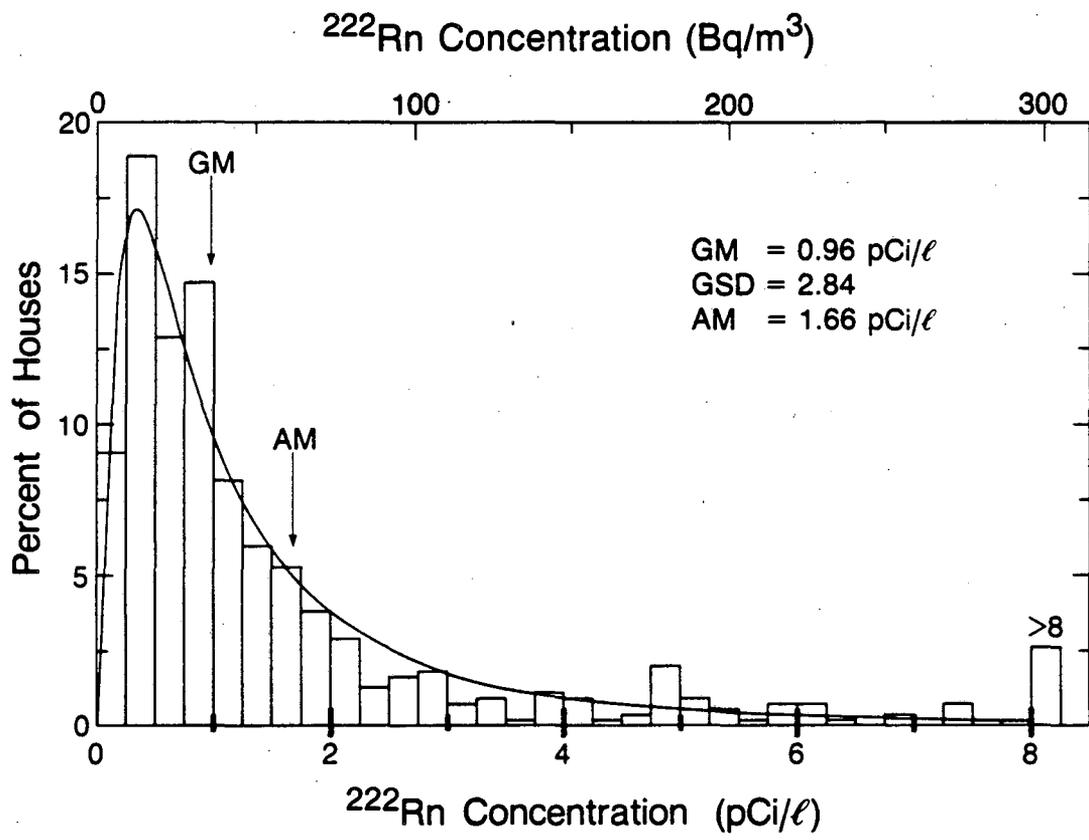
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XBL 842-10053

Figure 1. Cumulative Frequency Distributions of Radon Entry Rate Determined in Dwellings in Several Countries as the Product of Simultaneously-Measured Ventilation Rate and Radon Concentration.

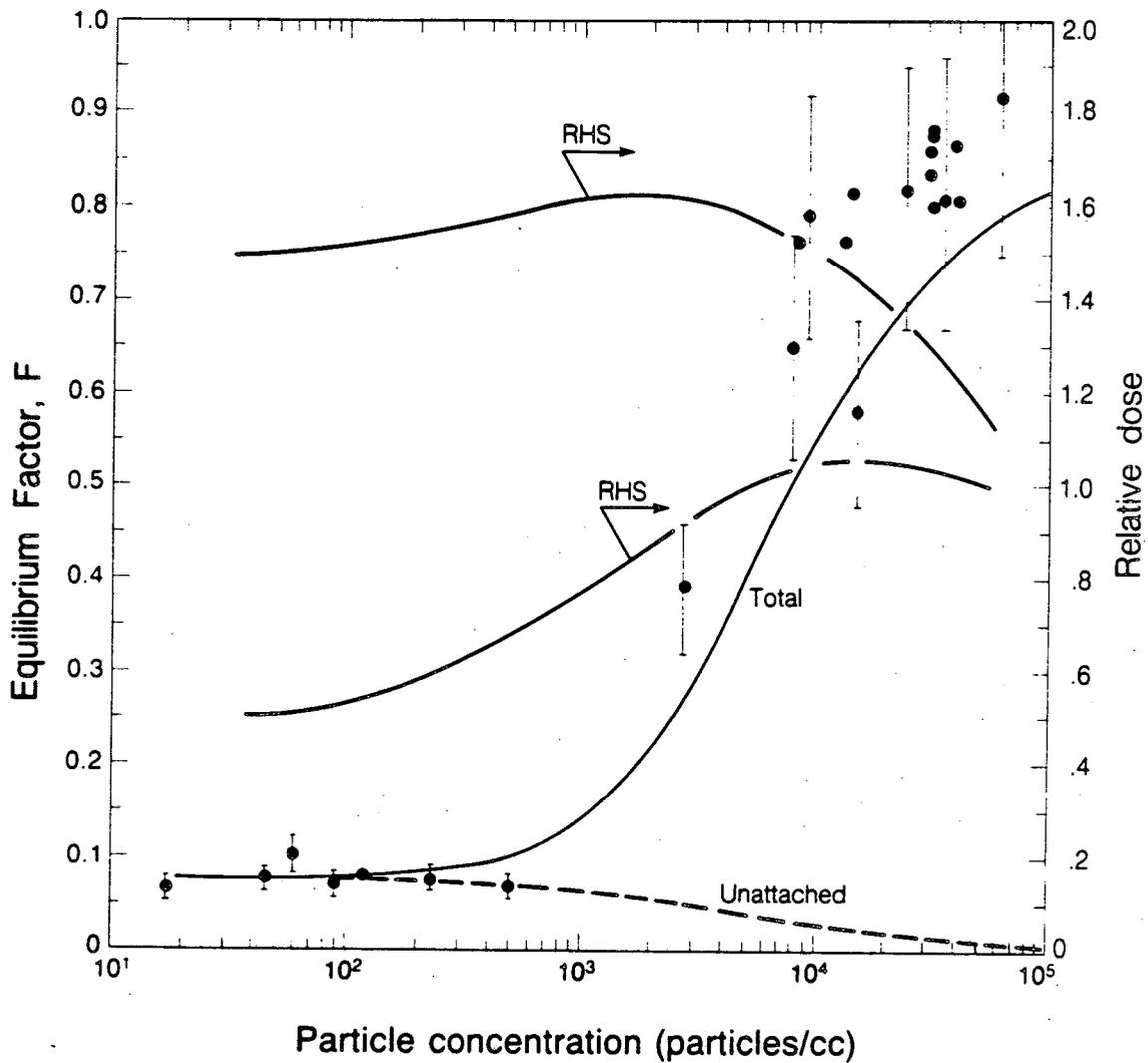
The number of residences in each sample is indicated in parentheses; the sources of these results are a) (ref. 20), b) (ref. 21), c) (refs. 2, 5, 10, 22-24), d) (ref. 3), e) (ref. 25), f) (ref. 26). The bars at the left indicate the range of contributions expected from a variety of sources, with assumptions indicated in brackets. For each source we have assumed a house having a single story of wood-frame construction with a 0.2-m-thick concrete-slab floor. The floor area and ceiling height are assumed to be  $100 \text{ m}^2$  and 2.4 m, respectively; water usage is assumed to  $1.2 \text{ m}^3$  per day, with a use-weighted transfer efficiency for radon to air of 0.55; the ventilation rate is assumed to be in the range  $0.2-0.8 \text{ h}^{-1}$ . (References for source contribution estimates: outdoor air (ref. 27); U.S. concrete (ref. 28); alum-shale concrete (ref. 29); water (ref. 30); soil flux (ref. 4).)  $37 \text{ Bq m}^{-3}$  equals  $1 \text{ pCi l}^{-1}$ .



XBL 8411-4824

Figure 2. Probability Distribution from Direct Aggregation of the 552 Individual Data in 19 sets.

The smooth curve is the lognormal functional form corresponding to the indicated parameters, calculated directly from the data.



XBL 8312-6710C

Figure 3. Equilibrium factor,  $F$ , versus particle concentration. The solid circles represent measured data; the solid line labeled Total represents calculated concentrations of total airborne decay products, while the dashed line shows calculated values for unattached decay products. The two curves referring to the right hand axis show the relative alpha radiation dose to lung tissue; the upper curve is based on dosimetric calculations for children undergoing light activity, while the lower curve is for adults at rest (see text).

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