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INDOOR CONCENTRATIONS OF RADON 222 AND ITS DAUGHTERS: SOURCES, RANGE, AND ENVIRONMENTAL INFLUENCES

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April 1985

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INDOOR CONCENTRATIONS OF RADON 222 AND ITS DAUGHTERS: SOURCES, RANGE, AND ENVIRONMENTAL INFLUENCES

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ABSTRACT

I here review what is presently known about factors affecting indoor concentrations of radon 222 and its daughters. In U.S. single-family homes, radon concentrations are found to average about 1.5 pCi/l, but substantially higher concentrations occur frequently: perhaps a million U.S. homes have concentrations exceeding 8 pCi/l (from which occupants receive radiation doses comparable to those now experienced by uranium miners). The major contributor to indoor radon is ordinary soil underlying homes, with this radon being transported indoors primarily by the slight depressurization that occurs toward the bottom of a house interior (due to indoor-outdoor temperature differences and winds). Water from underground sources contributes significantly in a minority of cases, primarily residences with private wells, with public water supplies contributing only a few percent of indoor radon, even when drawn from wells. The strong variability in indoor concentrations is associated primarily with variability in the amount of radon entering homes from these various sources, and secondarily with differences in ventilation rates. However, for a given entry rate, the ventilation rate is the key determinant of indoor concentrations. Human doses are also influenced strongly by the chemical behavior of the daughters (i.e., decay poducts of radon), and considerable progress has been made recently in investigating a major aspect of this behavior, i.e., the manner in which daughters attach to airborne particles, to walls, and - indeed - to the lining of the lung itself, where the key radiation dose occurs.

INTRODUCTION

The radiation dose from inhaled daughters of ^{222}Rn constitutes about half of the total effective dose equivalent that the general population receives from natural radiation. A variety of results from the United States suggests that ^{222}Rn concentrations in residences average about 1 pCi/l (37 Bq/m^3). Estimation of the incidence of lung cancer due to the daughters associated with this much ^{222}Rn yields thousands of cases per year among the U.S. population.

It is also clear that indoor levels are sometimes an order of magnitude or more higher than average: it is the common experience of the community performing measurements in homes that ^{222}Rn concentrations in the range of 10 to 100 pCi/l occur with startling frequency. And, whereas the risk associated with even 1 pCi/l 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%. For the extreme concentrations that have been found, risks appear to approach that from cigarette smoking.

Not surprisingly, the early work on indoor concentrations has given rise to a broad range of research characterizing ^{222}Rn and its daughters indoors. This work has included significant monitoring programs in homes (although, as noted below, not in a statistical sampling of U.S. homes), investigation of the sources of indoor radon, examination of the factors affecting indoor concentrations, study of the behavior of ^{222}Rn daughters, and - of course - development of techniques to control indoor concentrations. In addition, radiobiologists and epidemiologists have begun to apply dosimetric and dose-response data to the problem of environmental exposures to daughters of ^{222}Rn.

The international research effort in this area has been very substantial, beginning in the 1970's. For detailed information,

one must turn to the very large literature, which has - for example - recently culminated in two substantial collections of research papers on indoor radiation exposures (1,2). The purpose of the present paper is to distill the growing understanding of indoor concentrations and the factors - sources, ventilation rates, and daughter reactions - that affect them.

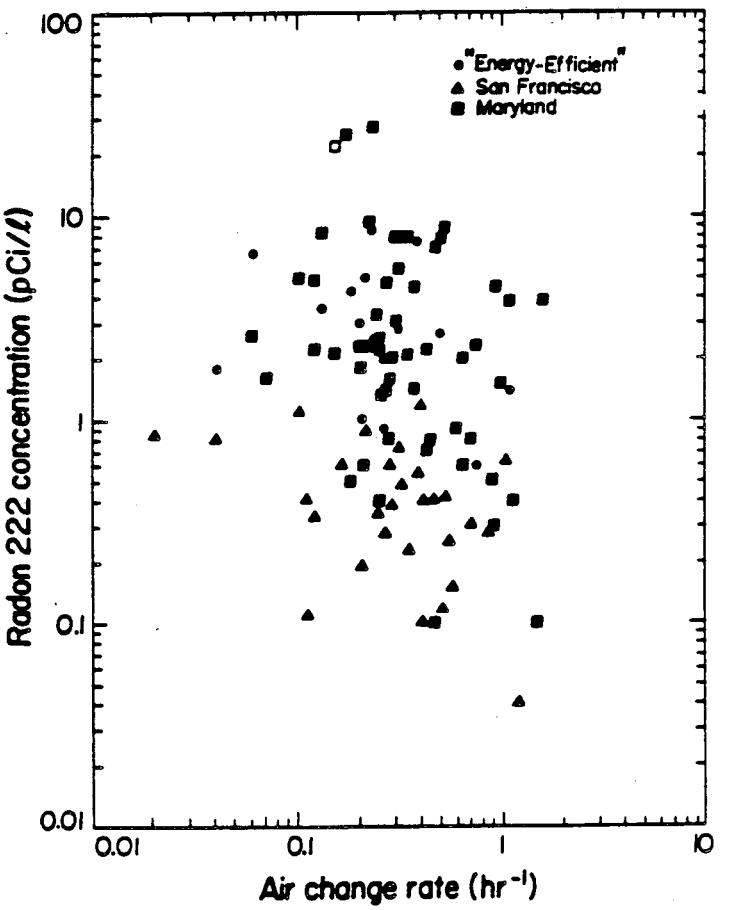

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Figure 1. Radon Concentrations and Air Change Rates Measured in 98 U.S. Residences.

The results shown are from three survey groups: "energy-efficient" houses in the United States and (one) in Canada; conventional houses in the San Francisco area; and conventional houses in a community in rural Maryland.

OVERVIEW OF IMPORTANT FACTORS

The initial observation of significant concentrations of radon (taken to be ^{222}Rn unless otherwise stated) in ordinary U.S. homes (3) 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. This expectation still has some merit, although -

for every major class of airborne pollution from indoor sources - it has become clear that the presence or absence of substantial emission rates is the major determinant of whether or not a building has excessive indoor concentrations.

The importance of differences in source strength became clear in initial investigations of the dependence of indoor concentrations on ventilation rate. It was found 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, as expected (4). However, at about the same time, paired measurements of radon concentration and ventilation rate in several housing samples showed no apparent correlation between these two parameters (5). As shown in Figure 1, for a given sample, the radon concentration and ventilation rates showed an approximately order-of-magnitude range; for the combined samples, the concentration 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.

These indications have prompted substantial work in understanding the size and variability of radon entry rates, as discussed below. However, it is important to emphasize that other factors still play an important role: ventilation rates vary substantially within the building stock, which - after all - is one major incentive for instituting major energy conservation programs (the adjunct incentive being that the average ventilation rate in U.S. buuildings is rather high as compared with rates in some countries). And, even for a given radon concentration, the concentrations and physical state of the daughters - which account for the health effects of interest - can vary significantly.

It is worth noting at this point the substantial tendency of the research community to measure radon concentrations in survey efforts, rather than daughter concentrations. This tendency arises, of course, from the availability of a reasonably reliable and very simple integrating radon monitor (6), a significant contrast to the state of daughter monitoring. And yet, given a reasonable understanding of the relationship between radon and its daughters and an awareness of the fact that the daughter-to-radon ratio does not vary as widely as radon concentrations, measurement of radon concentrations is a reasonable indicator of daughter concentrations and is certainly a very effective tool in survey efforts. This is entirely analogous to the situation for many other pollutants: for example, although any health effects associated with NO_2 exposures may have a substantial dependence on peak (as opposed to average) concentrations, an integrating sampler can be a very effective survey instrument, provided associated studies examine relationships between average and peak concentrations under well-characterized conditions.

As seen below, another incentive for emphasizing the radon concentration per se is that the very fact that this parameter shows the widest variability suggests clues to identifying and controlling excessive concentrations. Considering the origin of this wide variability, it will not be surprising that attention to radon sources and entry modes appears to have the greatest potential as a basis for control strategies.

SOURCES OF INDOOR RADON

Radon arises from trace concentrations of radium in the earth's crust, and indoor concentrations depend on access of this radon to building interiors. Radon can enter directly from soil or rock that is still in the crust, via utilities such as water (and in principle natural gas) that carry radon, or from crustal materials that are incorporated into the building structure in the form of concrete, rock, and brick. The relative importance of these pathways depends on the circumstances, but - for U.S. single-family homes - it has become clear that the first dominates the indoor concentrations that are observed.

Indications of this arose in early investigations, when it was found that measurements of radon emanating from structural materials could not account for observed indoor concentrations, based on estimates of the air exchange rate (3). A clearer picture emerged from the distribution of entry rates inferred from direct measurements of radon concentration and ventilation rate (such as those shown in Figure 1). Figure 2 shows such entry-rate distributions from various countries, as well as indicating the potential contribution of various sources. Although building materials were first suspected as the major source, based on experience in Europe (7), the initial U.S. results (5) strongly suggest that the soil must be the major source. Understanding 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) has been a major focus of research on radon entry, both in the United States and Europe.

Soil and Building Material

Establishing a radon mass-balance for a building requires consideration of the various sources. As indicated in Figure 2, a median (or geometric mean) entry rate for U.S. single-family homes is in the vicinity of 0.5 pCi l^{-1} h^{-1} (18 Bq m^{-3} h^{-1}). Based on emanation rate measurements from concretes (17), one might expect emissions from this source to account for a median of about 0.07 pCi l^{-1} h^{-1}, far below the total observed. On the other hand, the potential contribution from unattentuated soil flux, a median of

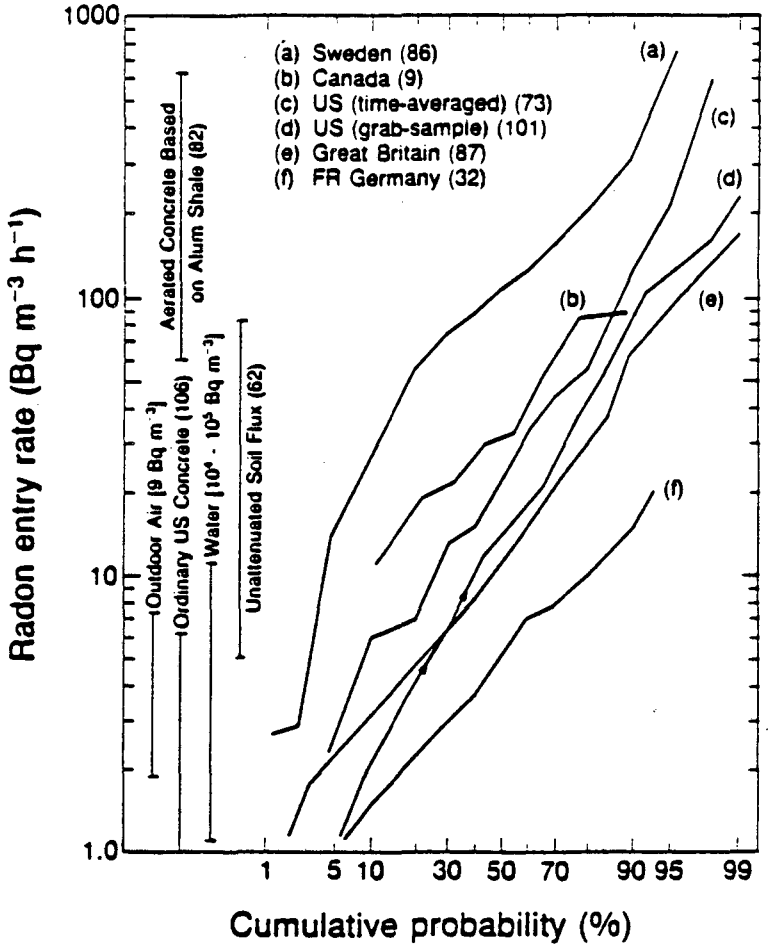

Figure 2. Cumulative Frequency Distributions of Radon Entry Rate Determined in Dwellings in Several Countries as the Product of Simultaneously-Measured Ventilation Rate and Radon Concentration.

The number of residences in each sample is indicated in parentheses; the sources of these results are a) (7), b) (8), c) (9, 4, 10-13), d) (5), e) (14), f) (15). 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 m^2 and 2.4 m, respectively; water usage is assumed to 1.2 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 h^{-1}. (References for source contribution estimates: outdoor air (16); U.S. concrete (17); alum-shale concrete (18); water (19); soil flux (20).) 37 Bq m^{-3} equals 1 pCi l^{-1}.

0.7 pCi l^{-1} h^{-1} (based on ref. 20), corresponds well with the indoor observations. However, houses have understructures that might be expected to impede substantially the ingress of radon, at least by diffusion. In fact, as discussed in more detail in a recent review of the source of radon indoors (19), although

transport via diffusion accounts well for observed fluxes from building materials and exposed soil, and could account for comparably small fluxes from the soil through some understructure materials (such as concrete), diffusion cannot account for the total entry rates observed specifically in single-family houses. Another mechanism must account for the efficiency with which radon from soil enters homes. It appears that this mechanism is bulk flow of soil gas driven by small pressure differences between the lower part of the house interior and the outdoors.

As it turns out, such pressure differences are precisely the cause of ventilation in homes during seasons when the windows are closed. These pressure differences arise from two environmental factors. First, the difference in temperature between indoors and outdoors causes small pressures across the building shell, with pressures at the bottom pointing toward the higher temperature (e.g., the heated interior) and pressures at the top causing air flow toward the colder temperature. This "stack" effect causes a convection pattern that exchanges indoor air with outdoor, with outdoor air being drawn from the vicinity of the understructure during the heating season. The second factor of importance is wind, which causes a depressurization of the house interior, as well as strong differences in pressure across the different walls that depend on the wind direction. The pressure differences caused by temperature differences and winds are roughly comparable in size, averaging on the order of a few pasqual (with higher values in relatively severe climates). These extremely small pressure differences account for air infiltration through walls (and other components of the house), the dominant contributor to home ventilation during heating season. These same pressure differences can, in principle, drive the small flows of soil gas that can account for the observed rate of radon entry into homes: soil gas contains enough radon that only 0.1% of infiltrating air would have to be drawn from the soil (19). Before proceeding to the studies specifically addressing this possibility, I note that it is often very useful to parameterize infiltration rates in terms of the temperatures differences and winds that drive them, and a very simple and useful model for doing so is now in wide use (21).

Recent work has begun to characterize directly the potential for pressure differences to cause entry of radon via soil gas, probably through imperfections and penetrations in the house understructure that permit passage of the relatively small amount of soil gas required. A study of radon entry in a single-family house with a basement analyzed the entry rate versus the ventilation rate, measured 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 (13). Moreover, the authors concluded that the observed pressure and soil parameters were consistent with the soil-gas flow rate that was

implied by the measured concentrations and ventilation rates. More detailed theoretical work (22) is helping to formulate a fundamental picture of the pressure and velocity fields in the soil surrounding homes with basements. Finally, recent experiments have directly observed, in two houses with basements, the underground depressurization implied by this picture, and have measured underground soil-gas movement by injecting and monitoring tracers (23). It is interesting to note that these results may also have significant implications for entry of other pollutants from the soil.

In respect to other housing types, the basement studies have given results that might also be expected to apply in large part to slab-on-grade structures, where the pressure difference generated can still draw soil gas through any penetrations in or around the slab. However, direct measurements in such structures have not been performed. The other understructure type of substantial importance is the crawl-space, which to some extent isolates the interior from the soil - at least in respect to pressure-driven flow between the two. Limited measurements of the transport efficiency of radon through crawl-spaces yield 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 (12).

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, which can retain radon from the soil in conditions where winds are not sufficient to flush it to the outdoors via the vents. Furthermore, for structures where the vents are sealed shut, e.g., to save energy, it is conceivable that the crawl-space still provides sufficient connection between the house interior and the soil that pressure-driven flow can enhance the flux from the soil above levels associated merely with diffusion; the work reported in ref. (12) may have observed this effect. 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, as a result of which indoor concentrations are little affected.

Thus sufficient mechanisms exist to account for the substantial amount of radon that appears to enter homes from the soil, apparently without great regard to understructure type. However, this does not imply that other sources of radon are unimportant. It is clear that materials utilized in a building structure <u>can</u> contribute substantial indoor concentrations, although this is not usually the case (even for natural stone that is higher than average in radium content). Moreover, in buildings that are relatively isolated from the ground, such as multi-story apartment buildings, indoor concentrations are expected to be lower than average - as is often the case in central European dwellings -

and to arise primarily from the building materials and, for typical U.S. infiltration rates, from radon in outdoor air.

Water

Probably more important than building materials, as a source of radon in certain parts of the housing stock, is domestic water drawn from underground sources. Surface waters have radon concentrations too small to affect indoor concentration when used indoors, but ground water is in a good position to accumulate radon generated within the earth's crust. As a result, very high radon concentrations can be found in associated water supplies: as an example, concentrations exceeding 100,000 pCi/l (3.7×10^6 Bq/m^3) have been found in wells in Maine (24). With normal water use, if the radon from such (admittedly rare) water enters the air of a typical house, an indoor concentration of about 10 pCi/l would result, among the higher levels observed.

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. 24). However, substantial data have recently become available on concentrations of radon in public water supplies drawn from ground water (25), indicating that the majority of such supplies have concentrations below 1000 pCi/l, but that a very small percentage has concentrations exceeding 100,000 pCi/l. Moreover, data are available to assess the effect of radon release to indoor air in a more comprehensive way: a very recent analysis 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 and that has an arithmetic mean of about 1.1×10^{-4}, close to the value cited above (26). The importance of these developments is that they permit quantitative assessment of the contribution of public water supplies to indoor radon concentrations. The preliminary result of such assessment is that such supplies contribute an average of approximately 0.03 pCi/l in homes served by ground water, about 3% of the average indoor radon concentrations in U.S. homes (see below). However, the very high water-borne concentrations that are <u>sometimes</u> found will contribute much larger airborne concentrations in the homes affected.

This distribution for the air-to-water ratio may also be used for assessing the contribution from private wells, to the extent that concentrations in water are known. Using data that are available for the approximately 18% of the population using private wells, ref. 26 calculates the indoor radon concentration from water

for this segment of the housing stock to average about 0.4 pCi/l. Moreover, about 1% of the entire housing stock would be expected to have indoor concentrations from water of about 1 pCi/l, due primarily to concentrations from private wells in high activity areas (such as Maine). The authors emphasize that these estimates for private-well contributions cannot be regarded to be reliable, but it is significant that, if they were approximately correct, the portion of the population using private wells would be experiencing significantly higher radon exposures than average, particularly in high-activity areas.

DISTRIBUTION OF INDOOR CONCENTRATIONS

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 may be made of the concentrations to which the U.S. population is exposed. The reason for this difficulty is that the studies that have been performed have varied significantly in incentives, scientific objectives, selection of homes, and measurement procedures. The results, not surprisingly, vary significantly, as may the conclusions that can be drawn from them. Thus, although the community has a general appreciation that the average indoor concentrations is in the vicinity of 1 pCi/l and that a notable number of homes exceed 10 pCi/l, no useful quantitative appreciation of the actual distribution in U.S. homes has been available. Knowledge of this distribution is essential in formulating a strategy for controlling excessive concentrations, as well as for making a reliable estimate of even the average population risk.

An obvious solution to this difficulty is to carry out measurements in a valid statistical sampling of U.S. homes. Given our current appreciation of typical concentrations and the incidence of high levels, it is thought that monitoring of perhaps 1000 to 2000 homes would determine mean concentrations very accurately and ascertain the fraction of homes at high concentrations (e.g., 10 times the mean) to a reasonable accuracy. However, although the Federal agencies interested in indoor air quality have been seriously considering the potential for a national survey of radon and other pollutants, this will not occur very rapidly, if at all. The main effect of the Federal evaluation may be to formulate a design that enhances the potential for aggregating results from smaller regional efforts.

Regardless of such efforts, the data already available are quite substantial and deserve careful evaluation, if only as a basis for proper design of subsequent monitoring efforts. In particular, although past studies have not been conducted with a consistent approach, the number of such studies is substantial, yielding some tens of data sets (with the precise number depending

on criteria for consideration). For this reason, a systematic analysis of U.S. results has recently been undertaken, explicitly considering the differences between studies and using lognormal representations as a basis 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 (27). Figure 3 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.

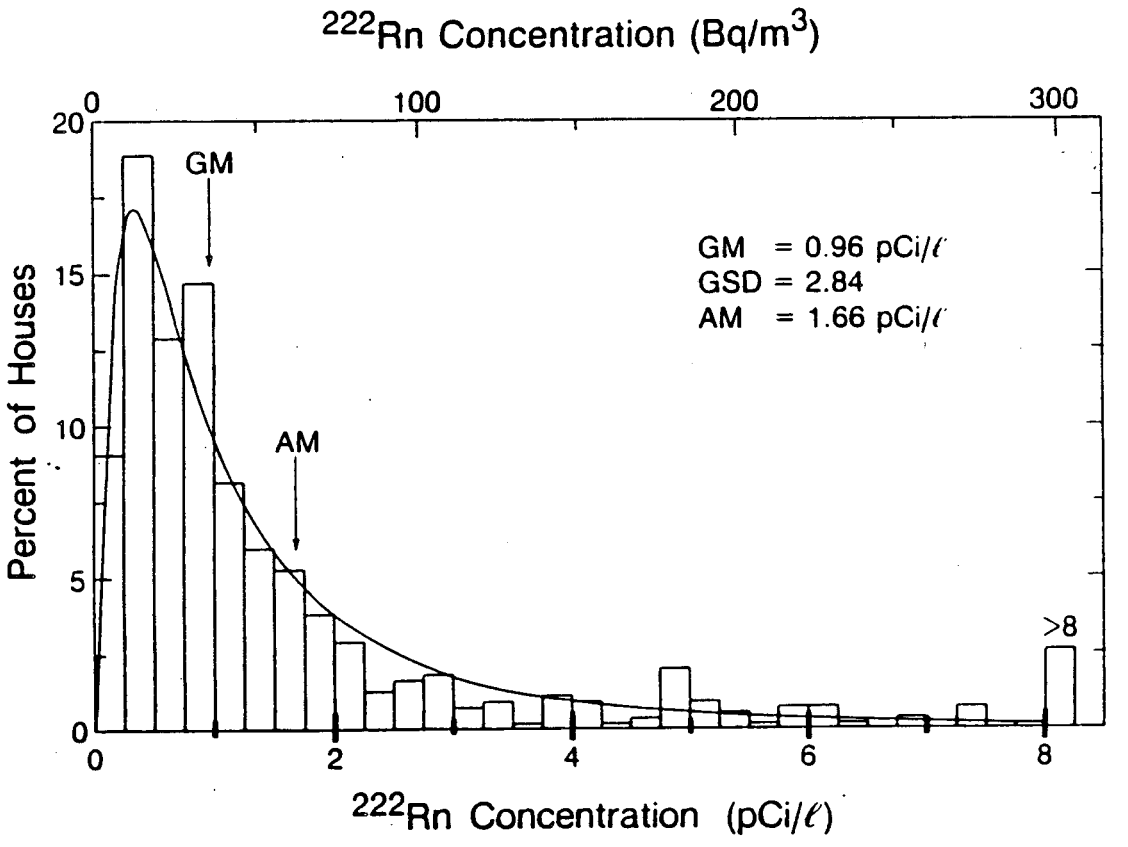

Figure 3. Probability Distribution from Direct Aggregation of the 552 Individual Data in 19 Sets.

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

To lay the basis for citing a principal result, I note that the analysis of ref. 27 utilized from 22 to 38 sets, corresponding to different areas (usually a state or urban area) of the country, with the larger number including monitoring efforts that were

prompted by 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 0.3 to 5.7 pCi/l (11 - 210 Bq/m^3), 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 0.9 pCi/l and a geometric standard deviation of 2.8, implying an average concentration of 1.5 pCi/l and 1 to 3% of houses exceeding 8 pCi/l. 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 housing stock, and the results of this analysis suggest that of the order of a million houses have annual-average concentrations averaging 8 pCi/l 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) (28). 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 index 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 unusually high mean concentrations.

We are still faced with difficulty in estimating concentrations in apartments (and, of course, in other types of buildings). Indications are, based on a few measurements (such as a single third-floor apartment in ref. 3), that concentrations are typically a few tenths of a pCi/l, substantially lower than concentrations in single-family houses. In fact, for an apartment or other building where the average space is relatively well-isolated from the ground, the major contributions to indoor concentrations may be expected to be outdoor air and building materials. The few measurements made to date in the United States are consistent with this expectation, which is also confirmed by the much larger European efforts monitoring concentrations in apartments.

Overall, the approximate contribution of various sources to U.S. residences may be summarized as in Table 1. There it is seen that 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 the building material and the outdoor air, which also contributes 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 large than in ordinary circumstances.

Table 1.

Approximate Contributions of Various Sources to Observed Average Indoor Radon Concentrations

	Single-Family Houses (pCi/l)	Apartments (pCi/l)
Soil potential (based on flux measurements)	1.5	< 1
Water (public supplies)	0.01*	0.01*
Building materials	0.05	0.1+
Outdoor air	0.2	0.2
Observed indoor concentrations	1.5	(0.3)

* Applies to 80% of population served by such supplies; contribution from water may average about 0.4 pCi/l in homes using private wells, with even higher contributions in high-activity areas.

\+ A higher contribution to apartment air is suggested on the presumption that, on the average, apartments have a higher amount of radon-bearing building materials per unit volume than do single-family houses.

BEHAVIOR OF RADON DAUGHTERS INDOORS

Basic Considerations

Even for a given indoor concentration of radon, the concentrations of its daughters (or "progeny") and their physical state can vary substantially. The behavior of the daughters is determined by their fundamental physical and chemical characteristics. Their chemical activity is what distinguishes the

daughters substantially from radon itself: the daughters are chemically active and can therefore attach to airborne particles, to indoor (macroscopic) surfaces, and - indeed - to the human tracheobronchial tract, where they can deposit either directly or after attachment to airborne particles. On the other hand, the detailed behavior and health significance of the daughters is influenced greatly by their half lives and decay modes, indicated in Figure 4. The two alpha decays that impart the radiation dose of greatest significance are shaded in the figure and are both the results of decay of polonium isotopes. The amount of (polonium) alpha energy that will ultimately be emitted from an arbitrary mixture of radon daughters in air is uniquely specified by the concentrations of the first three daughters, ^{218}Po, ^{214}Pb, and ^{214}Bi (known as Radium A, B, and C in earlier years). The next daughter, ^{214}Pb, has such a short half life that its activity concentration is, for practical purposes, identical to that of its parent, ^{214}Bi. Finally, ^{210}Pb has such a long half life that it is effectively cleared from indoor air - as well as from the lung - in contrast to the earlier daughters, whose 3, 27, and 20 minute half lives permit their accumulation in buildings with typical ventilation rates and their decay in the lung before they are cleared.

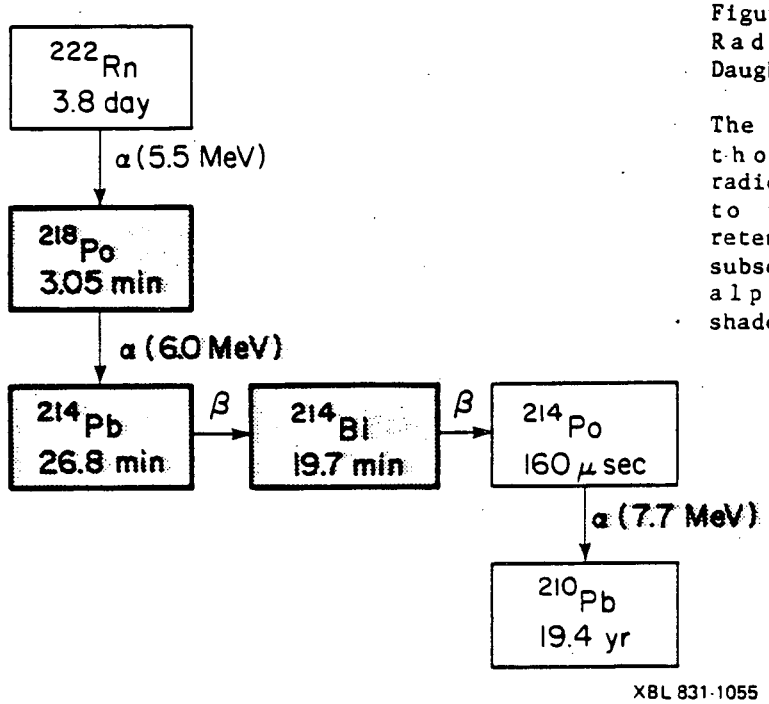

Figure 4. Decay Chain of Radon 222 and its Daughters.

The shaded isotopes are those of primary radiological interest due to the potential for retention in the lung and subsequent irradiation by alpha decays (also shaded).

A useful measure of daughter concentration, the potential alpha energy concentration (PAEC), is therefore determined from the individual daughter concentrations by the expression, PAEC = $K(0.10xI_1 + 0.52xI_2 + 0.38xI_3)$, where I_i is the activity

concentration of the ith daughter and the constant K has a value that yields a PAEC of approximately 1 WL (working level) - equal to 1.3×10^5 MeV/l - when all of the daughter concentrations are equal to 100 pCi/l. Alternatively, one can measure the concentration in terms of an equilibrium equivalent daughter concentration (EEDC), which is the concentration that - if attributed to every daughter - yields the same PAEC as the mixture that is actually present. In fact, equilibrium - a condition where all the daughters have the same concentration as radon - would only be attained if daughters were only removed by radioactive decay. It is useful, therefore, to define an equilibrium factor equal to the ratio of EEDC to radon concentration. In the real world, daughters are removed from the indoor air by several mechanisms, so that the equilibrium factor is always less than 1, most frequently in the range 0.3 to 0.7.

Were it not for the chemical activity of the daughters, the departure from equilibrium would depend solely on the ventilation rate. But the fact that the daughters can attach to particles or to surfaces, and that these attachment rates can vary with conditions, makes general characterization of the state of the daughters - and of its dependence on ventilation rate, particle concentrations, and other factors - exceedingly complex. However, since we are dealing only with a few species, whose rate of production from early members of the decay chain is determined solely by known half lives, it is possible to specify a relatively straightforward framework for considering the behavior of the daughters.

Figure 5 illustrates, for an unspecified daughter, various of the mechanisms for changing the state (or presence) of the daughter, other than radioactive decay itself. Because the deposition rates for the daughters depend strongly on whether or not they are attached to particles - and indeed on the particle characteristics - airborne particles play a crucial role in determining the concentrations that are present in the air, and potentially on the radiation dose that results from a given concentration. Given the parameters that are indicated in Figure 5, one can write down a system of mass-balance equations, following Jacobi (29), that determine the concentrations, based on given rate constants, or - conversely - that can determine specific rate constants on the basis of experiments that measure individual daughter concentrations. Practical application of such a theoretical approach usually requires assumptions that simplify the picture. One of the usual simplifications is consideration only of a single well-mixed space. Another is the lack of differentiation of rate constants on the basis of particle size or chemical composition.

These simplifications aside, key issues of interest are the rate of attachment of radon daughters to particles, as well as the rate at which free and attached daughters deposit on walls. (In many cases, deposition is parameterized in terms of the "deposition

velocity", which - for a given space - is proportional to the deposition rate.) By way of perspective, in contrast to typical ventilation rates on the order of 1 h^{-1} and daughter radioactive decay constants that are similar (or, in the case of ^{218}Po, one order of magnitude higher), rates of attachment to particles, for typical particle concentrations, appear to be on the order of 50 h^{-1}, with slightly lower rates - perhaps 15 h^{-1} - for plateout of unattached daughters onto interior surfaces. In contrast, rates for deposition onto walls of airborne particles (and of any daughters attached to them) are very low, on the order of 0.1 h^{-1}. As a result, an atmosphere with low particle concentrations tends to have a higher overall rate of deposition onto the walls - because a higher proportion of the daughters are unattached - and a lower equilibrium factor. This condition can, of course, be attained by use of particle-cleaning devices. However, the advantage indicated by the lower equilibrium factors (and hence lower PAEC) may be balanced by the fact that the detailed behavior of unattached daughters in the lung may cause a more significant radiation dose than that associated with attached daughters.

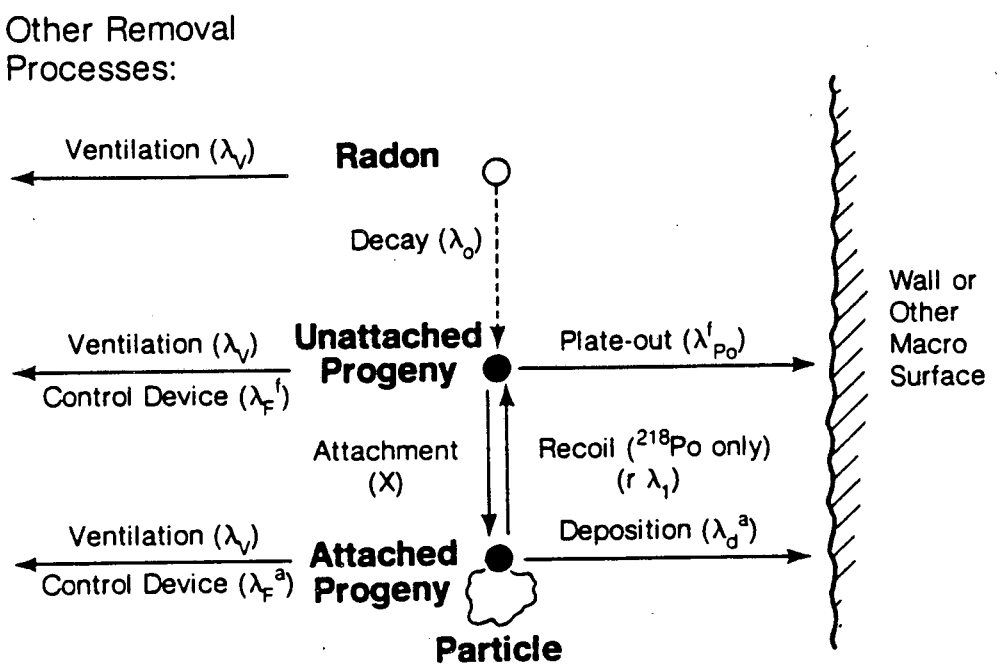

Figure 5. Daughter Removal Mechanisms (Other Than Radioactive Decay) and Associated Rate Constants.

Once created by decay of its parent, a daughter may attach to airborne particles, a process that is usually considered to be reversible for ^{218}Po because of the substantial recoil energy associated with alpha decay. Whether attached or not, a daughter can be removed from the indoor air by plateout/deposition on indoor surfaces, by ventilation, or by processing of the air through an air-cleaning system.

Recent Results

The complexity and importance of radon daughter behavior, as well as the potential interest in air-cleaning as a control technique, has given rise to a substantial amount of work - both experimental and theoretical - on characterization of radon daughters. A great deal of such work is reported in papers in references (1) and (2), as well as in other publications. References 30 - 32 constitute effective reviews. Here it is worth mentioning a few examples of important progress over the last several years.

Experiments in small and room-sized chambers, and related analysis in terms of the Jacobi model, have suggested the values for deposition rates indicated above (33). This and other work has demonstrated that the rate at which "unattached" daughters plate out, while very high as compared with particle deposition rates, is smaller than would occur if the daughters were present in the form of single unattached atoms (which would have a very high diffusion constant). The implication that an unattached daughter is actually a cluster of atoms including a daughter atom appears to be confirmed in experiments that measure the size distribution of daughters: the daughters appear to divide into two regimes, one mode having a size median diameter of about 100 nm, as might be expected based on the size distribution of particles typically present in the room, and a smaller fraction with median diameter in the vicinity of 10 nm, perhaps an order of magnitude greater than the size expected of a single ^{218}Po atom (34).

Considering what is known about the behavior of radon daughters, estimates have also been made of the effect of air cleaning techniques on the radiation dose to the lung. Such estimates suggest that the radical reduction in PAEC that is possible by air cleaning may not cause a corresponding decrease in estimated health effects; it is even possible that there is no decrease at all (35). On the other hand, a detailed review of dosimetric models yields the result that, although the PAEC is an imperfect measure of dose and - ultimately - of health effects, it is still a reasonably good indicator, assuming that parameters are in the normal range (36). These results would seem to suggest that - to the extent that air-cleaning devices result in particle concentrations outside the normal range - there is the potential that PAEC is no longer a good indicator of dose. On a related matter, the effectiveness of generally available particle-cleaning devices is highly variable, as indicated in Figure 6, from ref. 32, where it is found that - although systems based on HEPA filters and electrostatic precipitation can have a substantial particle cleaning rate (and an attendant substantial effect on PAEC) - many devices, especially the small and inexpensive ones that have recently become very popular, can have very small removal rates.

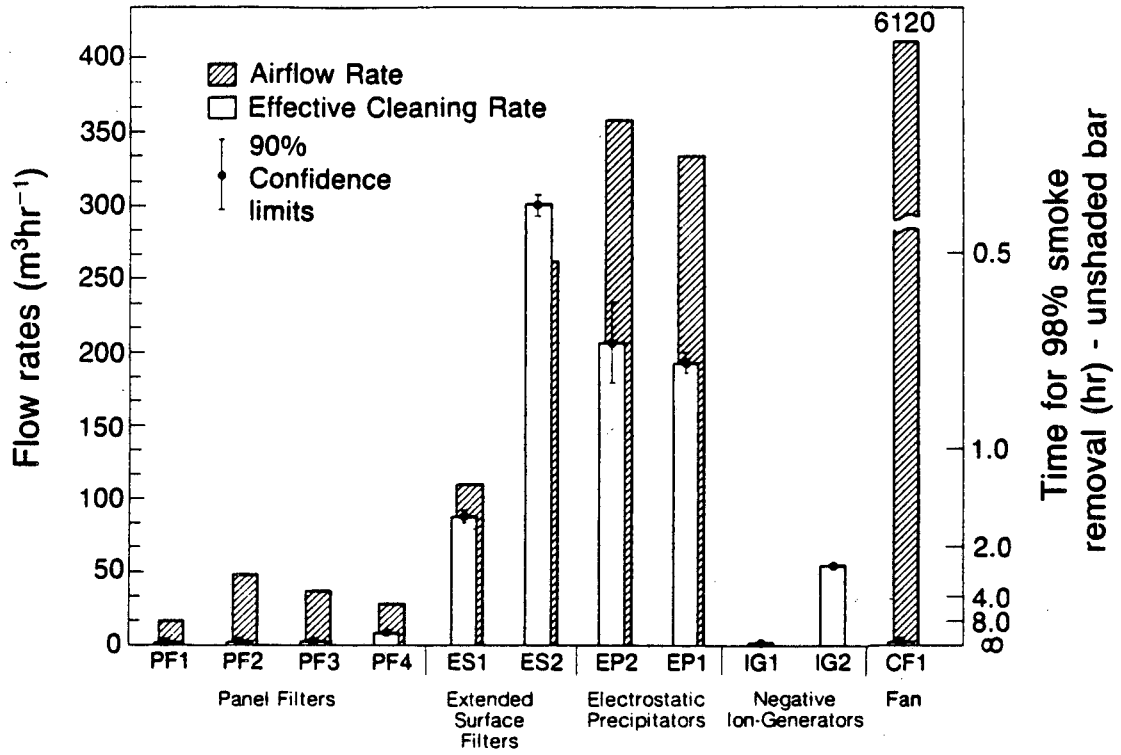

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Figure 6. Performance of Various Unducted Air-Cleaning Devices for Removal of Airborne Particulates (generated in these experiments by a cigarette-smoking machine).

Bars give flow rates: shaded is measured air flow rate, unshaded is effective rate at which air is cleaned of particles, calculated as the inflow of particle-free air required to produce the observed decay rate of cigarette smoke. (The right-hand vertical scale indicates 98% clearance times for each device.)

Finally, Figure 5 does not explicitly indicate one of the potentially substantial influences on daughter behavior, i.e., the fact that air within a room moves and that the pattern and rate of air movement can significantly affect plateout rates. Recent advances have made it possible to simulate air movement in an enclosure, thereby removing the simplifying assumption ordinarily used for simulation of radon-daughter behavior, that of a well-mixed room. The more detailed formulation permits treatment of the boundary layer more realistically, thereby providing a basis for determining the manner in which the plateout rate (or deposition velocity) depends on conditions in the room and, particularly, near the wall (37).

The importance, not only of ventilation rate, but also of other measures of air movement, indicates the need for a more complete understanding of the manner in which buildings operate if we are to understand how radon daughters behave. A similar conclusion arises from considering the manner in which radon enters

buildings, where it has become clear that the building is not a passive object into which radon diffuses, but actively contributes to the entry of radon indoors. And, indeed, both of these issues - i.e., radon entry and daughter behavior - are linked to the question of ventilation rate in a much more subtle way than was initially envisioned. Whereas the ventilation rate might be thought to influence indoor concentrations primarily in providing a means for removal of radon from the building interior, it is now clear that the same factors that account for infiltration affect radon entry decisively and - indeed - comparable factors drive air movement indoors, which has its influence on the behavior of the radon daughters. In a similar way, consideration of how to control excessive indoor concentrations requires attention to the several factors that determine indoor concentrations and that therefore offer the potential for reducing those that are deemed excessive.

STRATEGIES FOR CONTROLLING INDOOR CONCENTRATIONS

There is ample evidence that indoor radon concentrations constitute a significant portion of natural radiation exposures. As determined either by direct measurement of the radon daughters or by measurement of radon and assumption of typical equilibrium factors, even average concentrations yield estimated lung cancer rates that equal a substantial portion of the lung cancers that are not associated with cigarette smoking. Thus the average radon concentration in U.S. dwellings appears to account for thousands of cases of lung cancer annually. And, considering the apparent distribution of indoor concentrations, a substantial number of homes have very high concentrations. As noted above, perhaps a million U.S. homes have concentrations causing exposures that exceed the remedial action criterion recently recommended by the NCRP; occupants of these million homes have exposures comparable to those now received by uranium miners. Faced with a large number of homes apparently in need of help, we must consider how effectively to find these homes and to reduce concentrations significantly. A related question is how to lower the average exposure of the population, if this seems desirable.

Control strategies entail two basic elements: 1) the specific techniques for controlling concentrations, and 2) the framework within which such techniques are applied to homes found to be in need of action. The control techniques available correspond quite closely to the factors found to affect indoor concentrations. For several of the major classes of indoor pollution - whether radon, combustion products, or airborne chemicals - these factors may include the source strengths for the pollutants of interest, the ventilation rate (and ventilation effectiveness), and reactions of the pollutants with each other and with the buildings or its contents. For each pollutant class, concentrations appear to be distributed approximately lognormally,

and the largest contributor to the width of the distribution is typically the source strength, but - in each case - variability in ventilation rates and in reaction rates contribute significantly. Thus, the apparent ranking indicated above for the influence of these factors on radon concentrations is not anomalous.

Given our current understanding of the strong variability in radon entry rates and its origin, one technique for controlling indoor concentrations is to minimize entry rates, particularly in cases where they are unusually large. Considering the importance of pressure-driven flow of soil gas into houses through their understructures, substantial attention has been given in recent years to the potential for reducing this flow. It is clear that use of better barriers, sealants, and construction techniques can have a significant effect on the radon entry rate, but this potential appears - in most cases - to be limited, one reason being that slight movements of the house understructure can create small imperfections that appear to be adequate pathways for the entry of soil gas.

An alternative approach that has the potential for reduction of entry rates by large factors is to apply some technique that flushes radon from the region immediately below the house understructure, effectively presenting an alternative pathway for the radon flux from the ground. In certain cases, where the main entry route is highly localized, as through a drain tile and sump system, provision of local venting is highly effective. In the more general case of a basement or slab-on-grade, one or a few pipes that use a small fan to depressurize the soil (preferably gravel) immediately below the concrete floor can strongly reduce the radon entry rate, in effect by reversing the pressure differential that would ordinarily draw soil gas into the home. In the case of crawl spaces, active ventilation of the space below the house can easily be accomplished, although - as noted above - careful sealing of the floor may be quite practical in this case.

For situations where large entry rates are responsible for excessive concentrations, such entry reduction techniques appear to have the greatest potential effect. However, there are also circumstances where increases in ventilation rates are appropriate, whether because the ventilation rate in question is unusually low, because source reduction techniques do not appear effective for the case at hand, or because - in rare cases of extremely high concentrations - an immediate, if only temporary, solution is required. The primary limitation of increased ventilation rates, especially in homes that have very high concentrations, is that reduction of indoor concentrations by large factors will require increases in ventilation rate by large factors, which is often impractical, uncomfortable, or too expensive, at least for the long term. For homes where only modest reductions are sought, ventilation increases are quite practical, including systems that recover energy that would otherwise be lost - either by

incorporation of an air-to-air heat exchanger between incoming and outgoing air streams or by recovery of heat from an exhaust ventilation system. (However, an exhaust ventilation system must be used with some caution, since it may result in an increased depressurization of the house that leads to higher radon entry rates.)

An alternative means of control is, of course, use of air cleaning systems to remove radon daughters. The most common of these, as suggested above, employ particle removal techniques such as filtration and electrostatic precipitation. However, although such techniques can substantially reduce the daughter concentrations as measured by the potential alpha energy concentration, their effect on the actual dose to the lung is far from clear. As a result, the radon research community as a whole presently favors employment of source-reduction techniques as the first choice and use of increased ventilation rates as the next option.

The other element in a control strategy, formulation of a framework within which concentrations are controlled, is - if anything - more difficult to define. Even given an adequate understanding of the general occurrence of radon in homes (and other buildings) and of the attendant health implications, such a framework itself involves several interconnected elements. One is some agreement on objectives of the control strategy, including specification of concentration limits or guidelines. A related issue is allocation of responsibility, both for locating houses with excessive concentrations and for implementing the appropriate control techniques. Next is formulation and implementation of the actual scheme for identifying areas and individual houses with high concentrations. And finally is the logical structure that indicates what control approach should be used in each situation.

Formulating this framework will be no easy task. Parts of it are already being attacked, often helter skelter, but it is important to appreciate that no satisfactory and systematic treatment of the problem of indoor radon will be possible without conscious attention to each of these elements and to their interconnections. Some of the elements, e.g., the formulation of a scheme for finding cases in need of help, depend largely on our growing understanding of the factors that affect concentrations - a primarily scientific question. But other elements, such as that of responsibility, have very substantial social and even political components. As a result, attacking the problem of indoor radon goes far beyond the purely scientific or technical.

CONCLUSIONS

Due to work of recent years, the research community has made

substantial progress in understanding the factors that affect indoor concentrations of radon 222 and its daughters. A very wide range of concentrations is present in homes, e.g., in the United States, and these are found to depend, as expected, on source strengths, ventilation rates, and daughter reactions. The highest variability is found in source strengths, which - for single-family homes - are contributed primarily by radon from the soil, but with substantial contributions in some cases from water and building materials. Ventilation rates also affect indoor concentrations directly, although they do not appear to vary as much from house to house as do entry rates. Perhaps the more interesting aspect of ventilation rates is their indirect influence on source strengths and daughter removal because of common factors - such as temperatures and pressures - that affect source strengths, ventilation rates, and daughter behavior. As to the behavior of daughters themselves, considerable effort is being devoted to an understanding of the rates at which they attach to particles or to interior surfaces and to the behavior of "unattached" versus attached daughters.

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 the daughters 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.

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