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Publication Date

1985-05-01

UC-95d
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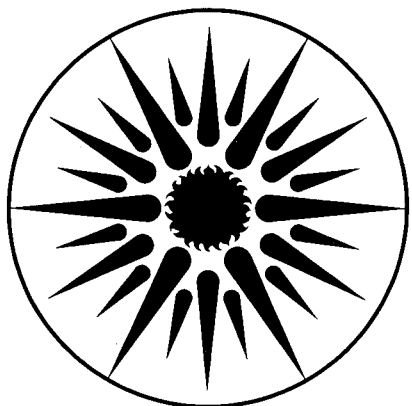
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R.G. Sextro

May 1985

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LBL-19726
EEB-Vent 85-12

CONTROL OF INDOOR RADON AND RADON PROGENY CONCENTRATIONS

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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 Research and Development, Building Systems Division of the U.S. Department of Energy (DOE) under Contract No. DE-AC03-76SF00098; and by the U.S. Environmental Protection Agency (EPA) through Interagency Agreement DW89930801-01-0 with DOE.

ABSTRACT

There are three general categories of techniques for the control of radon and radon progeny concentrations in indoor air -- restriction of radon entry, reduction of indoor radon concentrations by ventilation or air cleaning, and removal of airborne radon progeny. The predominant radon entry process in most residences appears to be pressure driven flow of soil gas through cracks or other openings in the basement, slab, or subfloor. Sealing these openings or ventilation of the subslab or subfloor space are methods of reducing radon entry rates. Indoor radon concentrations may be reduced by increased ventilation. The use of charcoal filters for removal of radon gas in the indoor air by adsorption has also been proposed. Concentrations of radon progeny, which are responsible for most of the health risks associated with radon exposures, can be controlled by use of electrostatic or mechanical filtration. Air circulation can also reduce radon progeny concentrations in certain cases. This paper reviews the application and limitations of each of these control measures and discusses recent experimental results.

keywords: air cleaning, deposition, electrostatic filtration, equilibrium factor, indoor air quality, mechanical filtration, radon, radon progeny, residential buildings, source control, ventilation.

INTRODUCTION

Radon and its immediate radioactive decay products are ubiquitous contaminants of indoor air. Radon isotopes ^{222}Rn and ^{220}Rn arise as part of the ^{238}U and ^{232}Th decay series, respectively. These radionuclides, and their eventual respective radium decay products ^{226}Ra and ^{224}Ra , are naturally occurring elements in the earth's crust. The relatively rapid decay of ^{220}Rn (often referred to as thoron, which has a half-life of 55 seconds compared with 3.8 days for ^{222}Rn) effectively limits the amount of this nuclide that can accumulate indoors in most situations; the average dose to the lung from ^{220}Rn progeny has been estimated to be about 25% of that from ^{222}Rn progeny (UNSCEAR 1982). Thus, while much of the discussion in this paper is generally applicable to either ^{222}Rn or ^{220}Rn , most of the details apply to ^{222}Rn (hereinafter referred to as radon) and its progeny.

Based on a recent compilation of measured indoor radon concentrations, radon levels in detached or semi-detached housing in the U.S. span two to three orders of magnitude. These concentrations appear to be lognormally distributed, with a geometric mean (GM) of 33 Bq m^{-3} (0.9 pCi L^{-1})* and a geometric standard deviation (GSD) of 2.8 (Nero et al. 1984). The corresponding arithmetic mean (AM) is 58 Bq m^{-3} (1.5 pCi L^{-1}). This can be compared with the ASHRAE guideline value of 0.01 working level (WL), or approximately 74 Bq m^{-3} (2 pCi L^{-1}) (ASHRAE 1981); based on this apparent distribution of radon concentrations in homes, almost 20% of the houses can be expected to exceed the ASHRAE guideline. The National Council on Radiation Protection and Measurements (NCRP) has recently proposed a 0.04 WL guideline, equivalent to approximately 300 Bq m^{-3} (8 pCi L^{-1}) (NCRP 1984). Even at this higher guideline value, approximately 1 to

*The SI units for airborne concentration of radionuclides are Bq m^{-3} ; the units in more common usage, at least until very recently, have been pCi L^{-1} , which are shown in parentheses throughout this paper.

2% of the U.S. housing stock -- 1 to 2 million homes -- can be expected to exceed this recommended level.

The health risks associated with radon are due to the alpha decay of two of its short-lived progeny, ^{218}Po and ^{214}Po . These polonium isotopes, and the lead and bismuth isotopes that constitute the immediate radon progeny, are shown in the radon decay chain in Figure 1. These progeny, unlike the chemically inert radon parent, are chemically active and can adhere to surfaces, such as airborne particles, room walls, and lung tissue. A number of models have been devised to estimate the lung dosimetry due to these radioactive decays. While a detailed review of these models is beyond the scope of this paper, the resulting dosimetric calculations indicate that the alpha dose from progeny not attached to aerosols is 9 to 35 times larger than the dose estimates for progeny attached to aerosols, depending upon the modeling assumptions (James et al. 1981).

Based on lung cancer incidence among uranium miners, estimates have been made of the lung cancer incidence due to radon exposures among the general population. Although there are a number of uncertainties, the expected lung cancer incidence in the U.S., based on the average radon concentrations just discussed, is between 1000 and 20,000 per year (Nero 1983). Exposures to higher radon concentrations increase the risk proportionately. This is an important health consequence, and efforts to reduce or control excessive exposures to radon and its progeny deserve attention.

CONTROL STRATEGIES

Background

Before discussing various methods of controlling or reducing radon and radon progeny concentrations indoors, it is worth reviewing the potential sources of radon in residential environments and noting the contribution each source can make to indoor concentrations. The major potential sources of radon in U.S. housing are the soil adjacent to the building substructure, domestic water supplies, and building materials. Other possible sources include natural gas and outdoor airborne radon. These various sources are illustrated schematically in Figure 2. The average source strength for radon from each of these sources is summarized in Table 1, and each source is discussed in greater detail below. The resulting *average* contribution to indoor radon concentrations can be estimated by dividing the source strengths listed in Table 1 by the air exchange rate, which is typically 0.5 to 1 hr⁻¹.

Soil. A primary source of radon indoors is the soil, where it is produced by the radioactive decay of radium found in trace quantities in all crustal materials. The average concentration of ²²⁶Ra in soil samples taken in 33 states in the U.S. was 41 Bq kg⁻¹ (1.1 pCi g⁻¹), with a range of 8.5 to 160 Bq kg⁻¹ (0.2 to 4.3 pCi g⁻¹) (Myrick et al. 1983). The radon gas, which is chemically inert, is then transported through the soil and into homes via cracks and other openings in the building substructure. Molecular diffusion of radon gas from the soil through these cracks or through building materials directly is a possible mechanism for radon transport into the building, although as noted in Table 1, the magnitude of the contribution to indoor concentrations does not appear to be sufficient to account for the levels often found indoors. Another process, pressure-driven flow of soil gas, is thought to be a major mechanism for the transport of soil gas into the house (Nero and Nazaroff 1984). Pressure differentials across the building shell

can arise due to wind loading and the thermally driven stack effect inside the building. These can create a slight depressurization relative to atmospheric pressure near the floor of the building shell on the order of a few pascals. This pressure differential can draw radon-bearing soil gas into the building. Thus the house, rather than being simply a passive accumulator of radon, has an active role in creating the forces responsible for a major radon entry mechanism.

Soil gas concentrations of radon range from 0.7 to $22 \times 10^4 \text{ Bq m}^{-3}$ (200 to 6000 pCi L^{-1}), with a typical concentration of $2 \times 10^4 \text{ Bq m}^{-3}$ (540 pCi L^{-1}) (Nero and Nazaroff 1984). The rate at which radon accumulates in the soil gas and the mobility of the soil gas in the soil is dependent upon a number of environmental and physical parameters. These parameters include the moisture content of the soil and other characteristics of the soil texture and structure which affect the soil permeability. Thus, while radium content of the soil is important, soil and environmental factors that affect gas flow also appear to be key components (Akerblom et al. 1984).

The cracks or other openings in the building substructure through which soil gas flow can occur may be due to the settling or aging of the building or simply a product of the construction or design practices used. The importance of this flow can be illustrated by estimating the flow needed to account for the average total entry rate for radon shown in Table 1. At the average soil gas concentration noted above, if only 0.2% of the typical building infiltration rate is due to flow through the soil, the incoming soil gas entry rate is sufficient to account for observed indoor radon levels. The radon entry rate could be much higher, due to either higher soil gas flow rates or higher radon concentrations in soil gas, or both. In many cases, the entry of radon-bearing soil gas appears to be the predominant source of radon in houses observed to have high indoor concentrations.

Building Materials. Because radium is a trace contaminant of crustal materials, it is present in all earth-based building materials. However, in the U.S. the emanation rate of radon from these materials is generally too small to cause elevated indoor radon concentrations. Ingersoll (1983) measured radon emanation rates for a variety of common building materials. For concrete, the emanation rate ranged from $(2.6 \text{ to } 19.8) \times 10^{-6} \text{ Bq kg}^{-1} \text{ s}^{-1}$ ($0.25 \text{ to } 1.93 \text{ pCi kg}^{-1} \text{ hr}^{-1}$), with an average of $7.7 \times 10^{-6} \text{ Bq kg}^{-1} \text{ s}^{-1}$ ($0.75 \text{ pCi kg}^{-1} \text{ hr}^{-1}$). For gypsum, the average emanation rate was found to be $6.3 \times 10^{-6} \text{ Bq kg}^{-1} \text{ s}^{-1}$ ($0.61 \text{ pCi kg}^{-1} \text{ hr}^{-1}$). Other materials, such as brick and rock, had lower emanation rates. The estimated source strength for indoor radon from concrete is shown in Table 1.

In certain locations in the U.S., materials high in radium were used in building construction or in the manufacture of building materials, such as the incorporation of uranium mill tailings in building materials or as backfill in areas of western Colorado, or the similar use of phosphate slag in areas of Florida, and Montana. In fact, it was this use of uranium mill tailings in Grand Junction, Colorado, that led to one of the first investigations of indoor radon concentrations in residential and commercial buildings in the mid-1960's. In some cases, high indoor radon concentrations were found (see, for example, USDOE 1979), and in others, particularly when phosphate slag was utilized, the radon emanation rates were low, although the indoor gamma radiation levels in buildings using these contaminated building materials are higher than background (Kahn et al. 1983; Lloyd 1983).

Most of the elevated indoor radon concentrations observed in the U.S. are not associated with "technologically enhanced" sources. It is the control of indoor radon levels due to natural radium concentrations that is the focus of this paper.

Water. Radon dissolved in water is a potential source of indoor airborne radon, although the average transfer factor relating the resulting concentration in air to the concentration in water is approximately 10^{-4} (Nazaroff et al. 1985b). Thus, in order to produce 40 Bq m^{-3} ($\sim 1 \text{ pCi L}^{-1}$) in air, the radon concentration in water must be $400,000 \text{ Bq m}^{-3}$ ($\sim 10,000 \text{ pCi L}^{-1}$). Surface water supplies, which provide potable water to almost half the U.S. population, contain very minimal concentrations of radon, averaging 1050 Bq m^{-3} (28 pCi L^{-1}). A recent survey of public drinking water supplies derived from groundwater and serving more than 1000 persons gives an average (AM) radon concentration of $13,000 \text{ Bq m}^{-3}$ (350 pCi L^{-1}) (Horton 1983). These public groundwater sources supply water to 32% of the population, while 18% obtain potable water from private groundwater sources. In some cases, private groundwater supplies have been found with radon concentrations exceeding $400,000 \text{ Bq m}^{-3}$ ($10,000 \text{ pCi L}^{-1}$), as found in certain granitic areas of Maine, for example (Hess et al. 1983). The data on radon concentrations in private well water are extremely limited; it seems likely that private groundwater supplies in other localities may also have radon concentrations exceeding $400,000 \text{ Bq m}^{-3}$ ($10,000 \text{ pCi L}^{-1}$).

Natural Gas. Like groundwater, natural gas can accumulate radon gas from radium in the rock structures surrounding the gas formation. Surveys of radon concentrations in gas distribution lines in various locations in the U.S. have shown a concentration range of 37 to 3700 Bq m^{-3} (1 to 100 pCi L^{-1}), with an average of around 740 Bq m^{-3} (20 pCi L^{-1}) (Johnson et al. 1973). At typical residential gas use and air exchange rates, even for unvented gas appliances, the contribution to indoor radon concentrations from natural gas is minor, less than 4 Bq m^{-3} (0.1 pCi L^{-1}).

Source Control

As with a number of indoor air pollutants, limiting production or entry of a pollutant is often easier and more cost effective than attempting to deal with the pollutant once it has been dispersed in the indoor environment. In some cases, source exclusion or elimination is the most straightforward approach. For example, the use of water with low radon concentrations will eliminate the possibility of substantial indoor radon release from water.

Where radon source elimination is not practical, such as when the source is soil gas or where no low-radon-bearing substitutes are practical, some source control methods are available. In the following section specific radon entry points are discussed, followed by a section on source reduction techniques. A general review of indoor air quality control techniques is provided by Fisk, et al. (1984).

Radon Entry. Entry points for the pressure-driven flow of radon-bearing soil gas depend on a number of factors, including the type of house substructure, the construction practices used, and the age and structural integrity of the house. Typical substructures in U.S. housing include concrete slab-on-grade, basement (partial or full), crawl-space (usually topped by a wooden floor), or some combination of these three basic designs. The first two of these substructures have similar potential entry paths, including cracks or other penetrations between the conditioned indoor space and the soil. These cracks may result from the aging and settling of the building or may be a design feature, such as the joint frequently found between the foundation walls and the floor. Penetrations for plumbing or electrical connections are also possible; often the hole surrounding the pipe or wiring is not filled or sealed.

Efforts to evaluate the differences in indoor radon concentration among houses with the various substructure types have only recently begun; thus the data are not conclusive. While homes with basements appear to have the greatest potential for high radon levels, the data collected thus far suggest that high indoor radon concentrations can also occur in houses with either a crawl space or slab-on-grade substructure. The variability in the components of the source terms for radon, such as radium content and soil permeability, may overwhelm any differences due to substructure type. In a survey of housing in the Pacific Northwest, for example, the average radon concentration in the first floor living area was 47 Bq m^{-3} (1.3 pCi L^{-1}) for 120 houses with basements, 33 Bq m^{-3} (0.9 pCi L^{-1}) for 93 houses with crawl spaces, and 43 Bq m^{-3} (1.2 pCi L^{-1}) for 7 houses having slab-on-grade construction (Thor 1984).

With regard to house substructure, clearly the potential coupling between the house substructure and the soil is largest for a basement simply on the basis of surface area alone. In many cases, concrete blocks are used for basement walls. Untreated, these can be fairly permeable to fluids, and chinking of the mortar between blocks can also occur. In addition, transport of soil gases can take place through the hollow core of the blocks. In some houses, open sumps are part of the basement construction. These sumps may be connected to a "weeping tile" system designed to remove water from beneath the basement floor and walls. This system can also serve as an effective entry pathway for soil gas when it is not occluded by water (Nazaroff et al. 1985a).

Houses built with crawl spaces appear to be less tightly coupled to the soil, although degree of coupling will depend upon features of the crawl space, such as whether the space is vented or unvented, and the number and size of penetrations between the living space and the crawl space. Many crawl spaces have open soil floors, thus radon entry into the crawl space is unimpeded. A recent study of crawl-space homes suggested that

about half the radon present in the crawl space entered the home, even with the crawl space vented. When the crawl-space vents were closed, the radon concentration in both the crawl space and the living space increased (Nazaroff and Doyle 1985). In a study of twenty-two homes in the Chicago area with unvented crawl spaces almost half of the houses were found to have radon concentrations above 185 Bq m^{-3} (5 pCi L^{-1}) and about a quarter had concentrations above 370 Bq m^{-3} (10 pCi L^{-1}) (Rundo et al. 1979).

Housing built using slab-on-grade construction can also have high radon concentrations; while the surface area of the building-soil interface is smaller than a house with a basement, the coupling between the house and the soil can still be substantial. Scott and Findlay (1983) found that cracks and penetrations through the slab floors were major sources of radon entry.

As discussed previously, water does not appear to be a major source of indoor radon for most housing in the U.S. However, in those localized situations where the radon concentration in water is high, domestic water use indoors may be the major source of radon. Not all water uses are equally effective at producing airborne radon, since the transfer process between water and air is dependent upon the degree of aeration of the water and the water temperature. Thus the use-specific transfer coefficient is highest for a shower and lowest for cold water use as in a toilet tank (Gesell and Prichard 1980). Measures to reduce indoor airborne radon concentrations due to water use could include localized ventilation in areas where hot water use is high (e.g., shower, laundry room), changing sources of water, aeration of the water before entry into the building, charcoal filtration of the water, or providing a large enough water storage capacity so that radon concentrations would be reduced by radioactive decay during the storage time.

Source Reduction Techniques. Reduction of radon entry from the soil into building interiors has generally involved (1) sealing specific leakage pathways, such as cracks,

joints or other penetrations, (2) application of a more general surface sealant and/or (3) sub-slab or subfloor ventilation. An important element in these procedures is the identification of likely entry points for soil gas; this is especially true for remedial work.

When the inner surfaces of the building foundation are finished with floor or wall covering materials, as is virtually always the case with slab-on-grade construction, identification and access to radon entry points may be particularly difficult. The task may be less complicated for an unfinished basement (though a larger surface area may be involved). Although the effectiveness of finding and sealing these entry pathways is dependent upon a number of factors, there is growing evidence from a variety of remedial projects to indicate that significant reductions in indoor radon concentrations can result (see for example, Scott and Findlay 1983; DSMA ACRES 1979; and DSMA ACRES 1980). This is not always the case, however, and measurements to assess the effectiveness of remedial techniques are usually necessary.

A common entry point is a sump system connected to a sub-slab drainage system, as described earlier. Radon can enter the building if there is no water trap to isolate the incoming drain line from the interior of the house. Installing or rebuilding the sump to accommodate a trap has been shown to be effective in reducing indoor radon concentrations, often by a factor of 4 to 5 (DSMA ACRES 1979; DSMA ACRES 1980).

Cracks or joints at the floor-wall interface can be sealed using epoxy patch material or flexible caulking that is then mechanically protected. Penetrations in wall or floors for plumbing or electrical service can be sealed in a similar fashion; often a flexible caulking material is used to accommodate thermal expansion or contraction (DSMA ACRES 1979; Keith 1980; Nazaroff and Doyle 1985; Ericson et al. 1984). As noted above, however, finding or having access to all such penetrations can be problematical in certain situations, and in some cases, it may not be clear whether cracks or chinks in mortar

actually provide a pathway between the interior of the house and the soil through the cement wall or floor. The long-term integrity of various methods of sealing cracks and other penetrations is not well documented.

Sealing entire surfaces can take a variety of forms, from brushed-on materials to flexible sheets and rigid barriers. For such sealants to be effective, they must have a low radon permeability and be able to withstand mechanical or chemical degradation. A general review of the effectiveness of surface barriers is given in Ketcham (1983). There are limited data on the performance of these various surface techniques, although in some cases, the average radon concentration has been reduced by a factor of 6 (Keith 1980). One frequently mentioned application of a surface barrier is the use of plastic sheeting to cover the open soil floor of a crawl space. This technique has been used to reduce moisture concentrations in crawl spaces. A recent but very limited experimental investigation of this technique for radon reduction has shown such a barrier to have only limited effectiveness, possibly due to pressure-driven flow of radon-bearing soil gas around the plastic sheet (Nazaroff and Doyle 1985).

Substructure ventilation is another technique to reduce radon entry into the living space. A common version of this is the ventilated crawl space, which can be used in conjunction with sealing of cracks and penetrations in the living space flooring and/or use of a radon barrier over the open soil floor. The effect of reducing crawl space ventilation has been examined in a few cases, and the radon concentrations in both the crawl space and the living space increased with crawl-space vents blocked. Sealing potential leakage pathways was found to reduce the flow between the crawl space and living area. (Nazaroff and Doyle 1985). In some cases, mechanical ventilation of the crawl space has also been used to reduce buildup of radon (Keith 1980).

Another type of ventilation that has had limited application is sub-slab ventilation, which can be accomplished in several ways. The use of a fan to draw air from an existing "weeping tile" and sump system and exhaust it outside has been shown to be effective, and in general, its cost should be low since extensive modifications to the slab or substructure are not required (Nitschke et al. 1984). Similarly, a tile and ventilation system could be installed beneath an existing floor, although this is likely to be expensive. A third method that has been demonstrated in several countries is to insert and seal a pipe or pipes through a penetration made in the slab. The use of a small fan to draw soil gas from this system produces a slight depressurization effect beneath the slab and inhibits pressure-driven flows of soil gas through other openings into the conditioned space. The backflow of air from the building into the surrounding soil induced by this pressure differential also helps dilute the radon concentration in the soil gas adjacent to the building. Reductions in average radon concentration of more than 90% have been reported (Ericson et al. 1984; Vivyurka 1979).

Removal of Indoor Radon

In some cases source reduction measures may not be feasible or may not sufficiently reduce radon concentrations. Ventilation of indoor spaces is widely used for control of indoor pollutants generally, though in some cases the energy and economic costs can be substantial. Another technique for reduction of indoor concentrations is the use of a pollutant-specific removal technique; one such method that has been suggested for radon is the use of an activated charcoal adsorbent. The effects of ventilation and charcoal adsorption on radon concentrations are discussed in more detail below. The effect of ventilation on radon progeny concentrations is reviewed in the next section.

Ventilation. The effects of ventilation are generally described using a fairly straightforward, well-mixed box model to estimate steady state concentrations of the pollutant of interest. In such a mass-balance model, the average indoor concentration, C_i , is equal to the various source terms divided by the removal terms:

$$C_i = \frac{S + \lambda_v P C_o}{\lambda_v + K + \lambda_0 + \lambda_F}, \quad (1)$$

where

S = source strength per unit indoor volume ($\text{Bq m}^{-3} \text{ hr}^{-1}$),

P = penetration fraction for outdoor airborne pollutants ($= 1$ for an inert gas such as radon),

C_o = outdoor concentration,

λ_v = air exchange rate ($=$ ventilation rate; hr^{-1}),

K = chemical or physical transformation rate (hr^{-1}),

λ_0 = removal rate due to radioactive decay of radon ($= 0.00758 \text{ hr}^{-1}$), and

λ_F = pollutant removal rate due to operation of an air cleaning device (hr^{-1}).

In the case of radon, several of these parameters have a negligible effect on indoor concentrations. The outdoor airborne concentration, C_o , is usually small compared with typical indoor levels. The chemical or physical reaction constant, K , is also zero, since radon is chemically inert (except under extreme circumstances not likely to be found in a residential environment) and is not significantly adsorbed on most building surfaces. The removal rate, λ_F , due to operation of an air cleaner is also zero, for essentially the same reasons (activated charcoal filtration appears to be ineffective, as discussed in the next subsection). And finally, the radioactive decay constant for radon, 0.00758 hr^{-1} , is quite small compared with typical ventilation rates. Thus Equation 1 essentially reduces

to the ratio of the source strength to the ventilation rate.

Use of this equation involves a number of simplifying assumptions, particularly the assumption of perfect mixing of the indoor air. The equation also does not account for any coupling between ventilation rate and radon source strength. As we have discussed earlier, pressure-driven flow is thought to be responsible for most of the radon entry into U.S. housing. Several recent studies have indicated that the radon entry rate is often a function of ventilation rate and that entry rates associated with air exchange can be significantly greater than diffusive transport alone. In some cases, it appears the additional ventilation or air exchange can result in further depressurization of the building shell, as in the case of exhaust ventilation, for example. In a detailed study of the entry of radon in a house near Chicago, it was observed that operation of the fireplace resulted in an increase in ventilation rate, as measured using a tracer gas. At the same time, the radon entry rate increased substantially, apparently because the fireplace use resulted in additional depressurization of the building shell, which in turn, increased leakage of soil gas into the building (Nazaroff et al. 1985a). Mechanical exhaust ventilation may have a similar effect, particularly if no supply vents are provided to admit outdoor air. Coupling between the rates of natural infiltration and radon entry, as discussed by Nazaroff et al. (1981b), for example, may be a consequence of the fact that wind and the thermal stack effect drive both infiltration and radon entry. Studies have also been done using residential air-to-air heat exchangers, which provide more balanced ventilation flows. No increase in radon entry rates was observed, presumably because there was no net increase in building depressurization with use of the air-to-air heat exchangers, which supply incoming air mechanically to make up for the mechanically vented exhaust air (Nazaroff et al. 1981a; Offermann, et al. 1982).

The effect of ventilation rate on indoor radon concentration is shown as a short dashed line in Figure 3, where several observations can be made. In order to achieve a reduction in radon concentration equivalent to those seen from application of some source control measures, a factor of 5 to 10, for example, the ventilation rate would have to increase by the same factor (neglecting any coupling between source strength and ventilation rate). A five-to-ten-fold increase in ventilation rate is substantial. At low initial air exchange rates, below about 0.5 hr^{-1} , such an increase may be feasible. If the initial air exchange rate is about average, from 0.6 to 1.2 hr^{-1} , a factor of 5 to 10 increase is much less practical. For example, most air to air heat exchangers used in residential applications will increase the air exchange rate 0.4 to 0.9 hr^{-1} (Fisk and Turiel 1983).

On the other hand, as illustrated by Figure 3, the indoor radon concentration rises steeply for ventilation rates below 0.5 hr^{-1} . While it is difficult to achieve ventilation rates this low on a retrofit basis, new homes can be constructed with natural ventilation rates close to 0.1 hr^{-1} . In doing so, it may be useful to provide for additional mechanical ventilation (using an air-to-air heat exchanger, for example) to bring the ventilation rate of the structure up to $\sim 0.5 \text{ hr}^{-1}$ if necessitated by indoor air quality problems.

Charcoal Adsorption. The adsorption of radon by activated charcoal is a well-known phenomenon, and its use for cleansing mine atmospheres of radon gas has been suggested by a number of authors (see, for example, Hopke et al. 1984, and references therein). Charcoal has also been suggested for control of indoor radon, although few evaluations of its use have been made. In two recent experiments, operation of an activated charcoal filtration unit produced negligible effects on indoor radon levels (Nitschke et al. 1984; Sextro et al. 1985). Both papers report that radon progeny concentrations were reduced by use of the charcoal filtration device. As noted in Sextro et al. (1985), airborne particle concentrations were also reduced by use of the charcoal

filtration unit. As discussed in greater detail in the following section, removal of particles contributes to the reduction in progeny concentration.

Radon Progeny Control

Radon progeny, the radioactive products of the radioactive decay of radon, are the main source of the radiological risks of exposure to radon. Because these elements, unlike their inert radon parent, are chemically active and can therefore attach to surfaces, such as airborne particles, room surfaces, or lung tissue, control of progeny concentrations presents a different set of considerations. In this section, equations describing progeny behavior and the associated health risks are first presented, followed by discussions of the effects of radon progeny control. Finally, estimates of the relative alpha radiation dose to the lungs under various control conditions are discussed.

Background. A commonly used method of parameterizing the airborne concentration of radon progeny with respect to their alpha decay properties is the Potential Alpha Energy Concentration (PAEC), which is given by

$$\text{PAEC} = k_1 A_1 + k_2 A_2 + k_3 A_3, \quad (2)$$

where the subscripts 1 to 3 refer to ^{218}Po , ^{214}Pb , and ^{214}Bi respectively. The coefficients, k_i , are a function of the potential alpha decay energy and the half-life of the nuclide of interest. For progeny concentrations, A_i , measured in Bq m^{-3} , the coefficients are $k_1 = 2.84 \times 10^{-5}$, $k_2 = 1.39 \times 10^{-4}$, and $k_3 = 1.03 \times 10^{-4}$, which gives the PAEC in units of working level (WL). One working level is defined as any combination of radon progeny in one liter of air such that the ultimate decay to ^{210}Pb will result in 1.3×10^5 MeV of alpha decay energy.

Another useful concept is the equilibrium factor, F , which is a measure of the degree of equilibrium established between radon and its decay products:

$$F = \frac{3700 \text{ PAEC}}{A_0}, \quad (3)$$

where A_0 is the radon concentration in Bq m^{-3} . If the radon progeny concentrations were those established solely by secular radioactive equilibrium (i.e., no other removal mechanisms exist other than radioactive decay) F would be unity. Since radon progeny are chemically active and can attach to room surfaces, the airborne concentrations are typically lower than expected from simple secular radioactive equilibrium; F is usually in the range of 0.3 to 0.7 for most indoor situations, depending upon the airborne particle concentration.

The behavior of radon progeny is illustrated in Figure 4, where the various processes contributing to the reduction of airborne concentrations are shown. The rate constant for each process is shown in parenthesis; radioactive decay as a removal process for each progeny nuclide is not indicated. While radon has two removal processes, ventilation and radioactive decay, progeny removal can occur in four ways: ventilation, air cleaning, deposition on macro surfaces, and radioactive decay. The progeny can also attach to the surfaces of indoor airborne particles, which, in turn, can be removed by ventilation, deposition, and air cleaning. As shown in Figure 4, the alpha decay of ^{218}Po can produce sufficient recoil momentum to detach the decay product from the particle; this detachment probability, denoted r in the figure, has been estimated to be 0.83 (most of this background discussion is based on Sextro et al. 1984 and Offermann et al. 1984 and references therein, unless otherwise noted). The rate of progeny attachment to particles, which is usually assumed to be independent of the chemical nature of the progeny, is dependent upon the particle concentration and size distribution. The mean attachment rate coefficient is $4.3 \times 10^{-3} \text{ hr}^{-1} \times (\text{particles cm}^{-3})^{-1}$ for particles typically found in

indoor air.

The overall progeny removal rate (excluding radioactive decay, which is accounted for separately), Λ_i , is the sum of the various removal terms:

$$\Lambda_i = \lambda_V + \lambda_F + f_i \lambda_D^f + (1-f_i)(\lambda_D^a) \quad (4)$$

where the removal terms are ventilation, λ_V ; direct removal by an air cleaning device, λ_F ; and deposition on room surfaces by progeny either unattached, λ_D^f , or attached, λ_D^a , to airborne particles. The parameter f_i is the fraction of progeny not attached to airborne particles (unattached fraction). Based on the steady-state mass-balance equations derived by Jacobi (1972) and Porstendoerfer et al. (1978) for the various radon progeny removal modes, the overall progeny removal rate, Λ_i , can be also determined by direct measurement of the progeny activities, A_i :

$$\Lambda_i = \lambda_i \left[\frac{A_{i-1}}{A_i} - 1 \right] \quad (5)$$

where λ_i is the progeny half life. Taken together, Equations 4 and 5 provide a means of estimating the deposition rate of unattached progeny. In a recent series of experiments in a room-sized chamber, the number-weighted particle deposition rate was found to be 0.16 hr^{-1} ; this rate is assumed to also be the average deposition rate of attached progeny. In the same set of experiments, the deposition rate of unattached progeny was estimated to be 15 hr^{-1} , almost a factor of 100 greater than for attached progeny (Offermann et al. 1984).

The various removal rates discussed thus far can be compared. At a particle concentration of $10,000 \text{ particles cm}^{-3}$, which is typical for homes without heavy cigarette smoking, the attachment rate for progeny to particles is 43 hr^{-1} , while the deposition rate for unattached progeny on macro surfaces is 15 hr^{-1} . Surface deposition of airborne particles (and of any progeny attached to them) is almost negligible, with a deposition

rate of 0.16 hr^{-1} . At lower particle concentrations, $1000 \text{ particles cm}^{-3}$ for example, the attachment rate to particles is 4.3 hr^{-1} , and by comparison, deposition of unattached radon progeny becomes a more important process. Similarly, at high particle concentrations, the attachment rate to particles is higher, and a greater proportion of progeny remain airborne, attached to particles.

Particle Reduction. The concentration of airborne particles is an important determinant of indoor radon progeny concentrations. Removal of particles by active air cleaning (i.e., operation of a mechanical or electrostatic filtration system) has two general effects on airborne radon progeny concentrations. First, air cleaning can remove radon progeny directly, either those attached to airborne particles which are captured by the air-cleaning system, or the unattached radon progeny which are also trapped by the air-cleaning device. Second, air cleaning also contributes to the reduction of radon progeny concentrations by reducing the particle concentration so that deposition of unattached progeny on indoor surfaces becomes a predominant removal mechanism.

There are a variety of air-cleaning devices available, including portable unducted devices that might be used for one or two rooms and devices that are installed in a forced-air space conditioning system. Although air-cleaning devices are also used in commercial and industrial applications, as part of heating, ventilating, and air conditioning (HVAC) systems, for example, this discussion focuses on residential systems since radon and radon progeny problems generally arise in residential buildings.

There are two broad categories of air-cleaning systems: mechanical fan-filters in which impaction, interception or diffusion are the major particle removal mechanisms and electrostatic filters, which rely on electrostatic forces between the particles and the collection surface. The effect of air cleaner operation can be parameterized in two ways, the effective cleaning rate (ECR) and the cleaning system efficiency. The ECR is the net

particle removal rate, determined as the difference in particle concentration decay rate as observed in a room with and without the air-cleaning device operating, multiplied by the room volume. Thus the ECR is the effective flow of particle-free air into the room that would produce the observed reduction in particle concentration. The system efficiency is the ECR divided by the air flow rate through the device itself.

The effectiveness of various ducted and unducted air cleaning systems was examined in a series of experiments using tobacco smoke as the test aerosol (Offermann et al. 1984; Sextro et al. 1985). The results for particle removal are displayed in Figures 5 and 6 for unducted and ducted devices, respectively. As can be seen in these figures, there is a wide range in the effectiveness of air cleaners; extended surface filters, such as a high efficiency particle air (HEPA) filter or a bag filter yielded the highest removal rates, while typical "furnace filters" or many of the small, portable fan-filter systems available produced essentially no reduction in particle concentrations for particles up to 3 μm in diameter.

Changes in radon progeny concentrations as a result of air cleaning show similar results. Air-cleaning devices that removed particles effectively had a commensurate effect on airborne radon progeny. The effect of particle concentration on the equilibrium factor, F , is shown in Figure 7, where the solid circles represent data from experiments with the unducted air-cleaning devices. As can be seen, the equilibrium factor decreases with decreasing particle concentration.

By combining Equations 2, 3, 4, and 5, an equation for the equilibrium factor can be derived in terms of the various removal or decay rates discussed previously.

$$F = \frac{\lambda_1}{\lambda_1 + \Lambda_1} \left[k_1 + \frac{\lambda_2}{\lambda_2 + \Lambda_2} \left(k_2 + k_3 \frac{\lambda_3}{\lambda_3 + \Lambda_3} \right) \right] \quad (6)$$

The solid line labeled Total in Figure 7 is the calculated equilibrium factor using this

equation. In order to calculate the overall removal rates, Λ_i , for use in Equation 6, the unattached fraction, f_i , must be estimated as a function of particle concentration. Figure 8 shows values of f_i inferred from the mass balance equations (Offermann et al. 1984). The equilibrium factor due to unattached progeny *only* can also be computed using Equation 6, where the k_i values are first multiplied by the respective unattached fraction, f_i . The calculated equilibrium factor values for unattached progeny are shown in Figure 7 as the dashed line. As can be seen, at particle concentrations below 500 particles cm^{-3} , the airborne progeny concentration is almost entirely associated with unattached progeny. The relative concentration of unattached progeny declines with increasing particle concentration. The total equilibrium factor, on the other hand, increases rapidly with particle concentration in the range of ~ 1000 to $\sim 25,000$ particle cm^{-3} , a concentration range typical of indoor environments.

Ventilation. We have discussed the effect of ventilation on radon concentrations indoors. However, radon progeny presents a more complex situation, since the progeny arise from the radioactive decay of radon, and because indoor concentrations are determined, in part, by deposition on surfaces and interactions with airborne particles. Rearranging Equation 3 to solve for the PAEC and utilizing Equations 1 and 6, the following equation can be derived, which describes the concentration of radon progeny (in terms of the PAEC) as a function of the various removal processes:

$$\text{PAEC} = \frac{S\lambda_1}{(\lambda_0 + \lambda_V)(\lambda_1 + \Lambda_1)} \left[k_1 + \frac{\lambda_2}{\lambda_2 + \Lambda_2} \left(k_2 + k_3 \frac{\lambda_3}{\lambda_3 + \Lambda_3} \right) \right], \quad (7)$$

where S is the radon source strength, as in Equation 1.

Results using this equation for two choices of indoor particle concentration are shown in Figure 3. In the case of no cigarette smoking, indoor particles are assumed to arise from other unvented combustion sources and from infiltrating outdoor air. At a

smoking rate of 4 cigarettes per hour, tobacco smoke is the predominant particle source. At each ventilation rate, the equilibrium particle concentration is calculated using Equation 1, followed by use of Equation 7 to estimate the corresponding progeny concentration. As the ventilation rate decreases, the PAEC increases more rapidly when particle concentrations are high (as in the case of cigarette smoking). As before, these calculations do not take into account any coupling between the radon source strength and the ventilation rate.

Air Circulation. Air circulation has been suggested as a control measure for indoor radon progeny concentrations, and a number of authors have reported observing a reduction in progeny concentration with increased air circulation (Nazaroff et al. 1981a; Windham et al. 1978; Rudnick et al. 1983). In most of these studies, however, the particle concentration was not measured during the course of the experiments. Recent experimental evidence suggests that the critical parameter in the effectiveness of air circulation in reducing progeny concentrations is the indoor particle concentration. At high particle levels, above $40,000 \text{ particles cm}^{-3}$, no measurable decrease in PAEC was observed with increased air circulation, and no reduction in particle concentration for particles less than $3 \mu\text{m}$ was seen (Offermann et al. 1984). It appears that as the particle concentration decreases, air circulation will begin to have an effect on PAEC.

This relationship between the effects of air circulation and the indoor particle concentration is consistent with the previous discussion regarding the effects of particle air cleaners. At high particle concentrations, most of the airborne progeny are attached to particles, which deposit on surfaces at a low rate. Increased air circulation has a negligible effect on the deposition rate of particles less than $3 \mu\text{m}$ in diameter. As the particle concentration decreases, the fraction of airborne progeny that remain unattached increases. Since deposition of progeny depends on their transport to surfaces, the depo-

sition rate of these unattached species is enhanced by air movement, which assists in circulating the unattached progeny close to the walls and increases the probability of deposition.

Lung Dose and Particle Concentrations. As noted earlier, the radiological effects of radon exposure are due to the alpha decay of the progeny and, based on dosimetric models, the unattached progeny produce a significantly larger lung dose than progeny attached to airborne particles. Effective air cleaning results in both particle and progeny removal, and as can be seen in Figure 7, the fraction of unattached progeny species increases with decreasing particle concentration. The lung dose expected from the resulting mixture of attached and unattached progeny can be estimated relative to the dose calculated assuming no unattached progeny (Harley and Pasternak 1972, as adapted by Jonassen 1982). Results from these calculations are shown in Figure 7, where the relative dose curves are shown as solid lines. The relative dose curves, both of which refer to the right axis, are based on two dosimetric cases; children undergoing light activity (top curve) and adults at rest (bottom curve). These two curves are reasonable representations of the limiting cases; more realistic assumptions regarding behavior patterns, breathing rate, air volume, etc., are likely to fall between these two lines. As these estimates illustrate, the radiological effects of reduced particle and progeny concentrations (without a commensurate reduction in *radon* concentration indoors) are not significantly smaller, even though the *total* radon progeny concentration (at a constant radon concentration) does decrease with reduced particle concentration.

SUMMARY AND CONCLUSIONS

Methods of control or reduction of indoor radon and radon progeny concentrations have been reviewed. These techniques may be categorized as radon source reduction,

radon removal, and radon progeny removal. There are a number of potential sources of radon in U.S. housing, including soil, potable water, and building materials. In most cases, it appears that flow of radon-bearing soil gas into houses, driven by a slight negative pressure differential across the building shell, is a major source of indoor radon; this pressure-driven flow appears to be the most likely source of radon that can account for the elevated radon concentrations observed in some houses. There are a number of radon source control techniques; their effectiveness will depend upon characteristics of the house substructure and the details of the specific application. While the results of such remedial measures have varied and the data base from which to generalize is small, five-to-ten-fold reductions in radon concentration have been reported.

In cases where source reduction is not possible or economically practical or is not entirely effective, concentration reduction measures can be employed, including ventilation or air cleaning. While, in principle, large changes in ventilation rate can be made, significant increases may not be economically or physically practical if the initial air exchange rate is in the 0.8 to 1.2 h⁻¹ range (typical of much of the existing U.S. housing stock). In many cases, reductions in indoor radon or radon progeny concentrations by a factor of 2 to 3 are possible through increased ventilation, although unbalanced exhaust ventilation procedures that lead to additional depressurization of the building shell are not likely to produce the expected reduction in radon concentration due to coupling between the additional air exchange and infiltration of soil gas bearing radon. In very tight houses with natural ventilation rates ~ 0.1 to 0.2 hr⁻¹ additional mechanical ventilation, as might be produced by an air-to-air heat exchanger, can be used to increase the ventilation rate of the structure to ~ 0.5 hr⁻¹ with a minimal energy penalty.

Progeny concentration reductions may also be achieved by air cleaning, which removes progeny by filtration of the unattached or attached airborne radon progeny and

also by reducing particle concentrations, thereby increasing the progeny deposition rate on indoor surfaces. However, the health risk from the alpha radioactivity of the remaining mixture of airborne radon progeny (both unattached and attached) may not be significantly reduced as a result of air cleaning.

It is clear that while an understanding of the efficacy of various radon and radon progeny control methods is emerging, substantial work remains in developing more general and systematic source control techniques. Because a large number of homes in the U.S. appear to exceed guideline levels for indoor radon concentrations, general indicators of the potential for high indoor radon concentrations need to be identified and investigated in order to locate geographical areas where either remedial or preventive control methods might be required.

ACKNOWLEDGMENTS

The author would like to thank William Fisk, David Grimsrud, and Barbara Moed for their very useful reviews of this paper. 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 Research and Development, Building Systems Division of the U.S. Department of Energy (DOE) under Contract No. DE-AC03-76SF00098; and by the U.S. Environmental Protection Agency (EPA) through Interagency Agreement DW89930801-01-0 with DOE. Although the research described here is partially supported by the EPA, it has not been subjected to EPA review and therefore does not necessarily reflect the views of EPA, and no official endorsement by EPA should be inferred.

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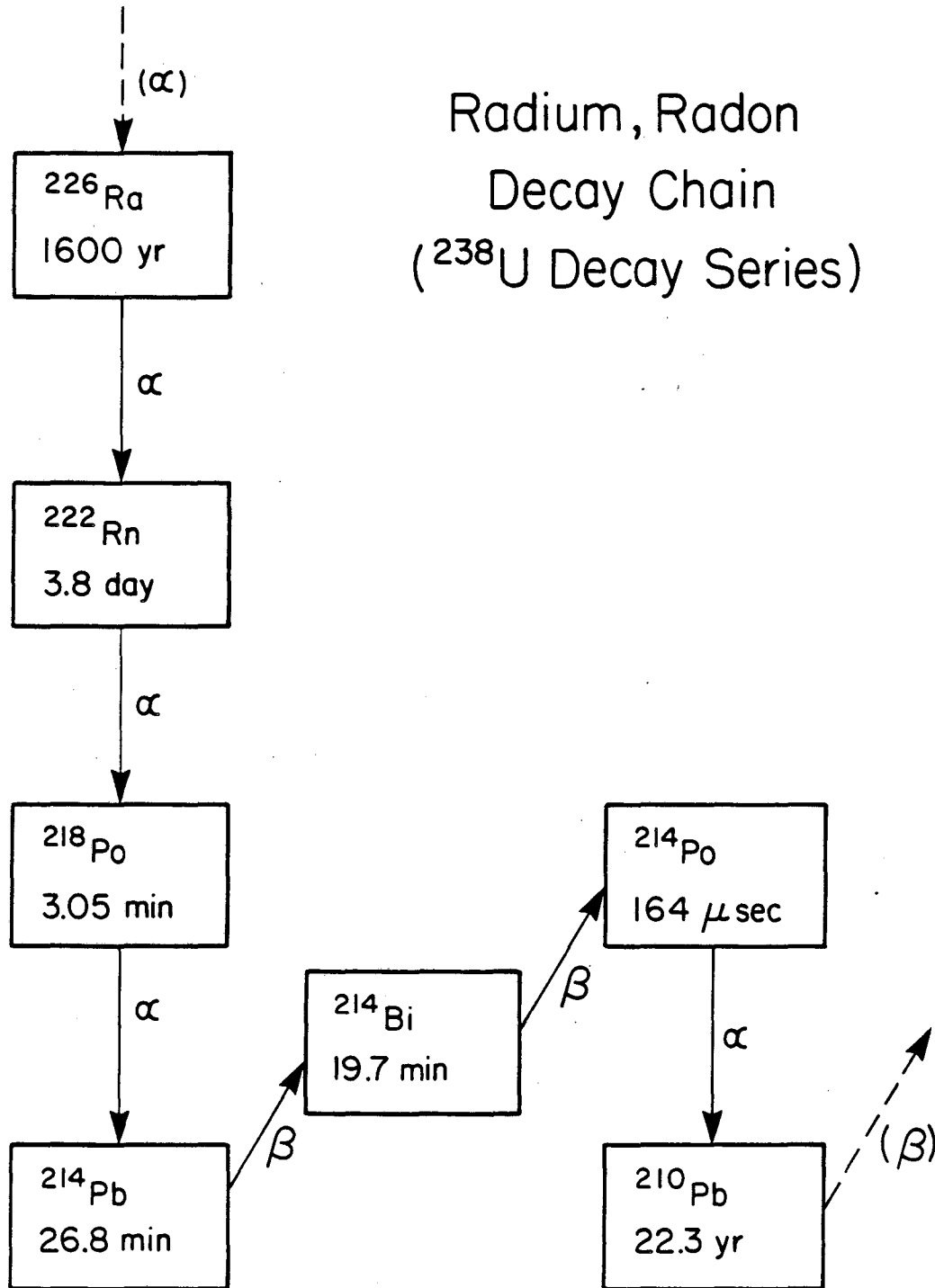
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TABLE 1

Typical Radon Source Contributions
for a Single-story Residence

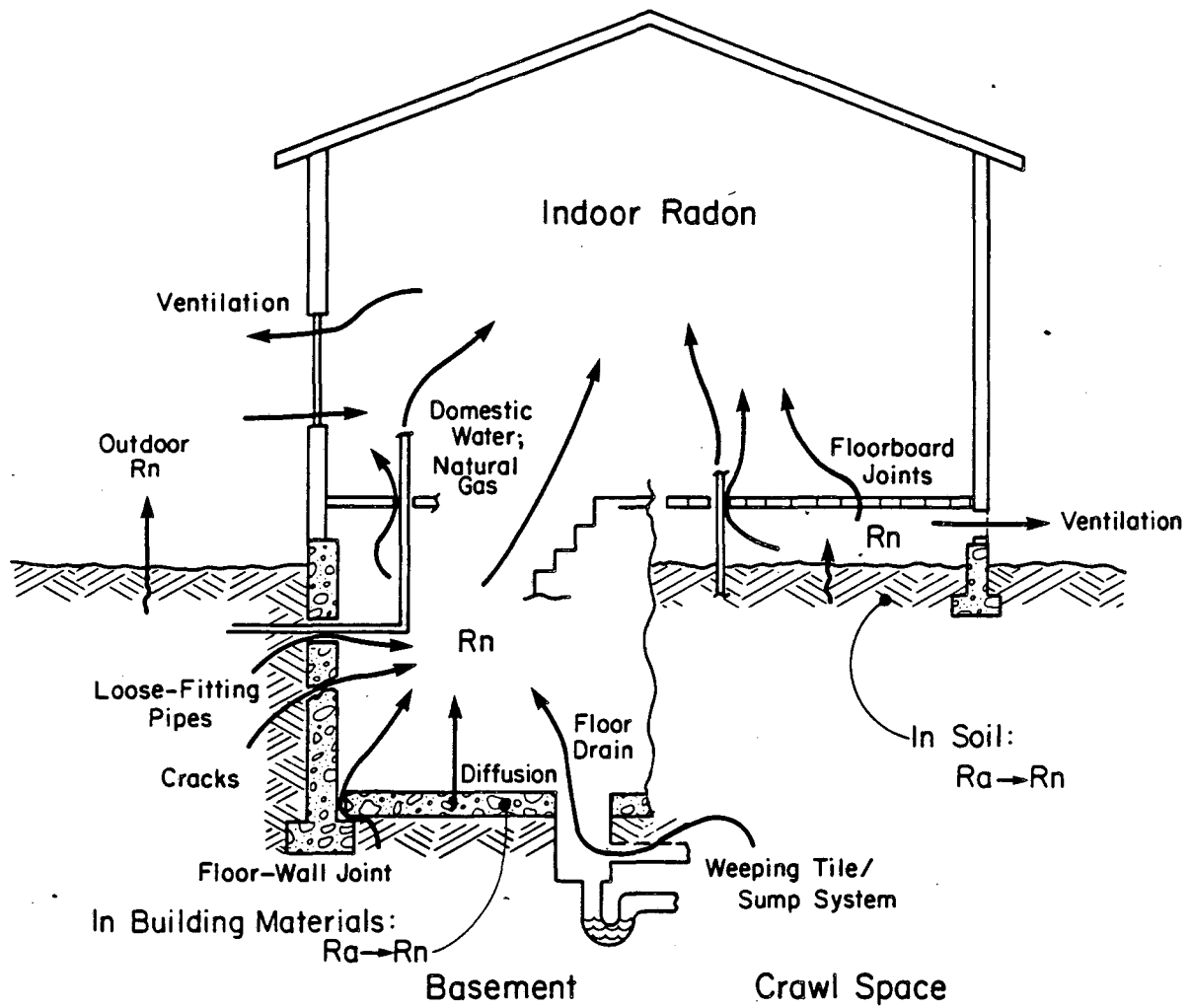
| Source | Average Source Strength $\text{Bq m}^{-3} \text{ h}^{-1}$ ($\text{pCi L}^{-1} \text{ h}^{-1}$) | Reference |
|--------------------------------|---|-------------------------|
| Outdoor air | 10.0 (0.3) | Gessel 1983. |
| Potable water | 1.0 (0.03) | a. |
| Concrete floor | 2.3 (0.06) | b. |
| Soil - diffusion through floor | 1.3 (0.04) | Nero and Nazaroff 1984. |
| Soil - uncovered soil | 32. (0.9) | Nero and Nazaroff 1984. |
| Total Entry Rate: | 52. (1.4) | c. |

- a. Potable water derived from public groundwater supplies (Nazaroff et al. 1985b).
- b. Assumes half the flux from a 100 m^2 , 20 cm-thick concrete floor enters the house (Ingersoll 1983).
- c. Arithmetic mean indoor radon concentration (Nero et al. 1984) divided by an average ventilation rate of 0.9 hr^{-1} .



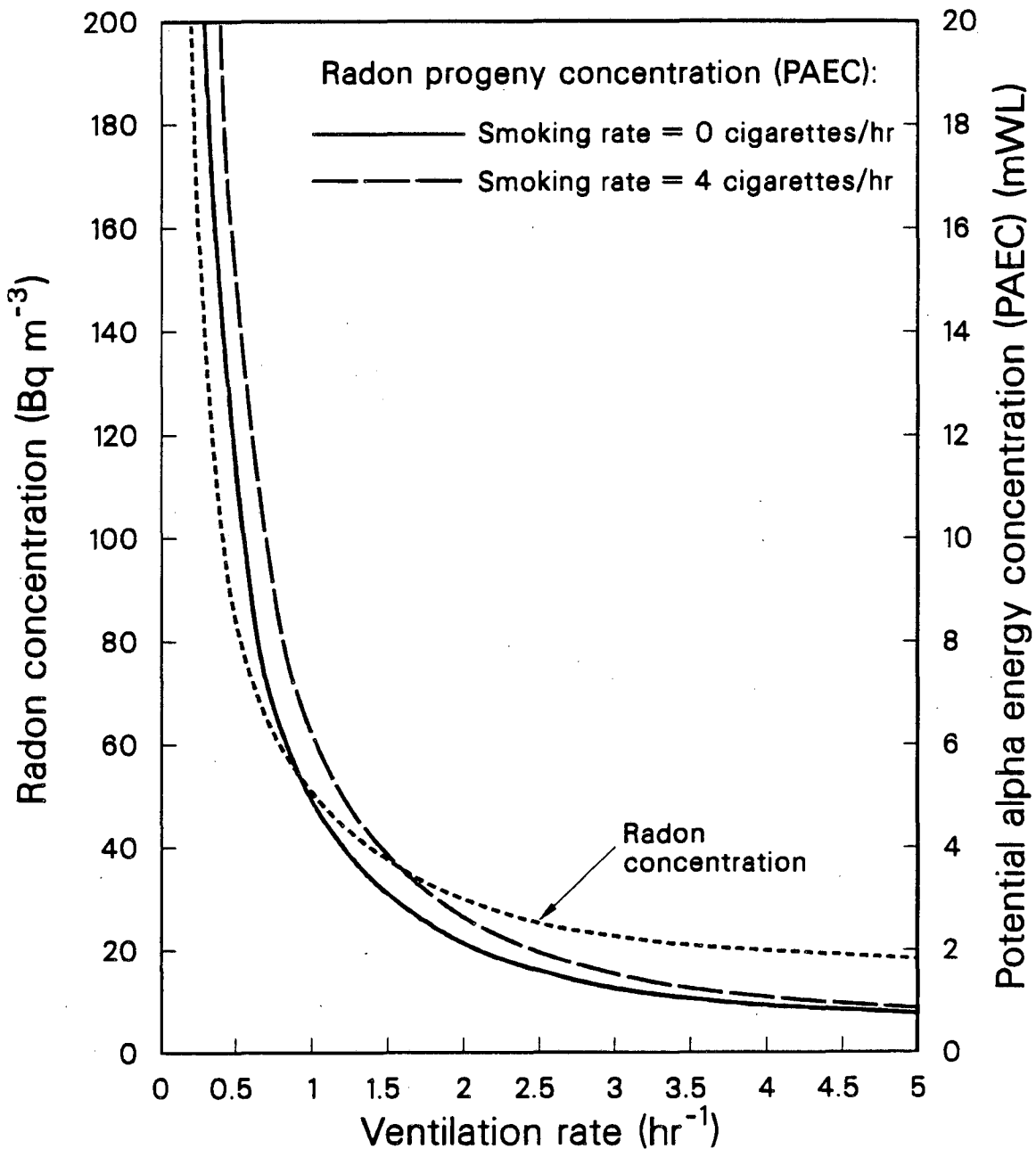
XBL 853-10395

Figure 1. Radon decay chain. The nuclides ^{218}Po , ^{214}Pb , and ^{214}Bi are of primary radiological concern due to inhalation and subsequent alpha decay.



XBL 853-10394

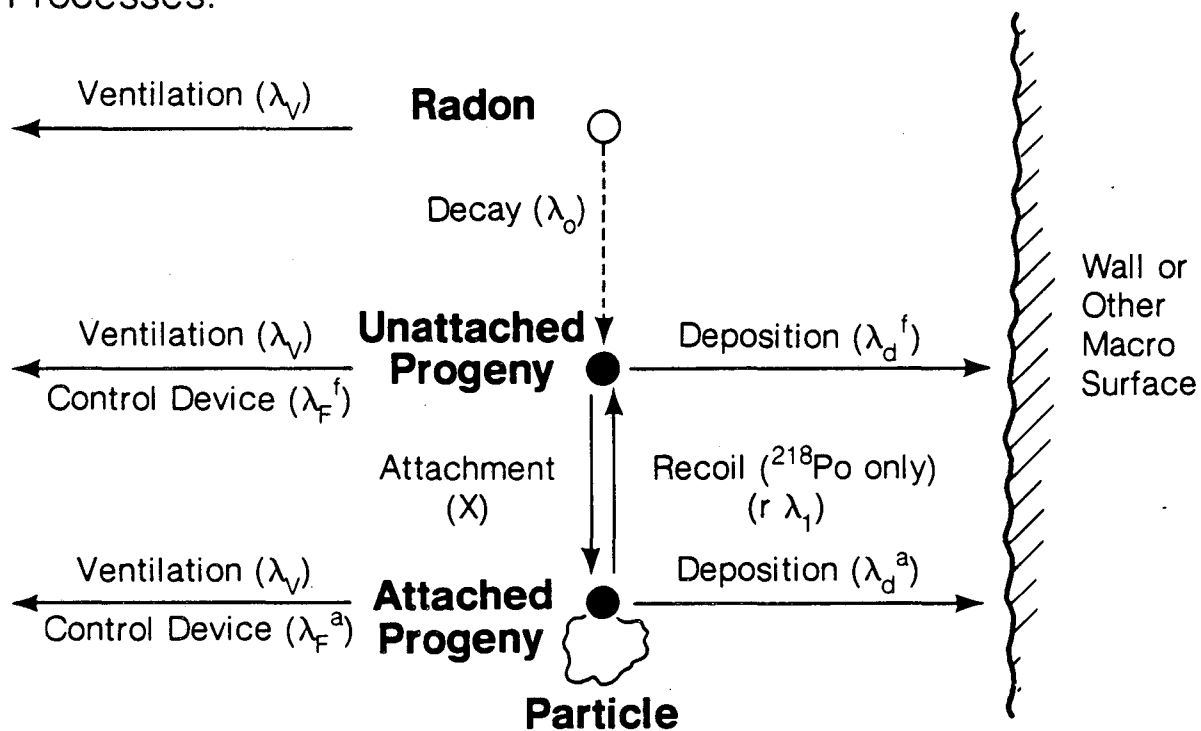
Figure 2. Pathways for radon entry into buildings.



XCG 854-202

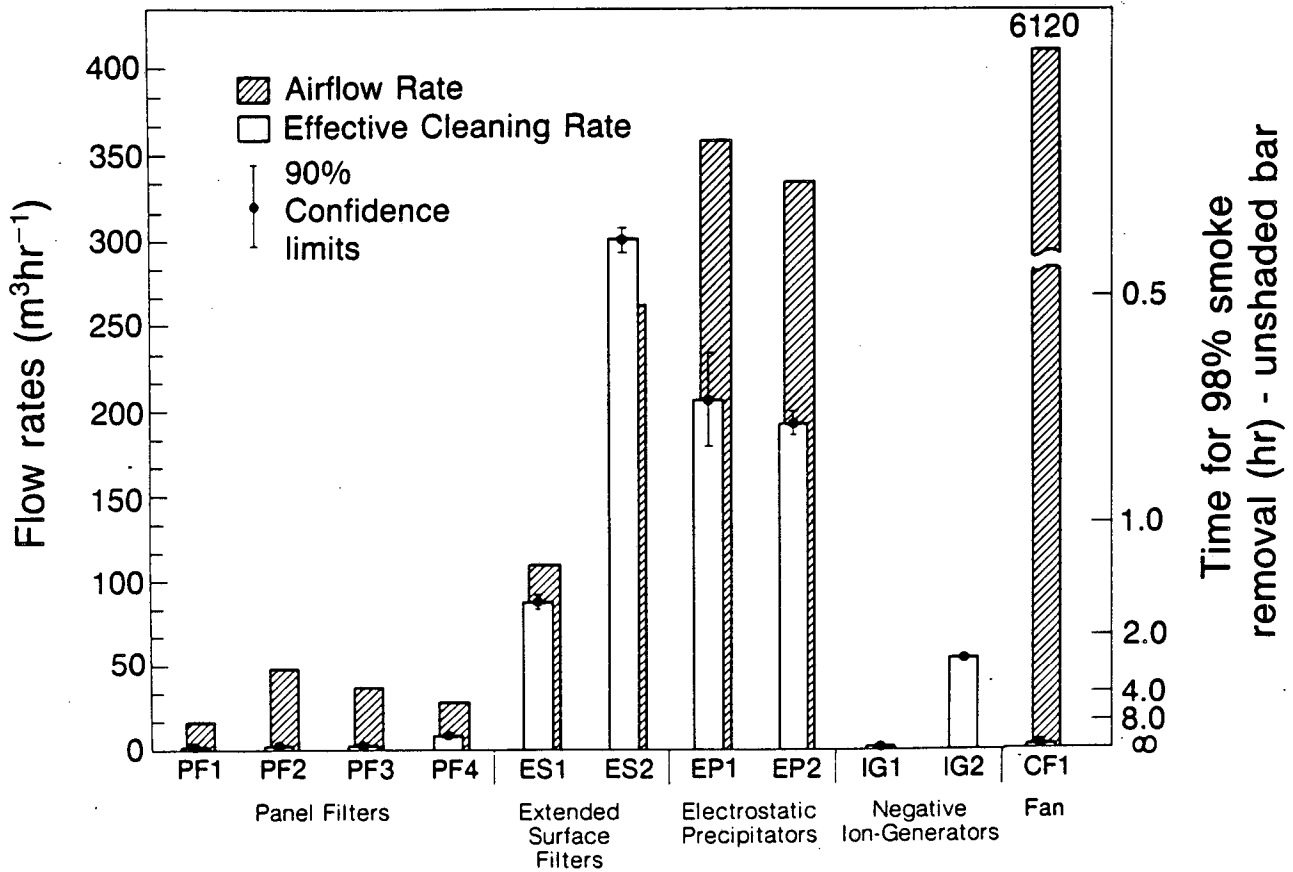
Figure 9. Radon and radon progeny concentrations versus ventilation rate. The calculated concentrations for both radon and radon progeny assume a constant radon source strength of $50 \text{ Bq m}^{-3} \text{ hr}^{-1}$. The PAEC curves are based on an assumed constant indoor source rate of 10^{12} particles hr^{-1} (from sources other than tobacco combustion) and infiltrating outdoor air with $20,000$ particles cm^{-3} and a penetration factor of 0.5 . For the case with cigarette smoking, each cigarette produces 9×10^{12} particles.

Other Removal Processes:



XBL 8311-647A

Figure 4. Schematic diagram of the removal processes (and their associated rates) affecting concentrations of radon and radon progeny. The radioactive decay pathways for radon progeny are not explicitly noted in this diagram.



XBL 8310-3343B

Figure 5. Effective cleaning and air flow rates for several types of unducted air-cleaning devices. The right axis indicates the time required (in hours) for removal of 98% of the airborne particles for each device (unshaded bar) operating in the 96-m³ chamber.

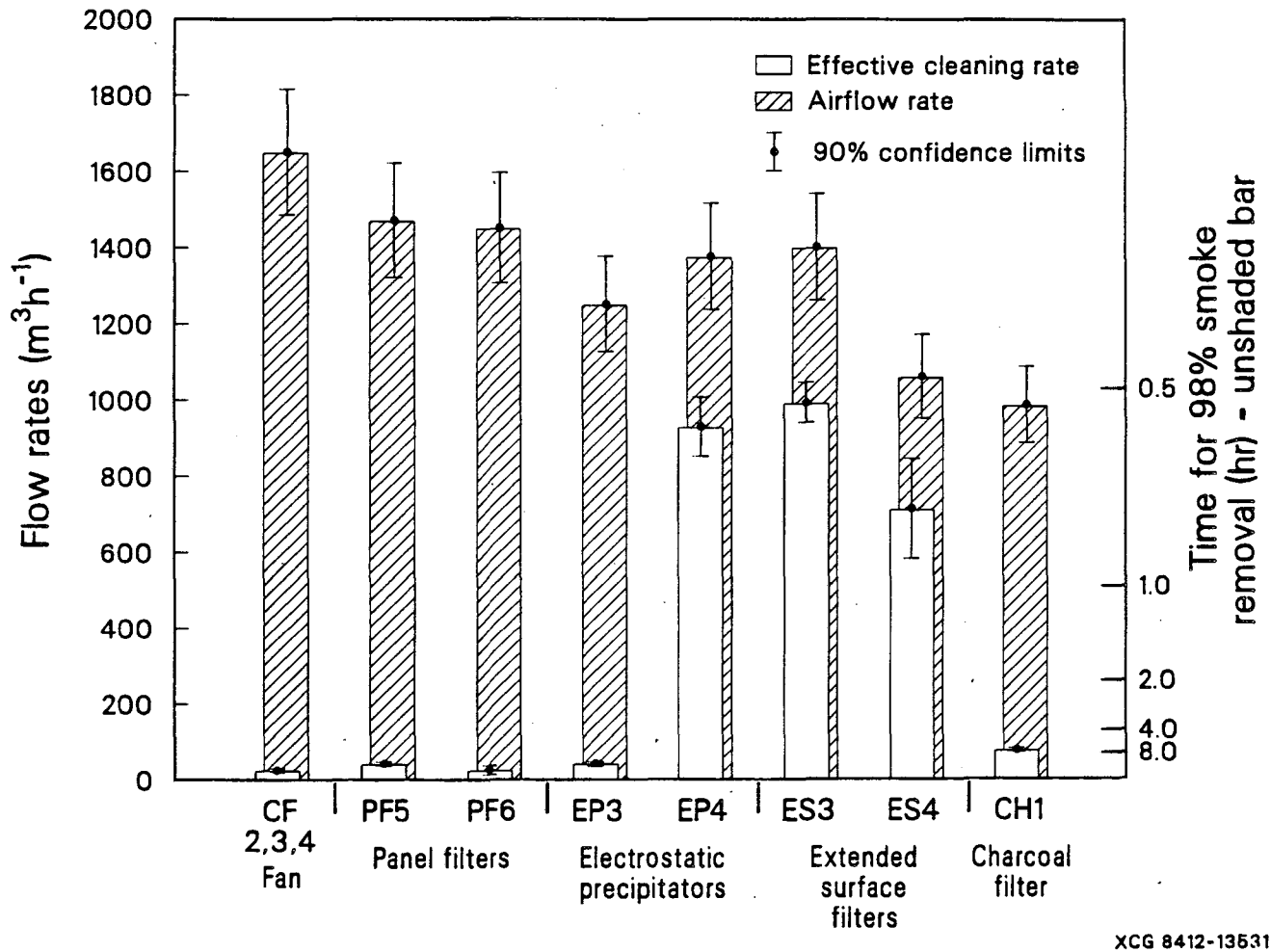
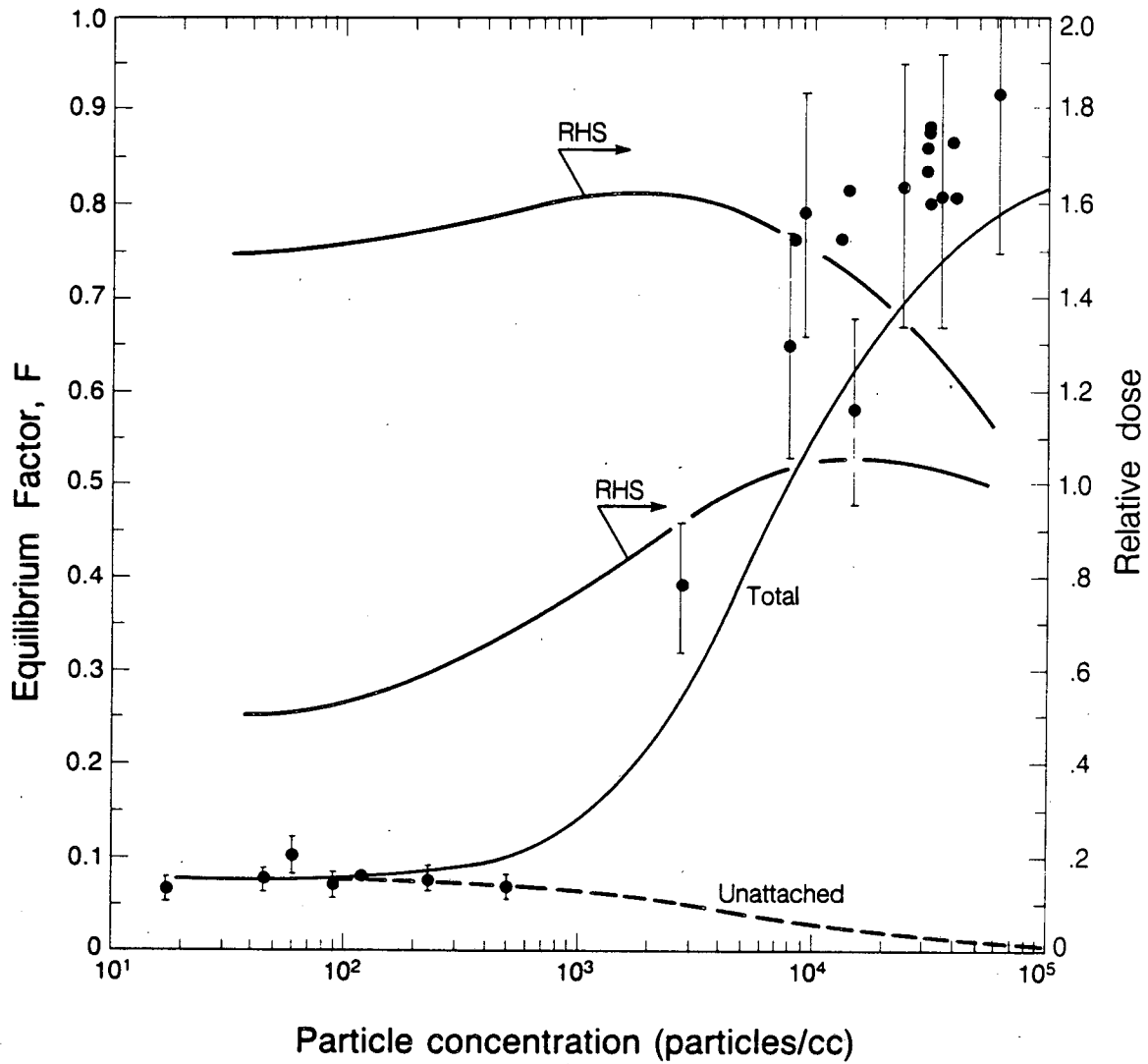
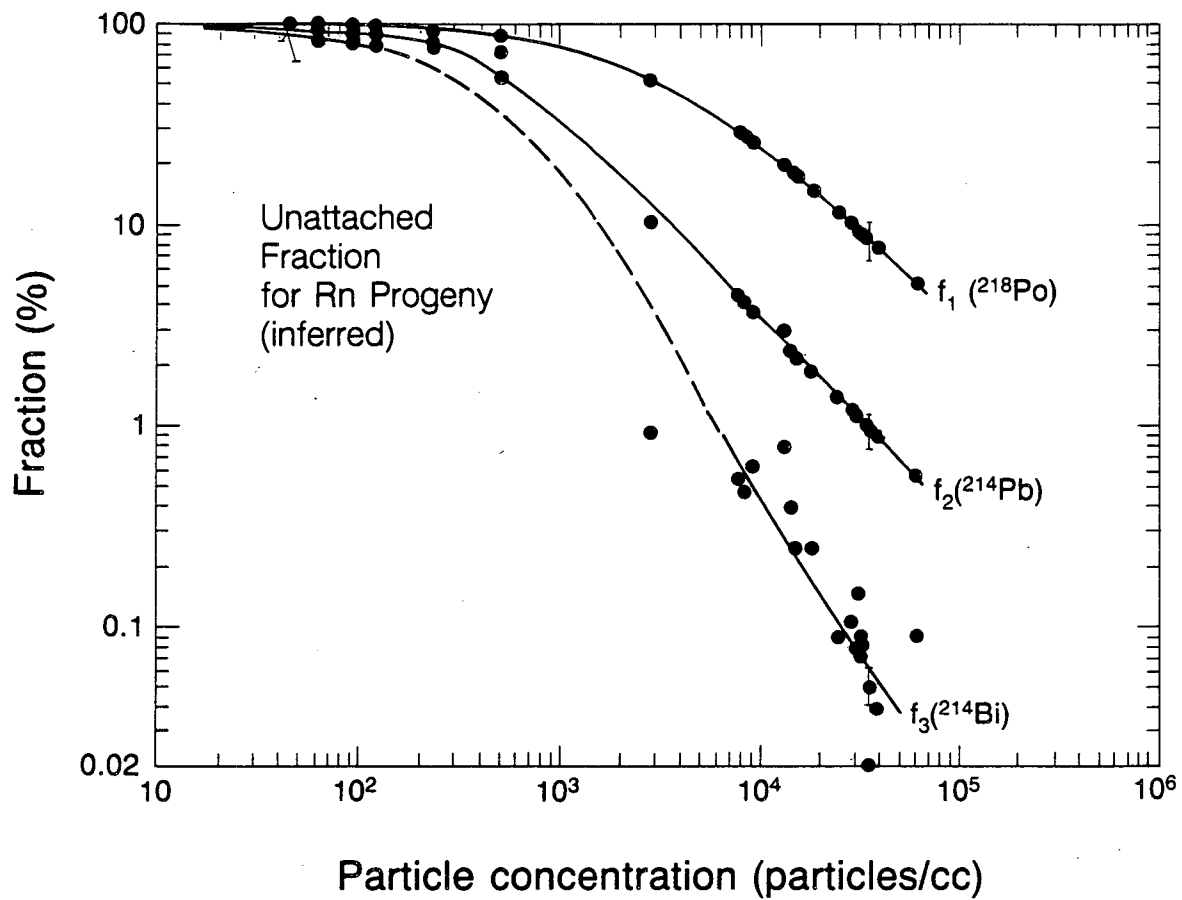


Figure 6. Effective cleaning and air flow rates for several ducted air-cleaning units. As in Figure 5, the right axis indicates the time required for 98% reduction in particle concentration for each device operated as part of the forced air system in the 140-m^3 test space.



XBL 8312-6710C

Figure 7. Equilibrium factor, F , versus particle concentration. Measured data and uncertainties are indicated by the solid circles and error bars. The solid line labeled *Total* represents calculated values for total airborne radon progeny, while the dashed line shows calculated values for unattached progeny. The two curves referring to the right hand scale (*RHS*) 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. The calculations further assume that at 10^5 particles cm^{-3} the progeny are all attached to airborne particles, and thus the relative dose is unity.



XBL 8312-6707A

Figure 8. Inferred values for the unattached fraction of ^{218}Po , ^{214}Pb , and ^{214}Bi as a function of particle concentration. The lines through the data serve to guide the eye.

This report was done with support from the Department of Energy. Any conclusions or opinions expressed in this report represent solely those of the author(s) and not necessarily those of The Regents of the University of California, the Lawrence Berkeley Laboratory or the Department of Energy.

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