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Evaluation of the Effectiveness of Several Air Cleaners for Reducing the Hazard from Indoor Radon Progeny

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In studies conducted in the early 1980s, it was suggested that the use of room-type air cleaners were relatively ineffective in reducing the hazards associated with the presence of radon decay products in indoor air. Some studies suggested that air cleaning could actually increase the dose delivered by the decay products by shifting the activity-weighted particle size distribution to smaller sizes to the point where dose increase even though exposure decreased. A recently developed automated, semicontinuous instrument now permits the direct measurement of activity-weighted size distributions in occupied homes so that the exposure to those occupants can be directly determined and the effect of the air cleaners on dose be estimated. Three different types of air cleaners were tested in this study; an electrostatic air cleaner (EAC), an ion generator/fan system (IG/F), and a filtration unit that was operated at two different fan speeds (LO-FIL and HI-FIL). The three units reduced the median exposure to radon progeny by 63% for the EAC, 34% for the IG/F unit, 66% for the filtration system at both low and high flow rates. Based on a model for lung dosimetry, the median reductions is estimated dose to the secretory cells of the bronchial epithelium are 50% for the EAC, 28% for the IG/F unit, and 68% for the filtration system at both fan speeds. In these studies, the performance of the EAC and filtration systems produce rather similar activity weighted size distributions while the lower dose reduction for the IG/F system is due to a small peak in activity in the size range of 1.5–5 nm. The source of this peak is not known, and requires further study to determine if the system is generating particles in this size range.

INTRODUCTION

Radon-222, an inert gas, belongs to the uranium-238 series of naturally occurring radionuclides that decay to stable lead-206 through eight alpha-ray and six beta-ray transitions. The first four radon progeny, polonium-218 (RaA), lead-214 (RaB), bismuth-214 (RaC), and polonium-214 (RaC′), are referred to as the short-lived decay products of radon because each has a half-life < 30 min. Lead-210 is the effective end decay product of the radon series. With a half-life of 22.3 years, its low specific activity effectively terminates the radon decay chain for most practical human exposure considerations. These decay products rather than radon itself are the active species responsible for the majority of the health hazard due to the release of ionizing radiation during their decay.

It has become recognized that inhalation and lung deposition of the decay products of radon-222 produce adverse health affects. The increasing radon concentration as well as the prolonged exposure periods related to indoor habitation make indoor radon a potential hazard. The U.S. Environmental Protection Agency (EPA) estimates that approxi-

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mately 13,000 lung cancer deaths per year in the United States may be a result of prolonged indoor radon exposure (EPA, 1992). This estimate includes the revised dosimetry proposed by the National Research Council (NAS/NRC, 1991) as well as the results of the national radon survey that EPA has now conducted (Marcinowski, 1992). Therefore, effective methods for lowering the risks from radioactivity in indoor air must be developed and thoroughly evaluated.

A concept that is important to the understanding of the health effects of the decay products of radon is that of the "unattached" fraction. It was first suggested in 1956 that there was a highly diffusive form of the radon and thoron decay products that could readily deposit in the respiratory system (Chamberlain and Dyson, 1956). This part of the size distribution has been referred to as the "unattached fraction." There are problems with the use of this term (Hopke, 1992) and it is a better practice to consider the actual particle size of the activity rather than combining them into an operationally defined size category. The decay products in the smallest size range have much higher mobilities in the air and can more effectively deposit in the respiratory system. Thus, for a long time the "unattached" fraction has been given extra importance in estimating the health effects of radon decay products. However, it should be noted that there are no strict definitions of exactly what size range of activity constitutes the "unattached" fraction (Hopke, 1992).

Previous investigations have demonstrated that these air cleaning systems can effectively remove radon decay products from indoor air. Reductions in the activity concentrations can be in the 60–80% range. At the same time, the airborne particles are also removed from the air. As a result, the "unattached" fraction increases in most of the cases. Although there are fewer decay products, they may be more effective in depositing their radiation dose to the lung tissue. The reductions in dose are, therefore, always smaller than reductions in the potential alpha energy concentration. In fact in some cases, the dose would be predicted to increase in spite of a lower airborne radioactivity concentration. These studies have been reviewed in detail by Hopke et al. (1990).

Two major problems can be identified in the previous investigations. The first one is that the measurement systems were not able to determine the full size distribution of radon progeny, especially in the range < 10 nm. Only estimates of the "unattached" fractions were made. However, the assumptions for defining "unattached" fraction in previous measurements are not always consistent with each other or with the definitions used in the dosimetric models, especially regarding the value of the diffusion coefficient assigned to the "unattached" fraction. Since the exact size of the most diffusive activity is important to its nasal penetration and bronchial deposition, the measurement of the actual size is critical to the full evaluation of the effects of air cleaning on dose.

The second problem is that the dose estimates in most of the studies have been made based on very simple lung models. In addition, the dose estimations have been made with different dose models without always reporting the results in such a way that newer models can be applied as they become available. Thus considerable differences in the resulting dose estimates have been obtained that make it difficult to compare one study or one cleaner with another. The important factor for these big discrepancies is that all of the models have much higher dose factors for "unattached" than for "attached" progeny but the actual coefficients vary significantly. Recent theoretical investigations showed that the dose conversion factor is strongly dependent on the actual activity size. The experimental separation of "unattached" and "attached" fractions in these prior studies is therefore insufficient to provide accurate dose estimation.

To determine the desirability of using air cleaners to mitigate the health effects from radon decay products in indoor air, studies are needed in which the concentrations and the full size distributions of the radon progeny activities are measured where air cleaners are employed in realistic environments. In this way the dosimetric implications of the changes in size distributions that are induced by air cleaning can be fully evaluated.

MEASUREMENT SYSTEM

The activity weighted size distribution was measured with the automated, semi-continuous graded screen array (ASC-GSA) described by Ramamurthi (1989) and Ramamurthi and Hopke (1991). The ASC-GSA measurement system involves the use of combination of six sampler-detector units operated in parallel. Each sampler-detector unit couples wire screen penetration, filter collection and activity detection with a solid state detector in a way as to minimize depositional losses. The system samples air simultaneously in all of the units, with a flow of about 15 L/min through the sampler slit between the detector and filter holder section in each unit. The sampled air is drawn through a Millipore filter (0.8 μm, Type AA). Complete details of the sampler are provided by Ramamurthi and Hopke (1991).

The computer control of sampling, counting, and analysis permits automated, semicontinuous operation of the system with a sampling frequency between 1.5 and 3 h. The activities of each radon progeny are estimated from alpha spectra collected during two counting intervals: the first one during sampling and the second 20 min after end of sampling. The observed concentrations of ^{218}Po, ^{214}Pb, and ^{214}Bi are used to reconstruct the corresponding activity-weighted size distributions using the Expectation-Maximization algorithms (Maher and Laird, 1985). The ASC-GSA system allows the determination of the activity weighted size distributions in six inferred size intervals in geometric progression within the 0.5–500-nm size range. The performance of the ASC-GSA system was tested in field (Hopke et al., 1991) intercomparison study with manual screen measurements showing very good agreement with systems from other leading laboratories in terms of the activity size distributions. However, small differences in the partitioning of activity in the 1.5–15-nm range led to poorer agreement in the calculated dose estimates. Although there may be uncertainties in the absolute values of dose, the relative values for the day-to-day changes and the effects of the air cleaners can be relied on.

In addition to the size distribution for each of the individual decay products, the total airborne activity concentration can be characterized by the potential alpha energy concentration (PAEC). The PAEC can be calculated from the individual progeny concentrations by

$$\begin{aligned}\mathrm{PAEC}(\mathrm{mJm}^{-3}) = 5.79\times10^{-7}\cdot c_1 \\ + 2.86\times10^{-6}\cdot c_2 \\ + 2.10\times10^{-6}\cdot c_3, \quad (1)\end{aligned}$$

where c_1, c_2, and c_3 are the activity concentrations of the three radon decay products in bequerels per cubic meter. The potential alpha energy concentration of any decay product mixture can also be expressed in terms of the so-called equivalent equilibrium concentration (EEC) of the radon. The EEC is the activity concentration of ^{222}Rn in radioactive equilibrium (equal activities) with its short-lived decay products which has the same potential alpha concentration as the actual nonequilibrium mixture.

$$EEC(Rn) = 1.81 \times 10^5 \cdot PAEC. \quad (2)$$

The value of 1.81×10^5 Bq mJ^{-1} is derived from the inverse of total energy in millijoules of the alpha particles emitted in the decay sequence of the radon progeny per bequerel of radon. The relationship between the concentration of ^{222}Rn and the potential alpha energy concentration is described by the equilibrium factor, F, given by

$$F = \frac{EEC(Rn)}{c_{Rn}} = \frac{1.81 \times 10^5 \cdot PAEC}{c_{Rn}}, \quad (3)$$

where c_{Rn} is the radon concentration in bequerels per cubic meter.

Since air cleaners will remove the decay products and not the radon, the effectiveness of the air cleaner in reducing exposure can be observed from the change in F value. If the radon concentration decreases because of changing conditions in or around the home, so will the decay product concentrations irrespective of the presence or absence of an air cleaner. Therefore, it is important that the measures of the air cleaner effectiveness be referenced to the radon concentration since the radon concentration will vary.

Another quantity, the unattached fraction, that has historically been reported is the fraction of activity in the smallest particle size range. The problem arises that the unattached fraction has never been clearly defined based on a specific range of particle sizes and thus, has been operationally defined (Ramamurthi and Hopke, 1989). Hopke (1992) has suggested that the term unattached be reserved for the size range < 2 nm that only includes molecules and clusters. In this study, the unattached fraction will be that portion of the size distribution that is found in our smallest size bin in the size distribution, 0.5–1.5 nm.

Dosimetric Model

In the most recent dose model developed by James (1992) and adopted by the National Research Council's Panel on the Dosimetric Extrapolation of the BEIR IV Risks to the General Public (NAS/NRC, 1991), particle size has been taken into consideration. The basal cell and the secretory cells in the bronchial epithelium were considered as target cells. The resulting dose conversion factors per unit exposure from monodisperse activity D_i, are presented in Figure 1 as a function of breathing rate.

The graph shows the dose to basal and secretory cells for three different breathing rates equivalent to sleeping (0.45 m^{3}h^{-1}), resting (0.54 m^{3}h^{-1}), and light activity (1.5 m^{3}h^{-1}). For all cases, the conversion factor is strongly dependent on the diameter especially for particles < 10 nm. Therefore, to calculate the dose per unit exposure to the cells at risk, the following formula applied:

$$\frac{D_s}{E_p} = \sum_{i=1}^{i=n} f_i D_{si}, \quad (4)$$

where E_p is exposure to PAEC [mJ h m^{-3}], D_s is total dose to secretory cells [Gy], D_{si} is dose to secretory cells per unit exposure to PAEC with size i [mGy m^3 mJ^{-1} h^{-1}], f_i is fraction of activity with size i, and n is number of size ranges considered.

A similar expression applies for the basal cells. Thus, any action influencing the physical parameters of indoor aerosols should be considered very carefully from the point of view of possible health risk. Because the major effect of any air cleaning system on the radon decay products in indoor air is the alteration of the activity

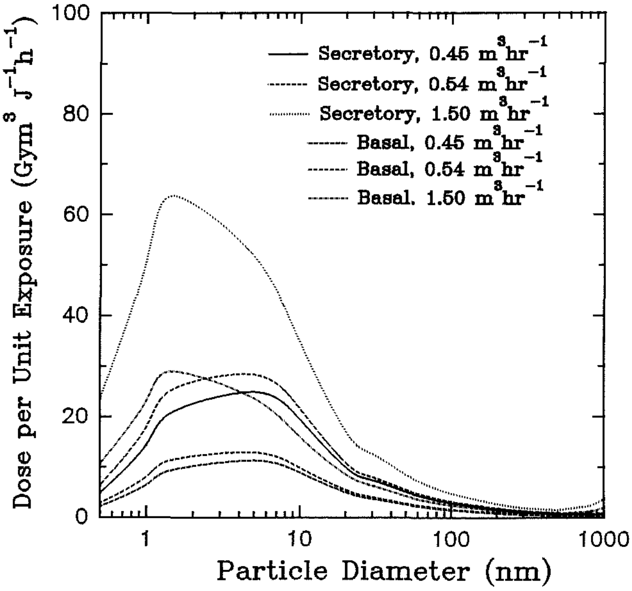

FIGURE 1. Dose per unit exposure to potential alpha energy concentration as a function of particle size (James, 1992; NAS/NRC 1991).

size distribution by reducing the particle concentration, the evaluation of such systems on the dose per unit exposure is of greater utility that the evaluation of the concentration reduction alone.

EXPERIMENTAL DESIGN

The measurements of the activity size distributions were made in Arnprior, Ontario where the occupants lived normally in their home during the period of the study. Arnprior is a community of 7000 inhabitants about 65 km northwest of Ottawa, Ontario. Prior experiments on the air filtration unit and an electrostatic air cleaner were performed in a single family home in Northford, CT (Li and Hopke, 1991a). In Arnprior, the house has a basement with an approximate area of 200 m^2 and the first floor has an area of about 210 m^2. A floor plan for this home is shown in Figure 2. The house was occupied by three people none of whom smoke. The presence of occupants that smoke cigarettes can have a significant effect on the size distributions, and exposure/dose relationship (Wasiolek et al., 1992). Measurements were made from May to July 1991. No heating or air conditioning was used during this period. It was not possible to measure condensation nuclei concentrations in these experiments. The ventilation rate was measured by the static decay of SF_6 using a MIRAN 1B2 portable internal reflection spectrometer and was found to be 0.49 ± 0.02 air changes per hour. The sampler and air cleaners were placed at the dining room end of the kitchen/dining area (23 m^2).

Three types of air cleaners were investigated: an ionization, filtration, and air circulation system (NO-RAD); an electronic air cleaner (EAC) with fan; and a multistage filtration unit with fan (Pureflow). The filtration unit and the EAC are described in detail by Li and Hopke (1991a). The NO-RAD system is described by Moeller et al. (1988). The details of the experiments are outlined in Table 1.

The air cleaner experiments were initiated in May 1991 in which almost 7 weeks of data have been collected; 3 full

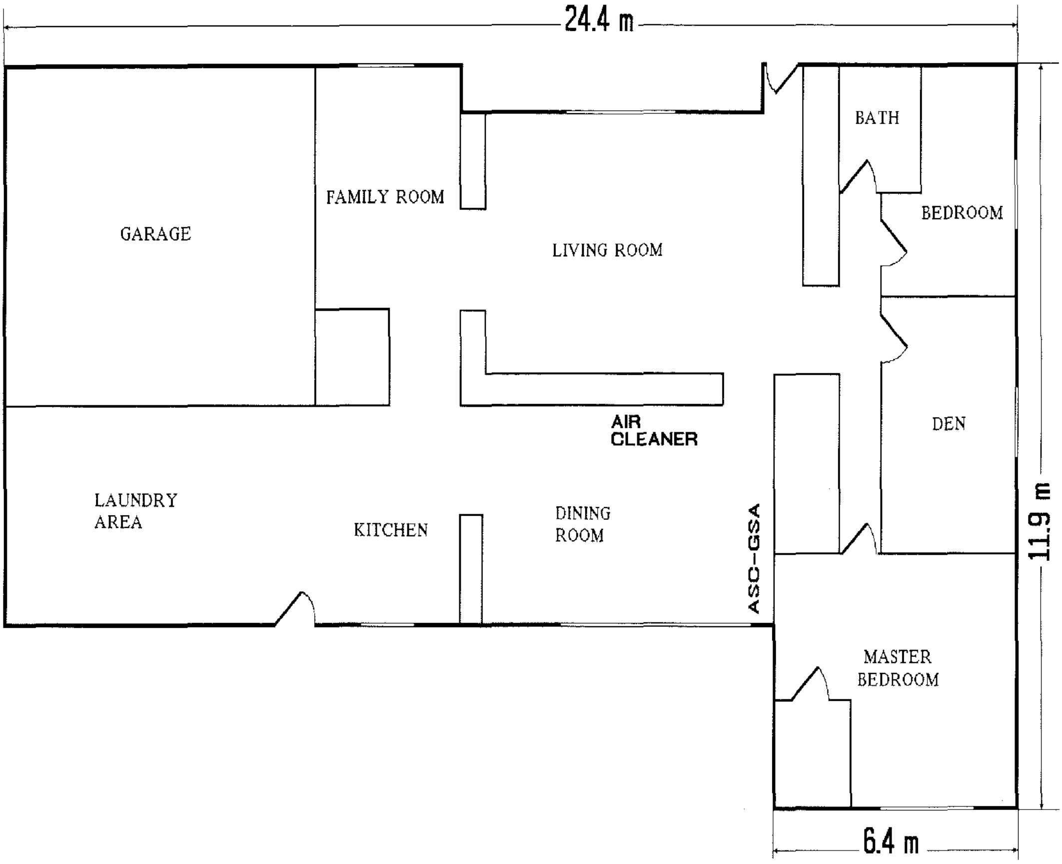

FIGURE 2. Floor plan of the first floor of the house in Arnprior, Ontario.

weeks of background exposure data as well as a few additional days at the end of the experiments, and 1 week each in which an ion generator/circulation fan system, the electrostatic air cleaner, and the filtration unit were operated. The EAC was operated at a flow rate of 3.86 m^3 min^{-1}. The NO-RAD was operated at its lowest fan speed which is approximately 2.83 m^3 min^{-1}. The filtration unit was initially set

TABLE 1. Experimental Conditions for the Air Cleaner Studies in Arnprior, Ontario

Exp.	Sampling period	Number of samples	Conditions
1	May 13–21	77	Background
2	May 21–28	50	NO-RAD ON
3	May 28–June 3	54	Background
4	June 3–10	77	EAC ON
5	June 10–17	69	Background
6	June 28–30	26	Pureflow ON (High Speed)
7	June 30–July 5	52	Pureflow ON (Low Speed)
8	July 5–6	11	Background

for its highest flow rate (4.25 m^3 min^{-1}). However, the occupants found the situation to be uncomfortable and then turned it down to 2.26 m^3 min^{-1}. Thus, there are separate sets of measurements made for each of these two different filtration unit fan speeds.

Radon concentrations were measured using a Pylon AB-5 monitor with a passive radon cell. Preliminary results of these experiments have been presented by Montassier et al. (1992a, b).

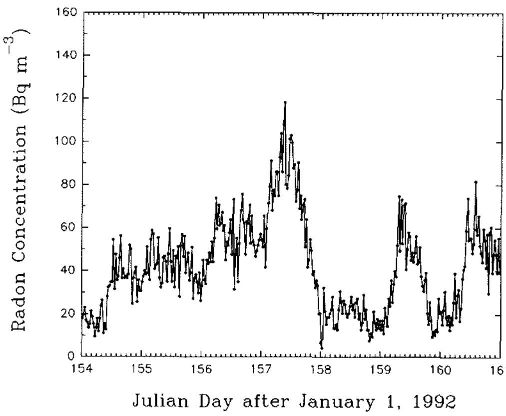

FIGURE 4. Radon concentration plotted as a function of time for the 4th week of the Arnprior house experiments.

RESULTS AND DISCUSSION

The frequency distribution for the radon concentration is given in Figure 3. Over 2400 radon measurements were made. To examine the radon variations somewhat further, 1 week of radon concentrations are plotted in Figure 4 and 2 days of data from that week are plotted in Figure 5. The diurnal variations often showed a peak concentration from the morning to the late afternoon. During this period, no heating or air conditioning was employed in the house. On the basis of modeling studies, Arvela et al. (1988) suggested that diffusion sources would contribute to the afternoon maximum when ventilation is at a minimum and pressure-driven flow contributes to an early morning maximum. However, it can also be hypothesized that the sun heated the air in the attic, causing expansion and a stack effect from the top of the house down. Thus, the maximum radon concentrations were during the day

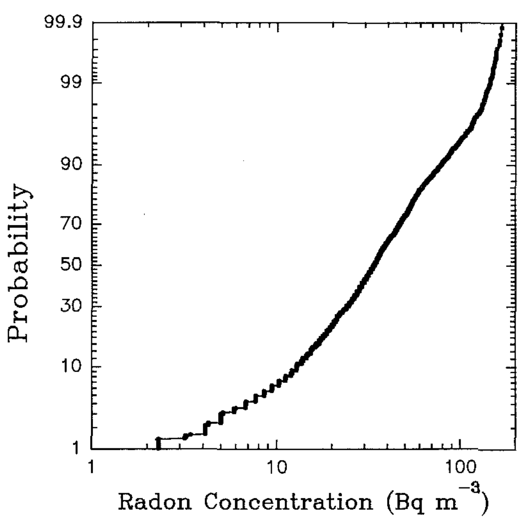

FIGURE 3. Cumulative frequency distribution for ^{222}Rn in the Arnprior home from May to July 1991.

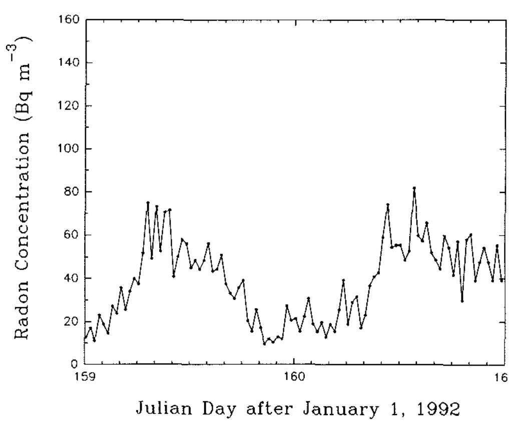

FIGURE 5. Radon concentrations in the Arnprior house during 2 days of the 4th week of experiments.

were rather than the more commonly observed maximum during the night when heating the house causes a bottom up stack effect.

The average activity size distributions for each of the experimental situations are presented in Figure 6. The average size distributions rather similar with the most notable differences occurring in the size ranges of 1.5–5 and 5–15 nm. In the distribution for the NO-RAD system, there is a noticeable peak in the distributions for PAEC and the longer lived decay products in the range of 1.5–5 nm. There are definitely lower fractions of all of the decay products over the range of 1.5–15 nm for the filtration system at the highest flow rate. The EAC and the filtration system at medium speed show activity-weighted size distributions that are quite similar to the average background distributions.

The PAEC reduction for each of the situation can be summarized by comparing the cumulative frequency distributions of the equilibrium factor for the periods when the air cleaners are working with the background distribution. Figure 7 shows the equilibrium factor distributions for each experimental condition. It can be seen that each of the air cleaners reduces the PAEC per unit radon relative to the background conditions. The reduction in median value for the EAC is 63%, 34% for the ion generator unit, and 66% for the filtration system at both low and high flow rates.

The filtration system curves deviate from one another at the higher end of the distributions where the higher flow rate is

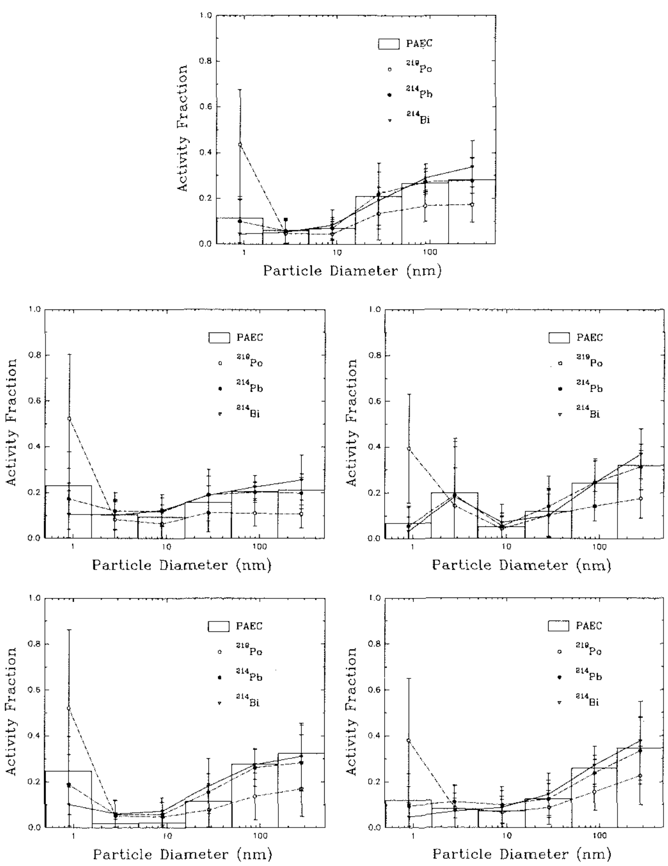

FIGURE 6. Average activity-weighted size distributions obtained during background periods (*top center*); with the EAC operating (*middle row, left*); with the NO-RAD operating (*middle row, right*); with filtration system, high speed (*lower row, right*); and with filtration system, low speed (*bottom row, right*).

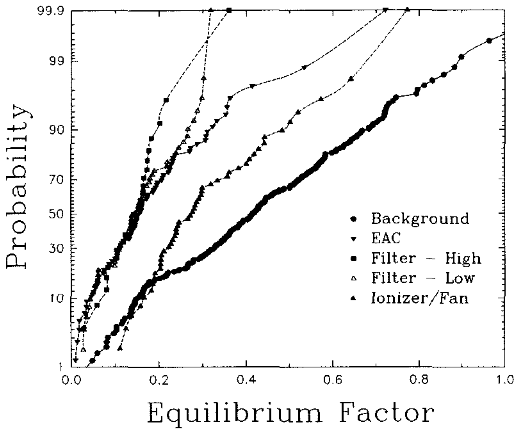

FIGURE 7. Cumulative frequency distributions for the equilibrium factors for each condition: background, filtration unit, electrostatic air cleaner, and ion generator.

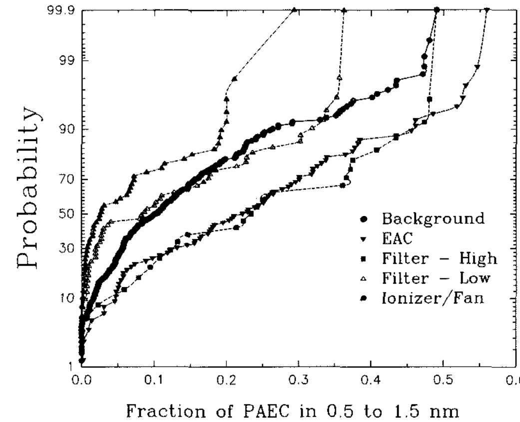

FIGURE 8. Cumulative frequency distributions for the fraction of PAEC found in the range of 0.5–1.5 nm for each of the experimental conditions.

more effective at reducing the exposure to radon progeny. The EAC also works as effectively at reducing exposure for the lower exposure conditions and is less effective than the filtration unit at the higher end of the distribution. The previous study (Li and Hopke, 1991a) has found the filtration unit to be significantly more effective than the EAC. That study was conducted with the fan always on high speed and in a home with a radon concentration about five times that present in the Arnprior home although the radon concentration is not expected to influence the performance of either of these cleaners. The NO-RAD system has consistently lower removal efficiency for the radon decay products and produced the peak in the average activity-weighted size distribution in the range of 1.5–5.0 nm.

To examine the effects of the air cleaners on the unattached fractions of PAEC and ^{218}Po, the cumulative frequency distributions for these quantities are plotted in Figures 8 and 9. The ionization/fan system reduced the fraction in the range of 0.5–1.5 nm, but increased the fraction in the next larger size interval. It may be that the ionizing units are creating particles in this size range. Further study of this observation is needed to determine what has caused the observed changes in the size distributions.

There are differences between Figures 8 and 9 showing that the cleaners have more effect on the unattached fraction of PAEC than on the corresponding fraction of ^{218}Po. The distributions for unattached

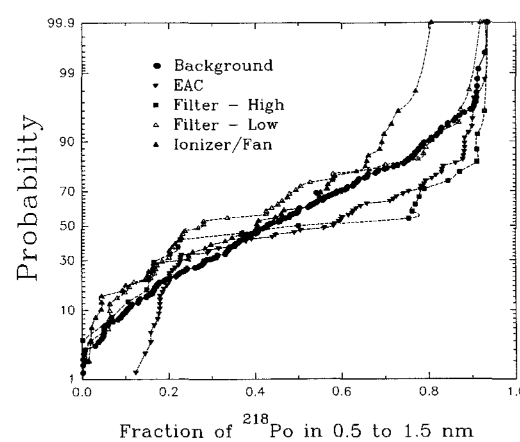

FIGURE 9. Cumulative frequency distributions for the fraction of ^{218}Po found in the range of 0.5–1.5 nm for each of the experimental conditions.

^{218}Po fractions are quite similar to one another with median values in the range of 0.4–0.5 nm. These results suggest a low particle concentration in the air coming into the room. Since this is an all electric home in a rural setting with non-smoking occupants, it is reasonable that there would typically be low particle concentrations. At low particle concentrations, deposition on the walls and other macrosurfaces can be significant in removing unattached ^{218}Po (Li and Hopke, 1991b; 1992).

There are increases in the unattached PAEC when the EAC and filtration system (high speed) were operating. At low speed, the filtration unit had little effect on the unattached PAEC and the NO-RAD reduced the unattached fraction. These results suggest that the NO-RAD has a quite uniform removal efficiency over the whole activity range. For the other systems, they appear to be more effective at removing particles than unattached progeny leading to increased unattached fractions of PAEC.

Figure 10 presents the cumulative frequency distributions for the estimated dose to secretory cells. A set of analogous distributions can be obtained for the dose to the basal cells and will have a similar shape, but at lower estimated dose values. It can be seen that for virtually all cases the distributions for the air cleaners show lower dose than during the background conditions. Only the upper tail of the NO-RAD distribution exceeds that obtained during the background experiments. However, there are considerably more background data (over 3 weeks) compared with any of the cleaners (1 week). It is likely that the higher dose may be due to the extra activity observed in the 1.5–5-nm size range (Figure 6). Because of the high dose/exposure ratio for this size range, even a small fraction of activity leads to a substantial dose contribution. Moeller, the inventor of the NO-RAD, indicated that problems were likely with its effectiveness at these low radon concentration (D. Moeller, private communication, 1992). It is clearly less effective than in the chamber studies reported by Moeller et al. (1988). Thus, it will be important in subsequent experiments to determine the likelihood of observation of this peak, the effectiveness of the unit as a function of radon concentration, and the relationships of these factors to the overall exposure-dose reductions. The median reductions in estimated dose to the secretory cells of the bronchial epithelium are 50% for the EAC, 28% for the IG/F unit, and 68% for the filtration system at both fan speeds.

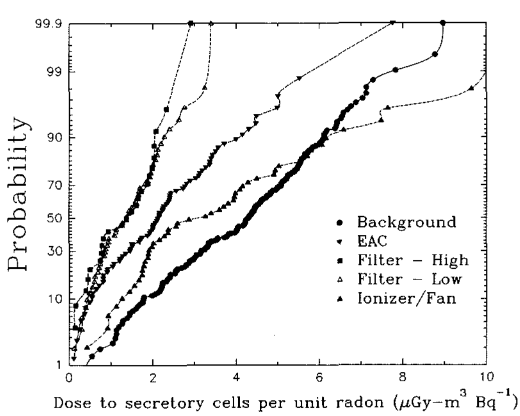

FIGURE 10. Cumulative frequency distributions for the estimated dose to secretory cells for each of the different experimental conditions in the Arnprior house.

CONCLUSIONS

In this home, both the filtration system and the electrostatic air cleaner provided substantial reduction in both exposure and dose. The filtration system was more effective at the higher flow rate for conditions in which higher exposures might occur although the median values for the exposure and dose reductions were essentially the same for both fan speeds. The ion generator/fan system was significantly less effective than the other systems or its performance in chamber studies. Further study is needed to investigate the occurrence of the small particle size peak in the activity distributions and the effect of radon concentration on the ion generator/fan system's effectiveness in radon progeny reduction. The reduction in exposure and dose observed in these studies suggests that these systems may have some utility in homes where the radon concentration is no more than two times the action level at which radon mitigation is recommended and could provide some assistance as an interim remedy while radon entry reduction techniques (i.e., subslab depressurization) are being constructed.

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