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Theoretical foundation for a simple method for simultaneous measurements of the unattached fraction and activity median diameter of attached radon progeny

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Abstract

Calculation of lung dose from established lung dosimetry models requires use of the unattached fraction of potential alpha energy concentration (PAEC) of radon progeny and the activity median diameter (AMD) of attached radon progeny, in addition to the total PAEC. In the present work, for indoor environments without the nucleation mode of aerosols, a method based on the wire screen penetration theory using two wire screens and a filter is proposed for simultaneous measurements of these two parameters. It is shown that the traditional wire screen method can overestimate or sometimes underestimate the unattached fraction, depending on the properties of the wire screen and the true unattached fraction. The present method eliminates such uncertainties.

Keywords: Radon progeny; Unattached fraction; Activity median diameter

1. Introduction

The unattached fraction of potential alpha energy concentration (PAEC) of radon progeny and the activity median diameter (AMD) of attached radon progeny are two of the key parameters in common lung dosimetry models; e.g., Jacobi–Eisfeld model (Jacobi and Eisfeld, 1980); James–Birchall (J–B) model (James et al., 1980); James model (James, 1988) and the National Research Council model (NRC model) (National Research Council, 1991).

The AMD can be measured using a variety of techniques, including direct measurement of the activity-weighted size distribution using cascade impactors supplemented with single screens or screen diffusion batteries (e.g., Porstendörfer, 1996). Alternatively, the number distribution of the aerosol can be measured using scanning mobility particle sizers followed by application of the Porstendörfer attachment coefficient formula (Porstendörfer, 1996). However, the equipment set-up and experimental procedures are not sufficiently handy and convenient to envisage their use in large-scale field surveys of the AMD. Therefore, in most cases, typical values of AMD are adopted.

The unattached fraction is commonly measured using wire screens. Early studies involving wire-screen measurements included those of James et al. (1972), Thomas and Hinchliffe (1972) and George (1972), while more recent investigations include those of Stranden and Berteig (1982), Bigu (1985), and Reineking et al. (1985). More recently, Ramamurthi and Hopke (1989) have demonstrated that the wire-screen method cannot separate the "unattached" and "attached" modes of radon progeny. A proposal was made for use of wire screens with 50% collection efficiency for particle diameter of 4 nm [$d_p(50\%)$], these wire screens collecting about 95% of the "unattached" mode while minimizing the collection efficiency of the "attached" mode to about

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1% and the "additional" mode due to cooking to less than 8%.

The unattached fraction of PAEC can be (although not necessarily) very small when compared to the attached fraction (e.g., 3 vs. 97% of the airborne alpha energy), and the strength of the "additional" mode, while known only imprecisely, can reach 15% of the airborne alpha energy (National Research Council, 1991). These quantities will still significantly affect measurement of the unattached fraction of PAEC even if we choose the optimum value of $d_p(50\%) = 4$ nm.

In the present work, a method based on the wire screen penetration theory is proposed using two wire screens and a filter to determine simultaneously the two parameters for lung dosimetry models: the unattached fraction of PAEC and the AMD of attached radon progeny.

2. Methodology

To provide a basis for present methodology, we refer to current understanding of the properties of radon progeny. Unless otherwise stated, the following particulars are extracted from a report of the National Research Council (1991) and references therein. It is assumed that the unattached mode has an activity median diameter (AMD) of 1.1 nm with a geometric standard deviation (GSD) of 1.5 [see for example, Birchall and James (1994)]. The attached mode for a typical indoor environment has an AMD of 150 nm with a GSD of 2.0, while that for an indoor environment with the presence of cigarette smoke as well as that for mine atmospheres has an AMD of 250 nm with a GSD of 2.5. These values of AMD and GSD are slightly different from those used by Ramamurthi and Hopke (1989), but the results should be qualitatively the same.

Castleman (1991) suggested the unattached radon progeny to contain between 5 and 8 water molecules. Although there are some arguments against the lognormal behavior of the unattached radon progeny based on the suggestion of Castleman (1991), the lognormal behavior has been generally accepted in the field of radon research (e.g., National Research Council, 1991; International Commission on Radiological Protection, 1994; Birchall and James, 1994; Porstendörfer, 1996; Yu and Guan, 1998; Yu et al., 1999; Porstendörfer and Reineking, 1999). If other types of distribution can be found in the future which better fit the behavior of unattached radon progeny, the formulations in the present paper can be easily modified to accommodate such changes.

Processes exist in an indoor environment (cleaning using a vacuum cleaner, cooking, using home heating systems, etc.) which can produce an additional mode (called the nucleation mode) with an AMD of around 20 nm. In the event of such processes, the theoretical formulations presented in this paper will not strictly apply. The main concern comes from home heating systems in which there can be leaky fireboxes that result in the release of particles in the nucleation mode each time the furnace fires. For other activities such as cleaning using a vacuum cleaner and cooking, the nucleation mode is transient with a lifetime of only a few hours. For the purposes of dose surveys, data gathering during transient events such as those listed above should be avoided.

In the following discussion, we will concentrate exclusively on indoor environments and the unattached and attached modes, i.e., we apply the formulations presented herein for indoor environments in which the nucleation mode is absent. Furthermore, we restrict present use of the methodology to typical indoor, non-industrial environments (such as residences or offices) without cigarette smoke, constituting the majority of sites for dose surveys. Derivations for an indoor environment with the presence of cigarette smoke and for a mine atmosphere are similar, but are beyond the scope of the present paper.

In the present methodology, two wire screens on top of a filter are housed in a sampler and air is drawn through them with a constant sampling face velocity. The top wire screen will be denoted by the subscript s1, the second wire screen by s2, and the filter by F. After sampling for a certain time, separate PAEC values in air are determined from the activities collected on the two wire screens and the filter, namely $PAEC_{S1}$, $PAEC_{S2}$ and $PAEC_F$. The total PAEC in air ($PAEC_T$) is given simply by $PAEC_T = PAEC_{S1} + PAEC_{S2} + PAEC_F$. Denoting the PAEC of the ith mode ($i = 1,2$ for unattached and attached modes, respectively) in air as $PAEC_i$, we have

$$PAEC_T = PAEC_1 + PAEC_2, \quad (1)$$

$$PAEC_{s1} = PAEC_1\, F_{11} + PAEC_2\, F_{21}, \quad (2)$$

$$PAEC_{s2} = PAEC_1\, F_{12} + PAEC_2\, F_{22}, \quad (3)$$

where F_{ij} is the fraction of the PAEC of the ith mode collected on the jth wire screen ($j = 1,2$ for wire screens s1 and s2, respectively; and $F_{i,2}$ represents the fraction of the PAEC of the ith mode in air collected on s2 after this mode has passed through s1). The unattached fraction of PAEC is then given by $PAEC_1/(PAEC_1 + PAEC_2)$. In practice, the activity measured for a wire screen should be corrected by the "front-to-total" (FT) activity ratio and the screen loss factor (SL) (Solomon and Ren, 1992; Solomon, 1997) to give the true activity collected by a wire screen (i.e., $PAEC_{S1}$ or $PAEC_{S2}$).

The fractions of PAEC of the unattached and attached modes collected on the jth wire screen, i.e., F_{1j} and F_{2j} respectively, can be determined using the wire screen penetration theory (Cheng et al., 1980; Cheng and Yeh, 1980) with the empirical correction of

Table 1
The values of F_{21} and F_{22} (scaled up by 10^3) for different AMD with GSD = 2.0 calculated for d_p (50%) = 3, 4 and 5 nm

AMD (nm)	3 nm F_{21}	3 nm F_{22}	4 nm F_{21}	4 nm F_{22}	5 nm F_{21}	5 nm F_{22}
130	8.296	8.160	12.030	11.750	16.030	15.530
140	7.622	7.508	11.060	10.820	14.740	14.320
150	7.049	6.953	10.230	10.030	13.640	13.280
160	6.558	6.476	9.518	9.345	12.690	12.390
170	6.133	6.061	8.902	8.753	11.880	11.610
180	5.761	5.699	8.365	8.234	11.160	10.930
190	5.435	5.380	7.892	7.776	10.530	10.330
200	5.145	5.097	7.473	7.370	9.975	9.793
210	4.888	4.844	7.100	7.008	9.478	9.315
220	4.657	4.618	6.765	6.683	9.033	8.886
230	4.450	4.414	6.464	6.390	8.632	8.499
240	4.262	4.230	6.193	6.125	8.270	8.149
250	4.092	4.062	5.946	5.884	7.941	7.831
260	3.937	3.910	5.721	5.664	7.641	7.540

Ramamurthi and Hopke (1989). Lognormal distributions are employed for the unattached and attached progeny. The unattached mode has less variation and is assumed to have an AMD of 1.1 nm with a GSD of 1.5 as mentioned above.

Under this distribution, values of (F_{11},F_{12}) are (0.8715,0.1013), (0.9394,0.05196) and (0.9707,0.02634) for d_p (50%) = 3, 4 and 5 nm, respectively.

On the other hand, the attached mode has larger variations in the AMD, and the AMD is required for common lung dosimetry models. Values of F_{21} and F_{22} are calculated for different values of AMD (from 130 to 260 nm in steps of 10 nm) and GSD = 2.0, which are shown in Table 1 for $d_p(50\%) = 3$, 4 and 5 nm. Linear regressions on these values give the following results:

$d_p(50\%) = 3$ nm:

$$F_{22} = 0.9756[0.9738, 0.9774] \times F_{21} + 7.408[6.399, 8.418] \times 10^{-5} \quad (R = 1.00000), \quad (4)$$

$d_p(50\%) = 4$ nm:

$$F_{22} = 0.9651[0.9628, 0.9673] \times F_{21} + 1.532[1.342, 1.723] \times 10^{-4} \quad (R = 0.99999), \quad (5)$$

$d_p(50\%) = 5$ nm:

$$F_{22} = 0.9534[0.9501, 0.9567] \times F_{21} + 2.734[2.368, 3.100] \times 10^{-4} \quad (R = 0.9998), \quad (6)$$

where the values in square brackets are the 95% confidence intervals and R are the correlation coefficients.

From measurements, $PAEC_{S1}$, $PAEC_{S2}$ and $PAEC_F$, and thus $PAEC_T$ are obtained. Given that F_{11} and F_{12} are fixed, and provided either F_{21} or F_{22} were known, then $PAEC_1$ and $PAEC_2$ could be solved by solving the simultaneous equations (1) and (2), or (1) and (3). However, since the AMD is unknown, both F_{21} and F_{22} are unknown. Our procedures in this situation are as follows. An initial value of AMD is assumed (say, 150 nm), and the corresponding F_{21} and F_{22} are read from Table 1. $PAEC_1$ and $PAEC_2$ can then be obtained by solving the simultaneous equations (1) and (2). These are then substituted into Eq. (3) to solve for a new F_{22}. If this new F_{22} is the same as the previous value, read from Table 1, then the value of AMD, and of F_{21} and F_{22} are correct, and the obtained values of $PAEC_1$ and $PAEC_2$ are the required values. However, if this new F_{22} is different from the previous value, then a new F_{21} is calculated from the suitable equation in Eqs. (4)–(6) and the new set of F_{21} or F_{22} is employed. In this way, a new set of $PAEC_1$ and $PAEC_2$ can be obtained by solving Eqs. (1) and (2). These are again substituted into Eq. (3) to solve for a new F_{22}. The iterations continue until the new F_{22} obtained from Eq. (3) in the present iteration is as same as that obtained in the last iteration. The AMD can be obtained from the final values of F_{21} or F_{22} using Table 1; the nearest AMD is chosen or interpolation can be used.

3. Example and dicussions

We employ the following example to illustrate the procedures and advantages of the present new method for determination of unattached fraction and AMD of attached radon progeny: $d_p(50\%) = 4$ nm, AMD = 190 nm, $PAEC_1 = 0.5$ mWL, $PAEC_2 = 9.5$ mWL. From these data, we have $F_{11} = 0.9394$, $F_{12} = 0.05196$, $F_{21} = 0.007892$, $F_{22} = 0.007776$. The PAEC values determined from measured activities on the two wire screens $d_p(50\%) = 4$ nm and the filter should then be $PAEC_{s1}=0.5447$ mW L, $PAEC_{s2}=9.985\times 10^{-2}$ mW L and $PAEC_F=10$ mW L-($PAEC_{s1}+PAEC_{s2}$) $=9.355$ mW L.

At the initiation of calculations we do not know the real AMD and we accordingly assume AMD = 150 nm, giving $F_{21}=0.01023$ and $F_{22}=0.01003$ from Table 1. Solving Eqs. (1) and (2), we have $PAEC_1=0.4761$ mW L and $PAEC_2=9.524$ mW L. When these PAEC values are inserted in to Eq. (3), we obtain a new F_{22} as 0.007882. From Eq. (5), the new $F_{21}=0.007904$. The new F_{21} is employed in Eq. (2) to obtain values for $PAEC_1$ and $PAEC_2$, which give another new F_{22} through Eq. (3). The fourth iteration yields new F_{21} and F_{22} and these are the same as those of the fifth iteration. These values accordingly represent the correct solution: $F_{21}=0.007899$, $F_{22}=0.007776$, $PAEC_1=0.50$ mW L and $PAEC_2=9.50$ mW L. The AMD is 190 nm from Table 1.

For the data in the last example, a traditional wire-screen method will report a $PAEC_{s1}/PAEC_T$ value of the unattached fraction of 5.4%. This is an overestimation of about 9%. Although this figure is not particulary large when compared to the typical uncertainty with which the counting of indoor airborne activity can be obtained (reaching up to ~10%), the result is still important in that we have arrived at a conclusion contrary to that of Ramamurthi and Hopke (1989). Ramamurthi and Hopke (1989) discussed the under-estimation of the unattached fraction measured using wire screens in terms of the inability of the wire screens to collect the entire unattached mode. Here, we have shown an overestimation (rather than an underestimation), being due to a net increase in the estimated PAEC caused by the extra collection of the attached mode.

Although the collection efficiency of the attached mode is low, the PAEC of the attached mode is much larger than that of the unattached mode (9.5 vs 0.5 in our example). As such, the collection of the attached mode has an effect on the collection of the unattached mode. From Eq. (2), overestimation exists because the true unattached fraction ($PAEC_1/PAEC_T$) $< F_{21}/(1-F_{11}+F_{21})$, which are 5.200, 14.433 and 31.765% for $d_p(50\%)=3$, 4 and 5 nm, respectively, for AMD = 150 nm. Overestimation exists in general for $d_p(50\%)=4$ and 5 nm, but both overestimation and underestimation can exist for $d_p(50\%)=3$ nm, depending on the true unattached fraction.

From the differences between F_{11} and F_{12} and from the above example, we note that the collection efficiency of the wire screen s2 is low compared to that of s1. This will lead to large uncertainties. To increase the collection efficiency of s2, two methods could be used in future studies. First, wire screens with different $d_p(50\%)$ could be used in our sampling system. Since a larger $d_p(50\%)$ corresponds to a larger collection efficiency, we could use a wire screen with a smaller $d_p(50\%)$ for s1 and a wire screen with a larger $d_p(50\%)$ for s2 to maximize the collection efficiency of s2. Second, multiple wire screens could be employed instead of a single wire screen for s2 (see, for example, Hopke et al. (1990)). The derivations for these two methods would be similar to those presented in the present work.

4. Conclusions

A simple method based on wire-screen penetration theory, using two wire screens on top of a filter and housed in a sampler is proposed for simultaneous measurements of two important parameters in common lung dosimetry models, the unattached fraction of PAEC and the activity median diameter (AMD) of attached radon progeny under indoor environments without the nucleation mode of aerosols. It has also been shown that the traditional wire screen method can both overestimate and underestimate the unattached fraction, depending on the properties of the wire screen and the true unattached fraction. The present method eliminates such uncertainties.

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