FIELD TESTS OF A RADON PROGENY SAMPLER
FOR THE DETERMINATION OF EFFECTIVE DOSE
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The International Commission on Radiological Protection (ICRP) recommends the use of a single conversion factor, derived from epidemiological studies of exposure to uranium miners, for the determination of the effective dose from inhalation of radon progeny. Dosimetric models of radon progeny inhalation predict that the dose conversion factors (DCF) are dependent upon the form of the radon progeny activity size distribution. The measurement of these activity size distributions is difficult and an alternative approach has been proposed. The so-called Effective Dosimeter uses a two-screen sampler with a collection efficiency matched to the particle size behaviour of the radon progeny DCF, as determined from the ICRP Human Respiratory Tract Model. For this present work an Effective Dosimeter was constructed as the second stage of a six-stage wire screen diffusion battery. This diffusion battery was operated at a continuous sampling rate of 0.8 lpm, with in-situ counting of the alpha particle activity from the progeny deposited on the filters providing an estimate of the radon progeny potential alpha energy concentration (PAEC). This hybrid system allowed two methods for the determination of the radon progeny DCF. The activity size distributions, measured using the diffusion battery, were combined with the values of the DCF as a function of particle size to obtain a size-weighted DCF. The DCF values were obtained from the ICRP respiratory tract model, as implemented in the computer code RADEP. The second determination of DCF was obtained directly from the fraction collected by the Effective Dosimeter. The hybrid diffusion battery was used to measure radon progeny in the Fairy Cave, Buchan, Victoria at 20-minute intervals over 30 hour period. This cave had radon concentrations exceeding 2000 Bq m⁻³, with low aerosol concentration and ventilation rate. The measurements were analysed to determine the radon progeny PAEC, the activity size distribution, the size-weighted DCF and the Effective Dosimeter collected fraction. The Effective Dosimeter DCFs were determined from the collected fraction using firstly a simple linear function and then using a more complex polynomial function to correct for residual errors. For the linear factor alone, the calculated Effective Dosimeter DCFs were on average 11% lower than the equivalent size-weighted DCF values. The agreement using the polynomial function was improved markedly, with a fitted ratio of 0.965, with a R value of 0.99.

Key words: Radon, radon progeny, activity size distributions, unattached fractions, ICRP Respiratory Tract Model, dose conversion factors, show caves, Effective Dosimeter

INTRODUCTION
The health risk associated with radon arises from the inhalation of the short-lived decay products, i.e. radon progeny (BEIR VI, 1999). The radiation dose from exposure to radon progeny can be derived from measurements of the potential alpha energy concentration (PAEC), combined with a dose conversion factor (DCF or dose per unit intake) which reflects the radiation dose to the respiratory tract from the deposition of the inhaled progeny. For exposure to ²²²Rn, both epidemiological and dosimetric modelling approaches have been used to derive appropriate dose conversion factors. The International Commission for Radiological Protection (ICRP) in its Publication 65 (ICRP, 1993) recommends the use of a single factor (1425 mSv/(J.h.m⁻³) or 5 mSv/WLM), determined from uranium mining epidemiological studies, as the preferred method converting radon progeny exposure to effective dose (radon progeny conversion convention). The
ICRP recognized that differences in aerosol conditions could modify the dose conversion factor, but considered the epidemiological approach to be simpler and more direct than the alternate dosimetric modelling approach.

However, while a single DCF value is operationally convenient, the use of the ICRP conversion convention does not provide conservative dose estimates in all circumstances. Dosimetric models based on the ICRP Human Respiratory Tract Model in Publication 66 (ICRP66, 1994) predict that the dose per unit intake is modified by the aerosol characteristics of the radon progeny, in particular the size and fraction of the nanometer-sized ultrafine mode (unattached fraction). (Birchall and James, 1995, Portsendorfer and Reineking, 1999). Increasing the value of the unattached fraction in these models increases the predicted dose conversion factors. The ICRP conversion convention is derived from data for uranium mining atmospheres, with relatively low unattached fractions. Any occupational exposure in an environment with low aerosol concentration or high ventilation rates would be expected have significantly increased dose conversion factors. Radon progeny exposure to tour guides in show caves with high radon levels is an example of a workplace where the models predict an increase to dose conversion factor of up to a factor of two or more (Portsendorfer and Reineking, 1999, Solomon, 1998). For exposure to the general public, the differences in aerosol conditions are offset by lower breathing rates, particularly for children. However, in assessing the efficacy of some radon remedial measures, such as air filtration and electrostatic precipitation, account needs to be taken of the change in aerosol conditions, in particular an increase in the unattached fraction and subsequent increase in the dose per unit intake.

Airborne radon progeny are associated with log-normally distributed particle size distributions with diameters in the range 0.6 nm to greater than 1 µm, with a predominant mode between 50nm and 500 nm (accumulation mode). A range of techniques for the measurement of size distributions of radon progeny associated with both accumulation mode and ultrafine particles have been developed. These have been extensively reviewed elsewhere (National Research Council, 1991). Many of these methods rely on the fractionation of the sub-micron radioactive particles by diffusion processes, using sets of wire screens, or by inertial and impaction processes, using cascade impactors. The latter systems are capable of resolving an additional coarse mode at particle sizes exceeding 1 µm. These systems are complex but have been used to measure radon progeny size distributions in homes, workplaces and uranium mines (BEIR VI, 1999). A simpler technique involving the measurement of unattached fraction has been widely used (Hopke, 1990). This method relies on the separation of the ultrafine and accumulation modes using a diffusion sampler, usually a single wire screen. These unattached fraction samplers poorly differentiate the two modes and provide little information on the particle size of either mode. To overcome some of the limitations of the single screen samplers, the Australian Radiation Protection and Nuclear Safety Agency (ARPANSA) has developed a two screen radon progeny sampler (Effective Dosimeter) designed to provide a direct measure of the size-weighted dose conversion factor.

The Effective Dosimeter comprises a two-screen sampler with a preseparator screen matched to nasal deposition and a second collection screen matched to the bronchial deposition, operated in conjunction with a reference filter. The operating parameters for the Effective Dosimeter can be selected to provide a collection efficiency that has the same particle size behavior as the radon progeny DCFs derived from the dosimetric modelling. The theoretical analysis showed that the Effective Dosimeter produced a better measure of the relative risk for exposure to radon progeny than a single screen (unattached fraction) sampler, and avoided the need to use complex multi-stage diffusion batteries or inertial impactors (Solomon, 1997). This paper describes a practical
implementation of the Effective Dosimeter and investigates the two-screen sampler response to the aerosol conditions in an underground cave over a period of some days.

METHODS

For this investigation, the radon progeny dose conversion factors were determined at a single site in the Fairy Cave, Buchan, Victoria, Australia over a three day period during June, 1997. This cave, one of two tourist caves in the area, is visited by over 100,000 tourists each year. Tours through the caves are conducted by permanent staff members. During the period of these measurement radon levels were consistently in excess of 2000 Bq m\(^{-3}\). Previous estimates of the radiation dose to the tour guides from radon progeny exposure in this cave, based on the ICRP 65 conversion convention, were in the range 3 to 9 mSv/year (Solomon et al., 1996). For the present study, the DCF values were determined using two methods:

- A size-weighted dose conversion factor was derived from measurements of radon progeny size distribution, combined with the particle-size dependent dose conversion factors. A six stage wire screen diffusion battery was used to determine the activity size distribution of radon progeny in the size range 0.6 to 1375 nm.
- A second determination was obtained from the second stage of the diffusion battery which was operated as an Effective Dosimeter.

Dosimetric Model

The extensive dosimetric model recommended by the ICRP in Publication 66 (ICRP, 1994) forms the basis for the derivation of radon progeny doses conversion factors for this study. Figure 1 shows the typical response functions for the dose conversion factor for an adult male with breathing rates of 1.2 m\(^3\).h\(^{-1}\) and 0.78 m\(^3\).h\(^{-1}\), for occupational and environmental exposure, respectively. These curves were derived using the computer program RADEP, which implements the ICRP66 Human Respiratory Tract Model (Birchall and James, 1995). The representative activity concentration ratios for 218Po : 214Pb : 214Bi were assumed to be 0.8 : 0.02 : 0.0 and 0.8 : 0.4 : 0.2, for particle sizes < 20 nm and –20 nm, respectively. The resultant doses are relatively insensitive to the exact choice of values for the equilibrium ratios. The ICRP66 model-derived values have been adjusted by a factor of 0.3 to provide consistency with the epidemiologically-derived risk estimates in ICRP65. This factor was chosen to match the RADEP-derived DCF values to the ICRP65 conversion convention for the uranium mine aerosol conditions ICRP65 ((Birchall and James, 1995).

Assessment of Radon Progeny Activity Size Distributions

The potential alpha energy concentration (PAEC), the unattached fraction of PAEC (f\(_p\)) and the PAEC activity size distributions were measured using a six-stage wire screen diffusion battery, which was operated with a continuous sampling rate of 0.8 lpm per stage. The diffusion battery used in-situ counting of alpha particles from the radon progeny activity deposited on the collector (filter or screen) in each stage. The mode of operation of the diffusion battery has been described previously (Solomon and Wilks, 1994). The collection efficiencies for each stage were calculated using the fan-filtration penetration theory applied to wire screens by Cheng and Yeh, 1980) and Cheng et al., 1980), with a semi-empirical diffusion coefficient equation in the molecular cluster size range (Ramamurthi and Hopke, 1989). The activity collected on the wire screen collector were corrected for alpha particle losses in the screens and for the fraction of activity on the front of the screen (front to total ratio)
using the functions in Solomon and Ren, (1992). The diffusion battery parameters are summarised in Table 1 and the stage configuration and the calculated stage collection efficiencies are shown in Figure 2.

The collection and analysis of the diffusion battery data were carried out automatically using a purpose-written computer program running on a PC based computer. For each 20 minute integration period, the set of six alpha activities were converted to PAEC and deconvoluted using both the Twomey (1975) and the Expectation Maximisation (EMax) algorithms (Maher and Laird, 1985) to derive two independent particle size distributions in 40 logarithmically spaced size intervals between 0.6 nm and 1375 nm. For each sample, a size-weighted dose conversion factor was derived from measured of radon progeny size distribution, combined with the particle-size dependent DCF values, calculated from RADEP.

**Effective Dosimeter**

The functional form of the DCF versus particle size response shown in Figure 1, is strongly affected by two sets of deposition processes. With size decreasing below 3 nm there is a dose reduction due to deposition losses in the nose (a pre-separation stage). Above 3 nm the dose decreases due to reduced deposition in the respiratory tract with increasing particle size (collection stage). This particle size response can be simulated with a sampler comprising two wire-screen collectors. A sampling system providing a close match to the DCF response function can be achieved using a wire screen pre-separator simulating the nasal deposition process (dashed curve in Figure 3) and a wire screen collector matching the respiratory tract collection (dotted curve in Figure 3). The particle size response for a two screen system using a 105 mesh, 0.7 cm diameter pre-separator and a 400 mesh, 4.0 cm diameter collector at a sampling rate of $1.36 \times 10^{-5} \text{ m}^3 \text{s}^{-1}$ (0.8 lpm) is shown by the solid curve in Figure 3.

The theoretical analysis showed that a two screen sampler with a size dependent collection efficiency $\varepsilon_{HE}(d_p)$ matched to the DCF response function $\text{DCF}(d_p)$, should provide a measure of the radon progeny DCF, irrespective of the exact form of the radon progeny particle size distribution. If $A_s$ is the activity measured on the second (collector) screen of the two-screen sampling stage and $A_f$ is the activity measured on a reference filter, then the Effective Dosimeter collection efficiency $\varepsilon_{HE}$ is given by;

$$\varepsilon_{HE} = \frac{A_s}{A_f} \quad (1)$$

A single linear scaling function can be used to derive a size-weighted dose conversion factor $\text{DCF}_\varepsilon$, (Solomon, 1997):

$$\text{DCF}_\varepsilon = \text{DCF}_{\text{min}} \times (1.0 + 35.0 \times \varepsilon_{HE}) \quad (2)$$

where $\text{DCF}_{\text{min}}$ is the value of the dose conversion factor at the minimum in the particle size response function (~ 500 nm). For the RADEP implementation of the ICRP66 respiratory tract model, normalised to epidemiological data,

$$\text{DCF}_{\text{min}} = 285 \text{ mSv/ (J h m}^{-3}) \quad (1.0 \text{ mSv/WLM}) \quad (3)$$

or

$$\text{DCF}_{\text{min}} = 513 \text{ mSv/ (J h m}^{-3}) \quad (1.8 \text{ mSv/WLM}) \quad (4)$$
for environmental exposure and for occupational exposure, respectively.

The theoretical analysis showed that an additional polynomial correction was needed to reduce the residual errors inherent in the linear scaling function. For the present Effective Dosimeter operating at 0.8 lpm the correction was determine to be (Solomon (1997)):

\[
DCF_{HE} = DCF \varepsilon \cdot \left( 1 + 1.25 \times 10^{-4} \cdot (\varepsilon_{HE} - 0.0181) \cdot (\varepsilon_{HE} - 0.281) \cdot (\varepsilon_{HE} - 0.625) / \varepsilon_{HE} \right)^5
\]  

**RESULTS**

The diffusion battery was operated at a powered in site in the Kings Chamber of the Fairy Chamber, from the 18th to 20th of June 1997. A total of 247 activity size distributions were measured and each of these was analysed to determine the geometric mean and proportion of each mode as well the size-weighted dose conversion factor. Figure 4 shows the derived variation with time of the geometric mean diameters (GMD) for the ultrafine and accumulation modes. The ultrafine GMD was in the range 0.6 nm to 10.1 nm, with a mean and standard deviation of 1.9 nm ± 1.6 nm. The deconvolution algorithms used for the analysis are approximate methods, capable of dealing with non-linear processes involved in the data analysis. Measurement errors in the input data can produce large variations in output results. Given that the time response of the sampling method is of the order of an hour, some of the more rapid variations in the ultrafine GMD are most likely due to the propagation of uncertainties in the data, particularly when the fraction in the ultrafine mode is small. In comparison, the variation of the GMD for the attached radon progeny was less than a factor of 2.5, ranging from 64 nm to 157 nm, with a mean and standard deviation of 120 nm ± 20 nm.

The unattached fraction (fp), as determined from the fraction in the ultrafine mode, varied from zero to 26%, with a mean and standard deviation of 11.7% ± 7%. The fraction collected by the Effective Dosimeter (fHE), ranged from 5.0% to 15.1%, with a mean and standard deviation of 10.4% ± 2.4%. The variation of both fractions over the measurement period is shown in Figure 5. Overall, the general form of the two fractions is in good agreement, except for the period when the unattached fraction is close to zero. For these measurements periods, the attached radon progeny provide the residual Effective Dosimeter response.

The size-weighted DCF and the Effective Dosimeter DCF values for the measurement period are shown in top and bottom graphs in Figure 6, for occupational exposure (breathing rate 1.2 m³ h⁻¹) and environmental exposure (breathing rate 0.78 m³ h⁻¹), respectively. The lower solid line on each graph shows the ratio of the two DCF values for each sample. Although the DCF values vary by up to a factor of two, this ratio was relatively constant for both exposure conditions. For occupational exposures, the size-weighted DCF values ranged from 1740 to 3530 mSv/(J.h.m⁻³) (6.1 to 12.4 mSv/WLM), with a mean and standard deviation of 2710 ± 460 mSv/(J.h.m⁻³) (9.5 ± 1.6 mSv/WLM). Using only the linear function in Equation (2), the Effective Dosimeter DCF values were on average 11% lower than the equivalent size-weighted DCF values. With Effective Dosimeter values adjusted by the polynomial correction in Equation (5) this difference was reduced to less than 4%. For environmental exposures, the size-weighted DCF values ranged from 1000 to 1940 mSv/(J.h.m⁻³) (3.5 to 6.8 mSv/WLM), with a mean and standard deviation of 1480 ± 230 mSv/(J.h.m⁻³) (5.2 ± 0.8 mSv/WLM).
DISCUSSION

The excellent agreement between the Effective Dosimeter and the diffusion battery DCF values confirms the theoretical predictions for the performance of Effective Dosimeter. Unlike the diffusion battery, the Effective Dosimeter does not provide information on the radon progeny activity size distribution. However, it is a much simpler device to operate and the analysis procedure to determine the DCF values is less complex. For the measurements in the Fairy Cave, the sampler was operated in a continuous sampling mode, but it is also possible to operate the two-screen sampler with short sampling periods and to collect samples for analysis by conventional radon progeny multi-count methods. The Effective Dosimeter measures the PAEC and the DCF simultaneously, and by combining these two parameters for each measurement, the effective dose per unit time for inhalation of radon progeny (µSv h⁻¹) can be estimated.

The application of the ICRP 65 conversion convention for estimating radon progeny effective dose and the use of a single factor provides a convenient and simple method for assessing the health risk from inhalation of radon progeny in most environmental and many occupational exposure situations. The significantly increased DCF values for this study pose a problem for radiation monitoring of tour guides working in the Fairy Cave. The systematic differences between the epidemiological and dosimetric modelling approaches, illustrated by the use of a normalisation factor of 0.3 for the RADEP DCF values used in this study, complicates the adjustment of DCF values for changes to aerosol conditions. The National Research Council study on comparative dosimetry in homes and mines (NRC, 1991) uses the dosimetric models to provide a relative measure of the change to DCF for different radon progeny size distributions. This approach was based on estimates of the uranium mine aerosol conditions for the epidemiological studies, and defines a parameter termed the K-ratio given by:

\[
K\text{-ratio} = \frac{DCF \ (Dosimetric \ Modelling)}{DCF \ (ICRP \ 65)} \quad (6)
\]

K-ratios were determined for the present measurements from the Effective Dosimeter and diffusion battery results. The top and bottom graphs in Figure 7 show the correlation between the two measurement data sets, for both occupational and environmental exposure, respectively. There is an excellent correlation between the two measurement methods across a range of K-ratio values between 0.7 and 2.5. For a linear regression of both data sets with the fitted line passing through zero, the slope for both exposure conditions was 0.96, with \( R = 0.995 \). For a non-zero intercept, the fitted slope and intercept was 1.008 and –0.091, respectively, for occupational exposure and 1.078 and –0.122, respectively, for environmental exposure. These linear fitting functions are shown on Fig 7.

CONCLUSION

Consistent with the theoretical simulation, the practical implementation of the Effective Dosimeter provided a measure of the RADEP-based dose conversion factor that were in excellent agreement with the size weighted dose conversion factors determined by the wire screen diffusion battery. If the results reported in this study are typical of conditions in the cave through year, then the use of the ICRP 65 conversion convention to estimate the occupational exposure to the tour guides working in the Fairy Cave would underestimate the effective dose by up to a factor of two.
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REFERENCES


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Table 1: Summary of Parameters for Wire Screens Diffusion Battery used by the Australian Radiation Protection and Nuclear Safety Agency. Dp50 values are the particle diameter corresponding to 50% collection efficiency.

<table>
<thead>
<tr>
<th>Stage</th>
<th>No. of Screens</th>
<th>Mesh (Note 1)</th>
<th>Screen diameter (cm)</th>
<th>Collector</th>
<th>Flow (lpm)</th>
<th>Dp50 (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0</td>
<td>NA</td>
<td>NA</td>
<td>Filter</td>
<td>0.8</td>
<td>NA</td>
</tr>
<tr>
<td>2</td>
<td>1</td>
<td>105</td>
<td>0.7</td>
<td>400 Mesh</td>
<td>0.8</td>
<td>(Note 2)</td>
</tr>
<tr>
<td>3</td>
<td>1</td>
<td>105</td>
<td>3.7</td>
<td>Filter</td>
<td>0.8</td>
<td>6.3</td>
</tr>
<tr>
<td>4</td>
<td>7</td>
<td>105</td>
<td>3.7</td>
<td>Filter</td>
<td>0.8</td>
<td>27</td>
</tr>
<tr>
<td>5</td>
<td>30</td>
<td>105</td>
<td>3.7</td>
<td>Filter</td>
<td>0.8</td>
<td>88</td>
</tr>
<tr>
<td>6</td>
<td>30</td>
<td>105</td>
<td>9.5</td>
<td>Filter</td>
<td>0.8</td>
<td>304</td>
</tr>
</tbody>
</table>

Note 1. 105 Mesh wire screen, Wire Diameter 72 µm, Screen Thickness 148 µm, Solid Fraction 25%

Note 2. Stage 2 operates as an effective dosimeter and has a collection efficiency optimised to match the size dependency of the radon progeny dose conversion factor.
Figure 1: Effective dose per unit intake of $^{222}\text{Rn}$ progeny as a function of the particle size, as determined from the computer program RADEP. The RADEP values have been adjusted by a factor of 0.3 to provide consistency with the epidemiologically-derived risk estimates.
Figure 2: Sampling configuration and calculated collection efficiency of ARPANSA six-stage wire screen diffusion battery, for a sampling rate $1.36 \times 10^{-5}$ m$^3$ s$^{-1}$ (0.8 lpm).
Figure 3: Sampling configuration and calculated collection efficiency of ARPANSA Effective Dosimeter for a sampling rate $1.36 \times 10^{-5}$ m$^3$ s$^{-1}$ (0.8 lpm), a 0.7 cm diameter, 105 mesh pre-separator and a 4.0 cm diameter, 400 mesh screen collector.
Figure 4: Time variation of the Geometric Mean Diameter (GMD) of the ultrafine mode (upper graph) and accumulation mode (lower graph) from diffusion battery measurements in the Fairy Cave, Buchan.
Figure 5: Time variation of fraction in radon progeny ultrafine mode ($\% f_p$) and collected fraction of Effective Dosimeter ($\% f_{HE}$) for period of measurements in Fairy Cave, Buchan.
Figure 6: Time variation of the size-weighted dose conversion factors (DCF), determined from diffusion battery measurements and the Effective Dosimeter DCF values for occupational and environmental exposure (upper and lower graphs, respectively).
Figure 7: Correlation between K-Ratios determine from Effective Dosimeter with values determine from size-weighted dose conversion factors based on diffusion battery measurements, for occupational (upper) and environmental (lower) exposures. Solid line is linear regression of measured data.