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REMOVAL OF RADON DAUGHTERS BY FILTRATION AND ELECTRIC FIELDS

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Abstract – The level of airborne activity of the short-lived radon daughter products in indoor air may be controlled by various means, like ventilation and filtration. While ventilation with radon-free air is a very effective measure, it usually also means introduction of colder air, which is in variance with energy saving efforts. Internal filtration, on the other hand, where the air is circulated through a filtration system, will not affect the radon level, but may reduce the level of the daughter activities, roughly inversely proportional to the filtration rate. The filtration may, however, also change the aerosol composition of the air, and usually increase the fraction of daughter products in the unattached state and hence, according to several dose models, increase the dose per unit of potential alpha energy concentration, delivered to various parts of the respiratory tract. The paper reports some results from an investigation of the effect of filtration on the composition of radon daughters in indoor air. It is demonstrated, that at moderate aerosol concentrations, filtration may at a given radon level, according to some dose models, effectively decrease the dose from the daughter products, while at least one model predicts very little variation in the dose with the filtration rate. Preliminary results are also reported on measurements of the state of electric charge on the various daughter products, and on the possibility of lowering the daughter concentrations by exposing the air to an electric field.

INTRODUCTION

The most effective means of reducing the concentration of radon and its airborne, short-lived progeny in indoor air is ventilation. If the air is replaced e.g. once per hour with radon-free air the radon concentration in the room can at the most reach 0.75% of the unventilated value and the daughter products even less than that. However, ventilation is not always acceptable or practical, and in such situations processes like filtration may be an alternative. When air is drawn through a filter, be it a mechanical or an electro-filter, radon, being an inert gas, will pass unhindered. The daughter products, on the other hand, may, depending on the type of filter used, the aerosol condition of the air etc, be removed with efficiencies often in the 90 - 100% range, and filtration may thus offer a convenient method of reducing the concentration of airborne radon daughters[1,2].

THEORY

Assume we have an unventilated room with a radon activity concentration, A_o ($Bq.m^{-3}$). Let the activities of the daughter products ^{218}Po (RaA), ^{214}Pb (RaB) and ^{214}Bi (RaC) be A_1, A_2 and A_3 respectively. The activity of the fourth daughter ^{214}Po (RaC′) is then also A_3. The concentration of potential alpha energy E_p is given by

$$E_p = (E' + E'')\,A_1/\lambda_1 + E''\,A_2/\lambda_2 + E''\,A_3/\lambda_3 \quad (1)$$

where λ_1, λ_2 and λ_3 are the decay constants of the first three daughters and E' and E'' are the energies of the alpha particles from ^{218}Po and ^{214}Po. If the energies are measured in joules (J), the activities in $Bq.m^{-3}$ and the decay constants in s^{-1}, the potential alpha energy concentrations will be measured in $J.m^{-3}$.

If the daughters are in equilibrium with the mother product, then

$$A_o = A_1 = A_2 = A_3 \quad (2)$$

The maximum potential alpha energy concentration E_{po} is then

$$E_{po} = A_o\,[E' + E'')/\lambda_1 + E''/\lambda_2 + E''/\lambda_3] \quad (3)$$

The degree of equilibrium of the daughter products in the airborne state is characterised by the equilibrium factor, F, defined by

$$F = E_p/E_{po} = 0.105a + 0.516b + 0.380c \quad (4)$$

where $a = A_1/A_o$, $b = A_2/A_o$ and $c = A_3/A_o$ are the relative concentrations of the three radon daughters (relative to radon).

The equilibrium factor, F, will reflect the effect of a given air treatment, such as filtration, on the potential alpha energy concentration independently of variations in the radon concentration. The effect on the individual daughter products, however, is better expressed by the removal rates

$$r_1 = (1/a\text{-}1)\,\lambda_1,\; r_2 = (a/b\text{-}1)\,\lambda_2 \text{ and } r_3 = (b/c\text{-}1)\,\lambda_3 \quad (5)$$

The removal rates express for each daughter product

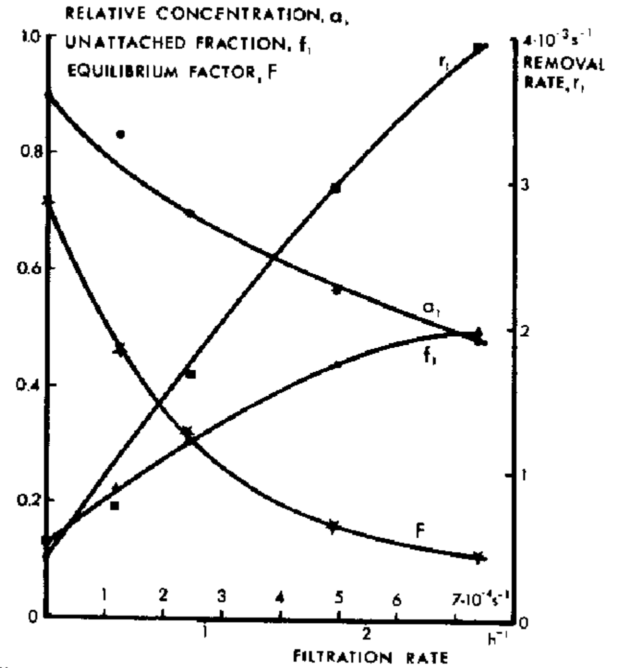

Figure 1. Equilibrium factor, F, relative concentration, a_1, unattached fraction, f_1, and removal rate, r_1, of RaA as a function of the filtration rate. Aerosol concentration 0.5 to 1.5×10^{11} m^{-3}.

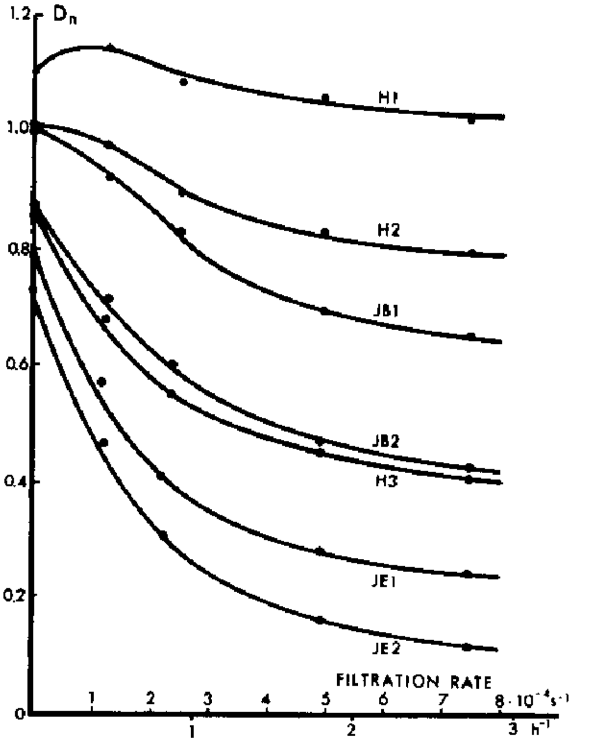

Figure 2. Normalised doses, D_n, (to bronchial basal cells) as a function of the filtration rate. H1: Harley-Pasternak, Children, light activity. H2: Harley-Pasternak, Adults, male, light actvity. H3: Harley-Pasternak, Children and Adults, resting. JB1: James-Birchall, Adults, 1.2 m^3/h. JB2: James-Birchall, Infants, 1 year, and Adults, 0.45 m^3/h. JE1: Jacobi-Eisfeld, Infants, 1 year. JE2: Jacobi-Eisfeld, Adults, 1.2 m^3/h.

the rate with which that particular nuclide is being removed from the air by other means than radioactive decay(2).

The radiological impact of being exposed to a given atmosphere is only partly determined by the potential alpha energy concentration but also by the likelihood of deposition of the inhaled activity at specific sites and thus upon the fraction of the daughters in the unattached state. Several models for the dose received over a given time by a given part of the respiratory tract have been proposed(3,4,5). Generally the dose D can be expressed as

$$D = Rt(B_1' f_1 A_1 + B_1 (1-f_1) A_1 + B_2' f_2 A_2 + B_2 (1-f_2) A_2 + B_3' f_3 A_3 + B_3 (1-f_3) A_3) \quad (6)$$

where t is the exposure time, f_1, f_2 and f_3 are the unattached fractions of RaA, RaB and RaC and R, B_1', B_1, B_2', B_2, B_3' and B_3 are constants characteristic for the model used.

In order to get a dose figure independent of fluctuations in the radon concentration, the doses given by Equation 6 may be normalised by dividing by the dose, D_o, corresponding to an atmosphere with the same radon concentration, A_o, in equilibrium with its daughters, Equation 2, and where the unattached fractions f_1, f_2 and f_3 are zero, i.e.

$$D_o = A_o R\, t\, (B_1 + B_2 + B_3) \quad (7)$$

and the normalised dose D_n is

$$D_n = D/D_o = (b_1 + (b_1' - b_1) f_1) a_1 + (b_2 + (b_2' - b_2) f_2) a_2 + (b_3 + (b_3' - b_3) f_3 a_3 \quad (8)$$

where

$b_1' = B_1'/(B_1 + B_2 + B_3)$, $b_1 = B_1/(B_1 + B_2 + B_3)$ etc.

EXPERIMENTS AND RESULTS

A series of measurements have been performed in an unventilated basement room with an average radon concentration of about 400-500 Bq.m^{-3}. The following quantities were measured: radon concentration, radon daughter concentration (alpha-spectroscopically), unattached fraction of individual daughters and aerosol concentration(2). The aerosol concentration was controlled by the use of gas (bunsen) burners. The air in the room can be passed through mechanical or electro-filters with filtration rates up to approximately 3 times per hour.

In Figure 1 are shown some of the results from one of the runs. The aerosol concentration was kept in the range 0.5–1.5 × 10^{11} m^{-3} (50,000-150,000 per cm^3) with a mean particle diameter about 0.05 μm. The curve shows the relative concentration, a_1, the unattached fraction, f_1, and the removal rate, r_1, for RaA together with the equilibrium factor, F, (see

Equation 4), all plotted as a function of the effective filtration rate. If filtration was the only removal process active, apart from radioactive decay, the removal rate would equal the filtration rate. However, since a fairly large fraction (and increasing with increasing filtration rate) of the RaA is in the unattached state, diffusion to the walls (plateout) plays a major role in the removal of RaA. Similar relations are also found for RaB and RaC, although the unattached fractions and consequently removal rates were lower (maximum unattached fractions were $f_2 = 0.18$ and $f_3 = 0.13$). These results are omitted for the sake of clarity. The plateout also affects the equilibrium factor. The experimental value of F at a filtration rate of $7 \times 10^{-4}\ s^{-1}$ is about 0.12 while filtration alone would give a value of 0.30.

It follows from Figure 1 and Equation 6 or 8, that the radiological dose received over a given period from an atmosphere with a given radon concentration depends upon the filtration rate in a rather complicated way. As an example of this the normalised doses, D_n, to the basal cells in the bronchial epitheleum, for an atmosphere with an average aerosol diameter about 0.10 micrometer, have been calculated according to three different dose models and for, essentially, two groups of individuals. The dose models used are:

1) the Harley-Pasternak model, where $b_2' = b_3' = 0$ and b_1' about 15 to 300 times b_1,

2) the James-Birchall model, where $b_3' = 0$ and b_1' and b_2' are about 10 to 15 times b_1 and b_2 respectively, and

3) the Jacobi-Eisfeld model, where b_1', b_2' and b_3' are about 0.7 to 4 times b_1, b_2 and b_3.

The results are plotted in Figure 2. It appears that the effect of filtration of the atmosphere on the dose delivered depends strongly both upon the type of individual exposed and upon the model used. It should be emphasised that it is not possible to compare values directly from one curve with those from another since the normalising factors are different; furthermore an intercomparison of the different dose models is not within the scope of this paper.

The figure shows, however, that filtration with rates of up to three times per hour in most of the cases considered decreases the doses with factors from two to six, but that even at the highest practical filtration rates a certain activity remains in the air and hence a certain dose is delivered to persons exposed. It has, however, been shown that a part of airborne radon daughters are electrically charged, and it should therefore be possible to extract them from the air by an electric field.

This possibility was demonstrated experimentally as follows. A metal disc with a diameter of 47 mm (same as the filters used) was placed on top of a 2m high stand placed in the middle of the experimental room (Figure 3). A negative voltage was applied to the disc and after a collection time of 10 min, the disc was analysed for activity in the same manner as a filter. In all cases a filter sample was taken simultaneously with the disc sample. The activity analysed is not the total activity collected by the disc, since only the activity deposited on the upper surface of the disc can be measured. Furthermore the counting of the disc activity assumes a uniform distribution of the activity over the surface, and obviously the activity will deposit more readily at the edges, where the field strength is maximum. It is therefore not unreasonable to assume that the disc

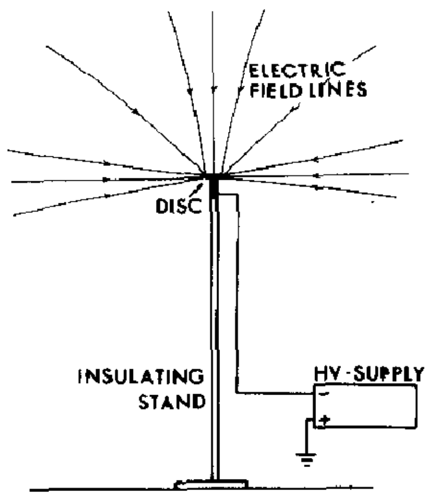

Figure 3. Arrangement for collection of radon daughters by an electric field.

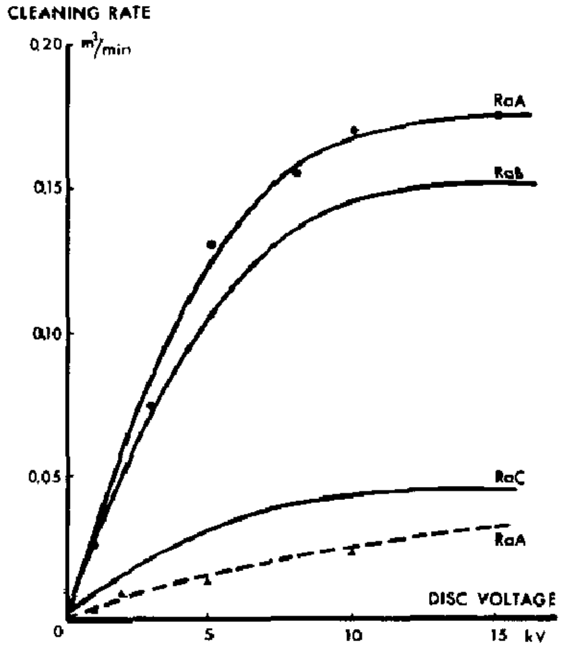

Figure 4. Cleaning rates for metal disc at negative voltage. Solid lines: filtration at 3 h^{-1}, broken line: no filtration.

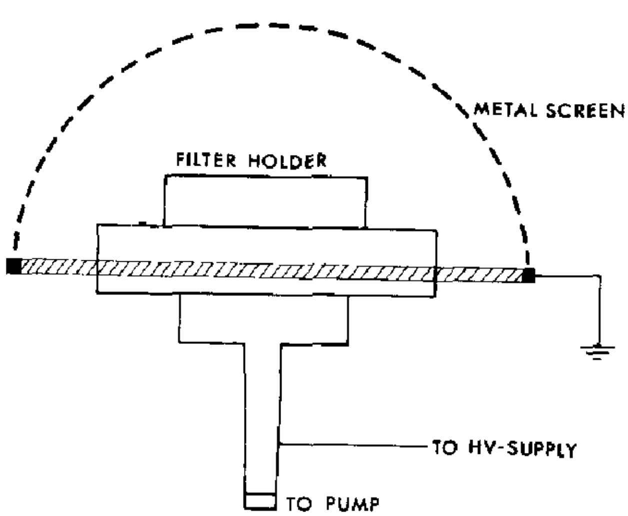

Figure 5. Arrangement for measurement of charged fraction of radon daughters.

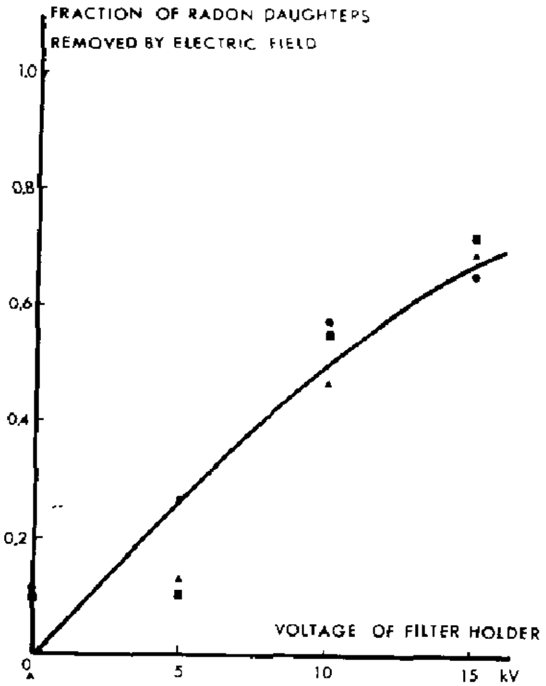

Figure 6. Removal of radon daughters by electric field. ●RaA ▲RaB ■RaC. Aerosol concentration 10^9 to 10^{10} m^{-3}. No filtration.

activity counted is only about half of the total activity collected.

The disc activity can be calculated for each daughter as activity collected per unit time (e.g. $Bq.s^{-1}$) and if this figure is divided by the corresponding activity concentration we find the rate with which the disc cleans the air for the daughter product in question.

In Figure 4 are shown the results of the measurements of collection of activity on discs at two different situations: filtration at 3 times per hour, and no filtration, in both cases with an aerosol concentration of about 10^9 to 10^{10} m^{-3}. It appears from the figure that the cleaning power of the discs at a given voltage is much higher when the air is simultaneously filtrated. This is probably caused by the radioactive ions (the charged daughter products) having higher mobilities than is the case when the air is not filtrated. It should be mentioned that discs at zero or positive voltage do not collect activities to a measurable extent.

In order to get an idea of the fraction of the individual daughter products which is charged the following experiment was done. The filter holder was kept at a positive voltage during collection and thus surrounded by a field which will repel the positively charged daughter products. In order to make the field better defined and stronger at a given voltage, the holder opening is placed in a grounded hemispherical metal screen (Figure 5).

Let u be the sampling flow rate, V the voltage difference between the filter holder and the screen, r and R the radii of the filter holder and the screen respectively; it can then be shown that only ions with mobilities higher than

$$\frac{u}{2\pi V}\cdot\left[\frac{1}{r}-\frac{1}{R}\right] \qquad (9)$$

will reach the filter.

With this arrangement it is possible to use higher voltages and the daughter products will not plate out as readily as with ordinary cylindrical capacitors. The grounded screen will to a certain extent collect unattached daughters, even when these are not charged, but it is fairly easy to correct for this effect. The arrangement was tested in two series of experiments, carried out in the same kind of atmospheres as used for the measurements of electrostatic deposition on metal discs. In each experiment two filter samples were taken simultaneously, using the same flow rate. The one filter holder was placed freely in the room at ground potential, the other as shown in Figure 5. For each individual daughter product the ratio between the activity collected with and without a voltage on the holder was calculated, and the average determined for all experiments at the same voltage. The difference between unity and this average value was calculated, and is plotted in Figure 6 for the case where no filtration took place. Similar results were found when filtration was applied, although the effect of increasing voltage was smaller.

Notwithstanding the fact that the results have a considerable degree of uncertainty, the figure clearly shows that it is possible with a voltage of 15 kV on the filter holder to repel from 50 to 70% of the daughter products. It is also clear that, at least in the experiments described in Figure 6, saturation is not achieved, even though the limiting mobility of the arrangement is as low as about 10^{-8} $m^2.V^{-1}.s^{-1}$.

The most surprising feature of Figure 6 is probably that RaB and RaC seem to be affected by the fields to more or less the same degree as does RaA. The results presented here should, as already mentioned, only be taken as a preliminary test of the possibility of affecting the level of airborne radioactivity by electrical means, probably as a supplement to other methods. A more detailed analysis of the electrical properties of the radon daughters is under way, including measurements at elevated aerosol conditions, and of the effect of electric fields on the room air as a whole.

ACKNOWLEDGEMENT

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