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Relationship between the ^{210}Po activity incorporated in the surface of an object and the potential α -energy concentration

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Abstract

The ^{210}Po activity in the surface layer of an object is determined by the total deposition flux of Rn progeny. The total deposition flux is mainly a function of deposition fraction and concentration of attached and unattached progeny. A linear relationship between the activity of ^{210}Po incorporated in the surface of a glass object and the average potential α -energy concentration (PAEC) in air is found by using the Jacobi-Porstendörfer predictive model for short-lived Rn progeny. The cumulative exposure to Rn progeny over a certain time period is also found to be related to the surface activity of ^{210}Po . The surface of a glass object can be used as an effective dosimeter for retrospective dose estimation, but with inherent uncertainties involved. © 1999 Elsevier Science Ltd. All rights reserved.

1. Introduction

Atoms of short-lived Rn progeny can deposit on available internal building surfaces. Due to recoil α decays of ^{218}Po and ^{214}Po nuclides, some of the newly formed atoms can be incorporated into the surface. The activity of the long-lived Rn progeny ^{210}Pb implanted into the surface increases with the time of exposure of the object to

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the short-lived Rn progeny. The second successor of ^{210}Pb is the α -emitting nuclide ^{210}Po , the activity of which in the subsurface layer can be determined by α measurements.

During the last 10 years, methods have been developed for determining retrospective Rn exposure based on the measurement of ^{210}Po activity in an object by using various techniques for α -activity measurements (Lively & Ney, 1987; Samuelsson, 1988; Mahaffey et al., 1993; Samuelsson & Johansson, 1994; McLaughlin, 1998). It is established that the α -activity of ^{210}Po is related to the cumulative Rn exposure. Various glass objects were found to be useful for this purpose. The influence of dust on the implantation of recoil Rn progeny atoms was studied by Johansson, Wolff, and Samuelsson (1994).

In the present work, an attempt is made to find out the relationship between the activity of ^{210}Po in the object surface and the average potential α -energy concentration (PAEC) during the period of exposure of the object to short-lived Rn progeny. The steady-state Jacobi's predictive model (Jacobi, 1972) for Rn and its progeny was used to determine the relationship.

2. Model calculation

We denote r_{tot} as the total deposition flux of Rn progeny onto an object surface in a room, and r_1, r_2 and r_3 as the deposition fluxes of ^{218}Po , ^{214}Pb and ^{214}Bi , respectively; $r_{\text{tot}} = r_1 + r_2 + r_3$. The deposition flux (r_i) is the number of Rn progeny atoms (N_i) deposited onto a unit surface of an object per unit time (in units of atoms $\text{m}^{-2} \text{s}^{-1}$).

A half of the deposited atoms are assumed to be incorporated into the surface after α -decay, so $r_{\text{tot}}/2$ of ^{210}Pb atoms will be stored per $\text{m}^2 \text{s}$; the other half will be detached from the surface and ejected into the air. Since ^{210}Pb is radioactive with the decay constant λ_5 (the subscript 5 is used as ^{210}Pb is the fifth progeny of Rn), the number of its atoms in unit surface N_5 (in m^{-2}) is governed by

$$\frac{dN_5}{dt} = \frac{r_{\text{tot}}}{2} - \lambda_5 N_5. \quad (1)$$

The solution of this equation for the initial condition of $N_5 = 0$ at $t = 0$ is

$$N_5 = \frac{r_{\text{tot}}}{2\lambda_5}(1 - e^{-\lambda_5 t}) \quad (2)$$

which gives the functional variation of the number of ^{210}Pb atoms in unit surface area with time. The corresponding activity A_5 changes with time in a similar way:

$$A_5 = \lambda_5 N_5 = \frac{r_{\text{tot}}}{2}(1 - e^{-\lambda_5 t}). \quad (3)$$

The measurable quantity is the surface α -activity A_7 of ^{210}Po (the subscript 7 is used as ^{210}Po is the seventh ^{222}Rn progeny). Since ^{210}Po is in radioactive equilibrium with

^{210}Pb , A_7 is assumed to be equal to A_5 . Therefore, the surface activity of ^{210}Po (in Bq m^{-2}) in the surface layer of an object is related to the deposition fluxes (r_i) of short-lived Rn progeny by

$$A_7 = \frac{r_1 + r_2 + r_3}{2}(1 - e^{-\lambda_5 t}). \quad (4)$$

The detailed derivation of Eqs. (3) and (4) is given by Nikezić and Yu (1998).

2.1. Total deposition flux r_{tot}

As mentioned above, the total deposition flux (r_{tot}) is the sum of the deposition fluxes of all short-lived Rn progeny, i.e., $r_{\text{tot}} = r_1 + r_2 + r_3$. The deposition flux (r_i) of the i th Rn progeny is related to the concentration of atoms C_i (atoms m^{-3}) of the species i , and the deposition fraction (λ_d) (in s^{-1}), which will be described in the next paragraph. We denote v_d^u (in m s^{-1}) as the deposition velocity of atoms of unattached Rn progeny (superscript u), v_d^a the deposition velocity of atoms of attached progeny (superscript a), S (in m^2) the surface area available for deposition and V (in m^3) the volume of a room. In this way, $v_d S$ is the volume from which atoms will deposit onto the surface area S in unit time, and $v_d S/V$ is the fraction of the volume from which the atoms will deposit in unit time.

Since a ‘well-mixed’ assumption is adopted, the atoms of Rn progeny are uniformly distributed within the indoor air, so the fraction of volume from which the atoms will deposit in unit time mentioned above is equal to the fraction of atoms which deposit onto the surface. This quantity is called the deposition fraction λ_d (in s^{-1}). Therefore, the deposition fraction of attached and unattached atoms are, respectively, given by

$$\lambda_d^a = \frac{v_d^a S}{V}, \quad (5)$$

$$\lambda_d^u = \frac{v_d^u S}{V}, \quad (6)$$

The deposition fraction gives the *fraction* of atoms deposited onto the surface in unit time. The number of atoms per unit volume deposited onto the surface in unit time is simply described as $\lambda_d C$, so the total number of atoms deposited onto available surfaces inside the room per unit time is $\lambda_d C V$. The deposition flux, r is then given as

$$r = \frac{\lambda_d C V}{S}. \quad (7)$$

The deposition flux r_1 of ^{218}Po is the sum of two components, i.e., the unattached flux r_1^u and the attached flux r_1^a :

$$r_1 = r_1^u + r_1^a = \frac{V}{S}(\lambda_d^u C_1^u + \lambda_d^a C_1^a). \quad (8)$$

The deposition flux r_2 of ^{214}Pb is similarly written as

$$r_2 = r_2^u + r_2^a = \frac{V}{S}(\lambda_d^u C_2^u + \lambda_d^a C_2^a). \quad (9)$$

and that of ^{214}Bi as

$$r_3 = r_3^u + r_3^a = \frac{V}{S}(\lambda_d^u C_3^u + \lambda_d^a C_3^a). \quad (10)$$

The total deposition flux, r_{tot} , is the sum of Eqs. (8)–(10):

$$r_{\text{tot}} = r_1 + r_2 + r_3 = \frac{V}{S}[\lambda_d^u(C_1^u + C_2^u + C_3^u) + \lambda_d^a(C_1^a + C_2^a + C_3^a)]. \quad (11)$$

It is also possible to express the total deposition flux in terms of activity concentrations A_i (in Bq m^{-3} ; $i = 1,2,3$) in the indoor air as

$$r_{\text{tot}} = \frac{V}{S} \left[\lambda_d^u \left(\frac{A_1^u}{\lambda_1} + \frac{A_2^u}{\lambda_2} + \frac{A_3^u}{\lambda_3} \right) + \lambda_d^a \left(\frac{A_1^a}{\lambda_1} + \frac{A_2^a}{\lambda_2} + \frac{A_3^a}{\lambda_3} \right) \right], \quad (12)$$

where λ_i ($i = 1,2,3$) are the decay constants of ^{218}Po , ^{214}Pb and ^{214}Bi , respectively. It can be seen that the total deposition flux is a linear function of the activity concentrations of the short-lived Rn progeny. Alternatively, Eq. (12) can be written as

$$r_{\text{tot}} = \frac{V}{S} g(A_i^u, A_i^a, \lambda_d^u, \lambda_d^a). \quad (13)$$

The function $g = r_{\text{tot}}S/V$ ($\text{atoms m}^{-3} \text{ s}^{-1}$), called here the ‘deposition factor’, is independent of the room dimensions.

3. Results

3.1. Relationship between total deposition flux and PAEC

All activity concentrations in Eq. (12) can be calculated from the Jacobi’s predictive model, which has been described elsewhere (Jacobi, 1972). The input parameters of the model include the Rn input strength P_{Rn} (in atoms s^{-1}) in the room of volume V , the attachment rate λ_a (in s^{-1}), detachment probability from an aerosol of atoms after α -decay p_1 , the ventilation rate λ_v (in s^{-1}), and the previously defined deposition fraction λ_d^u and λ_d^a . In order to establish the relationship between the total deposition flux and PAEC, a small computer package has been prepared to calculate all the activity concentrations $A_i^{u,a}$, PAEC (mWL) and the total deposition flux from Eq. (12). It is clear from Eq. (12) that the deposition flux is directly proportional to the deposition fractions λ_d^u and λ_d^a . These two deposition fractions have typical values and ranges, by which model calculations were performed. The deposition fraction of unattached progeny was varied between 10 and 30 h^{-1} (Porstendörfer, 1984), while

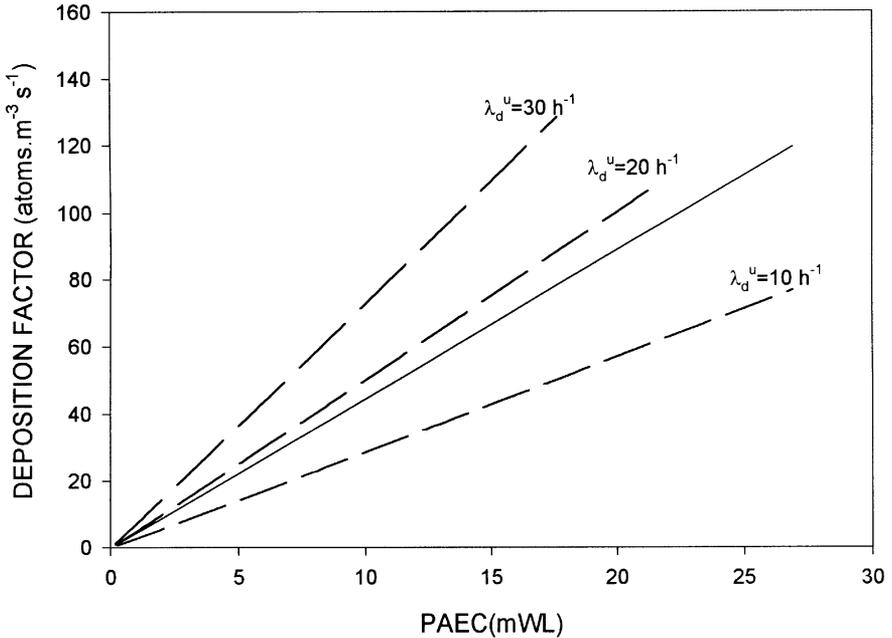


Fig. 1. Deposition factor as a function of PAEC for various deposition fractions of unattached Rn progenies. (Ventilation rate $\lambda_v = 0.55 \text{ h}^{-1}$, attachment rate $\lambda_a = 50 \text{ h}^{-1}$, deposition fraction of attached progenies $\lambda_d^a = 0.2 \text{ h}^{-1}$). The solid line is a linear fit to all the data in the graph.

that for attached progeny was kept constant ($\lambda_d^a = 0.2 \text{ h}^{-1}$) and the results are shown in Fig. 1.

The most convenient ways to vary the PAEC (as shown in Fig. 1) are to change the ventilation rate or the Rn source strength as input parameters of the model. Here, the Rn source strength was changed while the ventilation rate was kept constant. Very similar results are obtained when the ventilation is varied while the source strength is kept constant. It can be seen from Fig. 1 that the deposition flux is a linear function of the PAEC. The total deposition flux is very sensitive to λ_d^u . The best-fit line by linear regression of all the data in Fig. 1 is shown by the solid line assuming a uniform distribution of λ_d^u in the range of $10\text{--}30 \text{ h}^{-1}$, of which the slope is $4.45 \text{ atoms m}^{-3} \text{ s}^{-1}$ per mWL; the slope is between 2.86 and 7.31. The regression line is close to the line obtained for $\lambda_d^u = 20 \text{ h}^{-1}$. It can be assumed that the deposition fraction of unattached progeny will not be too far from this value in the long run.

Fig. 2 shows the results for different deposition fractions of attached progeny λ_d^a for a deposition fraction of 20 h^{-1} for unattached progeny. In this case the deposition fraction of attached progeny was varied between 0.1 and 0.3 h^{-1} (Porstendörfer, 1984), while that for unattached progeny was kept constant.

A linear relationship between the deposition flux and the PAEC is again observed. The total deposition flux is less sensitive to the deposition fraction of attached

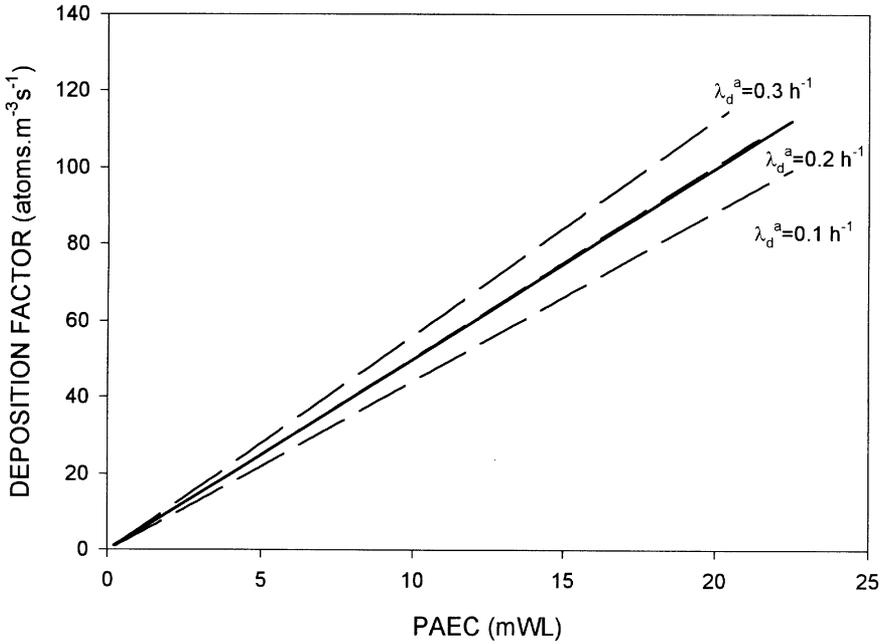


Fig. 2. Deposition factor as a function of PAEC for various deposition fractions of attached progenies. (Ventilation rate $\lambda_v = 0.55 \text{ h}^{-1}$, attachment rate $\lambda_a = 50 \text{ h}^{-1}$, deposition fraction of unattached progenies $\lambda_d^u = 20 \text{ h}^{-1}$). The solid line is a linear fit to all the data in the graph.

progeny. Regression analysis is again performed on all the data in this figure assuming a uniform distribution of λ_d^a in the range of $0.1\text{--}0.3 \text{ h}^{-1}$, and the best fit is shown by the solid line, which has a slope of $5.25 \text{ atoms m}^{-3} \text{ s}^{-1} (\text{mWL})^{-1}$ and overlaps the line for $\lambda_d^a = 0.2 \text{ h}^{-1}$. The slope of the lowest line is 4.42 and that of the uppermost line is 5.61.

Fig. 3 is the combination of the Figs. 1 and 2. The deposition fraction of the unattached progeny λ_d^u has been varied between 10 and 30 h^{-1} with a step of 1 h^{-1} ; the deposition fraction of the attached progeny λ_d^a has been changed from 0.1 to 0.3 h^{-1} with a step of 0.01 , and finally the Rn source strength was systematically varied between 0.1×10^5 and $20 \times 10^5 \text{ atoms s}^{-1}$ with a step of $1 \times 10^5 \text{ atoms s}^{-1}$, assuming a uniform distribution of each parameter in the described range. In the course of the calculation, the PAEC and deposition factor are determined. Each dot in the graph represents one combination of variables λ_d^u , λ_d^a and source strength. For example, the dot denoted by *A* in Fig. 3 is obtained for the following set of data: $\lambda_d^u = 10 \text{ h}^{-1}$, $\lambda_d^a = 0.1 \text{ h}^{-1}$ and source strength is $1.01 \times 10^6 \text{ atoms s}^{-1}$; for these data, $\text{PAEC} = 30.2 \text{ mWL}$ and the deposition factor is $71.3 \text{ atoms m}^{-3} \text{ s}^{-1}$. A linear fit has been applied to all the data in Fig. 3, and the best fit is represented by the solid line of slope $5.2 \pm 1.1 \text{ atoms m}^{-3} \text{ s}^{-1} (\text{mWL})^{-1}$. This slope is very close to that derived from

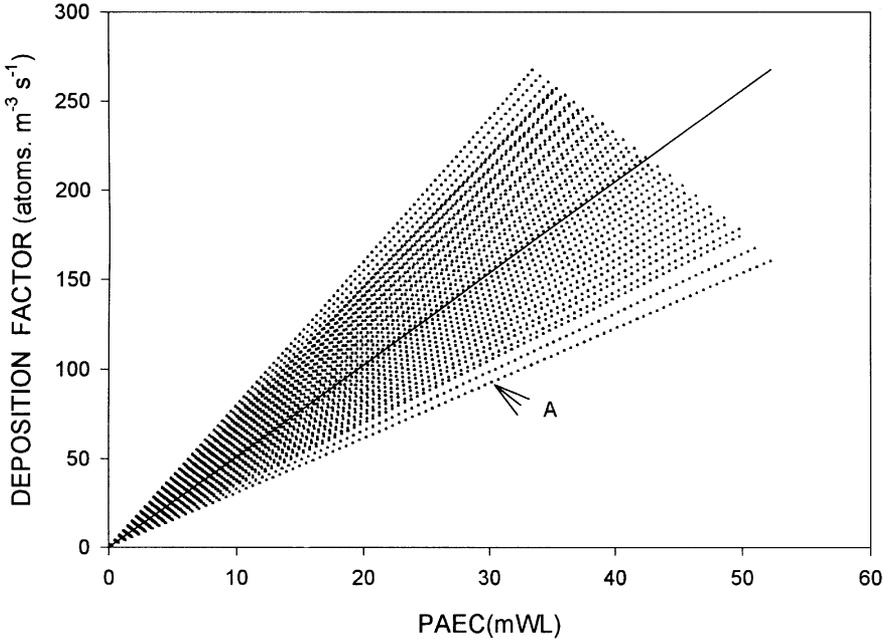


Fig. 3. Deposition factor as a function of PAEC. λ_d^a is varied from 10 to 30 h^{-1} with the step 1 h^{-1} ; λ_d^b is varied from 0.1 to 0.3 h^{-1} with the step 0.01 h^{-1} ; source strength is changed from 0.1×10^6 to 20×10^6 atom s^{-1} with the step 1×10^6 atom s^{-1} . The solid line is a linear fit of all points in the graph.

Fig. 2, and this value is adopted for further consideration. The uncertainty due to the unknown deposition fraction of the attached progeny is approximately 20%.

Realistic room dimensions (S and V) are needed for calculations of the total deposition flux of the short-lived Rn progeny. One can write

$$r_{\text{tot}} = \frac{V}{S} 5.2 \cdot \text{PAEC}. \tag{14}$$

In this way, a linear relationship between the total deposition flux and the PAEC is established, which is useful for the purpose of dosimetry.

3.2. Relationship between ^{210}Po activity and PAEC

We now go back to Eq. (4). The measurable quantity is A_7 , i.e., the surface activity of ^{210}Po incorporated into an object. The total deposition flux from Eq. (4) is simply written as

$$r_{\text{tot}} = \frac{2A_7}{(1 - e^{-\lambda_{st}})} \tag{4'}$$

By combining Eqs. (4') and (14), the relationship between the ^{210}Po activity and the average PAEC is

$$\text{PAEC} = \frac{0.19 \times 2A_7}{(1 - e^{-\lambda_7 t})} \times \frac{S}{V} = \frac{0.38A_7}{(1 - e^{-\lambda_7 t})} \times \frac{S}{V}, \quad (15)$$

where t is the age of the object, i.e., the duration of exposure of the object to deposition of short-lived Rn progenies, and the value of PAEC is the average for the period of exposure. Again, a linear relationship is obtained between PAEC and the activity of ^{210}Po implanted in the superficial layer of an object.

3.3. Surface of an object as an effective dosimeter

The total exposure to Rn progeny (X) is obtained by multiplying PAEC with the time of exposure t :

$$X = \text{PAEC} \times t. \quad (16)$$

The exposure to Rn progeny is usually given in WLM, which is equivalent to an exposure to a PAEC of 1 WL for 170 h. Therefore,

$$\begin{aligned} X(\text{WLM}) &= \text{PAEC}(\text{mWL}) \times 10^{-3} \times t (\text{h})/170 \\ &= \text{PAEC}(\text{mWL}) \times 10^{-3} \times t (\text{year}) \times 51.5 \end{aligned}$$

or

$$\begin{aligned} X(\text{WLM}) &= \text{PAEC} \times t = \frac{0.38 \times A_7}{(1 - e^{-\lambda_7 t})} \times \frac{S}{V} \times t (\text{year}) \times 51.5 \\ &= 19.6 \times 10^{-3} \frac{A_7}{(1 - e^{-\lambda_7 t})} \times \frac{S}{V} \times t (\text{year}), \end{aligned} \quad (17)$$

where t is in units of years, A_7 in Bq m^{-2} , and the term 10^{-3} comes from changing mWL to WL.

The drawback of Eq. (17) is the appearance of the ratio S/V . S is the surface area of all the available surfaces for deposition, which can be significantly larger than the room surface area. If we take the surface-to-volume ratio for a room without furniture to have a typical value of 2, and assume that the total surface area for deposition in a room with furniture is twice that for a room without any furniture, S/V would be approximately 4. Eq. (17) then becomes

$$X(\text{WLM}) \approx 80 \times 10^{-3} \frac{A_7}{(1 - e^{-\lambda_7 t})} \times t (\text{year}), \quad (18)$$

where the surface activity of ^{210}Po is in Bq m^{-2} , the exposure time t is in years, and the total exposure is in WLM.

The effective dose E (in mSv) can be obtained from the exposure to Rn progenies via a dose conversion factor (DCF) given in $\text{mSv}(\text{WLM})^{-1}$. There have been many discussions on the value of DCF, and published values differ by an order of

magnitude. Here, the most cited value of 5 mSv(WLM)^{-1} was adopted (James, 1983; Birchall & James, 1994). By using Eq. (18), one can find the effective dose as

$$\begin{aligned} E(\text{mSv}) &= 5 \frac{\text{mSv}}{\text{WLM}} \times X(\text{WLM}) \approx 5 \times 80 \times 10^{-3} \frac{A_7}{(1 - e^{-\lambda_a t})} \times t \text{ (years)} \\ &= \frac{0.4A_7}{(1 - e^{-\lambda_a t})} \times t \text{ (years)}. \end{aligned} \quad (19)$$

3.4. Numerical example

To verify Eq. (17), simple calculations were performed using the computer package previously mentioned. The input data are: Rn source strength of $4 \times 10^5 \text{ atoms s}^{-1}$; volume of the room 62.5 m^3 ; attachment rate $\lambda_a = 50 \text{ h}^{-1}$; detachment probability $p_1 = 0.83$; ventilation rate 0.55 h^{-1} ; surface area available for deposition $S = 250 \text{ m}^2$; deposition fraction of unattached atoms $\lambda_d^u = 20 \text{ h}^{-1}$ and deposition fraction of attached atoms $\lambda_d^a = 0.20 \text{ h}^{-1}$. The following results were obtained:

Rn concentration $A_0 = 86.65 \text{ Bq m}^{-3}$; $A_1^u = 13.80 \text{ Bq m}^{-3}$; $A_1^a = 48.80 \text{ Bq m}^{-3}$; $A_2^u = 1.17 \text{ Bq m}^{-3}$; $A_2^a = 31.02 \text{ Bq m}^{-3}$; $A_3^u = 3 \times 10^{-2} \text{ Bq m}^{-3}$; $A_3^a = 23.42 \text{ Bq m}^{-3}$; PAEC = 8.85 mWL, free fraction $f_1 = 6.49\%$; equilibrium factor $F = 0.37$, and finally the total deposition flux $r_{\text{tot}} = 10.76 \text{ atoms m}^{-2} \text{ s}^{-1}$. The total exposure X for 1 year to short-lived Rn progeny is $X = \text{PAEC} \times t(\text{h})/170 = 0.44 \text{ WLM}$.

We now look at the activity on the object surface. The deposition flux is $10.76 \text{ atoms m}^{-2} \text{ s}^{-1}$ in the numerical example, so the activity of ^{210}Po in the glass surface after 1 year is estimated to be $A_7 = 0.5 \times r_{\text{tot}}(1 - e^{-0.693 \cdot 1/22.3}) = 0.16 \text{ Bq m}^{-2}$ by Eq. (4). From Eq. (18), the exposure is then to be $X = 0.43 \text{ WLM}$ which is in good agreement with the value above.

3.5. Comparison with other authors

Samuelsson (1988) reported that 9 Bq m^{-2} of ^{210}Po in the surface corresponded to a Rn exposure of $8 \text{ kBq year m}^{-3}$ (^{222}Rn). (This value is recalculated by the present authors based on the data given by Samuelsson (1988).) Only the source strength is increased in the present model in order to have a Rn concentration of 8 kBq m^{-3} , and the total deposition flux is estimated to be $993 \text{ atoms m}^{-2} \text{ s}^{-1}$. After 1 year, the activity of ^{210}Po in the glass surface would be $A_7 = 15 \text{ Bq m}^{-2}$, which is 1.7 times greater than the 9 Bq m^{-2} . The value of 9 Bq m^{-2} can be reproduced when the deposition fractions in the model are assumed to be lower. For example, when the deposition fractions $\lambda_d^u = 10 \text{ h}^{-1}$ and $\lambda_d^a = 0.1 \text{ h}^{-1}$ are used, then A_7 is 9.3 Bq m^{-2} . However, this comparison is not completely appropriate because the ^{210}Po activity is related to the exposure to Rn progeny in the present work, but was related to Rn by Samuelsson (1988).

Pálfalvy Féher and Lörinc (1995) reported a ^{210}Po surface activity of 27.2 Bq m^{-2} measured by SSNTD detectors when the Rn concentration was 64000 Bq m^{-3} and the exposure lasted $\frac{1}{4}$ year. For this Rn concentration, when $\lambda_d^u = 20 \text{ h}^{-1}$ and

$\lambda_d^a = 0.2 \text{ h}^{-1}$, our model gives a deposition flux $r_{\text{tot}} = 7960 \text{ atoms m}^{-2} \text{ s}^{-1}$. The activity of ^{210}Po is then $A_7 = 0.5 \cdot 7960 \cdot (1 - e^{-0.693/4 \cdot 22.3}) \approx 30.8 \text{ Bq m}^{-2}$ which is in good agreement with the experimental value reported by Pálfalvy et al. (1995).

4. Discussion and conclusion

Many uncertainties exist in the determination of the cumulative exposure to Rn progenies from the ^{210}Po activity in the surface of a glass object (through Eq. (18)).

In the derivation of Eq. (4), there are a few assumptions. First, it was assumed that half of the recoil atoms were implanted in the object after α -decays; the numeral 2 in the denominator of Eq. (4) originated from this assumption. Secondly, humidity and dust particles settled on the object surface can reduce the implantation of the atoms after α -decay, which was not taken into account. In a very dirty atmosphere with a rather high aerosol concentration, the formulae derived here are not applicable. In addition, a constant deposition flux during the exposure was adopted, which may not be the real case.

Other assumptions were made in the derivation of Eq. (18). The slope of the regression line in Fig. 2, i.e., $5.25 \text{ atoms m}^{-3} \text{ s}^{-1}(\text{mWL})^{-1}$ which is taken as the most representative value, strongly depends on the deposition fraction of unattached and attached progenies. Figs. 1 and 2 show that the deposition flux is sensitive to the assumption on the deposition fraction. However, it might be assumed that the deposition fraction is approximately constant in a long time period.

The ratio S/V of the room has been taken to be 4 m^{-1} . Here, S is not the room's surface area, but it is the surface available for deposition including furniture surfaces, etc. It is obvious that the ratio can be different from the estimated value and can vary with the room.

The method for determination of effective dose based on measurements of ^{210}Po activity in the glass surface is not sensitive to the unattached fraction of Rn progeny. Only the total deposition flux of the unattached and attached progeny is responsible for the accumulated ^{210}Po activity. This introduces an additional uncertainty in the determination of the lung dose and the effective dose. The dose conversion factor of 5 mSv(WLM)^{-1} is also controversial but the discussion is out of the scope of this paper.

Despite the inherent uncertainties mentioned above, we can still conclude that the ^{210}Po activity in the glass surface is related to the exposure to short-lived Rn progeny; a linear relationship between ^{210}Po activity and PAEC is found. In this way, the glass surface can be used as a WLM meter.

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