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DISTRIBUTIONS OF THORON PROGENY CONCENTRATIONS IN DWELLINGS

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Abstract — The distribution of thoron (^{220}Rn) and thoron progeny indoors was derived from the diffusion equation in two-dimensions. In the ordinary living environment, thoron concentration decreases exponentially with the distance from the wall. The activity of ^{216}Po is almost in radioactive equilibrium with thoron due to the very short half-life of ^{216}Po. Comparing the average concentration in the room, the number of attached ^{212}Pb atoms is about seventy times larger than that of unattached ^{212}Pb. The concentrations of ^{212}Pb and ^{212}Bi are spatially homogeneous. However, in the case of a small effective diffusion coefficient, a concentration distribution appears. In such a case, the absorbed dose from the thoron progeny could vary according to its proximity to the wall or the centre of the room. The general characteristics of thoron and its progeny concentration distribution indoors is presented.

INTRODUCTION

The radiation exposure due to the inhalation of thoron progeny could be estimated to be of the order of 10–20% compared with short-lived radon progeny(1). In Japan, it has been reported that there are some dwellings which have a high thoron concentration(2,3). It was also reported that the interior walls of such dwellings are made of soil, which has a large exhalation rate of thoron, and therefore the dose from thoron progeny may not be neglected. Thoron concentration depends on distance from the wall which is the source of thoron, but less information is available concerning the distribution of indoor thoron progeny concentrations. It is necessary to study the characteristics of thoron progeny for estimation of the absorbed dose. Katase *et al*(4) have shown theoretically a relationship between the indoor thoron distribution and the effective diffusion coefficient, and have suggested a non-uniform distribution of thoron progeny. They have also calculated the distribution of unattached atoms of ^{212}Pb using a simple one-dimension diffusion equation. The concentration distributions of indoor thoron progeny have not been studied and calculated in detail, so that more research on thoron progeny is required.

The great difference in the half-life radon (^{222}Rn, $T_{1/2} = 3.8$ d) and thoron (^{220}Rn, $T_{1/2} = 55$ s) affects the exhalation rate from the wall and the concentration distribution in the room. A representative distribution of indoor radon concentration is uniform but that of thoron is non-uniform. It is therefore expected that there are different characteristics of the distribution between radon progeny and thoron progeny. Most of the radon and thoron progeny are attached to the aerosols and deposit on the surface of the room. Deposition is the most effective parameter of indoor radon and thoron progeny concentration as well as ventilation of the indoor air. Zarcone *et al*(5) compared their experimental results with model predictions that used first-order sink and source terms. The parameters of ventilation rate, deposition rate etc., were investigated separately, but how the thoron progeny concentration depends on the parameters has not been investigated.

It is here shown that distributions of indoor thoron progeny concentration can be predicted by a model calculation. The ventilation rate, deposition rate and indoor effective diffusion coefficient have been considered for the calculation of the characteristics of the distribution using the numerical method.

MODEL AND EQUATIONS

The behaviour of indoor radon and its progeny was formulated by Jacobi(6) and Porstendörfer *et al*(7) under the simple condition of the constant concentration in the room as the 'mass balance model'. For thoron and its progeny, the diffusion in the room should be required because of the very short half-life of thoron which emanates from the walls. For describing movement of the nuclides in the dwelling, an effective diffusion coefficient D ($m^2.s^{-1}$) is introduced which expresses the diffusion in the room macroscopically; the effective diffusion coefficients of thoron and its progeny are then assumed to have the same value. Distinguishing between attached (superscript 'a') and unattached or free (superscript 'f') atoms, the formation and extinction of the thoron decay chain is considered as follows:

$$\begin{array}{l} {}^{220}\mathrm{Rn} \xrightarrow{\lambda_T} {}^{216}\mathrm{Po}^f \xrightarrow{\lambda_A} {}^{212}\mathrm{Pb}^f \xrightarrow{\lambda_a} {}^{212}\mathrm{Pb}^a \xrightarrow{\lambda_B} {}^{212}\mathrm{Bi}^a \xrightarrow{\lambda_C} \\ \qquad\quad \downarrow \lambda_d \qquad\quad \downarrow \lambda_d \qquad \downarrow \lambda_d + \lambda_v \downarrow \lambda_d + \lambda_v \end{array}$$

The model for the calculation is shown in Figure 1. Thoron and its progeny concentrations were calculated by a two-dimensional and a steady state model. For considering the distribution of thoron and its progeny, the plane was divided into K × K cells. Thoron emanates from one of the walls and diffuses to the centre of the room. If thoron emanates from all the walls, we can obtain the concentration distribution by means of accumulating the result, which is estimated by the model in Figure 1. The emanation of thoron depends on the area of interior soil wall. Thoron decay products are formed and attach to indoor aerosol particles. The aerosol particles are removed in part by deposition on the surface of the wall and by ventilation through the window. It was assumed that ventilation occurred at all parts of walls instead of the window. The deposition and ventilation was considered at every cell next to the wall.

The cell number is indicated by i,j and N is the concentration in atoms.m^{-3}, the diffusion equations of the atom balance in each cell are classified into three groups. One is the group of the cells where no side is next to the wall. Then the equations are given as:

$$\frac{dN_{ij}^T}{dt} = D\frac{\hat{N}_{ij}^T - 4N_{ij}^T}{\Delta^2} - \lambda_T N_{ij}^T \qquad (1)$$

$$\frac{dN_{ij}^{Af}}{dt} = D\frac{\hat{N}_{ij}^{Af} - 4N_{ij}^{Af}}{\Delta^2} - \lambda_A N_{ij}^{Af} + \lambda_T N_{ij}^T \qquad (2)$$

$$\frac{dN_{ij}^{Bf}}{dt} = D\frac{\hat{N}_{ij}^{Bf} - 4N_{ij}^{Bf}}{\Delta^2} - (\lambda_B + \lambda_a)N_{ij}^{Bf} + \lambda_A N_{ij}^{Af} \qquad (3)$$

$$\frac{dN_{ij}^{Ba}}{dt} = D\frac{\hat{N}_{ij}^{Ba} - 4N_{ij}^{Ba}}{\Delta^2} - \lambda_B N_{ij}^{Ba} + \lambda_a N_{ij}^{Bf} \qquad (4)$$

$$\frac{dN_{ij}^{Ca}}{dt} = D\frac{\hat{N}_{ij}^{Ca} - 4N_{ij}^{Ca}}{\Delta^2} - \lambda_C N_{ij}^{Ca} + \lambda_B(N_{ij}^{Bf} + N_{ij}^{Ba}) \qquad (5)$$

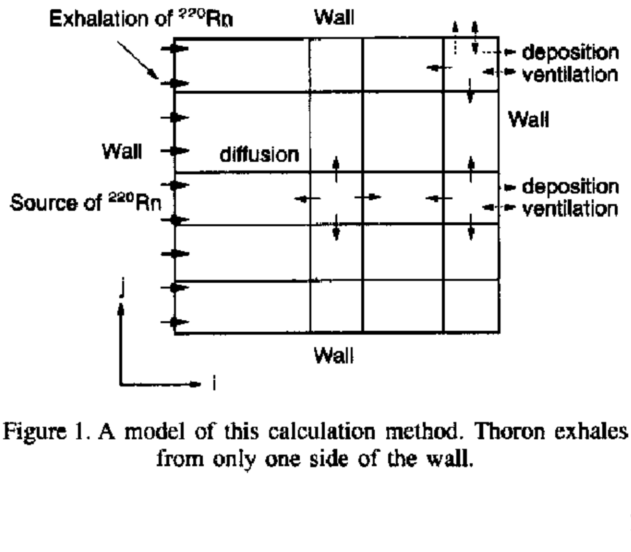

Figure 1. A model of this calculation method. Thoron exhales from only one side of the wall.

The other is the group of cells where only one side is next to the wall. Then the equations are given as:

$$\frac{dN_{ij}^T}{dt} = D\frac{\hat{N}_{ij}^T - 3N_{ij}^T}{\Delta^2} - \lambda_T N_{ij}^T + \frac{E}{\Delta} \qquad (6)$$

$$\frac{dN_{ij}^{Af}}{dt} = D\frac{\hat{N}_{ij}^{Af} - 3N_{ij}^{Af}}{\Delta^2} - (\lambda_A + \lambda_d^f)N_{ij}^{Af} + \lambda_T N_{ij}^T \qquad (7)$$

$$\frac{dN_{ij}^{Bf}}{dt} = D\frac{\hat{N}_{ij}^{Bf} - 3N_{ij}^{Bf}}{\Delta^2} - (\lambda_B + \lambda_a + \lambda_d^f)N_{ij}^{Bf} + \lambda_A N_{ij}^{Af} \qquad (8)$$

$$\frac{dN_{ij}^{Ba}}{dt} = D\frac{\hat{N}_{ij}^{Ba} - 3N_{ij}^{Ba}}{\Delta^2} - (\lambda_B + \lambda_v + \lambda_d^a)N_{ij}^{Ba} + \lambda_a N_{ij}^{Bf} + \lambda_v N^{Bo} \qquad (9)$$

$$\frac{dN_{ij}^{Ca}}{dt} = D\frac{\hat{N}_{ij}^{Ca} - 3N_{ij}^{Ca}}{\Delta^2} - (\lambda_C + \lambda_v + \lambda_d^a)N_{ij}^{Ca} + \lambda_B(N_{ij}^{Bf} + N_{ij}^{Ba}) + \lambda_v N^{Co} \qquad (10)$$

The last is the group of the corners in the plane where two sides of the cell are next to the wall. The equations are given as:

$$\frac{dN_{ij}^T}{dt} = D\frac{\hat{N}_{ij}^T - 2N_{ij}^T}{\Delta^2} - \lambda_T N_{ij}^T + \frac{E}{\Delta} \qquad (11)$$

$$\frac{dN_{ij}^{Af}}{dt} = D\frac{\hat{N}_{ij}^{Af} - 2N_{ij}^{Af}}{\Delta^2} - (\lambda_A + 2\lambda_d^f)N_{ij}^{Af} + \lambda_T N_{ij}^T \qquad (12)$$

$$\frac{dN_{ij}^{Bf}}{dt} = D\frac{\hat{N}_{ij}^{Bf} - 2N_{ij}^{Bf}}{\Delta^2} - (\lambda_B + \lambda_a + 2\lambda_d^f)N_{ij}^{Bf} + \lambda_A N_{ij}^{Af} \qquad (13)$$

$$\frac{dN_{ij}^{Ba}}{dt} = D\frac{\hat{N}_{ij}^{Ba} - 2N_{ij}^{Ba}}{\Delta^2} - (\lambda_B + \lambda_v + 2\lambda_d^a)N_{ij}^{Ba} + \lambda_a N_{ij}^{Bf} + \lambda_v N^{Bo} \qquad (14)$$

$$\frac{dN_{ij}^{Ca}}{dt} = D\frac{\hat{N}_{ij}^{Ca} - 2N_{ij}^{Ca}}{\Delta^2} - (\lambda_C + \lambda_v + 2\lambda_d^a)N_{ij}^{Ca} + \lambda_B(N_{ij}^{Bf} + N_{ij}^{Ba}) + \lambda_v N^{Co} \qquad (15)$$

where superscripts T, Af, Bf, Ba and Ca represent ^{220}Rn, ^{216}Po, unattached atoms of ^{212}Pb, attached atoms of ^{212}Pb and ^{212}Bi, respectively. The notation $\hat{N}_{ij}$ expresses the sum of concentrations in the cells adjoining N_{ij} (for example $\hat{N}_{22} = N_{12} + N_{21} + N_{23} + N_{32}$). Also λ_T, λ_A, λ_B and λ_C are decay constants of thoron, ^{216}Po, ^{212}Pb and ^{212}Bi in s^{-1}, respectively. For λ with other subscripts, λ_a is an attachment rate in s^{-1}, λ_v is a ventilation rate in s^{-1} and λ_d is a deposition rate in s^{-1} in the cells next to the wall. The notation Δ is the length of one side of the cell in m, and E is the exhalation rate of thoron from the wall in atoms.m^{-2}.s^{-1}. Superscript 'o' represents outdoor concentration.

The inflow of thoron into the room from outdoor air is neglected because of its short life-time. For the calculation of ^{216}Po and unattached atoms of ^{212}Pb in Equations 7, 8, 12 and 13, ventilation can be also neglected because $\lambda_A \gg \lambda_v$ and $\lambda_a \gg \lambda_v$. In Equations 1 and 6, there is a term for the exhalation from the walls. From Equation 6 to Equation 11, only diffusion to the next cell is considered besides the decay term. From Equation 6 to Equation 15, there are terms for diffusion, decay, deposition and ventilation.

Now under steady state conditions, all differential equations are equal to zero. Then we obtain 5 × K^2 sets of the simultaneous equations. These equations could be solved by the method of LU factorisation directly.

PARAMETERS

Various parameters should be determined for calculating the indoor thoron concentration using the present method. The effective diffusion coefficient, D($m^2.s^{-1}$), is defined as a diffusion in the room macroscopically. Guo *et al*(3) reported that its value ranged from 0.001 to 0.007 $m^2.s^{-1}$ in various dwellings and in our model dwelling, which was selected as the subject of this calculation, its value was about 0.005 $m^2.s^{-1}$. They also obtained the thoron exhalation rate E (atoms.$m^{-2}.s^{-1}$) from the wall and its value was 180 atoms.$m^{-2}.s^{-1}$ in the model dwelling. Concerning λ_a and λ_v, lots of values have been reported in various environmental conditions by various researchers(8–10). We choose the typical indoor values as follows: $\lambda_a = 50\ h^{-1}$ and $\lambda_v = 0.5\ h^{-1}$. In Equations 7–10 and 12–15, λ_d is related to the whole deposition rate $\overline{\lambda_d}$ reported by many researchers(9–13). The whole deposition rate expresses the average deposition rate in the whole room. The relationship equation between λ_d in the cell and $\overline{\lambda_d}$ in the room is

$$\lambda_d = \overline{\lambda_d} \cdot \frac{K}{4} \tag{16}$$

The representative $\overline{\lambda_d}$ was chosen to be 0.2 h^{-1} for unattached atoms and 20 h^{-1} for attached atoms because of the difference between unattached and attached atoms in the deposition velocity to the wall. In this calculation $\overline{\lambda_d^a}$ is a hundred times larger than $\overline{\lambda_d^f}$.

By increasing the number of cells, the distribution of thoron and its progeny concentration is obtained in detail. On the other hand, to increase the number of cells is a burden to the computer calculating LU factorisation. Average indoor concentration was hardly affected by the number of cells, as a result of calculating when K = 5, 10, 20 and 30. Thus a size of model dwelling of 3 × 3 m^2 was divided into 10 × 10 cells.

In this model dwelling, thoron progeny concentration has been measured using a potential alpha energy moni-

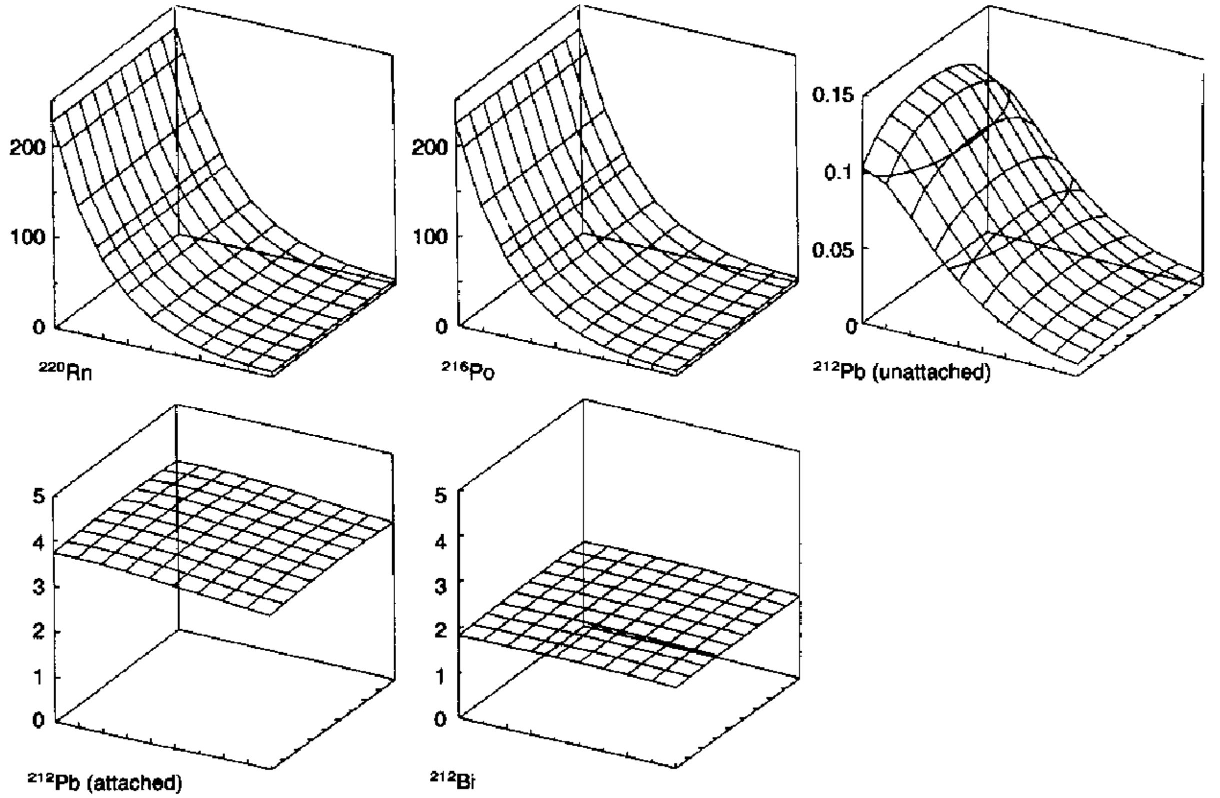

Figure 2. The general views of activity concentration distributions of thoron and its progeny at D = 0.005 $m^2.s^{-1}$ which is a normal value at any dwelling. The vertical axes show the activity concentrations given in units of Bq.m^{-3} in the room.

tor. The equilibrium equivalent thoron concentration was 1.1 Bq.m^{-3}(14).

RESULTS AND DISCUSSION

Figure 2 shows the distributions of thoron and its progeny concentration in dwellings calculated by the present method. Thoron emanates only from the whole left side wall and diffuses to the right. The effective diffusion coefficient, D = 0.005 m^2.s^{-1}, in Figure 2 is the normal value in the ordinary dwellings described above. Thoron concentration decreases exponentially with distance from the wall of thoron emanation. The activity of ^{216}Po is almost in radioactive equilibrium with thoron due to the very short half-life of ^{216}Po. Comparing the average concentration in the dwelling, the concentration of attached atoms of ^{212}Pb is about seventy times larger than that of unattached atoms of ^{212}Pb. Unattached atoms of ^{212}Pb can then be neglected when the concentration of ^{212}Pb in the dwelling is needed. Both the atoms of ^{212}Pb and ^{212}Bi, which are closely connected with human exposure, are the constant concentrations whether at the centre or the corner of the room. Therefore, it is easy to estimate the absorbed dose due to inhalation of thoron progeny which has uniform concentration in the room.

For a small D, for example D = 0.00005 m^2.s^{-1} that is ten times molecular diffusion coefficient of thoron in air, thoron and all thoron progeny have the distributions of concentration as shown in Figure 3. In such a case the dose estimation is complicated because it is not known what value of thoron progeny should be adopted. Fortunately, in human living spaces, there are few dwellings which have a small effective diffusion coefficient like a tightly closed room.

Figure 4 shows the relationship between equilibrium equivalent thoron concentration (EC$_T$) and the distance

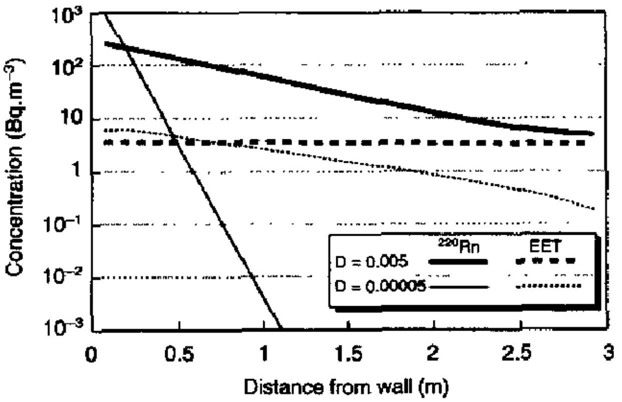

Figure 4. The dependence of equilibrium equivalent thoron concentration on the distance from the wall.

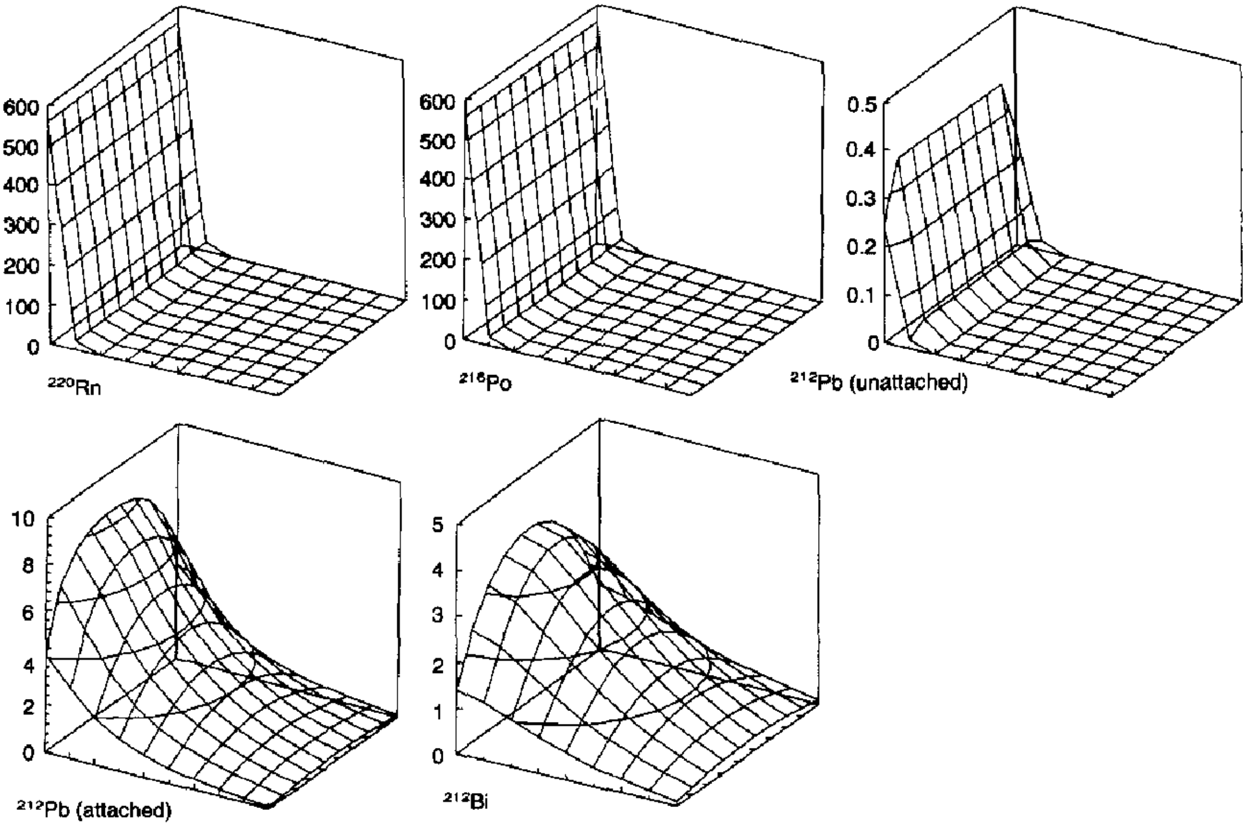

Figure 3. The general views of concentration distributions of thoron and its progeny at D = 0.00005 m^2.s^{-1} which is an extreme small value. The vertical axes show the activity concentrations given in units of Bq.m^{-3} in the room.

from the wall emanating thoron. Since thoron concentration never parallels EC_T, there is no sense in determining the equilibrium factor of thoron as discussed in radon exposure. Therefore, the dose estimation could not be carried out using the equilibrium factor of thoron and the absorbed dose should be calculated from the equilibrium equivalent thoron concentration after all. The equilibrium equivalent thoron concentration also has the distribution for small D. For small D, thoron is hard to diffuse, ^{216}Po and the unattached component of ^{212}Pb concentration near the wall is large, so the deposition to the wall before decay is increasing. In consequence, the equilibrium equivalent thoron concentration for small D is a little smaller than for the large D.

The effect of parameters of (a) effective diffusion coefficient, (b) deposition rate, (c) ventilation rate and (d) room size on the average concentrations of ^{212}Pb and ^{212}Bi were investigated in Figure 5. The average thoron progeny concentration is not affected by the effective diffusion coefficient (D) when $D \geq 0.001\ m^2.s^{-1}$ which is the normal value in the ordinary dwellings. Unless a particular situation is considered, the average concentration is constant and independent of D. The position of a window or the location of ventilation has no effect on the average concentration for the ordinary D, because the diffusion is sufficiently faster than the exchange between indoor and outdoor air. Since outdoor thoron progeny concentrations are nearly equal to zero, λ_d and λ_v are similar effects on the average concentration in Figures 5(b) and (c). As λ_d or λ_v increase, the ratio of ^{212}Bi and ^{212}Pb increases and the average concentrations are close to zero. The size of the room has no effect on the ^{222}Rn concentration, but it is expected that the room size does affect the thoron concentration because of the considerable difference in half-life. When the room size is bigger, the average thoron and its progeny concentration is smaller in Figure 5(d).

One-box, one-dimensional and two-dimensional models are compared in Table 1. The one-box model is what is called a 'mass balance model'. The equation for the one-box model is expressed by the mass balance in the whole room. The one-dimensional model is derived from the one-dimensional diffusion equation as a simple case of this two-dimensional model mentioned above. From Table 1, the average indoor thoron progeny concentrations are entirely in accord under the same conditions. The average could be estimated by the one-box model which was developed by Katase *et al*[(4)] and the quantity of the thoron emanation was incidental.

The actual dwelling has a three-dimensional structure, so it might be thought that a three-dimensional model is needed. However, the three-dimensional model is usually described by means of summing the distribution which also can be estimated by a two-dimensional model. Therefore this two-dimensional model is useful for the distribution of thoron and its progeny concentration when the effective diffusion coefficient is at an ordinary level. However, for unexpectedly small D there is no guarantee.

In the model dwelling, the equilibrium equivalent thoron concentration calculated by plausible parameters

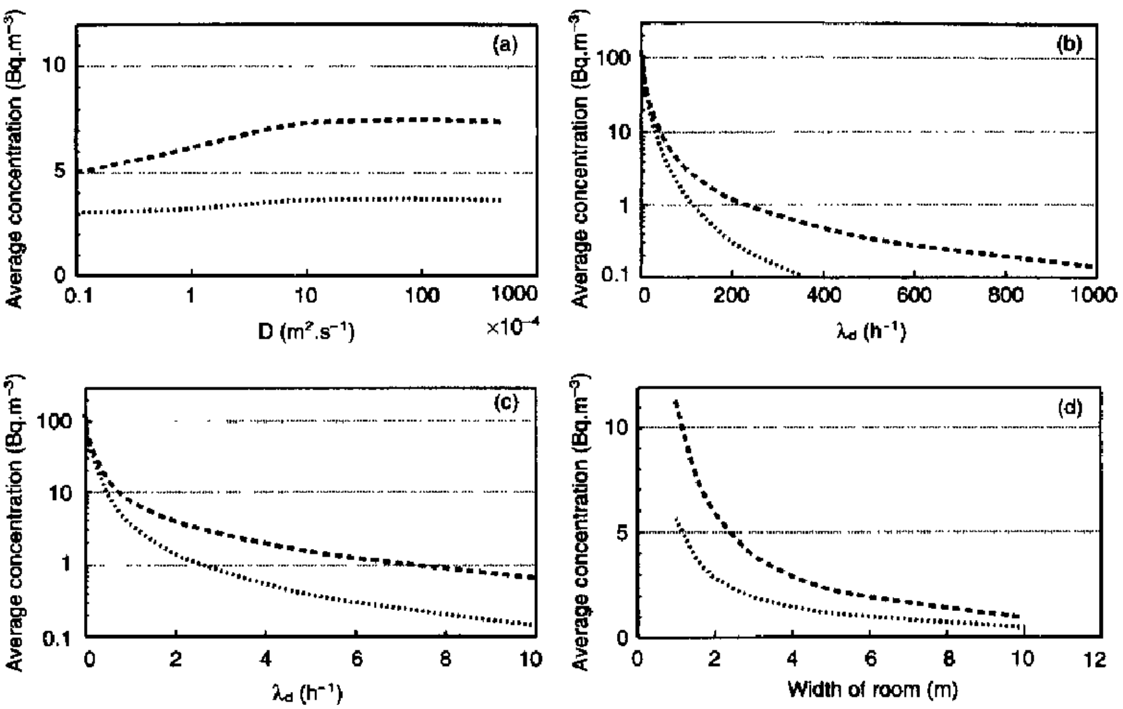

Figure 5. The effect of the parameters: (a) effective diffusion coefficient, (b) deposition rate, (c) ventilation rate and (d) room size. Heavy dashed line, ^{212}Pb; dotted line ^{212}Bi.

is compared with the experimental value. The calculated concentration was 3 Bq.m^{-3}. It is little higher than the experimental value of 1.1 Bq.m^{-3}. The difference is caused from the estimation of the ventilation rate or the deposition rate, because the room size, the exhalation rate of thoron and the effective diffusion coefficient are measured in the model dwelling. The ventilation rate tends to fluctuate more than the deposition rate. If the ventilation rate λ_v equals 2.0 h^{-1}, which is four times larger than the above calculation but the expected value, the calculated concentration becomes 1.1 Bq.m^{-3}. From the difference between calculated and experimental values, it is considered that λ_v may be underestimated.

CONCLUSION

The distributions and concentrations in a dwelling have been derived for thoron and its progeny using a two-dimensional steady state diffusion model. These results were quite different from those for radon because of the short half-life of thoron. From the exhalation rate of thoron and other parameters, indoor concentrations can be estimated for thoron and its progeny. Indoor thoron progeny concentrations which cause radiation dose to the public are constant in the room under ordinary environmental conditions. In human living spaces, one-box, one-dimension and two-dimension models give the same results about the average concentrations. Using a one-box model is enough to know the average concentration. For the small effective diffusion coefficient, indoor thoron and its progeny concentrations have a distribution in the room. In this case, various parameters are applied to affect the concentration. The calculated value is little higher than the experimental value. For agreement between calculation and experiment, is is necessary to investigate parameters in more detail. This method should be recognised as giving the general characteristics of indoor thoron and its progeny concentration distributions.

Table 1. A comparison of calculation models. The average indoor thoron progeny activity concentrations are given in units of Bq.m^{-3}.

	^{220}Rn	^{216}Po	^{212}Pb (unattached)	^{212}Pb (attached)	^{212}Bi
One-Box	66.5	66.4	0.057	3.73	1.88
One-dimensional	60.8	60.7	0.056	3.70	1.83
Two-dimensional	60.5	60.4	0.060	3.67	1.86

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